ABSTRACT

STOFOROS, GEORGE NIKOLAOS. Enhancement of continuous flow cooling of viscous foods using surface modified heat exchangers. (Under the direction of Dr. Josip Simunovic and Dr. K.P. Sandeep.)

Enhancement of continuous flow cooling of viscous products was examined numerically and experimentally, using computer simulation modeling and studies of surface modified heat exchangers, respectively.

Computer simulation studies were conducted using Multiphysics software system Comsol 5.2, in order to study cooling of applesauce and sweet potato puree. Simulation studies were used to compare the efficiency of cooling under two different flow regimes within the same tube in tube heat exchanger, with the food materials: i) flowing within the inner tube of the heat exchanger and ii) flowing within the annulus. The simulation results indicate that under an identical set of operating conditions, potential improvements in the cooling efficiency resulting from the movement of process material flow from the internal tube into the external annulus could be up to 14% and 23%, for applesauce and sweet potato puree, respectively.

Using the heat exchanger design selected based on results of the simulation studies, a preliminary study was conducted to examine the effects of surface treatment on cooling (97-57 °C) and thermal mixing via vibration of sweet potato puree, banana puree, and cheese sauce. Cooling performance and temperature uniformity were compared between two identical horizontal stainless steel tube in tube heat exchangers, one untreated and one with the food-contact surfaces chemically treated with a commercially available hydrophobic solution. A lower average product outlet temperatures ranging from 4 to 6 °C were observed for banana puree and cheese sauce with the treated heat exchanger, compared to the untreated case. However, for sweet potato puree cooling was slightly better (by 2-4 °C) using the untreated heat exchanger, compared to the treated case. Application of vibration at the resonance
frequency (20 Hz) of the mixing unit resulted in a more uniform cross sectional temperature distribution, which was observed for sweet potato puree and banana puree. However, application of vibration had no significant influence on cross sectional temperature distribution within cheese sauce.

In continuation of the previous studies, effects of different surface characteristics (wettability, roughness) on the adhesive behavior of sweet potato puree, banana puree, and cheese sauce, and the flow behavior of Carboxymethyl Cellulose (CMC) solution (conc. 1.5%) were examined for potential applications in cooling and mixing. Different engineered food-contact materials, including untreated and chemically treated stainless steel, polytetrafluoroethylene (PTFE), silicone sheets as well as treated and untreated knitted and woven polyester fabrics, were studied. Adhesive behavior of the tested foods and repellent behavior of the engineered surfaces were examined by comparing the residual product mass, during the standard methods of product depletion and the centrifuge adhesion tests. These tests were performed under two different temperature conditions; at ambient and at cooling processing temperature conditions. Results from the product depletion test showed that the hydrophobic surfaces with smooth characteristics (chemically treated stainless steel samples, PTFE, and treated knitted fabrics) retained the lowest amount of the foods, with the residual amounts of 35-45% at room temperature, and 45-65% at cooling conditions. In order to examine the cooling efficiency under cooling conditions, the temperature difference ($\Delta T$) between the initial and final highest product temperature was compared between the examined surfaces. The treated and untreated stainless steel, and PTFE surfaces presented the highest $\Delta T$ values, for all the foods tested. For the centrifugal adhesion test, the superhydrophobic knitted fabrics exhibited the best repellent behavior with all the foods tested, reducing the residual
product to 0-10% and 20-40% levels, under room and cooling temperature conditions, respectively.

Pressure drop and velocity profile of 1.5% CMC solution, flowing within the annulus of a tubular heat exchanger, were used to compare the effects of the examined surfaces on adhesive and flow behavior, respectively. Highly rough, hydrophobic surfaces (silicone sheets and untreated woven textiles), exhibited the highest pressure drop across the system with a value of 75842.4 Pa. While, the lowest pressure drop, at 48263.3-55158.1 Pa, was observed with the smooth surfaces (chemically treated stainless steel) and with the superhydrophobic samples (treated knitted and woven textile samples). The velocity profile of 1.5% CMC was also examined, using particle tracking velocimetry (PTV) as the testing method. The PTV results for the velocity profile of 1.5% CMC, showed a parabolic velocity profile for all the examined cases, with the fastest flowing particle located close to the center of the annulus flow.

The final study investigated the effects of two different food-contact materials; untreated stainless steel and engineered food-grade PTFE textile on cooling of sweet potato puree, banana puree, and cheese sauce. The PTFE textile was designed based on the surface characteristics observed in the previous studies. Using PTFE for its stability, the engineered surface was fabricated with hydrophobic properties, smooth characteristics, and knitted structure. Cooling experiments for both examined surfaces were conducted under steady state, identical operating conditions, with tested foods flowing within the annulus of a tubular heat exchanger. The outside surface of the inner tube of the heat exchanger was used as the tested food-contact surface, by the application of the knitted PTFE textile. Finally, results for all the tested foods showed a total improvement between 15-25% of cooling efficiency when using the inner tube of the heat exchanger covered with the PTFE textile.
Enhancement of Continuous Flow Cooling of Viscous Foods using Surface Modified Heat Exchangers

by
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DEDICATION

To my family.
BIOGRAPHY

George Stoforos was born on February 27, 1986, in Sacramento, USA, and he grew up in Lamia, Greece. He graduated from the School of Chemical Engineering of the National Technical University of Athens (NTUA) Greece, in November of 2008 (a five-year program of study). His Diploma thesis was in the kinetic studies of the effect of high hydrostatic pressure and temperature on pectinmethylesterase and polyphenoloxidase of strawberry juice under the supervision of Dr. Taoukis. From February of 2009 till January of 2010 he served in the Greek Army, a compulsory military duty for Greek citizens. Since then and until spring of 2012, he worked as a coach for a swimming team at his hometown, Lamia, (an activity/hobby aroused from his career as an active swimmer, for more than 15 years). In August 2014, he got his Master of Science degree in Food Science, from the Department of Food Science at North Carolina State University (NCSU) under the supervision of Dr. B.E. Farkas and Dr. J. Simunovic. His MSc thesis was on the acoustic enhancement of continuous flow cooling of multiphase food products. For his MSc research work, he was the recipient of 1st Place award in the Food Engineering Poster Competition, at IFT 15 (July 11-14, 2015, Chicago, IL, USA). In August 2014, he started his Ph.D. degree in Food Science, from the Department of Food Science at NCSU, under the supervision of Dr. J. Simunovic. His Ph.D. research topic has been focused on the enhancement of continuous flow cooling of viscous foods using surface modified heat exchangers. For his Ph.D. research work, he was the recipient of 2nd Place award at the Institute For Thermal Processing Specialists (IFTPS) Charles Stumbo Student Paper Competition of 2017. He has published one paper in the Food and Bioproducts Processing journal and has made five presentations at national and international meetings.
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CHAPTER 1:

Introduction

Advanced thermal technologies such as continuous flow microwave systems, ohmic and radio frequency heating have enabled food processors to provide uniform rapid volumetric heating resulting in reduction of come up time, reduction of fouling deposition, and minimization of product quality losses, during continuous thermal processing of viscous and multiphase food products. These include typical dairy products, fruit and vegetable purees and soups (Coronel et al., 2005; Steed et al., 2008; Cullen et al., 2012). However, due to the lack of advanced technologies, cooling of viscous products is still implemented using inefficient conventional cooling methods. During conventional cooling of viscous food products, laminar flow and low thermal conductivity, characteristic for these materials, lead to a non-uniform, slow cooling process, an increase of operation cost and degradation of final food quality (Stoforos et al., 2016).

Unfortunately, often predicted and anticipated developments of volumetric cooling technologies, such as magnetic field cooling (Sarlah et al., 2006; Kawanami et al., 2011) have been slow and expensive in their progress to large-scale commercial industrial applications. Additionally, food processors, in an effort to enhance the cooling rate of continuous flow thermal processing, utilize coolant mediums (like ethylene glycol based coolants) at a very low-temperature range, but without any significant improvement in cooling of viscous foods. Using this cooling approach, an additional problem has appeared, ethylene glycol and cold/chilled water piping can sweat or be covered with ice, resulting in dripping water, with a negative impact on sanitation conditions of the food production plant (Moerman, 2016). Moreover, thermal mixing, temperature equalization within the product, has been proposed as
an efficient method to enhance heat transfer and the overall continuous flow cooling process of highly viscous food products (Metcalfe and Lester, 2009; Stoforos et al., 2016). However, the efficiency of thermal mixing and cooling processing of viscous food products, such as fruit and vegetable purees, was affected by the formation and build-up of fouling product layers on the surfaces of processing equipment (heat exchangers, mixing units) (Stoforos et al., 2016).

During continuous flow thermal processing of foods and biomaterials, deposition and accumulation of unwanted food materials, such as proteins, minerals, carbohydrates and fats, in the form of fouling, on surfaces of processing equipment, has been a major challenge for food and dairy industries (Swartzel, 1983; Sandu and Singh, 1991; Balasubramanian and Puri, 2009; Kananeh et al., 2010; Mérian and Goddard, 2012; Murugesan and Balasubramanian, 2013; Barish and Goddard, 2013). Formation of fouling layers on processing equipment reduces the operating efficiency by increasing the resistance to heat transfer and thermal mixing and by creating fluctuation of pressure drop across the heat exchanger (Swartzel, 1983; Balasubramanian and Puri, 2009; Awad, 2011). Moreover, fouling increases the likelihood of biofilm formation, resulting in additional operating expenses, corresponding to frequent food processing plant shutdowns for cleaning and extensive use of chemical detergents and sanitizers (Sandu and Singh, 1991; Kananeh et al., 2010; Mérian and Goddard, 2012; Barish and Goddard, 2014).

Controlling product deposition and fouling formation on the processing equipment surface depends on several different parameters such as operating conditions (temperature, and flow rate), food properties (viscosity and food composition) and surface properties (material, surface wettability and topography) (Santos et al., 2004; Awad, 2011; Mérian and Goddard, 2012). Food composition and concentration of food components such as proteins, minerals,
lipids, and carbohydrates, and total solids concentration, are critical factors regarding fouling of foods to heating surfaces (heat exchangers, spray dryers, etc…) (Changani et al., 1997; Bansal and Chen, 2006; Wallhäußer et al., 2012). Fouling and deposition of unwanted material can occur in many different types, based on different physical, and chemical processes involved. Fouling in food processing is formed from the precipitation of partially soluble salts (scaling, Type-B milk fouling), from the physicochemical reactions of proteins (Type-A milk fouling) and carbohydrates, precipitation of suspended particles (colloids), crystallization and solidification of fat components (during freezing and cooling), biofouling and corrosion reactions (Bansal and Chen, 2006; Wallhäußer et al., 2012; Goode et al., 2013; O’callaghan and Hogan, 2013).

Moreover, this problem mainly has been observed during conventional thermal processing (heating, cooling, and thermal mixing) of highly viscous and multiphase food products, such as typical dairy products, fruit and vegetable purees and soups (Stoforos et al., 2016). Unfortunately, the adhesive behavior of highly viscous foods is more prone to occur on stainless steel, the most commonly used material for food processing equipment, such as heat exchangers and mixing units (Goode et al., 2013; Barish and Goddard, 2013; Stoforos et al., 2016). Stainless steel is an inert material, relatively resistant to corrosion, stable with temperature, and highly thermally conductive, ideal for food-contact surface material for thermal processing equipment units (Goode et al., 2013; Barish and Goddard, 2013). However, stainless steel has surface characteristics, such as partial wetting properties and high surface energy, that are prone to fouling and adhesive behavior of viscous food materials (Goode et al., 2013; Barish and Goddard, 2013; Stoforos et al., 2016).
Recently, several studies have examined surface modification techniques, to adjust stainless steel properties (wettability, roughness, and chemistry) and reduce or completely prevent fouling or biofouling of the food and dairy processing equipment (Beuf et al., 2003; Santos et al., 2004; Rosmaninho et al., 2007; Premathilaka et al., 2007; Balasubramanian and Puri, 2009; Kananeh et al., 2010; Mérian and Goddard, 2012; Murugesan and Balasubramanian, 2013; Barish and Goddard, 2013). Surface modification studies have been focusing on mimicking the properties of natural anti-fouling and self-cleaning materials, such as lotus leaf (*Nelumbo nucifera*). The superhydrophobic behavior, in combination with the roughness of the nanostructure of the lotus leaf, is associated with unique self-cleaning and water-repelling properties, known as the lotus effect (Latthe et al., 2014). Novel surface modification techniques, such as polymer and nanoparticle coatings (silica or fluorocarbon compositions), physicochemical surface modification, plasma-enhanced chemical vapor deposition, electro-chemical and ion implantations have enabled researchers to successfully lower the free surface energy of the food contact surfaces by altering wettability (hydrophobic and hydrophilic) and roughness characteristics. Another method to fabricate an antifouling surface is by modifying the chemistry of the surface, for example by using a hydrophilic (poly(ethylene glycol)) coating to stop hydrophobic interactions between whey protein and stainless steel (Premathilaka et al., 2006; Mérian and Goddard, 2012).

Antifouling materials, and surface modification techniques, with low surface free energy and smooth surface characteristics, have successfully been used in food research, improving the operation and cleaning efficiency and reducing the bacterial adhesion (Beuf et al., 2003; Santos et al., 2004; Premathilaka et al., 2006; Rosmaninho et al., 2007; Balasubramanian and Puri, 2009; Kananeh et al., 2010; Mérian and Goddard, 2012; Barish and
Goddard, 2013; Goddard et al., 2013). However, antifouling surface modification techniques have not extensively been used for commercial applications in food processing. The cost of fabrication and in some cases, incompatibility with food grade materials, in addition to the reported short-term stability of antifouling properties of modified surfaces still need to be improved to enable potential applications in advanced thermal processing of food products (Barish and Goddard, 2013).

The main objective of this research was to improve continuous flow cooling processes of viscous and poorly conductive food products, minimizing the cooling-related quality losses of the final products, and potentially improving the sanitation conditions, by using surface modified heat exchangers. It was anticipated that by determining the best flow configuration design, use of surface modified heat exchangers, with the desired surface properties based on the examined foods, would reduce product accumulation and fouling formation on processing unit surfaces. Subsequently, it was expected that the reduction of fouling formation and build-up on the surfaces of heat exchangers would enhance cooling and cleaning processes of viscous foods while maintaining good sanitation conditions in the food processing plants.
CHAPTER 2:

A Review of Adhesive Behavior and Fouling of Viscous Foods during Thermal Processing

(Review of Literature)

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ABSTRACT

This paper presents a review of fouling mechanisms during food processing, additionally with the challenges and opportunities associated with the new antifouling methods. Fouling formation on the surface of processing equipment, during thermal processing of foods, is a major problem in the industry, reducing the processing efficiency and increasing the operating costs. During thermal processing of foods, fouling formation depends on physical and chemical interactions between the flowing food and the contact surface. Fouling process can be influenced by operational conditions (fluid velocity and temperature; surface temperature), food composition (proteins, minerals, fats, carbohydrates) and surface characteristics (surface material, wettability, roughness). Food components such as proteins, minerals, fats, and carbohydrates, are critical factors regarding the fouling type formation on the food-contact surface. Fouling in food processing is formed by the precipitation of minerals (Type-B milk fouling), from the physicochemical reactions of proteins (Type-A milk fouling) and carbohydrates, precipitation of suspended particles (colloids), crystallization and solidification of fat components (during freezing and cooling), biofouling and corrosion reactions. Novel antifouling surface modification methods, such as polymer and nanoparticle coatings (silica or fluorocarbon compositions), physicochemical surface modification, plasma-enhanced chemical vapor deposition, electro-chemical and ion implantations have been applied on the stainless steel surface of processing equipment to adjust the surface characteristics and produce antifouling and self-cleaning materials. Antifouling materials, with characteristic low surface free energy and smooth surface, have successfully been used in food and dairy research, improving the operation and cleaning efficiency and reducing the bacterial adhesion.

Key words: Review of fouling process, effects of fouling, antifouling methods
2.1. Introduction

Advanced heating technologies, such as ohmic heating, continuous flow microwave, and radio frequency thermal treatment systems, have enabled food processors to maximize product quality during continuous thermal processing of viscous and multiphase food products, like typical dairy products, fruit and vegetable purees, and soups (Coronel et al., 2005; Steed et al., 2008; Cullen et al., 2012). However, cooling processing of viscous and multiphase foods is still implemented using inefficient conventional cooling methods. During conventional cooling of viscous food products, laminar flow and low thermal conductivity, characteristic for these materials lead to a wide temperature distribution within the product, resulting in a non-uniform, very slow cooling process and reduction of final food quality (Stoforos et al., 2016). Temperature equilibration within the product, thermal mixing, has been examined as an efficient method to enhance heat transfer and the overall continuous flow cooling process of highly viscous products (Metcalfe and Lester, 2009; Stoforos et al., 2016). However, the efficiency of thermal mixing and cooling of viscous foods was affected by the formation and accumulation of fouling product deposition on the surface of processing equipment (heat exchangers, mixing units) (Stoforos et al., 2016).

Fouling and accumulation of unwanted material, such as proteins, starches, and minerals, on the surface of processing equipment, mainly observed during conventional thermal processing of viscous products, is a major problem in the food industry, reducing the processing efficiency and increasing the operating costs. The formation and build-up of fouling layers on food processing equipment reduces the efficiency of heat transfer (Fig. 2.1), thermal mixing and increases the pressure drop across the processing unit, resulting in degradation of final food product quality and increasing the overall processing and cleaning operation time.
and cost (Balasubramanian and Puri, 2008; Balasubramanian and Puri, 2009; Barish and Goddard, 2013; Barish and Goddard, 2014; Stoforos et al., 2016). Moreover, accumulation of fouling layers on food processing surface is prone to form biofilms, resulting in extensive use of chemicals and sanitizers and frequent food processing plant shutdowns for sanitation and cleaning (Sandu and Singh, 1991; Kananeh et al., 2010; Mérian and Goddard, 2012; Barish and Goddard, 2014).

This study aims to provide a better understanding of the mechanism of fouling and product adhesion during food processing and address the recent developments in antifouling materials. Furthermore, this paper reviews the challenges and opportunities of the new antifouling developments, for potential applications in advanced thermal processing.

Figure 2.1. Fouling formation on the food-contact surface of stainless steel plate heat exchanger; the thick fouling layer (protein, carbohydrate, lipid) increases the resistance to heat transfer (ΔH) between the product and hot water and reduces the overall operational efficiency (Mérian and Goddard, 2012).
2.2. Fouling

Fouling is defined as the deposition and accumulation of unwanted food material on the surface of processing equipment having a negative impact on processing efficiency and food quality and safety (Balasubramanian and Puri, 2008; Balasubramanian and Puri, 2009; Fryer and Asteriadou, 2009; Awad, 2011; Barish and Goddard, 2013; Barish and Goddard, 2014). Fouling and adhesion of food materials is a complex phenomenon that can occur in any liquid-solid interface. Fouling is a time-dependent process and involves simultaneously the steps of deposition and removal of deposit material on the processing equipment surface (Awad, 2011; Kazi, 2012; Hou et al., 2017). The overall fouling process can be described in the following stages (Awad, 2011; Hou et al., 2017): 1) initiation of fouling formation in the bulk of the carrier; 2) transport of the formed fouling material to the fluid-solid interface; 3) attachment and formation of fouling layer to the surface; 4) removal (spalling or sloughing of the deposited layer) of the fouling material from the surface; 5) ageing at the surface, starts with commencement of fouling deposition (Fig. 2.2).
2.3. Fouling process

Fouling process begins with the formation of fouling material in the bulk of the fluid, with the first phase being a delay or induction period. During induction period, the basic mechanism of fouling process is heterogeneous nucleation close to the contact-surface. The process of nucleation and fouling formation is affected by temperature, velocity, and composition of the fluid, and the characteristics and condition of the contact-surface (Awad, 2011; Mérian and Goddard, 2012).

Following the initial phase of fouling formation, product fouling deposition and accumulation on the processing unit’s surface is associated with a two-step mechanism, that
is, transport, and attachment of the fouling material (Awad, 2011; Mérian and Goddard, 2012). The transport and attachment steps of fouling material from the bulk of the fluid to the surface involves one of the following mechanisms (Melo et al., 1988; Awad, 2011):

i. **Diffusion mechanism** involves the mass transfer of fouling materials from the flowing fluid to the heat transfer surface due to the concentration difference between the bulk of the fluid and the fluid streams close to the surface.

ii. **Electrophoresis mechanism** involves the action of electric forces between electrically charged fouling particles and surface. The charged particles will move toward or away from the charged surface depending on the polarity of the surface and the particles. Fouling deposition through the electrophoresis mechanism depends on the pH, temperature, velocity and electrical conductivity of the fluid. Deposition of the charged particles increases with an increased temperature and velocity of the fluid, and a decreased electrical conductivity. London–van der Waals and electric double layer interaction forces are usually the surface properties, responsible for electrophoretic effects.

iii. **Thermophoresis** is the transport mechanism of fouling product due to the temperature gradient in the fluid bulk and the processing surface. In particular, a high-temperature gradient close to a high-temperature contact surface will prevent the product from depositing, while, for the same absolute value of temperature gradient a cold contact-surface will promote product adhesion and deposition. The thermophoresis mechanism applies more for gases than for liquids.

iv. **Diffusiophoresis mechanism** involves condensation of gaseous streams onto the contact surface.
v. Sedimentation mechanism involves the deposition of large particulates such as colloidal particles, rust particles, clay, and dust on the surface due to the action of gravity. Sedimentation transport mechanism is important for large particles and low fluid velocities.

vi. Inertial impaction is a mechanism for large particles with sufficient inertia, deposited on the contact surface being unable to follow fluid flow.

vii. Turbulent down-sweeps mechanism appears in turbulent flow, with the fluid being transported toward the surface by turbulent down-sweeps acting as suction areas.

In the transport stage, there is mass transfer of additional fouling material to the surface, which facilitates the attachment stage of fouling process. During the attachment stage, fouling material adheres to the surface through different interactions such as electrostatic, hydrophobic, and surface nucleation (Awad, 2011; Mérian and Goddard, 2012). Attachment and adhesion of fouling material on the contact-surface depend on the surface properties (topography/roughness, chemical nature) (Awad, 2011; Mérian and Goddard, 2012). The surface properties, such as surface free energy, wettability (contact angle, spreadability), and heat of immersion are critical factors for product attachment and accumulation at the contact-surface of processing equipment (Awad, 2011). Surface wettability and heat of immersion increase as the difference between the surface free energy of the contact surface and the surface tension of the flowing fluid close to the surface increases.

Finally, fouling process includes also the removal and aging stages. Removal involves the detachment of the fouling material from the contact-surface that can be a result of auto-initiated detachment or through the action of external forces such as scouring, sloughing, or agitation (Awad, 2011; Mérian and Goddard, 2012). In later stages of fouling process, aging stage may occur, with the fouled layer to age, particularly in a heated surface such as a heat
exchanger. During the aging stage changes of the nature of the fouling material may occur over time, influencing adhesion and cleanability characteristics (Awad, 2011; Mérian and Goddard, 2012).

2.4. Fouling types

Product deposition, accumulation and fouling formation depends on several different parameters, such as operating conditions (bulk temperature of the fluid, surface temperature, flow rate), food properties (pH, viscosity, food composition) and surface properties (material, surface wettability, topography) (Bott, 1995; Santos et al., 2004; Awad, 2011; Mérian and Goddard, 2012). In the food industry, different types of fouling can occur based on the different physical and chemical processes involved, such as precipitation or crystallization fouling, particulate fouling (colloidal particles) or sedimentation (larger particles), chemical reaction fouling, solidification fouling, corrosion fouling, and biological fouling (Bott, 1995; Fryer and Asteriadou, 2009; Simões et al., 2010; Awad, 2011; Wallhäußer et al., 2012; Kazi, 2012; Goode et al., 2013).

2.4.1. Precipitation or crystallization fouling

Precipitation or crystallization fouling involves the crystallization of dissolved salts, such as calcium, on the surface of the processing surface. Crystallization fouling occurs from the dissolved salts from saturated solutions, at higher temperatures. This type of fouling occurs on the surface of heat exchangers during dairy processing and in water treatment during desalination process (Bott, 1995; Fryer and Asteriadou, 2009; Awad, 2011; Wallhäußer et al., 2012; Kazi, 2012; Goode et al., 2013).
2.4.2. Particulate or sedimentation fouling

Particulate or sedimentation fouling involves the deposition and accumulation of suspended particles on the heat transfer surface (Awad, 2011; Wallhäußer et al., 2012; Kazi, 2012). It is the deposition of suspended particles in the process streams onto the heat transfer surfaces. Sedimentation fouling is referring to large particles settling on the processing surface due to gravity.

Particulate fouling occurs from suspended solids in cooling water, soot particles of incomplete combustion, magnetic particles in economizers, deposition of salts in desalination systems, deposition of dust particles in air coolers (Bott, 1995; Awad, 2011; Kazi, 2012). An example of this type of fouling in food processing is during spray dry processing, where milk powder adheres to the surface of the drying chamber, usually near exit points (Ozmen and Langrish 2003; O’callaghan and Hogan, 2013; Sadeghinezhad et al., 2015). The stickiness of milk powder on the heat transfer surface is generally considered to result from the formation of liquid bridges from amorphous sugar or other dissolved material on the surface of the processing unit (Ozmen and Langrish 2003; O’callaghan and Hogan, 2013; Sadeghinezhad et al., 2015). Furthermore, particulate fouling is a common problem during membrane filtration of fermented beverages, such as beer (Blanpain and Lalande, 1997; Czekaj et al., 2000).

2.4.3. Chemical reaction fouling

Fouling formation for this type is the result of unwanted chemical reactions that take place during the heat transfer process between the components of the flowing fluid. Heat transfer surface is not a reactant during this type of fouling but can act as a catalyst for chemical reactions, such as cracking, cooking, polymerization, and autoxidation (Bott, 1995; Awad,
2011; Kazi, 2012). Decomposition and polymerization of proteins and hydrocarbons and crude oil fouling on heat transfer surfaces are characteristic examples in food and dairy processing for this type of fouling (Wallhäußer et al., 2012).

2.4.4. Solidification fouling

Solidification fouling involves deposition of fluid components on the cooled processing surface. During solidification or freezing, fluid components with a high melting point freeze form a solid fouling deposit layer onto a subcooled surface, that acts as a resistance to heat transfer between the coolant and the liquid (Fig. 2.3) (Sharma et al., 1982; Bott, 1995; Awad, 2011; Wallhäußer et al., 2012; Kazi, 2012; Goode et al., 2013). Subcooled processing surfaces are subject to solidification fouling from normally soluble salts, fats, and waxes (Sharma et al., 1982; Bott, 1995; Fryer and Asteriadou, 2009; Awad, 2011; Wallhäußer et al., 2012; Kazi, 2012; Goode et al., 2013). Solidification fouling occurs at low temperatures, usually in the range of ambient temperature or lower. Operation factors such as flow rate of the flowing fluid, temperature and crystallization conditions, surface conditions, and concentration of the fluid components are factors affecting solidifications fouling (Kazi, 2012).
Due to the low temperature of the contact surface ($T_s$) and the components of the fluid, a solid layer from the fluid components of thickness ($x_s$) and temperature ($T_f$), higher from $T_s$ is formed, acting as a resistance to heat transfer between the coolant and the liquid (Bott, 1995).

### 2.4.5. Corrosion fouling

Corrosion fouling is the result of chemical or electrochemical reactions between the processing surface and the flowing fluid. Corrosion may cause fouling in two ways; first by the accumulation of corrosion fouling material onto the surface of corrosion site, providing resistance to heat transfer. Second, corrosion particulate fouling products may be transported and be deposited as particulate fouling in another site of the processing surface system (Bott, 1995; Awad, 2011). Corrosion fouling in a metal surface during water treatment is a characteristic example of this fouling type (Bott, 1995; Fryer and Asteriadou, 2009; Awad, 2011; Wallhäußer et al., 2012; Kazi, 2012).
2.4.6. Biological fouling

Biological growth (biofouling) is the attachment and growth of biofilms; organic films consisting of microorganisms and their products (Bott, 1995; Simões et al., 2010; Fryer and Asteriadou, 2009; Awad, 2011; Wallhäußer et al., 2012; Kazi, 2012) (Fig. 2.4). Biological fouling occurs in two subtypes of fouling, microbial and macrobial. Microbial fouling is the most important type of biological fouling in food processing and involves the deposition of microorganisms such as fungi, yeasts, bacteria, and molds. Macrobial fouling refers to the accumulation of macroorganisms that can be found in seawater or estuarine cooling water, such as clams, barnacles, mussels, and vegetation (Awad, 2011).

Figure 2.4. Steps of biofilm formation process (Breyers and Ratner, 2004).
2.4.7. Milk fouling

Fouling in dairy processing is one of the most studied and affected sectors. Moreover, milk fouling on heat transfer surfaces (usually in a plate heat exchanger) is classified into two main categories, based on the processing temperature, Type-A, and Type-B (Fig. 2.5). Type-A involves the protein fouling (chemical fouling type) and takes place within the temperature range of 75 °C to 110 °C. During Type-A fouling, protein (β-Lactoglobulin) denaturation forms a white, soft, and spongy-like fouling film (milk film) on the heat transfer surface, with a composition of 50%-70% proteins, 30%-40% minerals, and 4%-8% fat. During Type-B fouling, accumulation of minerals (calcium phosphate, calcium citrate) forms a dense gray granular layer (milk stone) with a composition of 70%-80% minerals (mainly calcium phosphate), 15%-20% proteins, and 4%-8% fat (Bansal and Chen, 2006; Wallhäußer et al., 2012; Sadeghinezhad et al., 2015). Both milk fouling categories may occur simultaneously during Ultra high-temperature (UHT) (~138 °C) processing. However, mainly Type-A fouling may occur during High-Temperature Short-Time (HTST) (72–100 °C) processing, with possible localized Type-B fouling (for heat exchanger surface above 110 °C) (Bansal and Chen, 2006; Wallhäußer et al., 2012; Sadeghinezhad et al., 2015).
Figure 2.5. Image of milk fouling (Type-A and Type-B), on the surface of stainless steel plate heat exchanger (Barish and Goddard, 2013).

2.5. Factors affecting fouling

The fouling formation is an unsteady process and depends on physical and chemical interactions between the flowing liquid and the contact-surface. For that reason, fouling process can be influenced by fluid properties (velocity and temperature) and composition (protein, mineral etc...), surface characteristics (surface material, wettability, roughness) and
conditions (temperature) and the operational conditions (Bott, 1995; Awad, 2011; Mérian and Goddard, 2012).

2.5.1. Fluid velocity

Fluid flow velocity is one of the most important parameters on fouling formation, affecting both deposition and removal of fouling material. The hydrodynamic effects of high-velocity fluid flow, such as the eddies and shear stress at the surface, can increase the removal rate of the fouling material from the contact-surface. On the other hand, fouling components of the flowing fluid will deposit in low-velocity regions, characteristics of the laminar flow regime (Bott, 1995; Kukulka and Devgun, 2007, Awad, 2011).

2.5.2. Food Components

Food composition and concentration of food components such as proteins, minerals, fats, and carbohydrates, and total solids concentration, is a critical factor regarding fouling of foods to the heating surface (heat exchangers, spray dryers, etc…) (Changani et al., 1997; Bansal and Chen, 2006; Wallhäußer et al., 2012). Adhesion behavior of food components involves changes in the thermodynamic states which are different in nature for sugars, proteins, and fats, resulting in different types of fouling (Bansal and Chen, 2006; Wallhäußer et al., 2012; Goode et al., 2013; O’callaghan and Hogan, 2013). Fouling in food processing is formed from the precipitation of partially soluble salts (scaling), from the physicochemical reactions of proteins and carbohydrates, precipitation of suspended particles (colloids), crystallization and solidification (freezing and cooling), biofouling and corrosion reactions (Bansal and Chen, 2006; Wallhäußer et al., 2012; Goode et al., 2013; O’callaghan and Hogan, 2013).
Proteins

During thermal processing denaturation and aggregation can cause fouling on the processing surface. Fouling formation from proteins is due to the transport and attachment of protein aggregates on a surface. Protein and more specific whey protein, mainly β-lactoglobulin and secondly α-lactalbumin, are the major components of Type-A fouling.

Lipids

Lipid crystallization results in solidification fouling on the cold food-contact surface. The formation and accumulation of food-contact surface deposits can be a major problem in pipes distributing food fats, reducing processing efficiency (heat transfer, pressure drop, cleaning process) (Bansal et al., 2008; Huang et al., 2010). Solidification fouling formation is caused by higher melting point components crystallizing from the flowing solution to form a viscous gel, which hardens over time (Bansal et al., 2008; Huang et al., 2010). Food multi-component mixture fats, such as waxy crude oils, form crystals when their temperature falls below the cloud point; the temperature at which the oil begins to cloud resulting from crystallization during cooling (Huang et al., 2010). Temperature difference between bulk fluid and the cold surface is a critical factor for solidification fouling. Reducing the temperature difference between the flowing product and the food-contact surface can reduce the fouling formation (Bidmus and Mehrotra, 2004; Huang et al., 2010).

Carbohydrates

Carbohydrates are the food component responsible for fouling in evaporator systems, spray dryers, and filtration systems (O’callaghan and Hogan, 2013; Challa et al., 2015). Fouling characteristics of carbohydrate mixtures (starch, corn syrup solids and glucose) happen in the glass transition state of carbohydrates (O’callaghan and Hogan, 2013; Challa et al.,
As a result of that, an increase of the starch content will increase fouling, which depends on amylose and amylopectin content, and glass transition temperature (Challa et al., 2015).

Minerals

Dissolved soluble mineral salts in the flowing fluid, such as calcium carbonate, calcium phosphate, and calcium citrate, contribute to the formation of scales developed from the accumulation of precipitation of mineral crystal salts on the contact surface (Awad, 2011). In particular, milk fouling Type-B, or mineral fouling, is formed from the accumulation of minerals such as calcium phosphate and calcium citrate, on the surface of heat exchangers (Bansal and Chen, 2006; Wallhäußer et al., 2012; Sadeghinezhad et al., 2015).

2.5.3. pH of food

The effect of pH on fouling is not clear (Bansal and Chen, 2006). In general, a reduction in pH decreases the heat stability of proteins, while results in an increase of ionic minerals concentration, such as calcium (Foster et al., 1989; de Jong, 1997; Bansal and Chen, 2006). In milk fouling, a decrease in milk pH has a great impact on fouling, increasing the fouling build up due to the additional deposition of caseins (de Jong, 1997; Riverol and Napolitano, 2005; Bansal and Chen, 2006).

2.5.4. Food temperature

Food temperature is a critical factor in product adhesion and fouling process. It is worth mentioning that both the absolute (food) temperature and the temperature difference (between food and contact surface) are important for fouling. Increasing the food temperature results in higher fouling (Bansal and Chen, 2006; Kukulka and Devgun, 2007). In milk fouling, the
temperature of the milk is the most important control parameter, resulting in a different type of fouling, Type-A or Type-B, based on the particular product temperature (Bansal and Chen, 2006; Wallhäußer et al., 2012; Sadeghinezhad et al., 2015). Preheating of milk reduces the milk fouling on the heated surface. Preheating of dairy products results in denaturation and aggregation of proteins prior to UHT and HTST, reducing the fouling deposition (Foster et al., 1989; Bansal and Chen, 2006).

2.5.5. Surface temperature

Surface temperature is a critical parameter of the fouling process. In general, fouling rate increases as the temperature rises. At higher temperature, the rate of fouling chemical reactions, such as corrosion, crystal formation, and polymerization, increases (Awad, 2011). At higher temperatures, some of the anti-fouling surface characteristics, such as non-wetting behavior, tends to decrease. Lower surface temperature produces slower fouling buildup and usually deposits that are easily removable (Awad, 2011). However, low surface temperatures are prone to crystallization and solidification fouling. To overcome these problems, there should be an optimum surface temperature to be used for each situation. Furthermore, biofouling strongly depends on surface temperature. To reduce biofouling formation, low surface temperatures that will reduce the growth rate of the biological organisms, or high temperatures that will kill the microorganism cells are preferable (Mukherjee, 1996).

2.5.6. Surface material

Surface material selection is important, especially for corrosion fouling. Stainless steel (series 300) is the most commonly used material for food processing equipment. Stainless steel
is an inert material, relatively resistant to corrosion, stable with temperature, and highly thermally conductive, ideal for food-contact surface material for thermal processing equipment units (Goode et al., 2013; Barish and Goddard, 2013). However, stainless steel has surface characteristics, such as partial wetting properties and high surface energy, that are prone to fouling and adhesive behavior of viscous food materials (Goode et al., 2013; Barish and Goddard, 2013; Stoforos et al., 2016).

2.5.7. Surface wettability

In general surfaces with characteristic low free surface energy (partial non-wetting surface) exhibit longer induction periods, compared with high-energy surfaces (partial wetting surface), which promote nucleation process (Awad, 2011). Non-wetting or low-energy surfaces (such as polymer and ceramic coatings) have longer induction periods than wettable or high-energy surfaces, resulting in less product deposition. Depending on both the surface tension of the liquid and the surface free energy of the solid contact-surface, a characteristic contact angle ($\theta$ (°)) will form. Surface wettability expresses the interaction between the surface tension forces of the fluid and the attractive forces of the contact-surface (Fig. 2.6). Surface wettability is one of the parameters which differentiate the contact-surfaces, varying from hydrophilic ($\theta<0^\circ$ to $90^\circ$), hydrophobic ($\theta=90^\circ$ to $150^\circ$), up to superhydrophobic ($\theta>150^\circ$) behavior (Loibl et al., 2012; Korhonen et al., 2013). Superhydrophobic and hydrophobic surfaces exhibit a non-wetting or a partial non-wetting behavior with a low surface free energy while surfaces with hydrophilic characteristics, the surfaces with high free surface energy, exhibit a wetting or partial wetting behavior. Surface wettability can be quantified by measuring the equilibrium contact angle of a small volume of liquid at the liquid-
surface interface. The formed contact angle is expressed by the Young equation (Eq. 2.1), through the mechanical equilibrium of the interfacial tensions ($\gamma$ (N/m)) between the liquid, the contact-surface, and the surrounding vapor (Fig. 2.7) (Loibl et al., 2012; Korhonen et al., 2013). The mechanical equilibrium is between the liquid-vapor interfacial tension ($\gamma_{LV}$), the liquid-solid interfacial tension ($\gamma_{LS}$) and the solid-vapor interfacial tension ($\gamma_{SV}$).

$$\gamma_{SV} - \gamma_{SL} = \gamma_{LV} \cdot \cos\theta$$  \hspace{1cm} (2.1)

Figure 2.6. Liquid-surface interaction, for a non-wetting surface ($\theta>90^\circ$) (left) and a wetting surface ($\theta<90^\circ$) (right) (Chem1.com, 2017).
2.5.8. Surface topography/roughness

One other property of contact-surface is surface roughness. Rough surfaces increase the effective contact area of the surface, providing suitable sites for nucleation which promotes initiation of fouling, increasing particulate deposition and product adhesion. Moreover, increased surface roughness is associated with the creation of turbulence effects, resulting in instabilities in the viscous sublayer of the flowing fluid streams close to the contact-surface (Awad, 2011; Mérian and Goddard, 2012). The surface roughness of the contact-surface is usually determined using atomic force microscope (AFM). AFM is used to map the surface, providing a general description of the height variations in the surface (Mérian and Goddard, 2012).
2.5.9. Surface forces

Another surface property important for fouling formations is surface forces. Surface forces, such as London–van der Waals force and electric double layer interaction force can result in intermolecular attraction or repulsion between charged fluid particles leading to fouling formation (Awad, 2011). The electric double layer interaction force can be attractive or repulsive. In addition, viscous hydrodynamic force, such as drag or shear stress, of the contact-surface influences the attachment and removal of the fluid stream flowing close to the surface (Awad, 2011).

2.6. Methods for determining fouling

The fouling resistances can be measured either experimentally or by analytical methods. The main fouling detection methods include measurements of mass, pressure, temperature, and electric current differences (Awad, 2011; Wallhäußer et al., 2012).

2.6.1. Direct weighing

The simplest method to determine the extent of product and fouling deposition on the tested surface is by direct weighing. Direct weighing measurement detects the residual deposit on the contact surface based on the mass difference of the surface before and after the process (Awad, 2011).

2.6.2. Thickness

Another simple technique for fouling detection and measurement is the measurement of the thickness of the fouling deposit. Use of a micrometer or microscope to measure the
thickness of fouling deposition is a useful technique to evaluate and quantify crystallization fouling on the contact surface (Awad, 2011). For the fouling cases with a small sample thickness, AFM technique can be used to determine the changes in the roughness of the surface, resulted from product and fouling deposition (Goode et al., 2013).

**2.6.3. Pressure drop**

Pressure drop measurement is a standard, indirect method, for fouling detection. During fouling formation, the pressure drop across the processing equipment is increased due to the reduction of the flow cross-sectional area, and the rough character of the fouling deposit layer. Pressure drop changes over the time, during flow at a constant flow rate, is an indication of product and fouling deposition. In continuous flow processing systems, pressure measurement at the inlet and the outlet of the processing unit, for pressure drop detection, is a regular process, accompanied by other standard methods such as temperature measurements (Awad, 2011; Wallhäußer et al., 2012).

**2.6.4. Heat transfer parameters**

Similar to pressure drop measurements, the inlet and outlet temperature of the product is routinely measured. Temperature difference can be used as a method to detect product and fouling deposition, through the fluctuating of the measurements during the process (Awad, 2011; Wallhäußer et al., 2012). During fouling development, the efficiency of heat transfer is reduced, with the product outlet temperature to drop or remain high, for heating or cooling, respectively (Awad, 2011; Wallhäußer et al., 2012).
Moreover, changes in heat transfer parameters, such as heat flux or overall heat transfer coefficient can be used as an indirect measurement for fouling detection (Awad, 2011). These heat transfer parameters are affected by factors such as temperature changes, mass flow rate, and thermal conductivity of the product, the heating medium, and the fouling resistance, being affected by the fouling formation and deposition (Awad, 2011; Wallhäußer et al., 2012).

2.6.5. Electrical parameters

Electrical parameters such as electrical resistance and conductivity can be used as a fouling detection method. During mineral crystallization fouling, change in salt concentration due to precipitation induces a change in electric conductivity. Measurement of the changes in electrical and thermal conductivity can be used as a method to determine fouling formation on the contact-surface (Chen et al., 2004; Wallhäußer et al., 2012).

2.6.6. Acoustic methods

Fluctuation of acoustic parameters such as acoustic impedance, attenuation coefficients, and acoustic signal vibration amplitude can be used to detect fouling deposit on the contact-surface. The resistance of acoustic signal transmission due to the fouling deposit can be measured using transducers (one sender and one receiver) and accelerometer attached on the tested surface (Wallhäußer et al., 2011; Wallhäußer et al., 2012).

2.7. Removal and cleaning of fouling

Removal of the residual product in low-flow areas of processing equipment and cleaning of fouling deposition is a major challenge for the efficiency of food and dairy
processing (Fryer and Asteriadou, 2009; Goode et al., 2013). To remove the product and fouling deposition from the contact surface, mechanical removal due to fluid flow action or cleaning processes should overcome the cohesive forces that keep the fouling material together and the adhesive forces between the deposit and the contact-surface (Fig. 2.8) (Fryer and Asteriadou, 2009).

Moreover, in food and dairy processing, cleaning-in-place (CIP) is used to remove the residual product, fouling, and biofouling that remain in the process line after production. During CIP, water and/or chemical solution is circulated around plant process equipment under specific temperature and flow conditions (Fryer and Asteriadou, 2009; Goode et al., 2013).
Fryer and Asteriadou (2009) suggested a classification map of cleaning problems of fouling on the surface of processing equipment. The map relates the type of fouling material with the type of cleaning required (cold water, hot water, chemicals) (Fig. 2.9). The residual fouling materials, at the end of food processing, can be classified as:

i) Low viscosity fluids: the residual deposit is often water or has similar properties with water. This type of residual deposit is observed after cleaning between process runs.

ii) High viscosity fluids: the residual deposit material is a highly viscous fluid, such the residual product at the contact-surface of the processing equipment, after processing of foods with high starch content.

iii) Cohesive solids: the deposit is a solid-like fouling material, such as soft protein gel film (Type-A milk fouling) or very hard solids formed by precipitation of minerals (Type-B milk fouling).

Furthermore, Fryer and Asteriadou (2009) have divided fouling deposition into three different types, based on the fouling material and the cleaning process:

1. Type 1 is associated with residual fouling material of viscous foods and viscoelastic or viscoplastic fluids, such as yogurt, which is easy to remove, by rinsing the contact-surface with hot water.

2. Type 2 is associated with cleaning and killing microbial and biological material, such as biofilms and polymers (gel-like films), which are removed with the combination of hot water and chemical use.

3. Type 3 is associated with cleaning process of cohesive solids, the fouling materials of milk pasteurization and processing of fermented products (beer), which are removed with chemicals.
Mechanical fluid flow action (shear stress) is enough to disrupt the weak cohesive and adhesive forces of Type-1 fouling deposition and remove the deposit. For Type-2 fouling deposition, further to the hot water fluid flow, part of deposit removal is done by application of chemicals. For Type-3 fouling deposition, application of chemicals is necessary due to the strong cohesive and adhesive forces of the deposit. Chemicals break down the strong bonds of cohesive and adhesive forces and remove the difficult solid-like deposit from the surface.

![Cleaning Map](image)

Figure 2.9. Fryer and Asteriadou (2009) cleaning map; classification of fouling materials, based on the cleaning process.
2.8. Effects of fouling

Product and fouling depositions on the surface of processing equipment reduce the operating efficiency, by reducing the heat transfer and thermal mixing (temperature equalization) efficiency, and pressure fluctuation. (Bott, 1995; Awad, 2011; Wallhäuser et al., 2012; Kazi, 2012; Goode et al., 2013; Stoforos et al., 2016).

2.8.1. Effects of fouling on heat transfer

Product and fouling deposit layers on heat exchanger surfaces increase the resistance to heat transfer (Awad, 2011; Kazi, 2012; Goode et al., 2013). Accurate measurement of the effects of fouling on heat transfer is critical for process design. Changes in heat transfer efficiency may have a significant effect on food product safety and quality. Changes in heat transfer during fouling are designed by including a thermal fouling resistance coefficient or fouling factor, $R_f$ (m$^2$·K/W), in the equation relating the overall heat transfer coefficient (Eq. 2.2) (Awad, 2011; Kazi, 2012; Goode et al., 2013).

$$\frac{1}{U} = \frac{1}{U_o} + R_f \quad (2.2)$$

Where $U$ is the overall heat transfer coefficient (W/(m$^2$·K)), at a specific time ($t$ (s)) of the process and $U_o$ is the initial overall heat transfer coefficient (W/(m$^2$·K)) for the clean heat exchanger (i.e. without fouling).

Thermal fouling resistance coefficient, $R_f$ is a function (Eq. 2.3) of the thickness ($x$ (m)) and thermal conductivity ($k_f$ (W/(m·K))) of the fouling deposit layer and is a function of time as fouling process progresses (Fig. 2.10) (Awad, 2011; Kazi, 2012; Goode et al., 2013).

$$R_f = \frac{x}{k_f} \quad (2.3)$$
Figure 2.10, known as the fouling curves, presents the changes of thermal fouling resistance coefficient, $R_f$, during the fouling process. The delay time, $t_d$ (s), indicates the initial induction period where no fouling occurs. The value of $t_d$ depends on the properties and the nature of contact surface (non-wetting or wetting characteristics). After the induction, delay period, three different fouling factor time curves can be observed:

1. Linear fouling curve, where the deposition rate ($\Phi_d$) is constant and the removal rate ($\Phi_r$) is neglected or their difference is constant.

2. Falling rate fouling curve, where the fouling deposition rate increases over the time but not linearly, with a decreasing deposition rate over the time.

3. Asymptotic fouling curve, where both fouling deposition and removal rate are functions of fouling layer thickness. In this case the rate of fouling deposition gradually decreases, until the fouling process reaches a steady state $\Phi_d=\Phi_r$.

In particular, in industrial operations, asymptotic fouling is more common, with the asymptotic fouling factor ($R_f^*$) to be used for calculations (Awad, 2011).
Figure 2.10. Fouling curves presenting the changes of thermal fouling resistance coefficient, $R_f$, during the fouling process (Awad, 2011).

2.8.2. Effect of fouling in pressure drop

During product and fouling deposition, at a given flow rate, the pressure drop across the processing equipment will increase, due to the reduction of the flow cross-sectional area and the rough character of the fouling deposit layer (Awad, 2011; Kazi, 2012; Wallhäuser et al., 2012). The increase of pressure drop, during the fouling formation, results in additional pumping power requirement. Control and monitoring of pressure drop during fouling process is important to avoid excessive pressure and damage to the processing equipment (Awad, 2011; Kazi, 2012; Wallhäuser et al., 2012).
2.8.3. Effect of fouling in thermal mixing

Thermal mixing, or temperature equalization, has been proposed as an efficient method for enhancement of cooling process of highly viscous foods, such as sweet potato puree, banana puree, and cheese sauce (Stoforos et al., 2016). Stoforos et al. (2016) used acoustic and mechanical vibration to impose transversal vibration motion on a 180° bend pipe, used as the mixing unit, and generate thermal mixing of the viscous foods tested. However, the efficiency of thermal mixing was affected by the product and fouling deposition at the pipe wall for products such as banana puree and cheese sauce (Fig. 2.11) (Stoforos et al., 2016). The non-slip fouling layer deposit at the pipe wall for these products reduced the thermal mixing efficiency, acting as a resistance on the transmission of the acoustic and vibration signals, imposing the mixing (Wallhäußer et al., 2011; Wallhäußer et al., 2012; Stoforos et al., 2016).

Figure 2.11. Formation of thick fouling layer at the pipe stainless steel wall, during cooling and thermal mixing studies of banana puree (Stoforos et al., 2016).
2.9. Modified food contact surfaces for fouling reduction

Reduction of the deposition and adhesion of fouling components on the surface of processing equipment, like stainless steel heat exchangers, depends on different parameters such as operating conditions (temperature, flow rate), food composition/chemistry and processing surface properties (material, wettability, topography/surface roughness) (Santos et al., 2004; Mérian and Goddard, 2012; Goode et al., 2013). Surface modification has been a subject of research for many studies focusing on controlling, minimizing or elimination of fouling and biofilm formation in food and dairy processing (Santos et al., 2004; Rosmaninho et al., 2007; Balasubramanian and Puri, 2009; Kananeh et al., 2010; Mérian and Goddard, 2012; Goode et al., 2013; Barish and Goddard, 2013).

Unfortunately, the most commonly used material for food processing equipment, stainless steel (300 series), has surface characteristics, such as partial wetting properties and high surface energy, that are prone to fouling and adhesive behavior of food and dairy products (Goode et al., 2013; Barish and Goddard, 2013). Recent studies of fouling reduction, have adopted techniques such as polymer and nanoparticle coatings (polyethylene glycol, silica or fluorocarbon and polytetrafluoroethylene (PTFE)), and physico-chemical surface modification techniques such as plasma-enhanced chemical vapor deposition and electrochemical and ion implantations, in order to modify the surface characteristics of stainless steel, prevent fouling formation and improve hygienic properties of food-contact surfaces (Santos et al., 2004; Mérian and Goddard, 2012). Surface properties, such as surface wettability (contact angle, spreadability), morphology (roughness and topography), and surface chemistry, have been modified to produce functional surfaces that mimic the self-cleaning and repellent characteristics of natural antifouling materials, such as the lotus leaf (*Nelumbo nucifera*)
(Beuf et al., 2003; Santos et al., 2004; Premathilaka et al., 2006; Rosmaninho et al., 2007; Balasubramanian and Puri, 2009; Kananeh et al., 2010; Mérian and Goddard, 2012; Barish and Goddard, 2013; Goddard et al., 2013).

2.9.1. Surface modification techniques

Surface modification studies have been focused on mimicking the properties of natural anti-fouling materials, such as the lotus leaf (*Nelumbo nucifera*). The super-hydrophobic behavior, in combination with the roughness of the nanostructure of the lotus leaf, is associated with unique self-cleaning and water-repelling properties, known as the lotus effect (Latthe et al., 2014). Furthermore, novel surface modification techniques, such as polymer and nanoparticle coatings (silica or fluorocarbon compositions), physicochemical surface modification, plasma-enhanced chemical vapor deposition, electro-chemical and ion implantations have enabled researchers to successfully lower the free surface energy of the food contact surfaces by altering wettability (hydrophobic and hydrophilic) and roughness characteristics (Beuf et al., 2003; Santos et al., 2004; Rosmaninho et al., 2007; Premathilaka et al., 2006; Balasubramanian and Puri, 2009; Kananeh et al., 2010; Mérian and Goddard, 2012; Barish and Goddard, 2013). In food, and in particular dairy processing research areas, antifouling methods have been applied on the surface of stainless steel plate heat exchangers, resulting in significant reduction of the amount of product deposition and fouling formation, for foods such as milk, skim milk and tomato juice, improving the operation and cleaning efficiency of the heat exchangers and reducing bacterial adhesion (Beuf et al., 2003; Santos et al., 2003; Santos et al., 2004; Rosmaninho et al., 2007; Premathilaka et al., 2006;
Balasubramanian and Puri, 2009; Kananeh et al., 2010; Mérian and Goddard, 2012; Barish and Goddard, 2013).

2.9.2. Autocatalytic Ni–P–PTFE coating

One of the most used surface modification techniques is the coating of electroless (or autocatalytic) nickel-phosphorus and PTFE (Ni–P–PTFE). The process contains five steps: 1) alkaline cleaning bath, 2) pickling process, 3) galvanic deposition of nickel (activate substrate), 4) autocatalytic reaction (deposition of Ni–P plating on the sample), 5) adding PTFE particles in the coating bath to be incorporated in the Ni–P matrix (Fig. 2.12) (Santos et al., 2004). The incorporation of PTFE particles into the Ni-P matrix can take advantage of the different properties of Ni-P alloy and PTFE. Electroless nickel–phosphorous is widely used in the fouling studies because of its corrosion and wear resistance and its inherently uniform coating thickness. PTFE is chemically very inert and has a relatively high melting point, low friction coefficient and extremely low surface energy (~20-30 mN/m), resulting in non-stick properties of PTFE (Beuf et al., 2003; Santos et al., 2004; Zhao et al., 2007). The resulting properties of electroless Ni–P–PTFE coatings, such as non-stick, hydrophobic, partial non-wetting, anti-adhesive, lower friction, good wear and good corrosion resistance, have been used successfully in fouling and biofouling studies, in dairy process (Beuf et al., 2003; Santos et al., 2004; Rosmaninho et al., 2007; Zhao et al., 2007; Balasubramanian and Puri, 2009; Barish and Goddard, 2013). Ni-P-PTFE has the best cleaning efficiency comparing with the rest of the surface modification techniques. Moreover, Ni-P-PTFE is the most promising surface modification technique as an antifouling surface for non-microbiological deposits (calcium phosphate, β-lactoglobulin and milk-based product) and significantly better cleaning efficiency
comparing with the other materials (Beuf et al., 2003; Rosmaninho et al., 2007; Zhao et al., 2007; Balasubramanian and Puri, 2009; Rungraeng et al., 2012; Barish and Goddard, 2013).

2.9.3. Fluorocarbon based nanoparticle coatings

Using commercially available silica or fluorocarbon based nanoparticle coatings, and by applying one of the most commonly used surface modification techniques of electroless nickel (EN) plating with embedded PTFE nanoparticles, food and dairy process surfaces were successfully modified by lowering the surface energy via increasing the hydrophobicity and adjusting the roughness of the surface (Kananeh et al., 2010; Balasubramanian and Puri, 2008; Balasubramanian and Puri, 2009; Barish and Goddard, 2013). Similar with the autocatalytic
Ni–P–PTFE coating process, EN is an auto-catalytic chemical technique used to deposit a layer of nickel-phosphorus (Ni-P) alloy on the examined surface sample (Kananeh et al., 2010). The next step of EN process is the silica or fluorocarbon based nanoparticle coating (Kananeh et al., 2010). The modified surface through silica or fluorocarbon based nanoparticle coatings was successfully used for mitigation of fouling formation during thermal processing of milk based food (tomato sauce) products (Kananeh et al., 2010; Balasubramanian and Puri, 2008; Balasubramanian and Puri, 2009).

2.9.4. Silica coating

In food fouling and biofouling studies, commercially available silica coatings (SiOx), as well as silica coatings based on the sol–gel process have been tested. This approach is applied to produce a hydrophobic (commercially available samples) or hydrophilic (made with the sol-gel method) and a hydrated anionic surface (Beuf et al., 2003; Santos et al., 2004; Santos et al., 2006; Rosmaninho et al., 2007). AFM studies have shown that SiOx has higher surface roughness compared to other coating methods (fluorocarbon coatings). The SiOx surface modification technique has a promising performance in reducing fouling of dairy products; however, fouling deposition is higher compared to low surface free energy and hydrophobic coatings (Ni-P-PTFE) (Beuf et al., 2003; Santos et al., 2004; Santos et al., 2006; Rosmaninho et al., 2007).

2.9.5. Plasma enhanced chemical vapor deposition

Plasma enhanced chemical vapor deposition (CVD) or plasma enhanced CVD is a chemical vapor deposition process, used for surface modification by deposition of thin films
from a gas state (vapor) to a solid state on a substrate. The chemical reaction in plasma CVD technique is initiated through the formation of plasma, which is usually induced through radio frequency or microwaves. The coating material is introduced into the process as a solid target, using gaseous or vaporous precursors. Usually, in fouling studies, typical gases are CxHx for diamond-like carbon (DLC) films and Hexamethyldisiloxan (HMDSO) for SiOx films (Beuf et al., 2003; Santos et al., 2004; Rosmaninho et al., 2007; Premathilaka et al., 2007). Using Plasma CVD, there is also the possibility to implement additional atoms into the coating, by adding a silicon-containing precursor and oxygen to the working gases. Using this modification, additional coatings such as DLC–Si–O can be obtained. The disadvantages of this surface modification technique are the reduction of density and weaker adherence to the substrate material. Using CVD DLC films on the surface of the stainless steel a hydrophobic surface is produced with a promising antifouling behavior against protein fouling, with even better cleaning efficiency (Beuf et al., 2003; Santos et al., 2004; Rosmaninho et al., 2007; Premathilaka et al., 2007).

2.9.6. Surface preparation by sputter technique

Sputter deposition is a physical vapor deposition (PVD) process of thin film deposition by sputtering under sub-atmospheric pressure. This involves ejecting material from the examined coating material (the “target”) to the surface to be coated (the “substrate”). In a vacuum chamber, a high voltage is in the area between the substrate and the target, a reactive gas is introduced which gets ionized (usually argon gas). The applied high voltage makes the plasma ions to accelerate and hit the cathode, the target surface. On hitting the cathode surface,
plasma ions dislocate atoms from the target surface. Detached atoms are deposited on other parts of the surface (Fig. 2.13).

Figure 2.13. PVD sputter deposition method deposition (Santos et al., 2004). High voltage (RF) applied in the area between the target (cathode) and the substrate, ionizing the reactive gas (black circles). The ionized gas (blue circles) accelerates and hits the cathode, resulting in deposition of atoms (red circles) from the target to the substrate (Santos et al., 2004).

TiN (Titanium-nitrogen) coatings were produced by reactive sputtering of a titanium (Ti) with an unbalanced magnetron cathode and nitrogen (N₂) as the reaction gas. Results with milk and milk based products on fouling with TiN coatings were not as promising as with other
coating materials (Santos et al., 2004; Rosmaninho et al., 2007; Mauermann et al., 2009). TiN coating can be further modified to titanium carbide (TiC) and DLC coating films. Using physical vapor deposition (PVD) process with the TiN film in the presence of mixed N₂ gas and acetylene on top of a metal substrate, TiC film can be produced (Santos et al., 2004; Rosmaninho et al., 2007). TiC surfaces have been reported for their good repellent behavior against *Bacillus cereus* spores (Rosmaninho et al., 2007). Furthermore, DLC films can be produced with sputter deposition method, using argon and hydrogen plasma. The advantages of sputter deposition DLC coatings are the hard and very adhesive films, comparing with the CVD.

### 2.9.7. Ion implantation

There are two kinds of ion implantation techniques: direct ion implantation and turbulent ion implantation.

*Direct ion implantation (or ion beam implantation)*

During directed ion implantation (or ion beam implantation), the surface is bombarded with highly accelerated ions, with an average energy of several keV (~200 keV) (Beuf et al., 2003; Santos et al., 2003; Santos et al., 2004; Rosmaninho et al., 2007). The modifying material is directly implanted into the base substrate to form a surface alloy. The ions settled perpendicular to the surface in a depth of approximately of 0.2 µm (Beuf et al., 2003; Santos et al., 2003; Santos et al., 2004; Rosmaninho et al., 2007). In food and dairy fouling studies, sample surfaces with bean implantation of SiF₃⁺ ions have been studied. SiF₃⁺ ions implantation produces a surface with hydrophilic characteristics without affecting the surface topography (Santos et al., 2004). SiF₃⁺ ions implantation has a potential to reduce fouling
formation during milk processing, presenting a good approach to mitigate protein (β-lactoglobulin) fouling (Beuf et al., 2003; Santos et al., 2003; Rosmaninho et al., 2007).

**Turbulent ion implantation**

During turbulent ion implantation, a plasma is ignited in the vacuum chamber during turbulent ion implantation (Santos et al., 2004). The atoms hitting the surface have such a high energy that they can penetrate into the interior of the material up to the depth of about 100 µm (Beuf et al., 2003; Santos et al., 2004). In food and dairy fouling studies MoS$_2$ particles have been used (Beuf et al., 2003; Santos et al., 2004; Rosmaninho et al., 2007). MoS$_2$ particles promote the anti-adhesion behavior of the tested surface. Similar to the case of direct implantation of SiF$_3^+$ ions, MoS$_2$ particles do not affect surface topography of the samples and produce a hydrophilic surface (Santos et al., 2004). MoS$_2$ particles implantation surface modification technique looks promising for milk fouling reduction (Beuf et al., 2003; Rosmaninho et al., 2007). However, surface techniques, such as Ni–P–PTFE and SiF$_3^+$ ions implantation reduce fouling of milk better compared to MoS$_2$ particles (Beuf et al., 2003; Rosmaninho et al., 2007).

Antifouling materials and surface modification techniques, with low surface free energy and smooth surface characteristics, have successfully been used in food and dairy research, improving the operation and cleaning efficiency and reducing the bacterial adhesion (Beuf et al., 2003; Santos et al., 2004; Premathilaka et al., 2006; Rosmaninho et al., 2007; Balasubramanian and Puri, 2009; Kananeh et al., 2010; Mérian and Goddard, 2012; Barish and Goddard, 2013; Goddard et al., 2013; Stoforos et al., 2017a). However, antifouling surface modification techniques have not extensively been used for commercial application for food and dairy processing due to the cost and complexity of fabrication and, in some cases, the
incompatibility with food grade materials, in addition to the reported short-term stability of antifouling properties of the modified surfaces (Barish and Goddard, 2013). Based on these observations, there is a need for development of new food surface modifications techniques with lower cost and improved simplicity of fabrication.

During this research, chemical hydrophobic surface treatments and hydrophobic engineered textiles were examined as antifouling material in cooling of viscous foods. The results of these studies showed promising results in enhancement of cooling and cleaning processes of viscous foods. Additionally, a new surface modification technique was introduced, using textile construction methods, with low cost and with potential for extended use in the antifouling research area (Stoforos et al., 2017).

2.10. Conclusions

In this review paper, the mechanisms of fouling and product adhesion during food processing, as well as the challenges and opportunities of the new antifouling developments have been presented and discussed. Product and fouling deposition on the food-contact surface of processing equipment (heat exchangers, spray dryers, mixing units) during thermal processing of viscous foods and dairy products is a major problem in the food industry, reducing processing efficiency and increasing operating costs. The formation and build-up of fouling layers formed on food processing equipment reduce the efficiency of heat transfer, thermal mixing, and increase the pressure drop across the processing units, resulting in degradation of final food product quality and increasing the overall processing and cleaning operation time and costs, while also being prone to formation of biofilms.
During heating or cooling of food products, fouling formation depends on physical and chemical interactions between the flowing food and the food-contact surface. Fouling process can be influenced by fluid properties (velocity and temperature) and composition (proteins, minerals, fats, carbohydrates), surface characteristics (surface material, wettability, roughness) and conditions (surface temperature) as well as the processing operational conditions (High-Temperature Short-Time, Ultra high-temperature, microwave, ohmic heating).

Food composition and concentration of food components such as proteins, minerals, fats, and carbohydrates, and total solids concentration, is a critical factor regarding the fouling type formation on the food-contact surface. Fouling in food processing is formed from the precipitation of partially soluble salts (scaling, Type-B milk fouling), from the physicochemical reactions of proteins (Type-A milk fouling) and carbohydrates, precipitation of suspended particles (colloids), crystallization and solidification of fat components (during freezing and cooling), biofouling and corrosion reactions.

Surface properties, such as surface wettability (hydrophilic, hydrophobic, superhydrophobic), morphology (roughness and topography), and surface chemistry, of stainless steel heat, exchanges have been modified to prevent fouling formation and improve hygienic properties of food-contact. In food and dairy processing studies, novel antifouling surface modification techniques, such as polymer and nanoparticle coatings (silica or fluorocarbon compositions), physicochemical surface modification, plasma-enhanced chemical vapor deposition, electro-chemical and ion implantations have enabled researchers to successfully lower the free surface energy of the food-contact surfaces by altering wettability (hydrophobic and hydrophilic) and roughness characteristics to reduce fouling formation and
deposition, improving the operation and cleaning efficiency and reducing the bacterial adhesion.

Finally, reducing the product accumulation and fouling formation on the processing equipment, potentially can significantly improve the operation efficiency and reduce the total cost, by increasing the heat transfer and mixing of the process, while minimizing the product quality losses. However, antifouling surface modification techniques have not extensively been used for commercial applications in food processing. The cost, complexity of fabrication and the reported short-term stability of antifouling properties of the modified surfaces still need to be improved for potential applications in advanced thermal processing of food and dairy products.

**Nomenclature**

*Latin letters*

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>CIP</td>
<td>Cleaning-In-Place</td>
</tr>
<tr>
<td>CVD</td>
<td>Chemical vapor deposition</td>
</tr>
<tr>
<td>DLC</td>
<td>Diamond-like carbon films</td>
</tr>
<tr>
<td>EN</td>
<td>Electroless nickel plating surface modification technique</td>
</tr>
<tr>
<td>HTST</td>
<td>High-Temperature Short-Time process</td>
</tr>
<tr>
<td>HMDSO</td>
<td>Hexamethyldisiloxan</td>
</tr>
<tr>
<td>R_f</td>
<td>Thermal fouling resistance coefficient or fouling factor (m²·K/W)</td>
</tr>
<tr>
<td>t</td>
<td>Time (s)</td>
</tr>
<tr>
<td>U</td>
<td>Overall heat transfer coefficient (W/m²·K), at specific time during the process</td>
</tr>
<tr>
<td>U_o</td>
<td>Initial overall heat transfer coefficient (W/m²·K) for the clean, heat exchanger</td>
</tr>
</tbody>
</table>
UHT Ultra high-temperature process

\(x\) Thickness of fouling deposit layer (m)

Greek letters

\(\Delta H\) Heat transfer (J)

\(\Phi_d\) Deposition rate of fouling material

\(\Phi_r\) Removal rate of fouling material

Subscripts

d refers to the deposition process of fouling material

f refers to the fouling deposit layer

LV refers to the liquid-vapor interfacial tension (N/m)

LS refers to the liquid-solid interfacial tension (N/m)

o refers to the clean, without fouling heat exchanger

r refers to the removal process of fouling material

SV refers to the solid-vapor interfacial tension (N/m)
References


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CHAPTER 3:
Computer Aided Design of Continuous Flow Cooling of Viscous Foods

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Word count: 7,807

Short version of title: Computer Aided Design of Cooling of Foods…
ABSTRACT

This study presents the results of computer simulation modeling, using Computational Fluid Dynamics (CFD) and heat transfer software systems on continuous flow cooling of viscous food products, such as applesauce and sweet potato puree. Computer simulation modeling was used to compare two different flow regimes within the same tube in tube heat exchanger: i) cooling process of the food, while flowing within the internal tube cylinder of the heat exchanger and ii) cooling process of the food, while flowing within the annular passage of the heat exchanger. Results of the set of simulations indicate that under an identical set of flow rates, surrounding and initial temperature conditions (120 °C, 130 °C and 140 °C for the process material, 4 °C for the coolant) the flow regime in which the process material is flowing within the external annulus of the heat exchanger would theoretically result in a bulk product temperature at the outlet of the heat exchanger within a temperature range of ~80-95 °C for both examined products. When the process material is flowing within the internal tube of the heat exchanger, the calculated bulk product temperature at the outlet of the heat exchanger was in the temperature range of 91-102 °C for sweet potato puree and 85-99 °C for applesauce. Therefore, in this case, the potential improvement in the cooling efficiency resulting from the movement of process material flow from the internal tube into the external annulus could be up to 14% and 23%, for applesauce and sweet potato puree, respectively. Finally, the results showed that having the food product flowing within the annulus, cooling efficiency could be improved by increasing the heat loss to the environment, while maintaining the sanitation conditions in the processing plant.

Key words: Computer Simulation, Continuous flow cooling, sweet potato puree, applesauce
3.1. Introduction

Advanced thermal technologies such as continuous flow microwave (MW) systems, ohmic and radio frequency heating have enabled food processors to maximize product quality during continuous thermal processing of highly viscous and multiphase food products (Coronel et al., 2005; Steed et al., 2008; Cullen et al., 2012). However, due to the lack of advanced technologies, continuous flow cooling of these materials still depends on slow, predominately convection-based heat transfer methods. During continuous flow cooling of highly viscous foods, like fruit and vegetable purees, laminar flow, non-Newtonian flow behavior (characteristic for the most of the processed foods (Tokisangyo, 2016)), laminar flow and low thermal conductivity lead to a wide temperature distribution within the product and consequently in unequal cooling treatment and degradation of final food quality. This results in a non-uniform cooling of food material, very slow cooling processes and reduced food quality.

Unfortunately, often predicted and anticipated developments of volumetric cooling technologies, such as magnetic field cooling (Sarlah et al., 2006; Kawanami et al., 2011) have been slow and expensive in their progress to large-scale commercial applications. Additionally, food industries, in an effort to enhance the cooling rate of continuous flow thermal processing, utilize coolant mediums (like ethylene glycol based coolants) at a very low temperature range, but without any significant improvement in the cooling process of highly viscous food products. Moreover, using this type of coolants and considering the processing conditions (room air temperature and high relative humidity in the processing plant) and the "cold" temperature (below the dew point) of the surface of the heat exchanger, result in formation of dropwise and film condensation from moist air at the outside surface of the cooler, resulting in
dripping water (Moerman, 2016). As a result, condensation at the outside surface of the heat exchanger increases heat losses to the environment and also has a negative impact on sanitation conditions of the food production plan.

The main objective of this research was to study continuous flow cooling of highly viscous food products, such as applesauce and sweet potato puree, and determine the best operational conditions, that will enable the food processors to enhance cooling processing while maintaining the sanitation conditions in the food processing plant. Moreover, a computer simulation was used to compare two different flow configurations within the same tube in tube heat exchanger: i) product flowing inside the inner tube of the heat exchanger, conventional configuration, and ii) product flowing within the annulus side of the heat exchanger (Fig. 3.1). The anticipated outcomes of this study was that implementation of the second flow configuration could yield three potential benefits: a) reduction of cooling energy losses to the environment, b) movement of the colder, highly viscous layer of the food material into the more central flow segment where it will be easier to equalize the temperature profile and enhance the cooling of the flowing food, and c) reduction of condensate formation on the external surfaces of the cooling heat exchangers with an associated improvement in processing plant sanitary conditions.
3.2. Computer simulation model

Improvement of food processing procedures, such as thermal processing, is a major challenge in the food industry and food engineering. In the last decades, new developments in computational simulations have been used as a tool for better understanding and optimal design of different food processing applications such as sterilization, baking, drying, mixing, refrigeration (Xia and Sun, 2002; Norton and Sun, 2006; Yianniotis and Stoforos, 2014) and thermal processing (Lemus-Mondaca et al., 2011; Kechichiana et al., 2012). Moreover, new powerful modeling and simulation tools, such as Computational Fluid Dynamics (CFD), have enabled researchers to solve more complicated numerical and mathematical problems, involving the solution of complex partial differential equations describing fluid flow, heat and mass transfer, along with complex geometries and variable thermo-physical properties.
(density, thermal conductivity, heat capacity, viscosity, thermal diffusivity), related to applications in food processes.

In this study, *Multiphysics* software system *Comsol 5.2* (Comsol Inc., Burlington, MA, U.S.A.) was used to simulate a three-dimensional (3D) model of a tubular concentric heat exchanger, and study continuous flow cooling of highly viscous foods. The model comprises differential equations for fluid flow and heat transfer, solved by using CFD and heat transfer modules, to obtain results such as velocity profiles and temperature distributions, during cooling of viscous flowable food materials, such as applesauce, and sweet potato puree.

The basic geometry of the concentric tube in tube heat exchanger and the parameters of cooling, such temperature and flow rates of both the tested products and coolant, were modeled based on the experimental apparatus and the operational conditions used for cooling experiments with viscous foods. Moreover, the total length of the heat exchanger was 2.38 m, with an outer tube of 0.0762 m (3 in) diameter. The inner tube was modeled for two different study cases. For the first study case, the inner tube was designed identical to the existing experimental apparatus, with the diameter of the inner tube at 0.0508 m (~2 in), for validation of the model and comparison of experimentally obtained results of sweet potato puree with results obtained by computer simulation. The second study case was used to model and simulate different flow configurations, such as product flowing in the annulus or in the inner tube for both counter and co-current flow conditions. For this study case, the diameter of the inner tube was designed at 0.0381 m (1.5 in), to be identical with the hydraulic diameter \(D_H\) of the annulus flow, with the equal Reynolds number, for both examined flow configurations.

In this study, sweet potato puree and applesauce were chosen as the test materials. These products are characteristic highly viscous food products with a non-Newtonian flow
behavior. Published data from literature were used for the thermo-physical properties, such as density ($\rho$), specific heat ($c_p$), thermal conductivity ($k$), flow behavior index ($n$), fluid consistency coefficient ($K$), for both sweet potato puree (Coronel et al., 2005; Aqua-calc, 2016b) and applesauce (Steffe, 1996; Lozano, 2006; Toledo, 2007; Aqua-calc, 2016a) (Table 3.1). For these materials, identical volumetric flow rate of $6.3 \times 10^{-5}$ m$^3$/s (1 gal/min) was used for all the CFD simulations. For the heat transfer module, the inlet temperature of the materials was chosen based on the experimental data, at 92 °C for the validation study, while for the rest of the simulations the inlet temperature was at 120, 130 and 140 °C. Water was used as the cooling medium, with a constant volumetric flow rate of $56.9 \times 10^{-5}$ m$^3$/s (9 gal/min), while the inlet temperature was at 13 °C and 4° C, for validation studies and for all other simulations, respectively. Finally, stainless steel (300 series), with a thickness of 0.001651 m, was used as the structural material of the heat exchanger. Table 3.1, summarizes the thermo-physical properties (at room temperature of 20 °C, the initial temperature of the model) of all the materials used, with the properties of water and stainless steel used from the material library database of Comsol 5.2 Multiphysics.

Table 3.1. Properties of sweet potato puree, applesauce, water and stainless steel at 20 °C.

<table>
<thead>
<tr>
<th>Properties</th>
<th>Sweet potato puree</th>
<th>Applesauce</th>
<th>Water</th>
<th>Stainless steel</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\rho$ (kg/m$^3$)</td>
<td>1077</td>
<td>1031</td>
<td>1000</td>
<td>7850</td>
</tr>
<tr>
<td>$c_p$ (J/kg·K)</td>
<td>3750</td>
<td>3730</td>
<td>4182</td>
<td>475</td>
</tr>
<tr>
<td>$k$ (W/m·K)</td>
<td>0.54</td>
<td>0.58</td>
<td>0.60</td>
<td>44.50</td>
</tr>
<tr>
<td>$n$</td>
<td>0.39</td>
<td>0.25</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>$K$ (Pa·s$^n$)</td>
<td>18.78</td>
<td>12.70</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Viscosity $\mu$ (Pa/s)</td>
<td>-</td>
<td>-</td>
<td>1.002</td>
<td>-</td>
</tr>
</tbody>
</table>
3.2.1. Mathematical model

The purpose of the 3D heat exchanger model was to simulate and compute the velocity and temperature profiles of food materials for different flow configurations. The mathematical modeling for these simulations was based on continuous flow cooling of non-Newtonian, highly viscous food products, such as applesauce and sweet potato puree, under steady state processing conditions, laminar flow for the food samples and turbulent flow for the coolant. The simulations were conducted using CFD non-isothermal flow (weakly compressible flows) and heat transfer interfaces, which involve the solution of the differential equations of continuity (Eq. 3.1), momentum (Eq. 3.2) and energy (Eq. 3.3). Turbulent flow for coolant was modeled using the Reynolds-Averaged Navier-Stokes (RANS) equations (Eq. 3.4) (Shahsavand and Nozari, 2009).

\[ \nabla (u \cdot \rho) = 0 \]  \hspace{1cm} (3.1)

Continuity equation, also known as conservation of mass, describes the overall mass balance within the tube flow, where \( \rho \) (kg/m\(^3\)) is the fluid density, \( u \) is the fluid velocity component and \( \nabla \) is the del operator.

\[ \rho \cdot \left( u \cdot \nabla \right) u = -\nabla p - \left( \nabla \tau \right) + F \]  \hspace{1cm} (3.2)

Momentum or Navier-Stokes equation describes the motion of fluids, where \( p \) is the fluid pressure (Pa), \( \tau \) (or \( \sigma \) as it is referred in food rheology literature) is the stress component (Pa). The different terms in Eq. 3.2 correspond to the inertial forces (1), pressure forces (2), viscous forces (3), and the external forces (\( F \)) applied to the fluid (4).

\[ \rho \cdot c_p \cdot u \cdot \nabla T = \nabla \cdot (k \cdot \nabla T) + Q \]  \hspace{1cm} (3.3)
Eq. 3.3 describes the heat transfer in fluids, where $T$ is the temperature component (K) and $Q$ describes any other type of heat source (W/ m³), such as pressure work energy, viscous dissipation energy, and the energy from any external heat source.

The three-dimensional formulas of continuity, Navier-Stokes and energy equations apply for turbulent flow too. The following Eq. 3.4, RANS equation, can be used to give an approximate solution to Navier-Stokes equation, for turbulent flow.

$$\rho \cdot (u \cdot \nabla) \cdot u = \nabla \cdot \left[ - \rho \mathbf{I} + (\mu + \mu_T)(\nabla u + \nabla u)^T \right] + F$$

Where $U$ and $P$ are the time-averaged velocity and pressure, respectively; Eq. 3.4 has linear algebra terms, such as $\mathbf{I}$, which is the identity matrix and the term $(j)^T$, which is the outer product (the tensor product of two vectors). The term $\mu_T$ represents the turbulent viscosity. The turbulent viscosity, $\mu_T$, is evaluated using turbulence models. The most common model is the $k$-$\epsilon$ turbulence model (one of many RANS turbulence models), consists of solving two additional equations for the transport of turbulent kinetic energy $k$ (m²/s²) and turbulent dissipation $\epsilon$ (m²/s³) (Launder and Spalding; 1972).

For the evaluation of the model, Reynolds number ($N_{Re}$) (Eq. 3.5; Eq. 3.6) was calculated for both food products and coolant for all the different flow configurations (Madlener et al., 2009; Crespí-Llorens et al., 2015).

$$N_{Re} = \frac{\rho \cdot \overline{u} \cdot D_H}{\mu}$$

Eq. 3.5 was used to calculate Reynolds number for water (coolant), flowing in the inner tube and in the annulus side. Where $\overline{u}$ is the average velocity of water (m/s). The Reynolds number for water was at the turbulent regime, estimated at 15821.
Eq. 3.6, was used to calculate a generalized Reynolds number for non-Newtonian ($N_{GR\text{e}}$) foods (applesauce, sweet potato puree), flowing in the inner tube and in the annulus side. Where $\bar{u}$ is the average velocity of food products (m/s). The Reynolds number for all the tested products and for all the different flow configurations was very low ($N_{GR\text{e}}<<20$), in the laminar regime.

Additionally, bulk temperature ($T_{bulk}$ (°C)) was calculated at the exit length ($L$) of the heat exchanger for the tested food products (Eq. 3.7) (Whitaker, 1983). Bulk temperature (a function of radius ($r$), with $R$ to be the radius of the tube) was used as a measure to compare how efficient are the different flow configurations on continuous flow cooling of applesauce and sweet potato puree.

$$T_{bulk} = \frac{\int_{0}^{R} T(r, L) \cdot r \cdot u(r) \, dr}{\int_{0}^{R} r \cdot u(r) \, dr}$$ (3.7)

Moreover, knowing the inlet ($T_{inlet}$ (°C)) and outlet temperature ($T_{bulk}$), for both the coolant and examined foods, the energy gained from the coolant ($Q_w$ (W)) (Eq. 3.8) and lost by the product ($Q_p$ (W)) were estimated (Eq. 3.9). Finally, energy balance equation (Eq. 3.10) for the tubular heat exchanger was solved to calculate the heat losses to the environment ($Q_L$ (W)).

$$Q_w = m_w \cdot c_p \cdot \Delta T_w$$ (3.8)
Where \( m_w \) is the mass volumetric flow rate of water (kg·s) is, \( C_{p_w} \) is the specific heat (J/kg·°C) of water, and \( \Delta T_w \) (°C) is the temperature difference between the inlet and outlet temperature of water.

\[
Q_p = m_p \cdot c_{p_w} \cdot \Delta T_p
\]  

(3.9)

Where \( m_p \) is the mass volumetric flow rate (kg·s) is, \( c_{p_f} \) is the specific heat (J/kg·°C), and \( \Delta T_p \) (°C) is the temperature difference between the inlet and outlet temperature of the examined food.

\[
Q_p = Q_w + Q_L
\]  

(3.10)

### 3.2.2. Boundary Conditions

For the fluid flow modeling, for both laminar fluid flow and turbulent coolant flow, non-slip wall conditions were considered:

\[
u = 0
\]  

(3.11)

For the heat transfer modeling, the initial value of the temperature was set at room temperature, approximately at 20 °C. External natural convection heat transfer (\( Q \)) (W) on a long horizontal cylinder, was used as the boundary condition of the outer surface of the tubular heat exchanger:

\[
\dot{Q} = h \cdot A \cdot (T_{ext} - T)
\]  

(3.12)

Where \( A \) is the outer surface area of the heat exchanger, \( T_{ext} \) is the external temperature (air room temperature) at 20 °C, \( T \) presents the outer surface temperature of the heat exchanger.
and $h$ is the convection heat transfer coefficient (W/m$^2$·K), which is a function of the diameter of the heat exchanger, the $T_{\text{ext}}$ and absolute pressure $P_A$ at 101.33 kPa (1 atm).

For the heat transfer model, the boundary conditions at the pipe wall of the heat exchanger (all the stainless steel boundaries) were modeled using the conductive thin layer approach from *COSMOL Multiphysics*, where the heat flux $\vec{q}$ across the stainless steel pipe wall is given by the following equation:

$$\vec{q} = -k_s \cdot \frac{(\Delta T)}{d_s}$$  \hspace{1cm} (3.13)

For Eq. 3.13, $k_s$ is the thermal conductivity of stainless steel (W/m ·K), $d_s$ is thickness of the wall (m) and $\Delta T$ describes the temperature difference across the pipe wall (K).

### 3.3. Results and discussion

#### 3.3.1 Validation of model: Comparison of simulation results with experimental data

The results from computer simulations on cooling of sweet potato puree were compared with two different series of experimental data, under the identical assumed processing conditions, with the food flowing within the external annulus of the heat exchanger and the coolant flowing within the inner tube. Computer simulation results showed that sweet potato puree with an inlet uniform temperature distribution ($T_{\text{inlet}}$) at 92 °C was cooled down to a bulk temperature ($T_{\text{bulk}}$) of 64.6 °C, at the exit of the heat exchanger (Fig. 3.2).

Moreover, during cooling experiments with sweet potato puree, three single-point thermocouples (type-T) were used to measure the temperature of the food product at three different radial locations within the annulus of the heat exchanger (at the center of the annulus cross section area; close to the wall of the outer tube and at an intermediate point) (Fig. 3.3).
The cross-sectional product temperature was measured at three different locations across the length of the heat exchanger, at the inlet, exit and at the intermediate length of the heat exchanger. The experimental data points were compared against the simulated results, giving a good correlation (Fig. 3.4). Minor differences between the simulated data and the experimental results can be explained due to the non-uniform temperature distribution of the product at the inlet of the cooler during the experiments.

Figure 3.2. Simulation results of temperature distribution during cooling of sweet potato puree, with an inlet temperature of 92 °C and an outlet bulk temperature of 64.6 °C. Note that on temperature distribution graph different colors indicate different temperatures.
Furthermore, based on the temperature distribution graph, Figure 3.2, it is important to mention that the temperature of the product in the transition area of the product flow to/from the annulus passage, is significantly lower compared with the product bulk temperature at the same area of the heat exchanger. An observation, that is common for all the following cases where the tested food flowing within the annulus and indicates that the product in that transition region flows slower compared to the rest of the field. These ‘’static’’ or ‘’dead’’ zones, with minimal product flow, located in the transition area to/from the annulus side of the heat exchanger, generate the concern regarding dead zones and the efficiency of cleaning and sanitation efficiency.

Figure 3.3. Three thermocouples for a 3 in diameter tube, measuring the temperature at three different radial locations within the annulus side: center of the annulus area; close to the wall of the outer tube and at an intermediate point.
Figure 3.4. Comparison of simulation results against experimental data of product temperature distribution at the annulus cross sectional area of the heat exchanger, based on data at three different radial locations, namely center (0.0285 m), an intermediate point (0.0335 m), and close to the wall of the outer tube, (0.0365 m) for a) inlet, b) intermediate and c) exit. Note that radial distance starts measuring from the central axis of the concentric tubular heat exchanger.

3.3.2 Computer simulation results for cooling of sweet potato puree

Following the validation studies, the model was used to simulate the cooling processing of sweet potato puree under thermal processing conditions applicable in the food industry, using the advanced heating technologies. Computer simulation studies on cooling of sweet potato puree were conducted for three different product inlet temperatures 120, 130, 140 °C, under two different flow configurations, counter, and co-current: i) with the product flowing
in the inner tube and ii) with the product flowing in the annulus of the heat exchanger. Water was used as the coolant with an inlet temperature of 4 °C. The bulk temperature of the product at the exit of the heat exchanger was compared for all the different simulations to define the most efficient flow configuration for the cooling process.

Simulated data with sweet potato puree, using an inlet temperature of 120 °C, were used to compare the effects of counter and co-current flow modes on cooling for both flow configurations, using the equal hydraulic diameter for both inner and annular product flow. Based on the temperature distribution results and the calculated bulk temperatures of sweet potato puree at the exit of the heat exchanger, cooling was more efficient with the product flowing in counter-current flow, with an outlet bulk temperature at 81.1 °C, while the outlet bulk temperature for the case of co-current flow mode was at 82.1 °C (Fig. 3.5). On the other hand, for the case of sweet potato flowing within the inner tube, cooling in the co-current mode was slightly better with an outlet bulk temperature of 91.6 °C and 90.6 °C for the case of counter-current flow mode (Fig. 3.6).
Figure 3.5. Comparison of temperature distribution simulation data for a) counter-current and b) co-current flow modes, during cooling of sweet potato puree, flowing within the annulus, with an inlet temperature of 120 °C. Note the black arrows indicate the flow path for the product, while the blue arrows indicate the flow path for coolant.
Figure 3.6. Comparison of temperature distribution simulation data for a) counter-current and b) co-current flow modes, during cooling of sweet potato puree, flowing within the inner tube, with an inlet temperature of 120 °C. Note the black arrows indicate the flow path for the product, while the blue arrows indicate the flow path for coolant.
Furthermore, for a better understanding of the effects of cooling for the two different flow configurations, additional computer simulation studies were conducted focusing on the cooling performance and velocity profile of sweet potato puree. Velocity profile and cooling of sweet potato puree for both the examined flow configurations were tested in two different studies: i) having the same cross-sectional area, 0.0024 m² (adjusting the outside diameter (O.D.) of the inner tube at 0.0558 m (2.2 in)) for both inner tube and annulus product flow, ii) having the same Reynolds number for both cases, using the same $D_h$ value (O.D. of inner tube at 0.0381 m (1.5 in); inside diameter (I.D.) of inner tube 0.0762 m (3 in)) for both inner tube and annulus flow.

For the case with the same cross-sectional area, for both inner tube and annulus flow, similar velocity profiles were observed. A parabolic velocity profile was observed, with the maximum velocity ($u_{MAX}$) at 0.038 m/s, for the product flowing at the center of the cross-sectional area for both inner tube and annulus flow (Fig. 3.7). For the other case with the equal hydraulic diameter, a flattened velocity profile (plug flow type velocity profile) was observed for the annulus product flow, while a parabolic profile was observed for the product flowing within the inner tube (Fig. 3.8). Furthermore, for this case study, in order to obtain the same hydraulic diameter, the flow cross-sectional area of the inner tube had to be reduced and the corresponding area of the annulus had to be increased. A narrower cross-sectional area for the inner tube resulted in the highest velocity magnitude observed, at 0.087 m/s, while a wider flow cross-sectional area of the annulus resulted in a significantly slower flow, with the maximum velocity at 0.023 m/s.
Figure 3.7. Simulation results of the velocity profile for sweet potato puree flowing within the annulus and b) inner tube, using the equal cross-sectional area for both inner tube and annulus product flow.
Figure 3.8. Simulation results of the velocity profile for sweet potato puree flowing within the a) annulus and b) inner tube, using the same hydraulic diameter for both inner tube and annulus product flow.
Moreover, the temperature data for the two examined cases, indicated that cooling for sweet potato puree was more efficient for the case with the product flowing within the annulus compared to the case with the product flowing in the inner tube (Fig. 3.9; Fig. 3.10). Comparing the bulk product temperature at the exit of the cooler, for the case using the same flow cross-sectional area for both flow configurations, a $T_{bulk}$ of 78.7 °C and 93.1 °C was observed for the annulus and the inner tube product flow, respectively. For the same case, increasing the cross-sectional area of both the inner tube and the annulus, and hence increasing the heat transfer cross-sectional area for both flow configurations, resulted in an increase of the cooling performance. A higher $T_{bulk}$ for both the annulus and the inner tube flow were observed for the other case of study (same hydraulic diameter) with values at 87.7 °C and 95.0 °C for the annulus and the inner tube product flow, respectively.
Figure 3.9. Simulation results of temperature distribution for cooling of sweet potato puree, flowing in a counter-current flow mode: a) within the annulus side and b) within the inner tube, using the same cross-sectional area for both inner and annulus product flow. Note the inlet product temperature at 130 °C.
Figure 3.10. Simulation results of temperature distribution for cooling of sweet potato puree, flowing in a counter-current flow mode: a) within the annulus side and b) within the inner tube, using the same hydraulic diameter for both inner and annulus product flow. Note the inlet temperature at 130 °C.
Results of sweet potato puree with an inlet temperature of 140 °C were used to compare the temperature of the fastest moving (at the center of the flow cross-sectional area for both cases) and the least cooled fluid particle across the length of the heat exchanger (‘’hot spot’’), using the same $D_h$ for both cases. The ‘’hot spot’’ is located at the center of the inner tube, and close to the inside wall of the outer tube, for the case of the inner tube flow and the annular flow, respectively. The results showed that both the ‘’hot spot’’ and the fastest moving fluid particles were cooled down faster for the case of annular flow (Fig. 3.11). Finally, the temperature distribution data indicated that the cooling for the annulus flow was more efficient, with a $T_{bulk}$ at 93.8 °C, compared with a value of 104.7 °C for the other case (Fig. 3.12).

Figure 3.11. Comparison of simulation results of temperature across the heat exchanger length for the fastest moving (blue line for annular flow, red line for inner tube flow) and the least cooled food particles (black line for annulus flow, red line for inner tube flow) for both flow configurations, during cooling of sweet potato puree, with an inlet temperature of 140 °C.
Figure 3.12. Simulation results of temperature distribution for cooling of sweet potato puree, flowing in a counter-current flow mode: a) within the annulus side and b) within the inner tube. Note the inlet temperature at 140 °C.
Improvement of cooling for sweet potato puree flowing in the annulus was a result of heat transferred from the product to the coolant, and the additional heat loss from the product to the environment. Performing energy balance calculations, the additional thermal energy lost from the product flowing in the annulus to the environment was in the range of 174-699 W (Table 3.2). Heat transfer losses to the environment increased as the temperature of the product was higher. Moreover, heat losses to the environment were higher for the co-current flow mode and were significantly higher with increased product flow velocity. The most efficient cooling of sweet potato puree was observed with the product flowing within the annular passage of a heat exchanger, with the same cross-sectional area, 0.0024 m² (using an O.D. of the inner tube at 0.0558 m (2.2 in)) for both inner tube and annular product flow.

Finally, for the case with the product flowing within the inner tube, the energy losses between 13-84 W, associated with cooling energy lost from the coolant to the environment. The heat exchanged from the environment to the coolant, has a negative impact on cooling, resulting in a slower process and probably increasing the overall operational cost.
Table 3.2. Total thermal energy transferred by the product ($Q_P$) to the coolant ($Q_W$) and to the environment ($Q_L$) for counter and co-current flow configurations, during cooling of sweet potato puree.

<table>
<thead>
<tr>
<th>$T_{inlet}$ (°C)</th>
<th>Product flow configuration</th>
<th>$u_{MAX}$ (m/s)</th>
<th>$Q_P$ (W)</th>
<th>$Q_W$ (W)</th>
<th>$Q_L$ (W)</th>
</tr>
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<td>Co-current flow mode</td>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
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<td>annulus flow</td>
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<td>9097</td>
<td>8759</td>
<td>338</td>
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<td></td>
<td>inner tube flow</td>
<td>0.087</td>
<td>7054</td>
<td>7041</td>
<td>13</td>
</tr>
<tr>
<td></td>
<td>Counter-current flow mode</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
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<td>9333</td>
<td>9159</td>
<td>174</td>
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<tr>
<td>130</td>
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<td>12311</td>
<td>11612</td>
<td>699</td>
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<tr>
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<td>inner tube flow</td>
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<td>70</td>
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<tr>
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<td>annulus flow</td>
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<td>8395</td>
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<td>11081</td>
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<td>9070</td>
<td>9069</td>
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</table>

3.3.3. Computer simulation results for cooling of applesauce

The next stage in this research was to study continuous flow cooling of applesauce, another highly viscous food product with a non-Newtonian behavior, similar to sweet potato puree. The model was used to simulate cooling between applesauce and cold water using the equal flow rate and inlet temperature conditions (120, 130, 140 °C) as for the case of sweet potato puree. The studies for applesauce were conducted to examine and compare the cooling process between the two different flow configurations: i) with the product flowing in the inner
tube and ii) with the product flowing in the annulus of the heat exchanger. The two flow configurations were examined under different cases, comparing the effects of different flow modes, different heat exchangers designs, and different ΔT between the product and the coolant by using different product inlet temperatures. Similar to the case of sweet potato puree, bulk temperature of the product at the exit of the heat exchanger was calculated for all the different simulations to define the most efficient flow configuration for the cooling process.

Cooling efficiency of applesauce was compared during counter and co-current flow modes for both examined flow configurations, using an inlet product temperature of 120 °C. Cooling of applesauce during counter-current flow mode was more efficient for both the annulus and inlet tube flow patterns. Moreover, for the annular flow mode, the T_{bulk} was at 79.9 °C for the counter-current flow, compared with a T_{bulk} of 80.5 °C for the co-current flow (Fig. 3.13). For the inner tube flow pattern, higher T_{bulk} was observed compared with other flow configuration, with a T_{bulk} of 84.1 °C and 85.7 °C, for counter and co-current flow, respectively (Fig. 3.14).
Figure 3.13. Comparison of temperature distribution simulation data for a) counter-current and b) co-current flow modes, during cooling of applesauce, flowing within the annulus, with an inlet temperature of 120 °C.
Figure 3.14. Comparison of temperature distribution simulation data for a) counter-current and b) co-current flow modes, during cooling of applesauce, flowing within the inner tube, with an inlet temperature of 120 °C.
Similar to the sweet potato studies, cooling and velocity profile of applesauce were studied for the examined flow configurations. Velocity profile and cooling of applesauce were tested under two different studies: i) having the equal cross-sectional area, 0.0024 m², (adjusting the O.D. of the inner tube at 0.0558 m (2.2 in)) for both inner tube and annulus product flow, ii) having the equal Reynolds number for both cases, using the same $D_H$ value (O.D. of inner tube at 0.0381 m (1.5 in); I.D. of inner tube 0.0762 m (3 in)) for both inner tube and annulus flow.

Using the same volumetric flow rate of 0.000063 m³/s, for all the CFD models, the velocity profile of applesauce flow was studied, using the two different flow patterns for the counter-current flow mode, while the initial temperature of the food was at 130 °C. For the case with the equal cross-sectional area, for both inner tube and annular flow, similar velocity profiles were observed. A flattened velocity profile (plug flow type velocity profile) was observed, with a $u_{\text{MAX}}$ of 0.034 m/s, for the product flowing in the area near to the center of the cross-sectional area of both inner tube and annulus flow (Fig. 3.15). For the other case, with the equal hydraulic diameter, similar flattened velocity profile was observed for the annulus product flow, with $u_{\text{MAX}}$ of 0.022 m/s, while a parabolic velocity profile was observed for the product flowing within the inner tube, with a $u_{\text{MAX}}$ of 0.078 m/s (Fig. 3.16).
Figure 3.15. Simulation results of the velocity profile for applesauce flowing within the annulus and b) inner tube, using the same cross-sectional area for both inner and annular product flow.
Figure 3.16. Simulation results of the velocity profile for sweet potato puree flowing within the a) annulus and b) inner tube, using the same hydraulic diameter for both inner and annular product flow.
Furthermore, temperature data for the two examined cases indicated that cooling for applesauce was slightly more efficient for the case with the product flowing within the annulus compared to the case with the product flowing in the inner tube. Comparing the bulk product temperature at the exit of the cooler, for the case using the same flow cross-sectional area for both flow configurations, a $T_{\text{bulk}}$ of 84.2 °C and 91.4 °C was observed for the annulus and the inner tube product flow, respectively. A slightly higher $T_{\text{bulk}}$ for both the annulus and the inner tube flow were observed for the other case of study (equal hydraulic diameter) with values at 84.6 °C and 91.4 °C for the annular and the inner tube product flow, respectively (Fig. 3.17; Fig. 3.18).
Figure 3.17. Simulation results of temperature distribution for cooling of applesauce, flowing in a counter-current flow mode: a) within the annulus side and b) within the inner tube, using the same cross-sectional area for both inner and annular product flow. Note the inlet temperature at 130 °C.
Figure 3.18. Simulation results of temperature distribution for cooling of applesauce, flowing in a counter-current flow mode: a) within the annulus side and b) within the inner tube, using the equal hydraulic diameter for both inner and annular product flow. Note the inlet temperature at 130 °C.
Similar to the case of sweet potato puree, simulation models of applesauce with an inlet temperature of 140 °C were used to compare the temperature of the fastest moving fluid particle and the “hot spot” across the length of the heat exchanger. The temperature data showed that both the “hot spot” and the fastest moving fluid particles of applesauce were cooled down faster for the case of annular flow, with a bulk temperature of food at the exit of the heat exchanger at 91.9 °C, compared with a 99.4 °C for the case of the inner tube flow (Fig. 3.19; Fig. 3.20).

Figure 3.19. Comparison of simulation results of temperature across the heat exchanger length for the fastest moving (blue line for annular flow, red line for inner tube flow) and the least cooled food particles (black line for annulus flow, red line for inner tube flow) for both flow configurations, during cooling of applesauce, with an inlet temperature of 140 °C.
Figure 3.20. Simulation results of temperature distribution for cooling of applesauce, flowing in a counter-current flow mode: a) within the annulus side and b) within the inner tube. Note the inlet temperature at 140 °C.
The improvement of cooling for the annular flow of applesauce was a result of the heat transferred from the product to the coolant and the losses to the environment. Performing energy balance calculations, the additional energy lost from the product flowing in the annulus to the environment was in the range of 91-369 W (Table 3.3), significantly lower compared to the sweet potato puree results. Heat transfer losses to the environment were higher for co-current flow mode and increased as the intel temperature of the product increased and with increased product flow velocity. For the case of the applesauce, the design characteristics of the heat exchanger (cross-sectional area or D_H) did not have the same impact compared to sweet potato puree cooling. This result is probably due to the plug flow velocity type exhibited by applesauce, minimizing effects from the different examined flow patterns. Finally, similar to sweet potato puree studies, for the case with the product flowing within the inner tube, the values thermal energy lost to the environment were in the range 36-182 W, indicating the cooling energy lost from the coolant to the environment, having a negative impact on operational efficiency (heat transfer, cost).
Table 3.3. Total thermal energy transferred by the product \((Q_P)\) to the coolant \((Q_W)\) and to the environment \((Q_L)\) for counter and co-current flow configurations, during cooling of applesauce.

<table>
<thead>
<tr>
<th>T_{inlet} (°C)</th>
<th>Product flow configuration</th>
<th>(u_{MAX} ) (m/s)</th>
<th>(Q_P ) (W)</th>
<th>(Q_W ) (W)</th>
<th>(Q_L ) (W)</th>
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</tr>
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<td>9641</td>
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</tr>
</tbody>
</table>

3.4. Conclusions

In this study, a computer model was developed using *Multiphysics* software system Comsol 5.2, in order to study continuous flow cooling of viscous food products, such as applesauce and sweet potato puree. Using Computational Fluid Dynamics (CFD) and heat transfer software modules, computer simulation modeling was used to compare two different flow configurations within the same tube in tube heat exchanger: i) cooling process of the food, while flowing within the internal tube cylinder of the heat exchanger and ii) cooling process
of the food, while flowing within the annular passage of the heat exchanger. The model was used to estimate and compare the product bulk temperature ($T_{\text{bulk}}$) at the exit of the heat exchanger, temperature distribution, and velocity profile of the product, during continuous flow cooling studies for both examined flow configurations.

Results of the set of simulations indicate that under an identical set of flow rates, surrounding and initial temperature conditions (120 °C, 130 °C and 140 °C for the process material, 4 °C for the coolant) the flow configurations in which the process material flowing within the annular passage of the tube in tube heat exchanger, in a counter-current flow mode with the coolant, would theoretically result in a $T_{\text{bulk}}$ at the outlet of the heat exchanger of a temperature range of around 80-95 °C, for both foods tested. When the process material is flowing within the internal tube of the heat exchanger, the $T_{\text{bulk}}$ was in the temperature range of 91-102 °C for sweet potato puree and 85-99 °C for applesauce.

The improvement on cooling for the tested foods flowing in the annulus was a result of the heat lost from the product to the environment. Performing energy balance calculations, the additional energy lost from the product flowing in the annulus to the environment was in the range of 174-699 W and 91-369 W, for sweet potato puree and applesauce, respectively. For both examined foods, heat transfer losses to the environment were higher for co-current flow mode and increased as the inlet temperature of the product was higher and with increased product flow velocity. The most efficient cooling of sweet potato puree was observed with the product flowing within the annular passage of a heat exchanger, with the equal cross-sectional area (the outside diameter (O.D.) of the inner tube at 0.0558 m (2.2 in)) for both inner tube and annular product flow, while for the case of the applesauce, the design characteristics of the
heat exchanger (using the same cross-sectional area or hydraulic diameter) did not have the same impact.

Furthermore, the energy balance calculation data for the case with the product flowing within the inner tube presented energy losses from the coolant to the environment, in the range of 13-182 W, for both foods tested. The heat losses from the coolant to the environment, have a negative impact on the process, reducing the cooling efficiency and probably increasing the overall operational cost.

Comparing the temperature and velocity data for both applesauce and sweet potato puree, more efficient cooling was observed during cooling of sweet potato puree flowing in the annular flow pattern, presenting a parabolic velocity flow profile type. While less efficient cooling was observed with applesauce flowing in the annulus, with a slower flattened velocity profile (plug flow type velocity profile). The slower and flattened velocity profile of applesauce resulted in a less efficient cooling, compared with sweet potato puree, due to the lower product heat loss to the environment.

In conclusion, based on the results of this study cooling of highly viscous materials, with the product flowing within the annulus of the heat exchanger could yield potential benefits, of enhancement of conventional continuous flow cooling, while maintaining the sanitation conditions in the processing plant. Therefore, in this case, the potential improvement in the cooling efficiency resulting from the movement of process material flow from the internal tube into the external annulus could potentially be 14% and 23%, for applesauce and sweet potato puree, respectively. However, before industrial application of this cooling method, additional experiments and computational studies need to be conducted for better understanding and solving the problem of the static or "dead" spots observed in the transition
area of the product flow to the annular passage, regarding the potential cleaning and sanitation difficulties.

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Nomenclature

*Latin letters*

- **3D** Three-dimensional
- **A** Surface area of the heat exchanger (m²)
- **CFD** Computational Fluid Dynamics
- **cₚ** Specific heat (J/kg·K)
- **D_H** Hydraulic diameter (m)
- **F** External forces (N)
- **h** Convection heat transfer coefficient (W/m²K)
- **I.D.** Inside diameter (m)
- **K** Fluid consistency coefficient (Pa·sⁿ)
- **k** Thermal conductivity (W/m·K)
- **k** Turbulent kinetic energy (m²/s²)
- **L** Total length of the heat exchanger (m)
- **MW** Microwave system
n Flow behavior index (dimensionless)
\(N_{Re}\) Reynolds number (dimensionless)
\(N_{GRe_b}\) Generalized Reynolds number for non-Newtonian fluids
O.D. Outside diameter (m)
p Pressure (Pa)
P Time average pressure (Pa)
\(P_A\) Absolute pressure (Pa)
Q External heat sources (W/ m³)
QL Heat losses to the environment (W)
\(Q_p\) Energy lost by the product (W)
\(Q_w\) Energy gained by the coolant (W)
\(\dot{Q}\) Convective heat transfer (W)
\(\rightarrow q\) Heat flux (W/m²)
r Function of radius (m)
R Radius (m)
RANS Reynolds-Averaged Navier-Stokes equations
T Temperature (°C)
\(T_{bulk}\) Bulk temperature (°C)
\(T_{inlet}\) Inlet temperature (°C)
\(T_{ext}\) External temperature (air room temperature (°C))
\(u\) Velocity (m/s)
U Time average velocity (m/s)
\(u_{MAX}\) Maximum product velocity (m/s)
\_ \quad \text{Average velocity (m/s)}

\textit{Greek letters}

\Delta T \quad \text{Temperature difference (°C)}

\epsilon \quad \text{Turbulent dissipation (m}^2/\text{s}^3\text{)}

\mu \quad \text{Newtonian viscosity (Pa/s)}

\mu_T \quad \text{Turbulent viscosity (Pa/s)}

\rho \quad \text{Density (kg/m}^3\text{)}

\sigma \quad \text{Shear stress (Pa)}

\tau \quad \text{Shear stress (Pa)}

\textit{Subscripts}

T \quad \text{refers to turbulent flow}

\text{inlet} \quad \text{refers to the initial temperature at the inlet of the heat exchanger}

\text{bulk} \quad \text{refers to the bulk temperature of the food at the exit of the heat exchanger}

L \quad \text{refers to the heat loss to the environment}

P \quad \text{refers to the energy lost by the examined product}

W \quad \text{refers to the energy gained by the coolant}
References


CHAPTER 4:
Enhancement of Continuous Flow Cooling Using Hydrophobic Surface Treatment

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ABSTRACT

This work studied the effect of hydrophobic surface treatment on cooling (97-57 °C) and thermal mixing via vibration of viscous foods, namely, sweet potato puree, banana puree and cheese sauce. Cooling performance and temperature uniformity were compared between two identical straight horizontal tube-in-tube stainless steel heat exchangers, where one was untreated and one had food contact surfaces treated with a hydrophobic chemical treatment. With the treated heat exchanger, a visible minimization of product accumulation at the pipe wall was observed for all products tested. A lower average product outlet temperature ranging from 4 to 6 °C was observed for banana puree and cheese sauce with the treated heat exchanger, compared to the untreated one. In contrast, for sweet potato puree, a higher average product outlet temperature (by 2-5 °C) was observed for the treated heat exchanger compared to the untreated case. More uniform cross-sectional temperature distribution was recorded for the treated heat exchanger for all tested food materials. Application of vibration at the resonance frequency of the mixing unit (20 Hz), significantly improved the temperature uniformity within sweet potato puree and banana puree, reducing the maximum cross-sectional temperature difference within the food to 2-5 °C. However, no significant change has been observed for the temperature distribution within cheese sauce.

Key words: Hydrophobic surface treatment, cooling, heat transfer efficiency, thermal mixing, viscous foods
4.1. Introduction

Advanced heating technologies, such as continuous flow microwave, ohmic heating and radio frequency heating systems, have enabled food processors to minimize product quality degradation during continuous thermal processing of viscous and multiphase food products, such as typical dairy products, fruit and vegetable purees and soups (Coronel et al., 2005; Steed et al., 2008; Cullen et al., 2012). Unfortunately, the cooling stage of thermal processing of viscous products is still implemented using inefficient conventional cooling methods. During conventional continuous flow cooling of viscous foods, laminar flow, and low thermal conductivity, characteristic for these materials, lead to a wide temperature distribution within the product, resulting in a non-uniform, slow cooling process and degradation of final food quality. Recent studies proposed radial thermal mixing (cross-sectional temperature equalization), as an efficient method to enhance heat transfer and the overall continuous flow cooling process of highly viscous food products (Metcalfe and Lester, 2009; Stoforos et al., 2016). However, these studies on thermal mixing during the cooling stage of viscous foods, such as banana puree and cheese sauces, reported the formation, deposition and accumulation of gel and other food compounds on the surface of the pipe wall (Stoforos et al., 2016). The formation and build-up of this non-slip thick layer of low thermal conductivity material on the product-contact surfaces of thermal processing system components (heat exchangers, mixing units, hold tubes) has a very negative impact on heat transfer and thermal mixing efficiency, leading to the need for extension of system components and associated degradation of final food product quality (Stoforos et al., 2016).

During continuous flow thermal processing of foods and biomaterials, deposition and accumulation of “unwanted” food materials, such as proteins and minerals, especially in the
form of fouling, on the surface of processing equipment, has been a major challenge for food and dairy industries (Swartzel, 1983; Sandu and Singh, 1991; Balasubramanian and Puri, 2009; Kananeh et al., 2010; Mérian and Goddard, 2012; Murugesan and Balasubramanian, 2013; Barish and Goddard, 2013). Formation of fouling layers on processing equipment reduces the operating efficiency by increasing resistance to heat transfer and pressure drop across the heat exchanger (Swartzel, 1983; Balasubramanian and Puri, 2009; Awad, 2011). Moreover, fouling increases the likelihood of biofilm formation, resulting in additional operating expenses, corresponding to frequent food processing plant shutdowns for cleaning and extensive use of chemical detergents and sanitizers (Sandu and Singh, 1991; Kananeh et al., 2010; Mérian and Goddard, 2012; Barish and Goddard, 2014). Controlling the deposition and adhesion of food components on the surface of food processing equipment, like stainless steel heat exchangers, depends on different parameters such as operating conditions (temperature, flow rate), food composition/chemistry and processing surface properties (material, wettability, topography/surface roughness) (Santos et al., 2004; Mérian and Goddard, 2012). Recently, surface modification has been a subject of research for many studies focusing on controlling, minimizing or elimination of fouling and biofouling in food and dairy processing.

Surface modification studies have been focusing on mimicking the properties of natural anti-fouling materials, such as the lotus leaf (*Nelumbo nucifera*). The superhydrophobic behavior, in combination with the roughness of the nanostructure of the lotus leaf, is associated with unique self-cleaning and water-repelling properties, known as the lotus effect (Latthe et al., 2014). Furthermore, novel surface modification techniques, such as polymer and nanoparticle coatings (silica or fluorocarbon compositions), physicochemical surface modification, plasma-enhanced chemical vapor deposition, electro-chemical and ion
implantations have enabled researchers to successfully lower the free surface energy (or surface tension) of the food contact surfaces by altering wettability (hydrophobic and hydrophilic) and roughness characteristics. In food and in particular dairy processing research areas, antifouling methods have been applied on the surface of stainless steel plate heat exchangers, resulting in significant reduction of the amount of product deposition and fouling formation, for foods such as milk, skim milk and tomato juice, improving the operation and cleaning efficiency of the heat exchangers, while reducing the bacterial adhesion (Beuf et al., 2003; Santos et al., 2004; Rosmaninho et al., 2007; Premathilaka et al., 2006; Balasubramanian and Puri, 2009; Kananeh et al., 2010; Mérian and Goddard, 2012; Barish and Goddard, 2013; Cheng et al., 2015). Using commercially available silica or fluorocarbon based nanoparticle coatings, with the most commonly used technique of electroless nickel plating with embedded polytetrafluoroethylene nanoparticles, food process surfaces were successfully modified by lowering the surface energy via increasing the hydrophobicity and adjusting the topography or roughness of the surface (Kananeh et al., 2010; Balasubramanian and Puri, 2008; Balasubramanian and Puri, 2009). Furthermore, non-wetting surfaces with hydrophobic or superhydrophobic properties tend to promote slip-flow boundary conditions (in micro and nano-scale), which result in pressure drop and drag reduction during viscous laminar and increase of heat transfer coefficient (Watanabe and Udagawa, 2001; Kunugia et al., 2004; Choi and Kim, 2011; Srinivasan et al., 2013; Pastrello and Bonanno, 2015).

The main objective of this research was to enhance and improve the continuous flow cooling process of viscous and poorly conductive foods, minimizing the cooling-related quality losses of the final food product and potentially improving the sanitation conditions while preparing the foundation for future applications and development of advanced volumetric
cooling technologies. For these reasons, modification of the surface of a stainless steel concentric tubular heat exchanger was examined in this study by establishing a non-wetting behavior (hydrophobic, super-hydrophobic) as a method to minimize product deposition and accumulation at the pipe wall, anticipating the reduction of thickness and thermal resistance of the layer and therefore enhancement of cooling process, reduction of the pressure drop along the heat exchanger and a potential improvement of the sanitation conditions during processing of viscous and poorly conductive viscous foods. This is particularly relevant to the cooling stage of the process where the product in contact with the heat exchange surfaces typically has thermal conductivity minimized due to low temperatures and non-slip layer thickens maximized due to deposition of high viscosity (also maximized at low temperatures) material.

4.2. Materials and Methods

4.2.1. Test materials

For this series of experiments, different highly viscous food products, such as sweet potato puree (Yamco LLC, Snow Hill, NC, USA), banana puree (Aseptia/Wright Foods, Troy, NC, USA) and cheese sauce (Advanced Food Products, New Holland, PA, USA) were used as test materials. These materials were chosen because they show very good results during applications of advanced heating technologies, such as MW heating, while product deposition and accumulation at the pipe wall during the cooling stage have been reported for most of the tested products (Coronel et al., 2005; Steed et al., 2008; Stoforos et al., 2016). Approximately 8-10 gallons of food product samples were sufficient to fill the system and allow for recirculation of the test samples at flow rates of $5.8 \times 10^{-5}$ m$^3$/s and $6.3 \times 10^{-5}$ m$^3$/s ($\approx$1 gal/min). Water was used as the cooling medium, flowing within the inner tube of the heat
exchangers in a counter current flow configuration, with an inlet temperature between 13-24 °C and flow rates of $56.8 \times 10^{-5}$ m$^3$/s (9 gal/min) and $63.1 \times 10^{-5}$ m$^3$/s (10 gal/min).

4.2.2. Experimental methods

The effects of hydrophobic surface treatment on continuous flow cooling of highly viscous products were examined by comparing the efficiency of cooling between the untreated stainless steel heat exchanger against the performance of the heat exchanger where the food contact surfaces have been treated with a hydrophobic chemical treatment. To create the hydrophobic behavior on the surface of the treated heat exchanger, a non-toxic (but not food grade) hydrophobic chemical solution, Aculon (Aculon performance Surface Solutions, San Diego, CA) was applied. To study and understand the effects of hydrophobic surface modification on continuous flow cooling of viscous foods, two different series of experiments were conducted. The first series of experiments studied the surface properties, such as water and oil contact angles on the untreated and Aculon-treated, stainless steel (300 series) surfaces, followed by more comprehensive studies of continuous flow cooling with the untreated and treated heat exchangers, respectively.

4.2.3. Hydrophobic chemical surface treatment

Hydrophobic chemical treatment with Aculon was used as the surface modification technique. Aculon was chosen as the nanoscale hydrophobic surface modification treatment, due to its chemical properties as a multi-surface (including stainless steel, glass…) chemical solution stable at very high operating temperature conditions, up to 250 °C, with a uniform 20-100 nm thick surface treatment. The following steps were implemented for all the applications
of hydrophobic surface treatments. The first step of treatment was cleaning all of the tested stainless steel parts, to where they were visually clean and free of contaminants. The next step was the application of Aculon solution, by using a wet cotton cloth with the solution and then rubbing the treated surface, followed by wiping the surface with a clean cloth. The final step of hydrophobic treatment was drying the surfaces with dry air (at room temperature) until the surface was completely dry. Aculon chemical treatment was used for the treatment method for all the samples prepared for the contact angle measurements and food contact surfaces of the tube in tube heat exchanger, during the continuous flow cooling studies.

4.2.4. Contact angle measurements

The first case study of this research was to quantify the differences between the surface properties of untreated against hydrophobic chemical treated stainless steel samples. Stainless steel (300 series) 2 inches diameter tri-clamps end caps were used as the tested surfaces for the contact angle measurements, for both the untreated and Aculon-treated samples. Measurement of the equilibrium contact angle between the perimeter of a liquid drop and the contacting surface is a common method for evaluation of surface wettability, where the non-wetting surfaces are associated with hydrophobic and superhydrophobic (i.e. non-wetting surfaces where the surface tends to repel water) surfaces with a contact angle between 90-150° and higher than 150°, respectively (Korhonen et al., 2013). The equilibrium static contact angles of water and oil (n-dodecane) on untreated and chemically treated stainless steel sample surfaces were measured at room temperature (21 °C), by software-controlled measurement Sessile drop testing method, using laboratory instrumentation goniometer OCA 15 (Contact Angle Measurement Instrument, Future Scientific Corp, Garden City, NY, U.S.A.) and video-
based control software SCA 20 (Contact Angle Measurement Instrument, Future Scientific Corp, Garden City, NY, U.S.A.). Using a 500 microliter gastight syringe (Hamilton Company, Reno, NV, U.S.A), controlled 5 μL volume droplets of water and oil were formed in different points (3-5 points) across the tested surfaces to measure the contact angle and determine the homogeneity of the treated surfaces, via video image analysis using the computer software (Fig. 4.1).

Figure 4.1. Picture showing the video-based goniometer instrument OCA 15 and the micro-syringe SD-DM single direct dosing system, during contact angle measurements of 5 μL volume droplets of water in different locations across stainless steel 2 inches tri-clamps end caps.
4.2.5. Continuous flow cooling: experimental set up

The second study presented in this paper was focused on the effects of hydrophobic surface treatment during continuous flow cooling of viscous foods such as sweet potato puree, banana puree, and cheese sauce. To study the effects of the hydrophobic surface treatment during continuous flow cooling process of viscous food products, a recirculating complete thermal processing system was assembled, consisting of three basic sections: heating, cooling, and mixing. The thermal processing system consisted of a food grade progressive cavity pump Seepex MD-012 12 (Fluid Engineering Inc., Birmingham, AL, USA), a continuous flow modular microwave heating system consisting of a total of 13 bench-top Panasonic Inverter MW ovens, with a maximum power input of 16.350 kW, at the MW frequency of 2450 MHz, which have been used for the heating stage of the experimental trials.

Cooling section consisted of two stainless steel (300 series) horizontal concentric tube in tube heat exchangers, one untreated and one with the food contact surfaces treated with a hydrophobic chemical (Aculon), used separately and independently for different series of experiments, identical in dimensions with a 0.051 m (2 inches) and 0.076 m (3 inches) inner and outer tube diameter, respectively, and a total length of 1.83 m each. For both cases, the annulus side of the heat exchangers was used as the flow passage for the tested food product. Computer simulation studies have indicated that having the food flowing within the annulus, cooling efficiency would be improved, by increasing the heat losses to the environment, beneficial for cooling, while improving the sanitation conditions via elimination of the sweating pipe phenomenon (Stoforos and Simunovic, 2016). Moreover, for this study, using the annulus side of the heat exchanger as the flow passage for the tested material has additionally provided an easier access to food contact surface for the application of
hydrophobic surface treatments. To create the hydrophobic behavior on food contact surfaces of the heat exchanger, Aculon was applied on the inner and outer surfaces, of the outer tube and inner tube, respectively. Finally, at the exit of cooling section, the recirculating test system was closed with the mixing unit, which was used to equalize the cross-sectional cooling-induced temperature profile distributions perpendicular to the flow direction.

Apparatus and configuration of the mixing unit, described in a previously published continuous flow thermal mixing study (Stoforos et al., 2016) have been used here as well. Mixing unit flow-through surfaces remained untreated, during the studies for both the untreated and the treated heat exchanger. Moreover, the mixing unit, consisted of a 180° bend tube (formed from two silicone rubber-lined, reinforced flexible tube segments, each 0.53m long), and a stainless steel 180° elbow, mounted at the top surface of a low sonic frequency tactile audio transducer Buttkicker LFE (The Guitammer Company, Westerville, OH, USA). Frequency and duration of vibration for the tactile audio transducer were controlled through a computer frequency generator program, FreqGen 1.13 (Digital River Inc., Minnetonka, MN, USA), while the amplitude of vibration was achieved via a power amplifier type BKA1000-N (The Guitammer Company, Westerville, OH, USA). An accelerometer data logger device - SlamStickTM (Mide Inc., Medford, MA, USA) and a computer program Slamstick viewer (Mide Inc., Medford, MA, USA), were used to measure, record, review and process the vibration parameters (frequency, duration and amplitude of vibration), respectively.

T-type thermocouples were located at multiple positions through the MW heating system, cooling system and at the entrance and exit of the mixing unit. A specially customized T-type thermocouple probes - Keyhole Multipoint probes (Windridge Sensors LLC, Holly Springs, NC), were used to measure the temperature profiles of test materials, each with 3
different radially positioned temperature sensing points, one triple-point probe at the inlet and one at the outlet of each tube in tube heat exchanger. Furthermore, identical multipoint thermocouple probes as located at the outlets of the heat exchangers were also used to measure the cross sectional temperature profiles of the products flowing through the inlet and outlet of the mixing unit. The three sensing points of these probes were located at the center of the pipe, close to the pipe wall and at an intermediate point equidistant between the pipe wall and pipe center sensing points. Time-temperature data were recorded once per second using a 32-Channel temperature data acquisition system (IOTEch TempScan 1100, IOtech, Inc. 25971 Cannon Road, Cleveland, OH 44146-1833) (Fig. 4.2).
Figure 4.2. a) Schematic diagram and pictures of the closed-loop, recirculating experimental system for continuous flow thermal processing, consisted from b) MW-Heating Section, c) Cooling Section and d) Mixing Section.
4.2.6. Continuous flow cooling: experimental methods

For all the tested food materials, two different experiments were conducted, initially by using as cooler the untreated stainless steel tubular heat exchanger, followed by the second experiment with the Aculon-treated heat exchanger, under identical operating conditions, such as inlet temperature and flow rate. Moreover, by using the modular MW heating system all the products heated up to the same temperature range, with an average mean temperature entering the coolers at approximately 90-97 °C, which used as the initial inlet temperature for all the cooling experiments. Once the initial target temperature was reached, cooling studies were started while recirculating the product throughout the system and continue operating only 6 MW module units, to achieve and maintain steady state conditions (approximately constant inlet temperature) for a period of 200-450 s.

Furthermore, during cooling studies, thermal mixing and equalization of cross-sectional temperature at the exit of the cooler, were studied by employing acoustic/mechanical vibrations with a duration of 120 s at a range of frequencies close to the resonance frequency of the mixing system and at the maximum volume level of the used audio amplifier (Stoforos et al., 2016). The resonance frequency of the thermal mixing system was determined by following the methodology of previous studies (Stofros et al., 2016), through scanning acoustic frequencies in the low sonic range of 10–120 Hz and by applying 10s of vibration at the tested frequency, and the same fixed volume level of amplifier, followed by 5 s of silence (0 Hz), with incrementally increasing frequency steps of 10 Hz. SlamStick™ accelerometer was used to record the data (acceleration and frequency) of vibration and determine the resonance frequency, as the highest magnitude of acceleration via the one-dimension fast Fourier
transformation (1-D FFT) plot, obtained using the Slamstick viewer computer software (Stoforos et al., 2016).

Continuous flow cooling studies were not conducted for temperatures below 60 °C, where more uniform temperature distribution within the product is established and less product quality degradation occurs. The completion of the cooling trials was immediately followed by cleaning, via washing-down the system (modular MW heating, cooling and mixing sections) with hot water of approx. 60 °C at a flow rate of $6.3 \times 10^{-5} \text{m}^3/\text{s}$ (10 gal/min), for 300-1200s, depending on the tested food product. At the end of wash-down photos of both the untreated and treated heat exchanges, were taken for comparison of the efficiency of cleaning performance.

4.3. Results and discussion

4.3.1. Contact angle measurements

The first set of experiments for this study was to determine the differences in surface properties between the untreated and the Aculon chemically treated stainless steel surfaces. The contact angle (CA) was used as the parameter to define the differences between the untreated and the treated stainless steel surfaces, as a measure to quantify the wettability properties of the surface. Using the video analysis-based OCA 15 goniometer, the results on sessile drop static water contact angle at different points on the tested surfaces showed a homogeneous behavior for all the different samples, with the water contact angle with the Aculon treated stainless steel samples to be at 111°, while for the untreated stainless steel samples to be at 93°. The results of the contact angle measurements with water indicated that the stainless steel samples treated with Aculon presented a hydrophobic behavior, which
amplifies the non-wetting properties of the stainless steel, something that was clear visually, based on the size of the water droplets on the surface of treated and untreated stainless steel surface, respectively (Fig. 4.3).

On the other hand, sessile static contact angle measurements with oil (n-dodecane) showed an oleophilic behavior, with the contact angles of approximately 0° and 19°, for the chemically treated and untreated stainless steel samples, respectively. Oleophilic behavior for all the samples was easily visible through the video-analysis of the goniometer, where no droplets of oil were formed on the surface of the samples, with the liquid sample to spread across the stainless steel surface (Fig. 4.4).

Based on the contact angle measurements, surface modification with Aculon increased the hydrophobic behavior of the stainless steel samples while no significant changes were observed regarding the behavior of stainless steel samples against oil liquid samples, with the stainless steel samples maintaining a highly oleophilic behavior with and without chemical treatment.
Figure 4.3. Picture of sessile static contact angle measurements with water: a) Aculon treated stainless steel surface and b) Untreated stainless steel surface.

Figure 4.4. Picture of sessile static contact angle measurements with oil: a) Aculon treated stainless steel surface and b) Untreated stainless steel surface.
4.3.2. Resonance Frequency

The second part of this research studied cooling and thermal mixing performance during experiments with highly viscous foods flowing within the annular passage of the examined heat exchangers. The first set of tests was to determine the resonance frequency, the frequency with the highest magnitude of the acceleration (g) of vibration. Analyzing the collected data of acceleration, frequency (Hz), and time (s), from the accelerometer, similar results were observed for all the tested products, with the resonance frequency for the mixing system at approximately 20 Hz, as can be observed from the following acceleration data for cheese sauce (Fig. 4.5). The frequency with the highest magnitude of acceleration at 20 Hz, was used as the tested frequency during the studies for all the tested foods.

Figure 4.5. Effect of acoustic frequency on the acceleration (expressed as unit Gs) magnitude of vibration at different tested frequencies as determined from 1-D FFT plot. Not the different colors indicate the different axis, x (red), y (green) and z (blue).
4.3.3. Continuous flow cooling studies

To study the cooling and thermal mixing performance, time-temperature profiles were plotted; presenting the time-temperature plot of the flowing product, at the inlet and outlet of the cooling and mixing unit. In all cases, the outlet of the cooling and the inlet of mixing unit referred to the same measurement point within the thermal processing system. Time-temperature profiles at the inlet and the outlet of heat exchangers and the mixing unit, at 3 different cross-sectional points within the pipe, (center, wall and an intermediate point), were recorded to compare and present the differences in cooling and thermal mixing efficiency between the untreated and the chemically surface-treated heat exchangers. The maximum temperature difference between the 3 cross-sectional points was used as the measure of uniformity of temperature distribution within the tested product at the exit of the heat exchangers (ΔT_{MAX}) and at the exit of mixing unit (ΔT_{MAX,M}).

Finally, the effects on the cooling efficiency of untreated and hydrophobic surface-treated heat exchangers were examined by comparing temperature differences (ΔT_{HX}) between the average temperatures at the inlet of the heat exchanger against the average temperature at the outlet of the mixing unit. The average temperature at the outlet of the mixing unit was chosen as the more representative of the outlet cooling results because no extra active cooling occurs within the mixing section. Additionally, due to the thermal mixing in the unit, the average temperature at its outlet is more representative of the bulk temperature of the product. Finally, flow rate and pictures of the internal heat exchanger tube outer surfaces after experimental runs and washing-down process of the tested foods were compared to understand the effects of hydrophobic surface treatment on the cooling process.
4.3.4. *Sweet potato puree*

Studies on cooling and thermal mixing were conducted during the cooling stage of sweet potato puree processing, within the temperature range of 97-57 °C. Similar experimental conditions, under the steady state, were used, with sweet potato puree flowing within the annulus with an inlet temperature in the range of ~90-97 °C, and water flowing in counter-current mode, with a flow rate of $63.1 \times 10^{-5} \text{ m}^3/\text{s}$ and with an inlet temperature of 13 °C. The first step for all the various studies was to measure and compare the flow rate of the product while operating the pump at a fixed speed. A slightly higher flow rate of $6.3 \times 10^{-5} \text{ m}^3/\text{s}$ was measured for the treated heat exchanger, comparing with the untreated case where the flow rate was at $5.8 \times 10^{-5} \text{ m}^3/\text{s}$. For that reason, for the study case with the treated heat exchangers, two different experiments were conducted, the first at higher flow rate of $6.3 \times 10^{-5} \text{ m}^3/\text{s}$ and with application of vibration for thermal mixing studies and the second experiment with a flow rate of at $5.8 \times 10^{-5} \text{ m}^3/\text{s}$, (flow rate closely similar to the untreated case) without employing vibration for mixing, for better understanding of cooling and thermal mixing studies.

Comparing the time-temperature profiles between the results during cooling of sweet potato puree, better performance was observed for the case of the untreated stainless steel heat exchanger, with the $\Delta T_{\text{HX}}$ at 16-22 °C. Under the equal flow rates, slightly lower $\Delta T_{\text{HX}}$ at 16-20 °C was observed, for the case of hydrophobic surface treated heat exchanger, while significantly lower cooling performance was reported for the case of the treated heat exchanger operating at higher flow rate, with the $\Delta T_{\text{HX}}$ to be in the range of 10-13 °C (Fig. 4.6). Temperature distribution within sweet potato puree flow cross section was significantly more uniform during the cooling process with the treated heat exchanger, for all the tested flow rates. Moreover, for the study of sweet potato puree with the untreated heat exchanger, a $\Delta T_{\text{MAX}}$ of
13-17 °C was reported, at the exit of the cooler, while at the exit of mixing unit the $\Delta T_{\text{MAX, M}}$ was reduced to 5 °C (Fig. 4.7). For the treated heat exchanger, at the exit of the cooler the $\Delta T_{\text{MAX}}$ was at 7-9 °C and 5-6 °C, for higher and lower flow rates, respectively. Furthermore, by employing vibration, $\Delta T_{\text{MAX, M}}$ was significantly improved, reducing the maximum temperature difference to 3-5 °C, while no significant difference on the $\Delta T_{\text{MAX, M}}$ value was observed the for the other case (Fig. 4.8; Fig. 4.9).

Finally, the end of cooling experiments was followed by washing-down the thermal processing system, with hot water for a period of 1200 s. Comparing the photographs of the internal heat exchanger tube surfaces, that were taken at the end of wash-down process and after disassembly the system, significantly more efficient cleaning was observed for the case of the treated heat exchanger. A residual amount of product deposition of sweet potato puree product was still visible on the top surface of the inner tube of the heat exchanger (Fig. 4.10).
Figure 4.6. Comparison of $\Delta T_{HX}$ during cooling of sweet potato puree, for the untreated and the treated heat exchanger while (a) the product flowing at different flow rate of $5.8 \times 10^{-5}$ m$^3$/s and $6.3 \times 10^{-5}$ m$^3$/s, for the untreated and the treated heat exchanger, respectively; (b) the product flowing at the same flow rate of $5.8 \times 10^{-5}$ m$^3$/s for both cases.
Figure 4.7. Comparison of the temperature distribution within the product (at center, wall and an intermediate point) at the inlet (a) and the outlet (b) of the mixing unit for sweet potato puree after cooling in the untreated heat exchanger for a given product average temperature at the inlet of the heat exchanger. The vertical dashed lines represent the beginning and the vertical solid lines the end of vibration of 120 s at 20 Hz. Note that the average time for the fluid particle to move from the inlet to the outlet of the heat exchanger and mixing unit was 68s and 28 s, respectively.
Figure 4.8. Comparison of the temperature distribution within the product (at center, wall and an intermediate point) at the inlet (a) and the outlet (b) of the mixing unit for sweet potato puree after cooling in the treated heat exchanger for a given product average temperature at the inlet of the heat exchanger. Note that the average time for the fluid particle to move from the inlet to the outlet of the heat exchanger and mixing unit was 60 s and 24 s, respectively.
Figure 4.9. Time-temperature profiles for sweet potato puree, flowing in the annulus passage of the treated heat exchanger; comparing the average temperature at the inlet of the heat exchanger with (a) the inlet and (b) the outlet of the mixing unit, at 3 different points of the pipe: at the center, at the wall and at an intermediate point. Note that the average time for the fluid particle to move from the inlet to the outlet of the heat exchanger and mixing unit was 68s and 28 s, respectively.
Figure 4.10. Comparison of a) treated with hydrophobic chemical solution and b) untreated internal heat exchanger tube outer surfaces after cooling of sweet potato puree. Picture shows the disassembled tubes after washed-down with hot water.

4.3.5. Banana puree

Cooling and thermal mixing studies were conducted, during the cooling process of banana puree, within the temperature range of 97-75 °C, and by employing vibration of 120 s duration at 20 Hz, for mixing. Steady state experimental conditions were used, with banana puree flowing within the annulus with an inlet temperature in the range of ~90-97 °C, and water flowing in counter-current mode, with a flow rate of $63.1 \times 10^{-5} \text{ m}^3/\text{s}$ and with an inlet temperature of 18 °C.

Comparing the time-temperature profile between the results with cooling of banana puree, during the steady state period (for ~280 s), cooling processing using the treated heat exchanger was more efficient, with a $\Delta T_{\text{HX}}$ of 8-11 °C, while a $\Delta T_{\text{HX}}$ of 4-6 °C was observed,
for the untreated heat exchanger (Fig. 4.11). Temperature distribution within the product was significantly more uniform during cooling with the treated heat exchanger. For the study of banana puree using the untreated heat exchanger, a $\Delta T_{\text{MAX}}$ at 8-10 °C was observed at the exit of the cooler. With application of vibration, $\Delta T_{\text{MAX,M}}$ was reduced to 4-6 °C (Fig. 4.12). More uniform temperature distribution within banana puree was reported during processing with the treated heat exchanger, where at the exit of the cooler the $\Delta T_{\text{MAX}}$ was at 2-5 °C, without any significant improvement of temperature uniformity at the end of mixing using vibration (Fig. 4.13).

Figure 4.11. Comparison of $\Delta T_{\text{HX}}$ during cooling of banana puree, for the untreated and the treated heat exchanger.
Figure 4.12. Comparison of the temperature distribution within the product (at center, wall and an intermediate point) at the inlet (a) and the outlet (b) of the mixing unit for banana puree after cooling in the untreated heat exchanger for a given product average temperature at the inlet of the heat exchanger. Note that the average time for the fluid particle to move from the inlet to the outlet of the heat exchanger and mixing unit was 60 s and 24 s, respectively.
Figure 4.13. Comparison of the temperature distribution within the product (at center, wall and an intermediate point) at the inlet (a) and the outlet (b) of the mixing unit for banana puree after cooling in the treated heat exchanger for a given product average temperature at the inlet of the heat exchanger. Note that the average time for the fluid particle to move from the inlet to the outlet of the heat exchanger and mixing unit was 60 s and 24 s, respectively.
Washing down with hot water for 2400 s, followed the cooling experiments. Comparing the photographs between the internal heat exchanger tube surfaces, a significantly more efficient cleaning was observed with the hydrophobic surface treated heat exchanger. However, for the case of the untreated heat exchanger, the surface of the tube was fully covered with banana puree gel deposition, which formed during cooling (Fig. 4.14). The existence of the thick layer of gel at the pipe wall had a negative impact on heat transfer during cooling. Moreover, similar flow rate observed with the treated heat exchanger may have resulted due to the potential slip flow formed between the thick layer of gel at the wall and the hotter, lower viscosity food product.

![Figure 4.14. Comparison of a) treated with hydrophobic chemical solution and b) untreated internal heat exchanger tube outer surfaces after cooling of banana puree. Picture shows the disassembled tubes after washed-down with hot water.](image-url)
4.3.6. Cheese sauce

The final experiments have been performed during the cooling process of cheese sauce, within the temperature range of 95-68 °C, and by applying vibration of 120 s duration at 20 Hz, for mixing. Steady state operational conditions were achieved with cheese sauce flowing within the annulus passage with an inlet temperature of ~95 °C, and cooling water flowing in a counter-current mode, with a flow rate of $56.8 \times 10^{-5} \text{ m}^3/\text{s}$ and with an inlet temperature of 23 °C.

Time-temperature profiles for cheese sauce presented a significantly more efficient cooling process using the treated heat exchanger, with a $\Delta T_{\text{HX}}$ of 16-18 °C, compared to a $\Delta T_{\text{HX}}$ of 10-12 °C, for the case with the untreated heat exchanger (Fig. 4.15). A slightly more uniform temperature distribution within the food was observed with the treated heat exchanger with a $\Delta T_{\text{MAX}}$ of 8-10 °C, while for the untreated case, the $\Delta T_{\text{MAX}}$ was at 10-12 °C. Thermal mixing processing of cheese sauce via application of vibration at 20 Hz, presented a slight improvement on temperature distribution within the cross section of the flowing product, with a $\Delta T_{\text{MAX,M}}$ of 6-8 °C, for both cases (Fig. 4.16; Fig. 4.17).

For the case of cheese sauce, the wash-down step with hot water was for a shorter period of time 180 s, in order to study the differences regarding the existence and thickness of product deposition at the pipe wall. A significant difference was observed regarding the amount and thickness of product deposition at the pipe wall, with the worst case with the untreated heat exchanger, where the surface of the inner tube was covered with cheese sauce with a thickness of several millimeters (2-3 mm). For the case of the Aculon-treated heat exchanger, the surface of the inner tube is pictured with significantly less product accumulation at the pipe wall (Fig. 4.18). Like in the case of banana puree, the thick layer of cheese sauce
deposition at the pipe wall reduced the heat transfer efficiency. Moreover, similar flow rate observed with the treated heat exchanger may have resulted due to the potential slip flow formed between the thick layer of gel at the wall and the hotter, lower viscosity food product.

Figure 4.15. Comparison of $\Delta T_{\text{HX}}$ during cooling of cheese sauce, for the untreated and the treated heat exchanger.
Figure 4.16. Comparison of the temperature distribution within the product (at center, wall and an intermediate point) at the inlet (a) and the outlet (b) of the mixing unit for cheese sauce after cooling in the untreated heat exchanger for a given product average temperature at the inlet of the heat exchanger. Note that the average time for the fluid particle to move from the inlet to the outlet of the heat exchanger and mixing unit was 60 s and 24 s, respectively.
Figure 4.17. Comparison of the temperature distribution within the product (at center, wall and an intermediate point) at the inlet (a) and the outlet (b) of the mixing unit for cheese sauce after cooling in the treated heat exchanger for a given product average temperature at the inlet of the heat exchanger. Note that the average time for the fluid particle to move from the inlet to the outlet of the heat exchanger and mixing unit was 60 s and 24 s, respectively.
Figure 4.18. Comparison of treated with hydrophobic chemical solution and untreated internal heat exchanger tube outer surfaces after cooling of cheese sauce. Picture shows the disassembled tubes after washed-down with hot water.

Different results observed as far as the effects of hydrophobic coating on the cooling performance of the tested food products, was attributed to the different composition of the examined products. The improvement on cooling using the treated heat exchanger for products with high protein (cheese sauce) and carbohydrate (banana puree) was associated with the significantly lower amount of product deposition at the pipe surface, comparing with the untreated case. On the other hand, for products with a high starch content (sweet potato puree), the amount of product deposition was low for both treated and untreated heat exchangers, with hydrophobic treatment presenting no significant effects on cooling of sweet potato puree. More
focused studies will be required for better understanding of the interactions between contact
surface, implemented surface treatments, and the flowing food material properties.

4.4. Conclusions

In this work, the effects of hydrophobic surface treatment on continuous flow cooling
stage of thermal processing of sweet potato puree, banana puree, and cheese sauce were
studied. Cooling performance was compared between two identical horizontal stainless steel
tube in tube heat exchangers, one with untreated product contact surface and one with surfaces
treated with a hydrophobic Aculon chemical treatment, for identical operating conditions (flow
rate, inlet temperature), under steady state regime, with the tested food products flowing within
the annulus passage and with the coolant (water) flowing in counter-current mode.

The results for banana puree and cheese sauce showed a significantly more efficient
cooling performance when using the treated heat exchanger, with a lower average outlet
product temperature in the range of 4-6 °C, compared to the result obtained with the untreated
heat exchanger. However, for sweet potato puree, cooling using the untreated heat exchanger
was slightly better, with the average outlet product temperature been 2-4 °C lower, compared
to the treated case.

Furthermore, temperature cross sectional temperature distribution within the flowing
product during cooling and thermal mixing for all the tested materials was more uniform with
the treated heat exchanger. The maximum recorded temperature difference among the three
points, at the exit of the heat exchanger ($\Delta T_{\text{MAX}}$), was lower by 6-7 °C, 5-6 °C and 2 °C, for
sweet potato puree, banana puree and cheese sauce, respectively, for the treated heat exchanger
comparing to the untreated one.
Thermal mixing studies via application of vibration at the resonance frequency of the mixing unit of 20 Hz, showed good results for all the experiments with sweet potato puree and banana puree. A maximum recorded temperature difference among the three points, at the exit of mixing unit ($\Delta T_{\text{MAX},M}$), in the range of 2-4 °C, within sweet potato puree and banana puree was observed after the application of vibration. However, for all the studies with cheese sauce, no significant improvement in temperature uniformity was observed via application of vibration, with the $\Delta T_{\text{MAX},M}$ in the range of 8-10 °C.

Moreover, using the treated heat exchanger, the amount of product deposition at the pipe wall, after washing down with hot water, was significantly lower, which should be an indication of the reduced resistance to heat transfer.

Finally, this work showed that surface modification can be used as an effective method for potential applications in advanced thermal processing. Increasing the hydrophobic surface characteristics of the food contact surfaces resulted in enhancement of continuous flow cooling of viscous and poorly conductive foods and also result in the potential reduction of the cooling-related food quality losses while improving the sanitation processing conditions. However, results obtained using sweet potato puree as the process materials indicate the need for more studies in order to better understand the effects of surface characteristics on thermal processing, particularly cooling, of products with different food composition.

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Nomenclature

*Latin letters*

MW        Microwave system

T        Temperature (°C)

*Greek letters*

ΔT        Temperature difference (°C)

*Subscripts*

MAX       refers to the maximum temperature difference between the 3 cross-sectional points at the exit of heat exchanger

MAX_M     refers to the maximum temperature difference between the 3 cross-sectional points at the exit of mixing unit

HX        refers to the temperature differences between the average temperature at the inlet and at the outlet of the heat exchanger
References


CHAPTER 5

Part 1: Effects of Food-Contact Surface Characteristics on Adhesive Behavior of Viscous Foods for Potential Applications in Advanced Thermal Processing

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ABSTRACT

This research investigated the effects of surface characteristics on the adhesive behavior of sweet potato puree, banana puree, and cheese sauce. Different engineered food-contact surface samples, including untreated and chemically treated stainless steel, polytetrafluoroethylene (PTFE), silicone sheets as well as treated and untreated knitted and woven polyester fabrics, were studied. The examined surfaces were differentiated based on their surface wettability (hydrophilic, hydrophobic, or superhydrophobic) and surface roughness. The adhesive behavior of the tested foods and repellent behavior of the engineered surfaces were examined by measuring the residual product mass, remaining attached to the testing surface, during the standard methods of product depletion and the centrifuge adhesion tests. These tests were performed under two different temperature conditions; at ambient and at cooling processing temperature conditions. The results from the product depletion test showed that the hydrophobic surfaces with smooth characteristics (chemically treated stainless steel samples), PTFE, and treated knitted fabrics retained the least amount of the food products. The average residual mass for all the products on these surfaces was approximately at 35-45% at room temperature, and 45-65% at cooling temperature conditions. Under cooling conditions, to examine cooling efficiency, the temperature difference (ΔT) between the initial and final highest product temperature was compared between the examined surfaces. Treated and untreated stainless steel and PTFE surfaces presented the highest ΔT values, for all the foods tested. Finally, during the centrifugal adhesion test, the superhydrophobic knitted fabrics exhibited the best repellent behavior with all the food samples, reducing the residual product to 0-10% and 20-40% levels, under room and cooling temperature conditions, respectively.

Key words: Adhesive behavior, surface modification, viscous foods
5.1. Introduction

Accumulation, stickiness, and fouling of food components, such as protein, starches, and minerals, on thermal processing system components (heat exchangers, mixing units, holding tubes, spray dryers, food packaging) have a significant impact on processing efficiency and operating costs. Accumulation of fouling layers on food processing equipment increases the resistance to heat transfer, thermal mixing and pressure drop across the processing equipment, resulting in degradation of final food product quality and increasing the overall processing and cleaning operation time and cost (Balasubramanian and Puri, 2008; Balasubramanian and Puri, 2009; Barish and Goddard, 2013; Barish and Goddard, 2014; Stoforos et al., 2017).

Furthermore, fouling, adhesion, and accumulation of unwanted food material on the processing equipment surface depend on several different parameters such as operating conditions (temperature, and flow rate), food properties (viscosity, and food composition) and surface properties (material, surface wettability, and topography) (Santos et al., 2004; Awad, 2011; Mérian and Goddard, 2012). Fouling and deposition of unwanted material can occur in many different types, based on different physical, and chemical processes involved. Different main fouling types formed on the heat transfer surface are categorized into the following types (Awad, 2011): 1) particulate or sedimentation fouling (deposition of suspended particles), 2) crystallization or precipitation fouling (dissolved salts from saturated solution), 3) chemical reaction fouling (cooking, polymerization, etc...), 4) corrosion fouling (electrochemical reactions), 5) biological fouling or biofouling (attached and growth of microorganisms), and 6) solidification or freezing fouling (formation of a solid fouling deposit onto a subcooled surface, from fluid components with a high melting point freeze).
Moreover, this problem mainly has been observed during conventional thermal processing (heating, cooling, and thermal mixing) of highly viscous and multiphase food products, such as typical dairy products, fruit and vegetable purees and soups (Stoforos et al., 2016; Stoforos et al., 2017). Unfortunately, the adhesive behavior of highly viscous foods is more prone to occur on stainless steel, the most commonly used material for food processing equipment, such as heat exchangers and mixing units (Goode et al., 2013; Barish and Goddard, 2013; Stoforos et al., 2016). Stainless steel is an inert material, relatively resistant to corrosion, stable with temperature, and highly thermally conductive, ideal for food-contact surface material for thermal processing equipment units (Goode et al., 2013; Barish and Goddard, 2013). However, its high surface energy and surface wettability promote fouling, adhesion, and accumulation of highly viscous and dairy food products. The build-up of unwanted food material at the surface of heat exchangers results in increasing the heat resistance, flow resistance, and pressure drop, leading to reduced process efficiency and quality of the final food product (Beuf et al., 2003; Balasubramanian and Puri, 2008; Balasubramanian and Puri, 2009; Goode et al., 2013 Barish and Goddard, 2013; Barish and Goddard, 2014; Stoforos et al., 2017). Moreover, product accumulation and fouling are prone to biofilm formation, resulting in excessive use of chemical detergents and sanitizers, in addition to frequent processing plant shutdowns (Sandu and Singh, 1991; Kananeh et al., 2010; Mérian and Goddard, 2012; Barish and Goddard, 2014).

Recently, many studies have been devoted to using surface modification techniques to reduce or completely prevent fouling or biofouling of the food and dairy processing equipment (Santos et al., 2004; Mérian and Goddard, 2012). New developments in surface modification technologies, such as polymer and nanoparticle coatings, physicochemical
surface modification techniques such as plasma-enhanced chemical vapor deposition, and electro-chemical and ion implantations have enabled researchers successfully to modify stainless steel with antifouling characteristics (Santos et al., 2004; Rosmaninho et al., 2007; Balasubramanian and Puri, 2009; Kananeh et al., 2010; Barish and Goddard, 2013). Antifouling surfaces have been fabricated in a way to mimic the repellent characteristics of natural antifouling materials such as the lotus leaf \textit{(Nelumbo nucifera)}), by modifying surface properties (wettability (hydrophilic, hydrophobic, superhydrophobic) and topography), aiming to produce a low surface energy surface (Santos et al., 2004; Rosmaninho et al., 2007; Balasubramanian and Puri, 2009; Kananeh et al., 2010; Barish and Goddard, 2013). Another method to fabricate an antifouling surface is by modifying the chemistry of the surface, for example by using a hydrophilic (poly (ethylene glycol)) coating to stop hydrophobic interactions between whey protein and stainless steel (Premathilaka et al., 2006; Mérian and Goddard, 2012). Antifouling treatments have been shown to ease the operation and cleaning of the heat exchangers, while reducing the bacterial adhesion (Beuf et al., 2003; Santos et al., 2004; Premathilaka et al., 2006; Rosmaninho et al., 2007; Balasubramanian and Puri, 2009; Kananeh et al., 2010; Mérian and Goddard, 2012; Barish and Goddard, 2013; Cheng et al., 2015; Stoforos et al., 2017). However, high cost and complexity of their application as well as their reported short-term stability have impeded commercial application of such techniques (Barish and Goddard, 2013).

Developments of thermal processing with advanced heating technologies, such as continuous flow microwave (MW), ohmic heating, and radio frequency heating systems, have enabled uniform, rapid, and volumetric heating, which reduces come-up time, maximizes the final food product quality and reduces fouling deposition (Coronel et al., 2005; Bansal and
Chen, 2006; Steed et al., 2008; Cullen et al., 2012). On the other hand, cooling process of highly viscous and multiphase food products is still implemented using conventional cooling methods, resulting in a slow and unequal thermal treatment, and reduction of final food quality (Stoforos et al., 2016). Recent studies have shown that efficiency of cooling and cleaning process for viscous foods, such as cheese sauce and banana puree can be significantly improved by using hydrophobically treated stainless steel heat exchanger (Stoforos et al., 2017). However, the same study showed that hydrophobically treated heat exchanger has a negative impact on cooling performance of sweet potato puree (Stoforos et al., 2017). These observations highlight the importance of understanding and quantification of the surface parameters that affect the adhesive and heat exchange behavior of different food products. In addition to material properties, a successful alternative should also be cost-effective and industrially viable to attract commercial interest.

This study is the first part of a more comprehensive research project aiming to improve the efficiency of thermal processing, particularly cooling of viscous food material having poor thermal conductivity. This part of the research focuses on the influence of various surface characteristics such as wettability and surface topography of food-contact materials on the adhesive behavior of highly viscous foods and their potential importance in developing more efficient thermal processing systems.

5.2. Materials and Methods

5.2.1. Test materials

To study and understand the adhesive behavior of different viscous food products, namely sweet potato puree (Yamco LLC, Snow Hill, NC, U.S.A.), banana puree (Aseptia/Wright
Foods, Troy, NC, U.S.A.), and cheese sauce (Advanced Food Products, New Holland, PA, U.S.A.) different engineered food contact surfaces were fabricated and tested against the food samples. These food products were chosen because they exhibited different adhesive behavior when flowing against surfaces with different chemical and physical structures, during cooling and mixing processes (Stoforos et al., 2016; Stoforos et al., 2017).

To illustrate the effect of food-contact surface characteristics of the boundary walls on the adhesive behavior of the food samples, eleven different engineered food contact materials were examined for their wettability and surface roughness. Surface characteristics such as wettability and surface topography are not totally independent from each other and are related to the free surface energy (or surface tension) of food-contact surfaces, influencing of food and bacterial adhesion, cleaning, and heat transfer processes (Dürr, 2007; Choi and Kim, 2011).

The examined materials were chosen to cover a variety of different surface characteristics based on wettability and topography. The studied materials included (1) stainless steel samples, untreated and modified using chemical and lubricant surface treatments (Rain-X, ITW Global Brands, Houston, TX, U.S.A.; Aculon, Aculon performance Surface Solutions, San Diego, CA, U.S.A.; Sprayon LU206 Silicone Lubricate Spray, Sprayon, Cleveland, Ohio, U.S.A.), (2) fabrics (knitted and woven structures, with a base weight $2.9 \cdot 10^{-5}$ kg/m$^2$ and $2.3 \cdot 10^{-5}$ kg/m$^2$, respectively) untreated and treated with a commercial fluorocarbon-based resin, (3) PTFE sheets, (4) silicone sheets and (5) fiberglass reinforced silicone sheets (Table 5.1).

The following test methods were applied to study and understand the effects of surface wettability and topography on the adhesive behavior of food materials, for potential applications in advanced thermal processing.
Table 5.1. The list of the examined food-contact surface samples, with the base substrate and the applied treatment for each case.

<table>
<thead>
<tr>
<th>Food-contact surface</th>
<th>Base substrate</th>
<th>Treatment</th>
</tr>
</thead>
<tbody>
<tr>
<td>Untreated stainless steel</td>
<td>Stainless steel</td>
<td>Untreated</td>
</tr>
<tr>
<td>Rain-X</td>
<td>Stainless steel</td>
<td>Hydrophilic chemical treatment</td>
</tr>
<tr>
<td>Aculon</td>
<td>Stainless steel</td>
<td>Hydrophobic chemical treatment</td>
</tr>
<tr>
<td>PTFE</td>
<td>PTFE</td>
<td>Untreated</td>
</tr>
<tr>
<td>Sprayon LU206</td>
<td>Stainless steel</td>
<td>Surface lubricant</td>
</tr>
<tr>
<td>Silicone sheets</td>
<td>Silicone</td>
<td>Untreated</td>
</tr>
<tr>
<td>Silicone fiberglass sheets</td>
<td>Silicone</td>
<td>Untreated</td>
</tr>
<tr>
<td>Fluoropolymer coated polyester knitted textiles</td>
<td>Polyester fabric</td>
<td>Fluorocarbon-based resin</td>
</tr>
<tr>
<td>Untreated polyester knitted textiles</td>
<td>Polyester fabric</td>
<td>Untreated</td>
</tr>
<tr>
<td>Fluoropolymer coated polyester woven textiles</td>
<td>Polyester fabric</td>
<td>Fluorocarbon-based resin</td>
</tr>
<tr>
<td>Untreated coated polyester woven textiles</td>
<td>Polyester fabric</td>
<td>Untreated</td>
</tr>
</tbody>
</table>

5.2.2. Experimental methods

To characterize and identify the differences between the examined surfaces contact angle measurements and atomic force microscopic tests were conducted to measure surface wettability and roughness of the examined surfaces, respectively. Furthermore, to study the behavior of food materials in contact with the engineered surfaces, two different quantitative and reproducible tests were implemented, namely product depletion and centrifugal adhesion tests.
5.2.3. Contact angle measurements

Surface wettability was one of the parameters used to differentiate the examined surfaces, varying from hydrophilic (0-90°), hydrophobic (90-150°), up to superhydrophobic (150°) behavior. Measurements of water (θ_w) and oil (θ_oil) contact angles is a common method to evaluate surface wettability (Korhonen et al., 2013). The equilibrium static contact angles (CA) (the Sessile drop method) of water and oil (n-dodecane) on the tested samples were measured at room temperature (18-20 °C), using OCA 15 (Future Scientific Corp, Garden City, NY, U.S.A.), an optical contact angle goniometer, and via SCA20, its software interface. Using a 500 microliter gastight syringe (Hamilton Company, Reno, NV, U.S.A), controlled (3-5 μL) volume droplets of water and/or oil were deposited on different points (3-5 points) across the tested surfaces to measure the contact angle and determine the homogeneity of the treated surfaces.

5.2.4. Surface roughness measurements

Surface roughness measurements of tested surfaces were conducted using a multi-mode atomic force microscope (AFM) (Dimension 3100, Veeco Digital Instruments by Bruker, Billerica, MA U.S.A.). On a small scale (μm and nm) AFM is the method with the highest resolution among other optical and microscopy methods and is essential for studying and comparing surface roughness, surface porosity, and fouling detection between surface samples (Santos et al., 2004; Boussu et al., 2005; Liu et al., 2006). AFM measurement parameters, such as scan area, tip size and tip motion (contact, noncontact and tapping modes) and resolution, influence the surface roughness values and it is meaningful to compare roughness data only under the same measurement parameters (Boussu et al., 2005). In AFM studies, the most
commonly studied roughness parameter is the arithmetic average roughness (Ra (nm)), that is, the average of the absolute values of the roughness profile ordinates, providing a general description of the height variations in the surface. In food and fouling research area, AFM is the most commonly used method to characterize and compare the surface roughness of different untreated and treated stainless steel samples, with reproducible Ra results in the range of 10-300 nm (Santos et al; 2004; Barish and Goddard, 2013).

In the present study, AFM was used to map the surface of tested samples in tapping mode using a high frequency (~300 kHz) silicon probe having a nominal spring constant of 40 N/m. The tip of the probe, with a diameter of less than 10 nm, scanned an area of 5 × 5 μm with a tip velocity of 10 μm/s. The scanning resolution was 512 pixels per line and 256 lines of data per scanned area. Digital Instruments Nanoscope (Veeco Digital Instruments, Plainview, NY, U.S.A.) software was used to analyze the digital images from AFM and to determine the Ra values of the examined surface samples.

5.2.5. Product depletion test

Product depletion test is a common test method, used in food packaging studies for designing the best food packaging material to reduce the product remnants from the food packaging (Eleya and Hardy, 1993; Michalski et al., 1998; Loibl et al., 2012). For this study, a customized consistometer was built (based on the principles of Bostwick consistometer), to compare and quantify the effects of different surface characteristics, on the adhesive behavior of different viscous foods, by measuring the weight mass difference. The modified consistometer unit was built from stainless steel 300 series, with a length (L) of 0.241 m (9.5 in), width (W) 0.0381 m (1.5 in) and a trough height (H) of 0.0381 m (1.5 in). The modified
consistometer consisted at one end of a sample reservoir, closed off by a thin rigid PTFE gate, while the other end was open to allow unimpeded product flow and exit from the unit upon the PTFE gate removal. One additional part of the modified consistometer was a removable stainless steel strip \((L=0.241 \text{ m (9.5 in)}; W=0.036 \text{ m (1.4 in)}; \text{ thickness of } 0.0012 \text{ m (0.047 in)})\), which was attached to the surface of the unit and used as the food-contact surface. The food-contact surface of the stainless steel strip was used as the sample for the surface modification applications. Eleven different consistometer units were built, one for each testing material. The thickness of the stainless steel strip remained unchanged after application of the chemical treatments. However, it was increased by 0.01 m (0.4 in) and 0.02 m (0.9 in), with the addition of textile and polymer samples, respectively. All the tested consistometer units were attached to the top of a laboratory support jack, which was used to adjust the height and change the inclination \((0-90^\circ)\) of the consistometer units, and to enable the gravity-driven flow of the examined food samples (Fig. 5.1).

Moreover, for each examined food product, the initial step during these series of experiments, was to find the incline that would initiate and maintain an easy and smooth flow of the viscous food material. Using as the reference food contact surface of the consistometer unit (untreated stainless steel of 300 series), the reservoir of the consistometer was filled with 50 g of food sample at the examined temperature, with the reservoir door closed. Starting with the consistometer unit at horizontal \((0^\circ \text{ angle of incline})\) position, the reservoir gate was opened, allowing the food sample to initiate flow. The height of the laboratory jack supporting one end of the consistometer was steadily increased, increasing the inclination until an easy and smooth flow of the food sample was observed.
Figure 5.1. Parts of the customized consistometer unit: a) Consistometer unit body, stainless steel sheet and reservoir PTFE door, b) top view of the unit, c) side view of the unit and d) consistometer unit attached to the laboratory support jack to change the inclination of the unit. A magnetic protractor shows the angle of the incline.

During all the different experiment series, adhesive behavior of tested food samples was examined for different food-contact surface treatments and surface materials, comparing primarily the residual product mass remaining on the examined surface after the completion of the experiments, and secondly by measuring and comparing the exit time \( t_{\text{exit}} \). Initially, the weight of each consistometer was measured before and after introducing 50 g of food sample into the reservoir of the unit. The next step was to place the top edge of the tested consistometer unit, with the sample reservoir filled and the reservoir gate closed, against the top of the laboratory support jack, with the predetermined angle of incline \( \theta \). At time \( t \) of 0 s, the reservoir gate was opened and starting recording the \( t_{\text{exit}} \). After a period of 900 s (15 min),
sufficient amount of time to ensure that flow of the tested food sample has stopped, the residual weight of each consistometer unit was measured again.

The above experiment steps were followed for all the different food samples and all the food-contact surfaces examined, under two different temperature conditions. Initially, the first set of experiments was conducted having both the tested food sample and the examined surface at room temperature (~20 °C). For each food material, a sequence of three consecutive experimental runs was conducted, against every food-contact surface, to study the performance decay, for each surface sample. The sequence of the experimental steps was first to apply the examined surface modification treatments to the food-contact surface, conduct product depletion test, wash-down the surface, and then repeat the same steps twice but without re-treatment of the surface. The average residual product mass and standard deviation of the results were compared for all the examined food contact surfaces, for each food sample.

While the experiments described above were performed at room temperature, in real continuous flow cooling systems, temperature of the entering food material is usually around 95 ± 3 °C and the cooling wall is kept at 5 ± 2 °C. A MW work station bench-top Panasonic Inverter MW oven (operating at MW frequency of 2450 MHz, with a maximum input power at 1200 W), was used to heat up the food samples to the designed temperatures. The examined surface samples were kept in a cooler prior to the experiments, with a preset temperature at 5 °C, to achieve the desired food-contact surface temperature. In addition to the measurements of the residual mass, this experiment also monitored the temperature of the flowing food product. Changes in the tested product temperature, due to the contact with the cold food-contact surface and the exposure to the ambient air were measured using an infrared camera (FLIR ONE, FLIR Systems, Inc., Wilsonville, OR). Temperatures of the tested food sample
were recorded every 30 s for a total time of 360 s, during the product depletion tests. Temperature recording interval was chosen based on the required time for the infrared camera to capture the image and recalibrate for the next image. Total time of recording was selected based on preliminary tests, regarding the time required for the food samples to cool down from ~95 °C to room temperature (~20 °C).

To study the effects of different material compositions, modification techniques and surface characteristics on adhesive behavior and cooling performance of the tested foods, various substrates spanning a wide range of physical and chemical properties were either obtained from retail stores, purchased from suppliers or made in-house to be used as the testing samples in the product depletion experiments. The collected residual mass and temperature data were used to compare the adhesive behavior and cooling performance of the tested foods in contact with the examined food-contact surfaces, for potential use in continuous flow cooling applications, with highly viscous foods.

5.2.6. Centrifuge adhesion test

A second test, the centrifuge adhesion test, was conducted in order to quantify the effects of surface characteristics on the adhesive or repellent behavior between the engineered food-contact surfaces and tested food materials. This testing method uses centrifugal forces for adhesion studies. The test was initially introduced as a method for testing anti-icing materials (Laforte and Beisswenger, 2005). In its original application, the method applies the required centrifugal force to detach the ice from the tested surface, resulted by adjusting the rotating speed and calculates the corresponding adhesive stress (Laforte and Beisswenger, 2005). In the present study, the adhesive behavior of the tested food samples at different rotating speeds,
against all different examined food contact surfaces was determined by comparing the mass of the detached products. All centrifugal product adhesive reduction tests, for all the different food samples and all food-contact surfaces examined at different rotating speeds, were implemented under two different temperature conditions. Firstly, the experiments were conducted by having both the tested food and the examined surface at room temperature (20-23 °C), and secondly within the temperature range of the cooling process conditions. For the second case, food samples were heated using the MW within a temperature range of 95-100 °C, while the examined contact surfaces were at 15-20 °C.

The centrifuge adhesion tests were performed using a rotating tube unit. The rotating tube unit consisted of a tube segment, removable and different for each surface treatment examined, and a motor, having the capability to control the rotating speed. The outside surface of the rotating tube was used to apply the tested food-contact materials. A stainless steel tube segment, of 0.1524 m (6 in) in length and with an outside diameter (O.D.) of 0.038 m (1.5 in) was used. The O.D. of the tested tube samples remained unchanged after application of the chemical surface treatments, while the O.D. of the tested tubes was slightly increased, being 0.039 m (1.54 in) and 0.040 m (1.59 in) for the tested tubes coated with textile and polymer samples, respectively. Water, at a controlled temperature, was flowing within the tube with a constant flow rate of 63.1×10−5 m³/s (10 gal/min) to control the food-contact surface temperature.

Centrifuge adhesion tests, for all the examined food samples against the modified food-contact surfaces, were performed at three different rotational speeds of 50%, 75% and 100% of the maximum capacity of the motor. Lower rotational speeds were not examined due the to predominance of the gravitational force on the adhesive response of the food materials.
A tachometer was used to record the exact angular velocity of the testing rotating tube sample. Knowing the rotating speed or \( \omega \) (rad/s)), the mass of the tested food product sample or \( m \) (kg) and the outside radius or \( r \) (m) of the testing rotating tube, the applied centrifugal force or \( F_c \) (N) was calculated from the following equation (Eq. 5.1) (Laforte and Beisswenger, 2005):

\[
F_c = m \cdot r \cdot \omega^2
\]  

(5.1)

Before the beginning of each test, part of the surface of the removable tube segment was covered with a specific volume of the tested food product. To achieve the precision and the repeatability of the volume of the food samples, three different plastic (Acrylonitrile butadiene styrene (ABS) plastic) molds, based on different O.D. of the coated tube segments, were designed and built, using a 3D-printer (Stratasys, Eden Prairie, MN, U.S.A.). The plastic molds were precisely designed with a cavity at the center of one of the surfaces and extended edges, capable of attaching to the outer surface of the testing tubes and allowing for the cavity to be filled with a specific volume of food sample. The volume of the cavity of the plastic molds was designed with a depth of 0.0127 m (0.5 in), a width of 0.0127 (0.5 in), and length of 0.11 m (4.3 in). The depth of mold's cavity was designed based on the annulus passage of the heat exchanger used in one of the previous studies (Stoforos et al., 2017). The width and the length of the cavities of the molds were designed based on the O.D. and the length of the examined rotating tube. After filling the cavity of the plastic mold, and hence the top surface area of the testing tube, with the food sample, the initial mass of the total added product was measured, the tube segment was attached to the rotating tube unit and the plastic mold was removed. The tube was rotated clockwise for 60 s (Laforte and Beisswenger, 2005) at the selected speed. During the rotation of the tested sample, detached product was collected and the final mass of the removed product was determined (Fig. 5.2; Fig. 5.3).
5.3. Results and discussion

5.3.1. Contact angle measurements

This study tested the effects of different surface properties such as surface material, wettability and topography (roughness) on the adhesive and deposition behavior of different
food products, namely sweet potato puree, banana puree, and cheese sauce. Surface wettability and roughness were measured to quantify the differences between the modified food-contact surface samples. Surface wettability was examined, by measuring the contact angles of water ($\theta_w$) and oil ($\theta_{oil}$) for each surface sample. Table 5.2 presents the results of the $\theta_w$ and $\theta_{oil}$ measurements for the examined surface samples, showing a wide range of different wettability properties. Using the video-based OCA 15 goniometer, the results on sessile drop static water and oil contact angle at different points on the tested food-contact surfaces showed a homogeneous (small standard deviation) behavior for all samples. The Rain-X treated stainless steel samples showed hydrophilic behavior ($\theta_w \sim 7^\circ$), while the Silicon Lubricated Spray coated as well as the untreated stainless steel samples showed near-hydrophobic behavior ($\theta_w$ of 93-96$^\circ$). The fluorocarbon-treated polyester fabrics showed superhydrophobic (140-145$^\circ$) behavior (Fig. 5.4.). A hydrophobic behavior, with a $\theta_w$ of 105-130$^\circ$, was observed for the rest of the samples.

In the case of wetting properties of the oil liquid (n-dodecane), most samples showed oleophilic behavior ($\theta_{oil} = 0-33^\circ$), while all the polyester treated and untreated textile samples had an oleophobic behavior (100-115$^\circ$).
Table 5.2: Water and oil contact angles of the examined food-contact surfaces.

<table>
<thead>
<tr>
<th>Food-contact surface samples</th>
<th>( \theta_W ) (°)</th>
<th>( \theta_Oil ) (°)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Untreated stainless steel</td>
<td>93</td>
<td>17</td>
</tr>
<tr>
<td>Rain-X</td>
<td>7</td>
<td>11</td>
</tr>
<tr>
<td>Aculon</td>
<td>110</td>
<td>0</td>
</tr>
<tr>
<td>PTFE</td>
<td>105</td>
<td>33</td>
</tr>
<tr>
<td>Sprayon LU206</td>
<td>96</td>
<td>14</td>
</tr>
<tr>
<td>Silicone sheets</td>
<td>122</td>
<td>33</td>
</tr>
<tr>
<td>Silicone fiberglass sheets</td>
<td>114</td>
<td>25</td>
</tr>
<tr>
<td>Fluoropolymer coated polyester knitted textiles</td>
<td>140</td>
<td>100</td>
</tr>
<tr>
<td>Untreated polyester knitted textiles</td>
<td>130</td>
<td>100</td>
</tr>
<tr>
<td>Fluoropolymer coated polyester woven textiles</td>
<td>145</td>
<td>115</td>
</tr>
<tr>
<td>Untreated coated polyester woven textiles</td>
<td>130</td>
<td>105</td>
</tr>
</tbody>
</table>

Figure 5.4: Water contact angle measurements on a) Rain-X treated stainless steel, b) untreated stainless steel, c) Aculon treated stainless steel, and d) fluoropolymer coated polyester knitted textiles. Note that the needle on the pictures has an O.D. of \( 7.2 \times 10^{-4} \text{ m} \) (0.72 mm).
5.3.2. Surface roughness measurements

Surface roughness was examined as the second parameter to quantify the differences between the examined food-contact surfaces samples. Analysis of the AFM micrographs determined the arithmetic average surface roughness ($R_a$ (nm)) of the examined food-contact surfaces. Table 5.3 presents the results of $R_a$ for the tested surfaces. The $R_a$ value for the reference untreated stainless steel sample was 7.9 nm. Smooth surfaces were observed for the samples treated with Sprayon LU206, and Aculon with $Ra$ values of 0.3 nm and 0.7 nm, respectively. On the other hand, the average surface roughness of stainless steel treated with Rain-X, the hydrophilic formulation, was 16.2 nm. For polymer coated samples and textiles, the lowest $Ra$ value was observed for the fiberglass reinforced silicone sheets, at 2.5 nm. Among all the samples, the silicone sheet sample exhibited the roughest surface with the highest observed $Ra$ of 85.7 nm. The rest of the textiles and PTFE samples showed intermediate $Ra$ values, compared to other samples, in the range of 15-40 nm. Figure 5.5 depicts the atomic force micrographs of the samples.

Finally, it is important to clarify that $Ra$ value should not be confused with the absolute roughness, $\varepsilon$ (μm or mm), used in fluid mechanics for friction factor calculation in pipe flow, which is given from the mean peak-to-valley height surface roughness, $R_{ZD}$ (Farshad et al., 2001). Also, it is worth mentioning that AFM is not the recommended method for surface roughness measurements of textiles substrates. However, for the purposes of this study and to maintain testing consistency, a small localized scanned area of $5 \times 5$ μm was used to enable us to measure the roughness of knitted and woven fabric samples using AFM.
Table 5.3. AFM measurements of surface roughness ($Ra$ (nm)) for the different examined food-contact surfaces.

<table>
<thead>
<tr>
<th>Food-contact surface</th>
<th>$Ra$ (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Untreated stainless steel</td>
<td>7.9</td>
</tr>
<tr>
<td>Rain-X</td>
<td>16.2</td>
</tr>
<tr>
<td>Aculon</td>
<td>0.7</td>
</tr>
<tr>
<td>PTFE</td>
<td>37.3</td>
</tr>
<tr>
<td>Sprayon LU206</td>
<td>0.3</td>
</tr>
<tr>
<td>Silicone sheets</td>
<td>85.7</td>
</tr>
<tr>
<td>Silicone fiberglass sheets</td>
<td>2.5</td>
</tr>
<tr>
<td>Fluoropolymer coated polyester knitted textiles</td>
<td>30.4</td>
</tr>
<tr>
<td>Untreated polyester knitted textiles</td>
<td>15.3</td>
</tr>
<tr>
<td>Fluoropolymer coated polyester woven textiles</td>
<td>39.8</td>
</tr>
<tr>
<td>Untreated coated polyester woven textiles</td>
<td>33.8</td>
</tr>
</tbody>
</table>
Figure 5.5. AFM surface topography images of a) Aculon treated stainless steel, b) untreated stainless steel, c) PTFE, and d) Silicone sheets. The shadings correspond to the Z height variation. Darker shades represent lower Z value and brighter regions represent higher Z value.
5.3.3. Product depletion test

Adhesive behavior, of the tested food products, namely sweet potato puree, banana puree, and cheese sauce, was tested against the contact-surface samples, using product depletion methods. The effects of the food-contact material were examined primarily on the product mass retained on the surface during product depletion test and by comparing the $t_{exit}$. The average percent of the residual product mass was used as a measure to compare the different examined food contact surfaces, at two different temperature conditions, at room temperature and cooling process conditions, respectively.

5.3.4 Product depletion test: room temperature conditions

Starting at room temperature, the initial step for the product depletion tests was to determine the incline angle ($\theta$) that resulted in a smooth flow of different food materials. Using the untreated stainless steel as a baseline, $\theta$ of 30°, 60°, and 75°, was found sufficient to generate such flow for sweet potato puree, banana puree, and cheese sauce, respectively. Using the predetermined $\theta$, three consecutive experimental series were conducted for each tested food and each food contact surface sample. The data of residual mass of these experiments were used to compare the product depletion performance of each examined surface. Additionally, the standard deviation of the residual mass was calculated to evaluate the consistency and the stability of each used surface modified technique (untreated surfaces; chemically treated, and surfaces with coatings applied).

Standard deviation data showed that the untreated surfaces such as stainless steel, PTFE, silicone sheet, and silicone fiberglass sheet materials were very consistent with their performance on product depletion test, for all the tested foods. The chemically treated surfaces
(Rain-X, Aculon, Sprayon LU206) presented an inconsistent product depletion behavior. Moreover, the collected $t_{exit}$ data indicated that the lower the $t_{exit}$, the less product retained on the surface.

Results showed that the average residual product mass was comparable for sweet potato puree and cheese sauce. For these two products, superhydrophobic surface with knitted textiles structure presented the best performance, followed by hydrophobic smooth surfaces (Aculon, Sprayon LU206) and PTFE, with a residual product mass of 30% and 35-55%, respectively. The food-contact surface samples with the highest observed roughness (silicone sheets, untreated polyester woven textiles) were the samples with the highest amount of retained product, around 70%. An average performance was observed for the rest of the surface samples, including the reference sample of the untreated stainless steel (Fig. 5.6; Fig. 5.7; Fig. 5.8; Fig. 5.9).

Figure 5.6. Images of residual sweet potato puree, at the end of product depletion experiment 1, on the surface of (from top to bottom): untreated stainless steel, Aculon, PTFE, and fluoropolymer coated polyester knitted textile.
Figure 5.7. Effect of food-contact materials on the product mass retained on the surface during the product depletion tests for sweet potato puree, at room temperature. Bars represent the average % residual product of three consecutive experiments. Error bars indicate the standard deviation.

Figure 5.8. Images of residual cheese sauce, at the end of product depletion experiment 1, on the surface of (from the top to the bottom): untreated stainless steel, Aculon, PTFE, and fluoropolymer coated polyester knitted textile.
Figure 5.9. Effect of food-contact materials on the product mass retained on the surface during the product depletion tests for cheese sauce, at room temperature.

Furthermore, no significant difference was observed between the food-contact surfaces effects on banana puree depletion behavior, with a reported value of 60% as the average percentage of the product mass retained. Banana puree had the highest amount of residual product mass, with most of the surface area of the examined samples, remaining covered with the product at the end of the experiment runs (Fig. 5.10). Hydrophobic smooth surfaces presented the lowest percentage of residual mass with a value of 50% (Fig. 5.11).
Figure 5.10. Images of residual banana puree, at the end of product depletion experiment 1, on the surface of (from the top to the bottom): untreated stainless steel, Aculon, PTFE, and fluoropolymer coated polyester knitted textile.

Figure 5.11. Effect of food-contact materials on the product mass retained on the surface during the product depletion tests for banana puree, at room temperature.
5.3.5. **Product depletion test: cooling processing temperature conditions**

Product depletion tests were also conducted under cooling processing temperature conditions. The initial temperature of the samples and contact surfaces for this test were 95 ± 3°C and at 5 ± 2°C, respectively. Under these conditions, two independent experiments were performed to study product depletion and cooling efficiency for each of the food samples and used food-contact surfaces. Average percent of residual product mass and temperature difference (ΔT) between the initial and the final (360 s) product temperature, were used to compare the effects of the different examined food-contact materials on cooling of viscous foods.

Identical incline angles as in the previous experiments were used for sweet potato puree and banana puree, at 60°, and 30°, respectively. Comparison of $t_{exit}$ between the two different temperature conditions showed a significant decrease in the $t_{exit}$ for banana puree at higher product temperature. Similar for sweet potato puree, at higher product temperature, a decrease of $t_{exit}$ was observed for most of the examined food-contact surfaces, while an increase in $t_{exit}$ was observed for the two untreated polyester textiles. A quantitative observation was that the viscosity of these two foods decreases at a higher temperature. For the case of cheese sauce, using the same incline $\theta$ as in the case of room temperature conditions, solidification of the flowing material on the food-contact surface occurred within the first 60-120 s of the test run, ceasing the product flow and depletion (Fig. 5.12). The only exception was with the treated textiles, where a $t_{exit}$ of 40-50 s was observed. Moreover, to perform the product depletion tests for cheese sauce, under the cooling temperature conditions, an incline $\theta$ of 90° was implemented. Under these conditions, a $t_{exit}$ within 5-49s was observed for all the used surfaces.
Figure 5.12. Images taken at 360 s during product depletion test for cheese sauce, under cooling processing temperature conditions and using an incline $\theta$ of 75°; showing the product solidification on the surface of: a) untreated stainless steel, b) Rain-X, c) Aculon, and d) fluoropolymer coated polyester knitted textiles.

Results of the product depletion experiments showed that hydrophobic, and smooth, Aculon treated stainless steel surface presented the lowest value of the residual product, for all the tested foods. Furthermore, banana puree presented the lowest residual product on the examined surfaces. For the banana puree, no significant difference was observed in the results of product depletion, with Aculon, Sprayon LU206, and PTFE presenting the best performance.
with a residual mass of 35-40%. Untreated polyester textiles and treated woven textile samples presented the highest residual mass with a value above 60%. The rest of the examined samples were in the range of 45-50% (Fig. 5.13). Similar to the case of room temperature experiments, surface area of the examined surface samples remained covered with banana puree, even at the end of the experiment runs. Furthermore, during the study of product depletion, the highest temperature of the product was recorded, every 30 s for the first 360 s of the experiment. Compared to other two food materials, banana puree showed an instant temperature drop (from 95°C to about 72°C) soon after it contacted the chilled surface. At the end of the experiment (t=360 s) the measured product temperature was within the range of 25-31°C (Fig. 5.14).

Figure 5.13. Effect of food-contact materials on the product mass retained on the surface during the product depletion tests for banana puree, at cooling process temperature conditions.
Figure 5.14. A set of three images per sample, during depletion test of banana puree, under cooling temperature conditions, showing the product temperature at the start of the test (~10 s) (bottom, right side) and at 360 s (bottom, left side) the final residual product (top) on the surface of: a) untreated stainless steel, b) Aculon, c) PTFE, and d) fluoropolymer coated polyester knitted textile.

Higher values of residual sweet potato puree on the examined food-contact surfaces were observed, compared with the results for banana puree. Moreover, Aculon treated surface and PTFE samples presented the lowest percentage of residual product within the range of 62-64%. Sprayon LU206 treated stainless steel and silicone fiberglass sheet samples were next, with a value of 68-70%. The rest of the examined surfaces had an amount of residual product
higher than 70%, with the hydrophobic rough surface of silicone sheets showing the highest percentage at 86% (Fig. 5.15).

Similar to the case of banana puree, initial temperature of sweet potato samples were ~95°C, while at t = 0 s, the highest measured product temperature dropped to 75-78°C. The final measured temperature was in the range of 35-38°C, for most of the samples, except for the surfaces coated with the polyester textiles, with a final measured temperatures above 40°C (Fig. 5.16).

Figure 5.15. Effect of food-contact materials on the product mass retained on the surface during the product depletion tests for sweet potato puree, at cooling process temperature conditions.
Figure 5.16. A set of three images per sample, during depletion test of sweet potato puree, under cooling temperature conditions, showing the product temperature at the start of the test (~10 s) (bottom, right side) and at 360 s (bottom, left side) the final residual product (top) on the surface of: a) untreated stainless steel, b) Aculon, c) PTFE, and d) fluoropolymer coated polyester knitted textile.

Product solidification of cheese sauce on the surface of the examined samples had a negative impact on product depletion and time-temperature results. Moreover, most of the examined surfaces presented a percentage of residual product higher than 70%. Aculon hydrophobic treated stainless steel surface and superhydrophobic knitted and woven textile samples presented the best cheese sauce depletion performance with the amount of the residual product mass of 52%, 55%, and 62%, respectively (Fig. 5.17).
Figure 5.17. Effect of food-contact materials on the product mass retained on the surface during the product depletion tests for cheese sauce, at cooling process temperature conditions.

Similar to the previous products, cheese sauce was introduced in the reservoir area of consistometer at temperature of 95°C. At $t=0$ s, product temperature has dropped to 75-78 °C. Solidification of the material on the examined surface resulted in high amount of residual product and slowed the cooling process resulting in higher final product temperature compared with banana puree and sweet potato puree. An average final product temperature around 42 °C, was observed for most of the examined surfaces. Aculon treated food-contact surface was the only one with a value below 40 °C, with a final product temperature of 33 °C (Fig. 5.18). It should be mentioned that no specific study was conducted regarding the solidification temperature of sweet potato. However, based on the current time-temperature data and the
observation during product depletion test, cheese sauce solidification might have occurred within the first 60 s of product depletion tests, hence within the temperature range of 55-75 °C.

Figure 5.18. A set of three images per sample, during depletion test of cheese sauce, under cooling temperature conditions, showing the product temperature at the start of the test (~10 s) (bottom, right side) and at 360 s (bottom, left side) the final residual product (top) on the surface of: a) untreated stainless steel, b) Aculon, c) PTFE, and d) fluoropolymer coated polyester knitted textile.
Collected time-temperature data were used to calculate the value of ΔT between the initial measured temperature (t=0 s) and the final measured product temperature (360 s). The average value of ΔT between the two independent experiments was used as the measure to estimate the effects of surface material on cooling of the tested foods. Small standard deviation of the results indicates that there is no significant difference between the time-temperature results between the two experiments.

Moreover, the ΔT values were affected by parameters such as thermal conductivity of the tested food and the examined surface. For most of the samples, the ΔT values for cheese sauce were lower than the rest of the food products. Banana puree, the product with the lowest retained material on the examined surface, presented the highest ΔT values. Similar with product depletion results, Aculon treated stainless steel surface presented the best performance (highest ΔT), regarding cooling for all the tested foods. However, the maximum ΔT was observed for the case of Sprayon LU206 treated stainless steel surface with sweet potato puree with a total temperature drop of 40 °C. On the other hand, polyester textiles presented the lowest values of ΔT. This result can be explained by the low thermal conductivity and thickness of the textile samples. Additionally, during the test with the superhydrophobic textiles, the tested foods covered a small surface area compared with the rest of the examined samples (Fig. 5.19). Moreover, the superhydrophobic characteristics of these samples resulted in a reduced product flow path, covering a small surface area, usually near to the stainless steel wall of the consistometer, a more hydrophilic surface compared with the textile samples (Fig. 5.20). In a continuous flow cooling environment, performed under pressure, within constricted flow profiles and driven by the conveyance of pumps, flowing product would more fully cover the
food contact surfaces, potentially improving the heat transfer exchange efficiency of the cooling process for these hydrophobic surfaces.

Finally, based on the product depletion results and the time-temperature data, hydrophobic surfaces with smooth characteristics, such as the hydrophobic treated samples, presented the best performance. However, for potential cooling processing applications, to mimic these surface characteristics, more long-term stable and food-grade surface modification techniques are required.

Figure 5.19. Effect of food-contact materials on ΔT between the initial measured temperature (t= 0 s) and the final measured product temperature (360 s), for sweet potato puree, banana puree, and cheese sauce. Error bars indicate the standard deviation, between the results of ΔT for the two independent experiments.
Figure 5.20. Comparison of the covered surface area with cheese sauce (under cooling conditions) between treated (left) and untreated polyester (right): a) knitted, b) woven textiles.

5.3.6. Centrifuge adhesion test

Adhesive and repellent behavior between the tested foods and the examined food-contact surfaces were studied using the centrifuge adhesion testing method. Similar to the product depletion test, percent of the residual product mass on rotating tube sample was used as the measure to compare the adhesive and repellent behavior of different samples at different rotating speeds and temperature conditions. All the tested food and surface samples were examined at three different rotation speeds of 50%, 75%, and 100% of motor speed volume that correspond to, respectively, 44 rpm, 66 rpm, and 88 rpm measured using a tachometer. All the experiments were conducted at two different temperature conditions: at room temperature...
(20-23 °C), and under cooling process conditions, with the food samples being at a temperature range of 95-100 °C, and the examined contact surfaces at 15-20 °C.

Approximately 18 ± 2 g of the used food samples was sufficient to fill the fabricated cavity, and cover the area at the top of the examined rotating tube. The initial product mass of the food samples tested at room temperature was 20 g while the weight of the food material was about 17 g at the cooling temperature conditions. Having all the information of product mass, rotating tube O.D., and speed of rotation, centrifugal force was estimated as a measure of the required adhesion force to reduce or detach the product from the surface of the examined rotating tube. An applied centrifugal force of 0.010 ± 0.003 N, 0.018 ± 0.003 N and 0.030 ± 0.005 N, was calculated for the 44 rpm, 66 rpm, and 88 rpm speeds, respectively.

For all the tested samples, repellent behavior was better at room temperature conditions and at the increased rotating speed, resulting in a lower residual product on the examined rotating tube samples. Sweet potato exhibited the least adhesive behavior against all the examined surfaces and under all the tested temperature conditions. Results obtained with sweet potato puree indicate a significant reduction of the residual product mass for all the examined food-contact surfaces and the rotation speeds. All the samples showed an amount of residual product lower than 24% and 33%, at room temperature and cooling temperature conditions, respectively. Furthermore, at room temperature conditions, increasing the rotating speed from 44 rpm to 66 rpm reduced the amount of the residual sweet potato puree on the surface of the examined rotating tubes, by 5-15%. No significant difference was observed between the results of residual product at 66 rpm and 88 rpm. For the experiments at cooling temperature conditions, no significant difference was noticed between the amounts of residual sweet potato puree, for all the examined rotating speeds. Moreover, superhydrophobic knitted
textile samples presented the best repellent behavior, with a residual sweet potato puree between 6-0%, and 21-10%, at room temperature and cooling temperature conditions, respectively. The hydrophilic treated surface sample exhibited the highest amount of residual product at 20-13% and 28-32%, for room temperature and cooling temperature conditions, respectively (Fig. 5.21; Fig. 5.22; Fig. 5.23; Fig. 5.24).

Figure 5.21. Images of residual sweet potato puree, at the end of centrifuge product adhesion tests, at room temperature, on the surface of: a) untreated stainless steel, b) Rain-X, and c) fluoropolymer coated polyester knitted textile; with a rotating speed of 44 rpm (1st row), 66 rpm (2nd row), and 88 rpm (3rd row).
Figure 5.22. Effect of food-contact materials on the product mass remained on the surface after the centrifuge adhesion tests for sweet potato puree, at room temperature. Bars represent the % residual product at different rotating speeds of 44 rpm, 66 rpm, and 88 rpm.

Figure 5.23. Images of residual sweet potato puree, at the end of centrifuge product adhesion tests, at cooling temperature condition, on the surface of: a) untreated stainless steel, b) Rain-X, and c) fluoropolymer coated polyester knitted textile.
Figure 5.24. Effect of food-contact materials on the residual product mass on the surface during the centrifuged product adhesion tests for sweet potato puree, under cooling temperature conditions

For the experiments with banana puree, superhydrophobic polyester textiles with knitted structure and Aculon, hydrophobic treated stainless steel surface samples exhibited the best repellent behavior. Moreover, at room temperature and with the rotating speed of 66 rpm or higher, these surfaces, as well as the untreated knitted textile, could repel the whole mass of banana puree from the surface of the rotating tube, with the amount of residual product between 4-0%. Increasing the rotating speed had no significant effect on the amount of residual banana puree on the surface of the Rain-x, hydrophilic treated stainless steel, and the silicone based surface samples, with an amount around 45-60%. For the rest of the surface samples the
amount of residual product was reduced significantly at rotating speeds of 66 rpm and higher, with the residual product between 20-45% (Fig. 5.25; Fig. 5.26).

Under cooling conditions, Aculon treated surface and superhydrophobic knitted textile samples retained the least amount of residual banana puree. For these surfaces, increasing the rotating speed reduced the amount of the residual product from 43-48% at 44 rpm to 22-32% at 66 rpm and to 14-20% for the maximum speed of 88 rpm. Similar behavior, with a reduction of the residual product by increasing the rotating speed, but with higher overall product retention, was observed for the surface samples of PTFE, the superhydrophobic woven textile, and the two untreated textile samples. PTFE and superhydrophobic woven samples had a residual product of 58% and 42% at the maximum speed, respectively. The untreated textiles at the maximum rotating speed of 88 rpm, had a residual product at 63% and 78%, for the knitted and the woven textile samples, respectively. No influence of rotating speed was found for the rest of the samples, with banana puree being higher than 70%, with the highest amount of retained product was to be at ~91% for the rough hydrophobic surface of silicone sheet (Fig. 5.27; Fig. 5.28).
Figure 5.25. Images of residual banana puree, at the end of centrifuge product adhesion tests, at room temperature, on the surface of: a) untreated stainless steel, b) Rain-X, and c) fluoropolymer coated polyester knitted textile.

Figure 5.26. Effect of food-contact materials on the product mass retained on the surface during the centrifuged product adhesion tests for banana puree, at room temperature.
Figure 5.27. Images of residual banana puree, at the end of centrifuge product adhesion tests, at cooling temperature condition, on the surface of: a) untreated stainless steel, b) Rain-X, and c) fluoropolymer coated polyester knitted textile.

Figure 5.28. Effect of food-contact materials on the product mass retained on the surface during the centrifuged product adhesion reduction tests for banana puree, under cooling temperature conditions.
Lastly, among the three tested food materials, cheese sauce exhibited the highest adhesive behavior. For both temperature conditions tested, at the lowest rotating speed of 44 rpm, the retention of cheese sauce was 100% for all the tested samples except the superhydrophobic knitted textile sample. At the same rotation speed, superhydrophobic knitted textile surface had a residual product of 46% and 64%, at room temperature and cooling temperature conditions, respectively. At the rotating speed of 66 rpm, only the textile samples reduced the amount of cheese sauce from the surface of the rotating tube. At room temperature, superhydrophobic textile surface samples exhibited the best repellent behavior, with only 35% and 42% residual product for the knitted and the woven textiles, respectively. Similar to the treated textiles, the untreated knitted textile sample had a lower amount of retained product sample at 70%, compared with an 81% for the untreated woven textile samples.

Under the cooling temperature conditions and at rotational speed of 66 rpm, only the knitted textile samples reduced the amount of cheese sauce, compared with the rest of the surfaces tested. Treated superhydrophobic textile sample was found to have the lowest percentage of retained cheese sauce at 47%, while 80% was the amount of the residual product on the untreated knitted textile sample. Similar results were observed for the tests with the rotating speed of 88 rpm and at cooling temperature conditions. Under these experimental conditions, only the knitted textile samples reduced the amount of the tested cheese sauce. The residual mass of the food product on the treated and the untreated knitted samples was 39% and 80%, respectively. On the other hand, at room temperature condition and with the maximum rotating speed, additional examined food-contact surface samples exhibited a repellent behavior towards cheese sauce. Superhydrophobic treated knitted textile sample had the lowest amount of residual cheese sauce with a value of 11%. Superhydrophobic treated
woven fabric and untreated knitted fabric had the amounts of the residual product of 33% and 36%, respectively. A higher percentage of the residual food product, around 80%, was observed for PTFE, silicone fiberglass sheets and the untreated woven textile samples. Finally, of the amount of residual cheese sauce on Rain-X treated stainless steel was 94% while of 100% of cheese sauce was retained on the surface of the rest of the examined samples (Fig. 5.29; Fig. 5.30; Fig. 5.31; Fig. 5.32).

Figure 5.29. Images of residual cheese sauce, at the end of centrifuge product adhesion tests, at room temperature, on the surface of: a) untreated stainless steel, b) Rain-X, and c) fluoropolymer coated polyester knitted textile.
Figure 5.30. Effect of food-contact materials on the product mass retained on the surface during the centrifuged product adhesion reduction tests for cheese sauce, at room temperature.

Figure 5.31. Images of residual cheese sauce, at the end of centrifuge product adhesion tests, at cooling temperature conditions, on the surface of: a) untreated stainless steel, b) Rain-X, and c) fluoropolymer coated polyester knitted textile.
Figure 5.32. Effect of food-contact materials on the product mass retained on the surface during the centrifuged product adhesion reduction tests for cheese sauce, under cooling temperature conditions.

Results of these experiments were similar to the observation of Stoforos et al (2016) on thermal mixing using mechanical vibration with the same selection of examined food materials. Similar to the previous study, sweet potato puree, among other tested food materials, was easier to mix or repel, independent from the food-contact surface characteristics. For banana puree, surfaces with more hydrophobic characteristics, namely Aculon treated stainless steel and the treated knitted textile samples, exhibited better repellent behavior. Cheese sauce, particularly under the cooling processing temperature conditions showed the most adhesive behavior. Finally, for all the products tested, textile samples with hydrophobic/superhydrophobic behavior, and knitted surface topography showed the best
repellent behavior, an indication of the surface characteristics needed for potential applications in thermal mixing or for a combination of mixing and cooling processes.

5.4. Conclusions

This work studied the effect of different surface characteristics on the adhesive behavior of highly viscous food products, namely sweet potato puree, banana puree, and cheese sauce, for potential applications in cooling and mixing processing. To examine the effect of the surface characteristics on the adhesive behavior of these foods, different engineered food-contact materials were examined for their surface wettability and roughness. The examined food-contact surface samples included stainless steel samples, untreated and modified using chemical and lubricant surface treatments (Rain-X, Aculon, Sprayon LU206), fabrics (knitted and woven structures, untreated and treated with a commercial fluorocarbon-based resin), polytetrafluoroethylene (PTFE) sheets, silicone sheets and fiberglass-reinforced silicone polymer sheets.

The examined surface samples covered a wide range of surface characteristics, measured by contact angle and Atomic Force Microscopy tests. Surface wettability was one of the parameters used to differentiate the examined surfaces, varying from hydrophilic (0-90°) (Rain-X treated stainless steel), low hydrophobic scale (90-100°) (untreated stainless steel, Sprayon LU206 treated stainless steel), hydrophobic (Aculon treated stainless steel, PTFE, silicone sheets, fiberglass silicone sheets, untreated textiles) up to superhydrophobic (150°) (fluorocarbon treated textiles) behavior. Surface roughness was the second parameter used to quantify the differences between the surface samples, having samples with a very smooth surface such as Aculon and Sprayon LU206 treated stainless steel samples and with the highly
rough surface of silicone sheets. The rest of the samples had an intermediate surface roughness but higher than the untreated stainless steel surface used as the reference sample for this study.

The adhesive behavior of the tested food materials against the engineered surfaces was examined by comparing the percent of residual mass product on the food-contact materials, using the product depletion and the centrifuge adhesion tests. The two testing methods were performed under two different temperature conditions, at room temperature (20-23 °C) and at cooling processing temperature conditions with the initial temperature of the examined food samples at 95-100 °C. During the experiment at cooling temperature conditions, the initial temperature of the food-contact surface samples was at 5 °C for product depletion tests and while the surface temperature was maintained within a temperature range of 17-20 °C during the centrifugal adhesive tests.

Results of product depletion tests showed that sweet potato puree and cheese sauce exhibited similar adhesive behavior, but with a higher amount of residual product retained on the examined surfaces comparing with banana puree. The results of product depletion test at room temperature showed that for hydrophobic smooth surfaces (Aculon and Sprayon LU206 treated stainless steel samples), hydrophobic PTFE sheets and superhydrophobic knitted textiles all the tested foods presented the least adhesive behavior, with an average residual mass of 35-40%. Similar, at cooling conditions, the same food-contact materials retained the least amount of product, with an average residual product of 35-45%, 60-70%, and 55-75%, for banana puree, sweet potato puree, and cheese sauce respectively.

Time-temperature data of the highest surface temperature was recorded during product depletion tests at cooling temperature conditions. Comparing the temperature difference (ΔT), between the initial and the final temperature, for each tested product and examined food-
contact surface, the untreated and chemically treated stainless steel samples, additionally with PTFE presented the highest $\Delta T$ values for banana puree and sweet potato puree at 35-38 °C. Cheese sauce was the food product with lowest $\Delta T$ values for all the food-contact surfaces used, with only the hydrophobic smooth Aculon treated stainless steel sample exhibiting a $\Delta T$ higher than 35 °C. Product solidification observed for cheese sauce on the surface of the cooled examined surface samples, had a negative impact on product depletion, also resulting in lower $\Delta T$ values.

Furthermore, the adhesive behavior of the tested foods and repellent behavior of the engineered food-contact surfaces were examined using the centrifugal adhesive test. Under two different temperature conditions, three different rotating speeds at 44 rpm, 66 rpm, and 88 rpm were tested for each food sample and contact surface material. Increasing the speed of rotation, the applied centrifugal force also increased, resulting in a lower amount of retained product on the surface of the rotating surface sample. Independent from the speed of rotation and the examined food-contact surface, sweet potato puree exhibited the least adhesive behavior, with the residual product of 0-10% and 10-20%, at room temperature and cooling conditions, respectively. Repellent behavior with banana puree, for the hydrophobic surfaces treated with Aculon stainless steel and the knitted fabric samples, retained the least amount of product on the surface. For these surface samples and with the rotation speed of 66 rpm or higher, the residual banana puree product was at 0-4% and 15-32%, at room and cooling temperature conditions, respectively. Finally, for cheese sauce, only the superhydrophobic and hydrophobic knitted fabric samples reduced the amount of the residual product on the surface of the rotating sample, under all temperature conditions. The least amount of retained cheese sauce was observed with the superhydrophobic knitted fabric samples, at the maximum speed
of rotation, with the residual at 11% and 39%, at room temperature and cooling temperature conditions, respectively.

In conclusion, this study showed that the commonly used food-contact surface of untreated stainless steel probably does not have the ideal food-contact surface characteristics for thermal processing and mixing applications, especially cooling under continuous flow conditions. Moreover, the results of this research showed that hydrophobic surfaces with smooth characteristics and the more rough surfaces, with more complex topography, and hydrophobic/superhydrophobic behavior are the ideal surfaces for cooling and mixing application, respectively. However, for potential processing applications, to mimic these surface characteristics, more long-term stable and food-grade surface modification techniques or treatments will be required.

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Nomenclature

*Latin letters*

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>AFM</td>
<td>Atomic Force Microscope</td>
</tr>
<tr>
<td>CA</td>
<td>Contact Angle</td>
</tr>
<tr>
<td>(F_c)</td>
<td>Centrifugal force (N)</td>
</tr>
<tr>
<td>H</td>
<td>Height (m)</td>
</tr>
</tbody>
</table>
L  Length (m)

mass  Mass (kg)

MW  Microwave system

O.D.  Outside Diameter (m)

r  Radius (m)

Ra  Arithmetic average surface roughness (nm)

RZD  Mean peak-to-valley height surface roughness (μm or mm)

T  Temperature (°C)

t  Time (s)

t<sub>exit</sub>  Time required for the food samples to exit the consistometer apparatus

W  Width (m)

Greek letters

$$\Delta T$$  Temperature difference (°C)

$$\varepsilon$$  Absolute roughness (μm or mm)

$$\theta$$  Incline angle of the consistometer units (°)

$$\theta_w$$  Equilibrium static contact angle of water (°)

$$\theta_{oil}$$  Equilibrium static contact angle of oil (n-dodecane) (°)

Subscripts

w  refers to the equilibrium static contact angle of water

oil  refers to the equilibrium static contact angle of oil (n-dodecane)
References


CHAPTER 6:

Part 2: Effects of Food-Contact Surface Characteristics on Flow of Viscous Products for Potential Applications in Advanced Thermal Processing

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Short version of title: Effects of Food-Contact Surface Characteristics on Flow of Viscous Products…


**Abstract**

This research involved the study of the effects of different surface characteristics such as wettability, roughness, and topography on adhesive and flow behavior of Carboxymethyl Cellulose (CMC) solution (conc. 1.5%). Pressure drop and velocity profile of highly viscous 1.5% CMC solution, flowing within the annular passage of a tube in tube heat exchanger, were used to compare the effects of the examined surfaces on adhesive and flow behavior, respectively. Different food-contact materials, such as untreated and chemical treated stainless steel, polytetrafluoroethylene (PTFE), silicon-based sheets, and untreated and treated with fluorocarbon-based resin, knitted and woven fabrics were studied. The examined surfaces were differentiated based on surface wettability (hydrophilic, hydrophobic, or superhydrophobic) and surface structure (roughness and topography). The pressure drop across the heat exchanger was estimated and used as an indirect measurement of the presence of product deposit at the pipe wall. Highly rough hydrophobic surfaces (silicone sheets and untreated woven textile samples), exhibited the highest pressure drop across the system with a value of 75842.4 Pa (12.0 psia). While smooth surfaces (Aculon and Sparyon treated stainless steel) and samples with superhydrophobic characteristics (treated knitted and woven textile samples) presented the lowest pressure drop at 48263.3-55158.1 Pa. Finally, the velocity profile of 1.5% CMC was studied, using particle tracking velocimetry (PTV) as the testing method. The PTV results on the velocity profile of 1.5% CMC, showed a parabolic velocity profile for almost all the examined cases, with the fastest flowing particle be close to the center of the annulus flow.

**Key words:** Surface modification, pressure drop, particle tracking velocimetry
6.1. Introduction

Fouling and accumulation of unwanted food material, such as proteins, starches, and minerals on the surface of thermal processing units (heat exchangers, mixing units, holding tubes, spray dryers, food packaging) reduces the processing efficiency and increases the operation cost. Product deposition and fouling can occur in any liquid-solid surface and has a significant impact on heat transfer efficiency and pressure drop across the processing unit (Balasubramanian and Puri, 2008; Balasubramanian and Puri, 2009; Barish and Goddard, 2013; Barish and Goddard, 2014; Stoforos et al., 2017). Formation of unwanted fouling layer, with a low thermal conductivity, on the surface of processing equipment, such as heat exchangers, increases the resistance to heat transfer reducing the processing efficiency and reduces the cross-sectional area increasing the pressure drop across the processing unit (Swartzel, 1983; Balasubramanian and Puri, 2009; Awad, 2011). Moreover, accumulation of unwanted material on food processing surface is prone in formation of biofilm, resulting in an extensive use of chemical detergents and sanitizers and frequent food processing plant shutdowns for sanitation and cleaning (Sandu and Singh, 1991; Kananeh et al., 2010; Mérian and Goddard, 2012; Barish and Goddard, 2014).

In food and dairy industries, different types of fouling can occur due to the different physical, chemical interaction between the fluid-solid interfaces and involve problems such as protein aggregation, minerals deposition, fat burn-on in the heat exchanger, product solidification on cooled surfaces and growth of biofilm (Awad, 2011, Goode et al., 2013). The different types of fouling in the area of food and dairy processing can be categorized in the following list (Awad, 2011, Goode et al., 2013): 1) particulate or sedimentation fouling (deposition of suspended particles), 2) crystallization or precipitation fouling (dissolved salts
from saturated solution), 3) chemical reaction fouling (cooking, polymerization, etc...), 4) corrosion fouling (electrochemical reactions), 5) biological fouling or biofouling (deposition and growth of organic films from microorganisms), and 6) solidification or freezing fouling (deposition and formation of a solid fouling deposit onto a subcooled surface, from fluid components with a high melting point freeze).

Moreover, fouling and product build-up is a problem mainly been observed during conventional thermal processing (heating, cooling, and thermal mixing) of highly viscous and multiphase food products, such as fruit and vegetable purees, soups, and typical dairy products, (Stoforos et al., 2016; Stoforos et al., 2017). Unfortunately, surface characteristics (high surface energy and surface wettability) of stainless steel, the most commonly used material for food processing equipment are prone to the adhesive behavior of highly viscous foods (Goode et al., 2013; Barish and Goddard, 2013; Stoforos et al., 2016). To prevent fouling formation and improve hygiene of food-contact surfaces, surface modification techniques, such as polymer and nanoparticle coatings (poly (ethylene glycol, silica or fluorocarbon and polytetrafluoroethylene (PTFE)), and physicochemical surface modification techniques such as plasma-enhanced chemical vapor deposition, and electro-chemical and ion implantations have enabled researchers successfully to modify stainless steel with antifouling and shelf cleaning characteristics (Santos et al., 2004; Mérian and Goddard, 2012). Surface properties such as wettability (contact angle, spreadability), surface roughness and topography, have been modified to produce functional surfaces that mimic the self-cleaning and repellent characteristics of natural antifouling materials, such as the lotus leaf (Nelumbo nucifera) (Santos et al., 2004; Rosmaninho et al., 2007; Balasubramanian and Puri, 2009; Kananeh et al., 2010; Barish and Goddard, 2013). Antifouling modified surfaces, with characteristic low
surface free energy (weak attractive forces), and smooth surfaces with specific finish topography, have successfully been used for food and dairy products, improving the operation and cleaning efficiency of the processing units (plate heat exchangers, tube in tube heat exchangers), while reducing the bacterial adhesion (Beuf et al., 2003; Santos et al., 2004; Premathilaka et al., 2006; Rosmaninho et al., 2007; Balasubramanian and Puri, 2009; Kananeh et al., 2010; Mérian and Goddard, 2012; Barish and Goddard, 2013; Goddard et al., 2013; Cheng et al., 2015; Stoforos et al., 2017). Low surface free energy (related with hydrophobic and superhydrophobic characteristics), such as polymer and ceramic coatings, have longer induction periods than wettable (high free surface energy, hydrophilic characteristics), resulting in lower amount of product deposition. On the other hand, the higher roughness of the surface increases the contact area of the surface, providing suitable sites for nucleation, which are more favorable for the initial steps of fouling (Awad, 2011, Goode et al., 2013).

Moreover, the developments in thermal processing with advanced volumetric heating technologies (continuous flow microwave, ohmic heating, and radio frequency heating), provide uniform rapid volumetric heating which reduces the come up time, minimizes the final food product quality losses and lowers the product and fouling deposition (Coronel et al., 2005; Bansal and Chen, 2006; Steed et al., 2008; Cullen et al., 2012). However, cooling part is still implemented using inefficient conventional cooling methods, resulting in a slow and unequal thermal treatment, and reduction of final food product quality (Stoforos et al., 2016; Stoforos et al., 2017). A recent study on the enhancement of cooling processing has compared the cooling and cleaning process for viscous foods, such as sweet potato puree, banana puree and cheese sauce between an untreated stainless steel and a hydrophobic treated stainless steel heat exchanger (Stoforos et al., 2017). This study used a chemical hydrophobic treatment on the
surface of stainless steel to reduce the amount of product accumulation and solidification, on the wall pipe wall, and improve the performance of the heat exchanger, by enhancing the cooling and cleaning efficiency. The results showed a significant improvement on cleaning processing with the hydrophobically treated heat exchanger for all the products tested. The cooling process was significantly enhanced using the treated heat exchanger for cheese sauce and banana puree, while the untreated heat exchanger exhibited the best performance for sweet potato puree (Stoforos et al., 2017). Based on these observations and the different behaviors observed for the different viscous foods, the first part of this research project examined the effects of different surface characteristics on the adhesive behavior of sweet potato puree, banana puree, and cheese sauce at room temperature and at cooling processing temperature conditions. (Stoforos et al., 2017b). The examined surfaces were differentiated based on surface wettability (hydrophilic, hydrophobic, or superhydrophobic) and surface roughness and topography. The results of this study showed that the hydrophobic and superhydrophobic surfaces with smooth characteristics (chemically treated stainless steel samples), and non-stick materials such as PTFE, retained the least amount of product and recommended as the food-contact surfaces for potential cooling applications.

This study is a continuation of the first part of this research project (Stoforos et al., 2017b), focusing on the effects of different surface characteristics on pressure drop and velocity profile of highly viscous products, for potential applications in continuous flow cooling processing. For that reason, in the current research, pressure drop and velocity data were collected and used as the testing methods to compare the effects of the different examined food-contact surface samples on the adhesive behavior and velocity profile, respectively. Anticipating that the results from this research project will quantify the surface parameters for
potential design and development of new engineered surfaces that will reduce the product accumulation at the pipe wall and result in enhancement of heat transfer and reduction of pressure drop across the apparatus.

6.2. Materials and methods

The current research examined the effects of different surface characteristics on pressure drop and velocity profile of viscous products. For that reason, different food-contact surfaces were fabricated, varying on surface wettability, roughness and topography. To study and quantify, the effects of the examined food-contact surfaces on the adhesive behavior and flow of the viscous samples, experiments were conducted to collect data on pressure drop ($\Delta p$ (Pa), volumetric flow rate ($Q$ (m$^3$/s)), and flow visualization across the apparatus, a tube in tube heat exchanger.

6.2.1. Test materials

To study and understand the effect of the chemical and physical characteristics of the surface of the boundary walls on the adhesive behavior and velocity profile of viscous fluids, eleven different engineered food contact materials were fabricated and tested, the same as in the previous study (Stoforos et al., 2017 b). The studied materials included (1) stainless steel samples, untreated and modified using chemical surface treatments (Rain-X, ITW Global Brands, Houston, TX, U.S.A.; Aculon, Aculon performance Surface Solutions, San Diego, CA, U.S.A.; Sprayon LU206 Silicone Lubricate Spray, Sprayon, Cleveland, Ohio, U.S.A.), (2) fabrics (knitted and woven structures, with a base weight 2.9·10-5 kg and 2.3·10-5 kg, respectively) untreated and treated with a commercial fluorocarbon-based resin, (3) PTFE
sheets, (4) poly-silicone sheets and (5) fiberglass reinforced silicone polymer sheets (Stoforos et al., 2017b).

The examined surface samples covered a wide range of different surface characteristics, based on surface wettability, roughness, and topography. The differences between the examined surfaces were quantified using contact angle and Atomic Force Microscopy (AFM) measurements, for surface wettability and roughness, respectively. Surface wettability was one of the parameters differentiate the examined surfaces, varying from hydrophilic (0-90°) (Rain-X treated stainless steel), low hydrophobic scale (90-100°) (Untreated stainless steel, Sprayon LU206 treated stainless steel), hydrophobic (Aculon treated stainless steel, PTFE, silicone sheets, fiberglass silicone sheets, untreated textiles) up to superhydrophobic (~150°) (fluorocarbon treated textiles) behavior. Moreover, surface roughness was the second parameter differentiate the examined surface samples, having materials with a smooth surface (Aculon and Sprayon LU206 treated stainless steel) and with high surface roughness such as the silicone sheets samples. The rest of the examined materials had an intermediate surface roughness but higher than the untreated stainless steel used as the reference sample for this study (Stoforos et al., 2017b).

Furthermore, to examine the effects of the different engineered surface samples on pressure drop and velocity profile of viscous products, Carboxymethyl Cellulose (CMC) (TIC Gums, Inc., Belcamp, MD, U.S.A.) solution (conc. 1.5%) was used as the examined liquid. CMC at 1.5% concentration was chosen as the carrier fluid for this part of the experiments, due to the characteristic high viscosity and the transparency that required for the velocity profile measurements. At room temperature (~20 °C), the tested 1.5% CMC solution had a density of
1051 kg/m³. At the same temperature conditions, rheological parameters of 1.5% CMC solution were measured in order to calculate the flow behavior parameters of the solution.

6.2.2. Experimental methods

This research studied the effects of different surface properties (material, wettability, topography) on product deposition and flow in a heat exchanger during continuous flow of viscous products. Data of pressure drop and velocity were collected and used as the testing measurements to compare the effects of the different examined surface samples on the adhesive behavior and the flow of 1.5% CMC solution at room temperature (18-20 °C), respectively. The experiments with CMC were conducted at room temperature, having the solution at the highest viscosity, for more accurate calculation and comparison of pressure drop data. Pressure data were collected at the inlet and the exit of the heat exchanger, to calculate the pressure drop across the heat exchanger system, (Δpᵟ). While the pressure drop across the annulus passage of the heat exchanger was calculated, using the measurements of volumetric flow rate, and rheological parameters of 1.5% CMC solution. The volumetric flow rate of 1.5% CMC solution, was measured during the different experiments and used to compare the flow of the viscous solution against the different surfaces. The volumetric flow rate of CMC was measured for all the experiments while having the pump speed constant. Finally, for the velocity data, Particle Tracking Velocimetry (PTV) technique was used as the testing method. During PTV tests, particles were introduced to the carrier fluid, capturing the particle motion through video. The video recorded data was analyzed and used to determine the velocity profile of the tracked particles, through a Matlab software algorithm.
For the experiments of this research, a heat exchanger system, in recirculation mode, was assembled. It consisted of a food grade progressive cavity pump (Seepex MD-012 12, Fluid Engineering Inc., Birmingham, AL, U.S.A.), and a tube in tube heat exchanger. The tube in tube heat exchanger was built with a total length (L), of 1.11 m and with a 0.051 m (2 inches) and 0.076 m (3 inches) inner (I.D.) and outer (O.D.) tube diameter, respectively. Approximately 0.011 m³ (3 gallons) of CMC solution, were sufficient to fill the system and allow for recirculation of the test samples, in the annulus passage of the heat exchange, while water, at room temperature (~20 °C), was flowing, in a counter current mode, within the tube with a constant flow rate of $63.1 \times 10^{-5}$ m³/s (10 gal/min), in order to control the food-contact surface temperature. For all the different experiments with the different surface samples, the annulus side of the heat exchangers was used as the flow passage for the carrier fluid, a flow configuration recommended for improving cooling efficiency of food products, by increasing the heat loss to the environment, beneficial for cooling, while improving the sanitation conditions via elimination of the sweating pipe phenomenon (Stoforos and Simunovic, 2016).

Furthermore, the outside surface of the inner tube of the heat exchanger was used as the tested food-contact surface, applying on it all the different surface modification material and methods examined. The O.D. of the tested tube samples remained unchanged after application of the chemical surface treatments, while the O.D. of the tested tubes was slightly increased for the tested tubes coated with textile and polymer samples. The thickness of the inner tube was measured for each examined surface, to determine the O.D. of the tube for each case. On the other hand, the same outer tube apparatus of the heat exchanger was used for all the different experiments for the different examined surface treatments. The outer tube apparatus was built using 3 inches O.D. side tube glasses and sanitary stainless steel tee fitting
pipe. The sanitary stainless steel tee fitting pipe was used as the fitting of a 3 inches stainless steel end cap, which was modified to attach two 2M endoscope cameras (Depstech, Shenhai yuanhang Investment Co, Shenzhen, China) for the in-line video recording of particle motion. The two endoscope cameras were placed in the annulus of the heat exchanger, in a position to record the side cross sectional view of the fluid flow, with frames per second (FPS) rate of 30.

Furthermore, during the PTV experiments, particles were injected into the carrier fluid at the inlet of the heat exchanger, using three different 5 mL syringes. The syringes were attached on a modified gasket used for the mechanical fitting of the outer tube of the heat exchanger with the processing system (Fig. 6.1). The tip of each syringe was located at a different radial location of the annulus passage of the heat exchanger, close to the wall of the inner tube, at the center of the annulus cross-sectional area and close to the outer tube pipe wall, respectively. The particles were introduced at three different cross-sectional points, in order to study and compare the velocity profile of the flow and to determine the type of the flow near the wall (slip or non-slip), for all the different examined surface treatments. The particles for the PTV studies were made by mixing at room temperature of 40 mL of banana puree (Aseptia/Wright Foods, Troy, NC, U.S.A.), 5 mL of fluorinated red water tracking dye (Cole-Paramer instrument company, Vernon Hills, IL, U.S.A.), and 0.02 g of sodium alginate (Chem Center, La Jolla, LA, U.S.A.). Banana puree was used as the base ingredient of the mix, due to the similar density with the test 1.5% CMC solution, approximately 1030 kg/m³. Fluorinated color dye was added to the mixer to increase the brightness intensity of the particles for the PTV Matlab analysis, and finally, the small amount of sodium alginate was used for the formation of the particles, with a diameter of ~1mm.
Finally, two pressure gauges were installed, at the inlet and the outlet of the heat exchanger system, to measure the pressure at each point, and calculate the pressure drop across the heat exchanger ($\Delta p_s$) based on the collected data.

Figure 6.1. a) and b) Schematic diagram and picture of the closed-loop, recirculating experimental system installation for testing and recording, PTV and pressure drop data during experiments with 1.5% CMC solution flowing within the annulus passage and the examined modified food-contact surfaces. c), and d) the location of endoscope cameras and particle injection point in the experimental system.

6.2.3. Pressure drop

Pressure drop data were used to study the effect of the different examined food-contact materials on the adhesive behavior of viscous products under continuous flow conditions.
Pressure drop and pressure drop changes over time can be used as an indirect measurement of the presence of product deposit at the pipe wall (Awad, 2011). Product deposition, at given flow rate, will increase the pressure drop across the apparatus, by reducing the flow area. Pressure drop data across the system and across the annulus passage of the heat exchanger were used to compare the adhesive behavior of 1.5% CMC solution against the examined surface samples.

The pressure data collected at the inlet and the outlet of the heat exchanger system were used to calculate the pressure drop across the system. While the pressure drop across the flow in concentric annular passage was estimated using Eq. 6.1 (White, 2011):

$$\Delta p = 2 \cdot f \cdot \left(\frac{L}{D_h}\right) \cdot (\rho \cdot u^2)$$

(6.1)

Where $L$ is the length of the annulus side of the heat exchanger at 0.50 m, $\rho$ is the density (kg/m$^3$) of the fluid flowing within the annulus (1.5 % CMC solution for this study), $D_h$ is the hydraulic diameter (m) given by Eq. 6.2, and $u$ is the velocity of fluid (m/s) (Eq. 6.3) and $f$ is the friction factor, which for laminar flow in concentric annulus is given by Eq. 6.4. (White, 2011).

$$D_h = 2 \cdot (r_2 - r_1)$$

(6.2)

Where $r_1$ (m) and $r_2$ (m) are the outside radius of the inner and the inside radius of the outer tube of the heat exchanger, respectively. The O.D. of the inner tube was different for each experiment, varying based on the different examined material. While the inside diameter of the outer tube was the same for all the experiments, at 0.073 m (2.86 in).

$$u = \frac{Q}{\pi \cdot (r_2^2 - r_1^2)}$$

(6.3)

Where the denominator of Eq. 6.3 is the cross sectional area of the annulus passage.
\[
f = \frac{64 \cdot \zeta}{N_{GRe}} \quad (6.4)
\]

Where \(\zeta\) is a correction factor for the hydraulic diameter, given from tables (White, 2011) and depends on the aspect ratio between the inner and the outer tube radius. For this case, \(\zeta\) had value of 1.498, for all the tested materials. \(N_{GRe}\) is the generalized Reynolds number for non-Newtonian fluids for a concentric annulus and is given by the following equation (Eq. 6.5):

\[
N_{GRe} = \frac{\rho \cdot u^{2-n} \cdot D_h^n}{K \cdot \left[ \frac{3n+1}{4n} \right]^n \cdot 8^{n-1}} \quad (6.5)
\]

Where \(K\) is the flow consistency index (Pa·s\(^n\)), and \(n\) the flow behavior index (dimensionless) for 1.5 \% CMC solution. Rheological parameters, shear stress, shear rate and apparent viscosity, of 1.5 \% CMC solution were measured with a MCR 302 rheometer (Antoon Paar AB, Lund, Sweden), at room temperature, using the serrated bob and cup measuring system. The rheological parameters of 1.5 \% CMC solution, were measured, at room temperature conditions. To estimate flow consistency index and flow behavior index the following equation was used (Eq. 6.6):

\[
\sigma = K \gamma^n \quad (6.6)
\]

For \(\sigma\) being the shear stress (Pa), \(\dot{\gamma}\) the shear rate (s\(^{-1}\)), \(K\) the flow consistency index (Pa·s\(^n\)), and \(n\) the flow behavior index (dimensionless). Applying the natural logarithms on both sides of Eq. 6.6, we get the following equation (Eq. 6.7):

\[
\ln \sigma = K + n \cdot \ln \dot{\gamma} \quad (6.7)
\]
The flow consistency index and the flow behavior index were estimated from a plot of ln(σ) versus ln(γ). The slope of the straight line drawn on this graph is equal to the flow behavior index while the intercept gives the value of the ln(K).

6.2.4. Velocity profile

To determine the velocity profile of 1.5% CMC solution flowing in the annulus passage of the heat exchanger, during the different experiments, with the outer surface of the inner tube having a different examined food-contact surface sample, PTV was used as the testing method. Fluorescent particles were introduced at the inlet of the heat exchanger in the three different locations in the annulus flow. Video microscopy technique of PTV was used to capture the movement of the tracer particles. The video microscopy analysis started by converting the recorded video into a sequence of gray-scale frames, using the Matlab software (PTVlab_GUI (Brevis et al., 2011)). From the resulted sequence, 15-20 of the frames, with the brightest and clearest images were selected for the PTV data analysis, using a Matlab algorithm code (Appendix A). The Matlab algorithm was used to locate the tracing particles in each frame examined, based on the pixel brightness. The next step was to identify the same particles in neighboring frames and create particle trajectories, as a function of time (Furst, 2017). The data of the particle trajectories from the previous step was used to calculate, in pixels\(^2\), the mean-square displacement (msd) of the tracing particles over a lag time (expressed as a number of frames) (Furst, 2017). Finally, calibration is required to convert the \(\sqrt{msd}\) term from pixels in real displacement (m) and the lag time to real time (s) based on the frame rate (FPS is 30 for this case) of the camera used (Furst, 2017). After calibration and knowing the real displacement
(MSD) of the particle over the real lag time ($\Delta t$), velocity of each particle ($u_p$) was estimated from the following equation (Eq. 6.8) (Monnier et al., 2012):

$$u_p = \frac{MSD}{\Delta t} \quad (6.8)$$

Knowing the velocity and the location of each tracing particle, velocity ($u$) versus the distance from the outer surface of the inner tube ($d$) was plotted to get the velocity profile of 1.5% CMC flowing in the annulus, for each examined food-contact surface sample.

**Locating particles**

The resulted images, from the conversion of the recorded video into gray-scale frames, have an 8-bit depth, with the brightness intensity of the gray-scale pixels to be between 0 and 255. The Matlab algorithm identifies as particles the pixels with a brightness intensity value at 255. Moreover, to correctly identify and locate the tracing particles in each examined frame, the following Matlab functions were used: 1) bpass.m was used before the particle location process, to clear the noise from the background of the examined frames, 2) pkfnd.m was used to identify and locate candidate particles, if no other pixel within a specific distance (defined in the algorithm as the expected radius of the particles), is brighter. 3) cntrd.m was applied for refinement of particle location from the results of pkfnd.m, by calculating the centroid location of the intensity distribution around the central brightest pixel (intensity of 255) (Fig. 6.2).

**Particle trajectories**

In the previous step, using the bpass.m, pkfnd.m, and cntrd.m functions, a list of particles position was created for each frame. The data with particles position list is used from the function track.m to generate particle trajectories as a function of time. Track.m uses the
particles position list for each frame to identify the same particles between neighboring frames and link the particle positions in each frame into particle trajectories, as a function of time.

**Particle mean squared displacement calculation**

The output data from the track.m, an array of particle position as a function of lag time steps, are processed with the MSD.m function to estimate the mean-squared displacement of the trajectories estimated. MSD.m calculates the mean-squared displacement from a trajectory, by dividing this trajectory into displacements, at the corresponded lag time. MSD.m repeats this calculation for all the trajectories. Note that MSD.m function estimates the mean-squared displacement using not-overlapping lag times. The output data of the MSD.m, an array consisted of three columns i) the estimated msd ii) the corresponded lag time, and iii) the number of observation per data point (lag time). The estimated results of msd, as a function of time, were statistically processed with the position data from the particle trajectories to link the msd data with the tracer particles and calculate the velocity of each particle. Finally, calibration is required to convert the displacement from pixels to real distance (m) and the lag time to real time, using the FPS from the camera used.

**Calibration**

To express the $\sqrt{msd}$ results from pixels to real displacement (m), a reference image is used, with a known real distance inside the frame. The matlab function imtool is used with the reference image to measure and express the known real distance into pixels.
6.3. Results and discussion

6.3.1. Pressure drop

This study tested the effects of different food-contact materials on the adhesive and flow behavior of 1.5% CMC solution by studying and comparing pressure drop across the system and the annulus, and the volumetric flow rate of the product for each case. To calculate pressure drop across the annulus passage of the heat exchanger for each examined food-contact surface, the outside radius of the inner tube and the volumetric flow rate of 1.5% CMC were measured for each case. Additionally, the rheological parameters of 1.5% CMC were measured, to determine the flow consistency index and the flow behavior index of the solution.
Initially, rheological tests were performed to calculate the flow consistency index and the flow behavior index of the 1.5% CMC solution. The tests were performed in triplicate. Based on the plot of $\ln(\sigma)$ versus $\ln(\dot{\gamma})$, 1.5% CMC solution exhibited a non-Newtonian, shear thinning behavior with a flow behavior index of $0.361 \pm 0.003$, and a highly viscous behavior with a flow consistency index of $28.7 \pm 0.2$ Pa s (Fig. 6.3). One should notice the low standard deviation values for both flow consistency and flow behavior indices, being less than 0.1 and 0.05, respectively, indicating good repeatability of the rheological tests.

Figure 6.3. Rheological behavior of 1.5% CMC solution, at room temperature.
The outer surface of the inner tube of the heat exchanger was used as the applied surface for all the examined food-contact materials. Stainless steel tube samples with a total length of 1.11 m and O.D. of 0.51 m (2 in), were used as the samples to apply on the different examined food-contact surfaces. The outside radius of the inner tube remained unchanged, at 0.2555 m (1.00 in), for the tested samples of the untreated stainless steel, and for all the chemical treated samples of Rain-X, Aculon, and Sprayon LU206. The outside diameter of the inner tube was increased at 0.0263 ± 0.1 m for the all the textile samples, PTFE, and silicone fiberglass sheets surface samples. Silicone sheets were the examined material with the highest value of outside radius of the inner tube at 0.0269 m (Table 6.1).

The flow rate of 1.5% CMC solution was measured during each experiment with the different food-contact surfaces tested while having the speed of the pump constant. The highest volumetric flow rate of 1.5% CMC, at 6.31×10^{-5} \text{ m}^3/\text{s} (1 \text{ gal/min}), was observed in the samples with the lowest surface roughness (untreated, the Aculon and Sprayon LU206 treated stainless steel samples). The lowest volumetric flow rate of 1.5% CMC was observed for the surface samples of untreated woven and knitted textiles and for the surfaces with the highest surface roughness, (silicone sheets, silicone fiberglass sheets) with a flow rate of 5.01×10^{-5} \text{ m}^3/\text{s} (~0.8 gal/min), 5.24×10^{-5} \text{ m}^3/\text{s} (~0.8 gal/min), 5.12 ± 0.1 ×10^{-5} \text{ m}^3/\text{s} (~0.8 gal/min), respectively. While, an intermediate volumetric flow rate of 1.5% CMC was measured for the samples of PTFE and the treated textile values, at 5.84×10^{-5} \text{ m}^3/\text{s} (~0.9 gal/min), and 5.54 ± 0.2×10^{-5} \text{ m}^3/\text{s} (~0.9 gal/min), respectively (Table 6.1).

The pressure drop across the heat exchanger system was estimated by using the data collected at the inlet and the outlet of the heat exchanger system, during the experiments with the examined food-contact materials. No difference over the time on the measured pressure
was observed for all the cases. The highest pressure drop across the system was observed for high rough hydrophobic samples of silicone sheets and untreated woven textile samples with a value of 75842.4 Pa (12.0 psia). The pressure drop across the system for the rest of the surface samples was approximately around 48263.3-55158.1 Pa (7-8 psia), with the lowest value to be observed for the samples with the smooth surface (Aculon and Sparyon treated stainless steel) and superhydrophobic (treated knitted and woven textile samples) characteristics.

The pressure drop across the concentric annular passage was estimated based on the collected data of volumetric flow rate and the rheological parameters of 1.5% CMC, and the dimensions of the annulus side of the heat exchanger. Using the Eq. 6.1-6.5, for a laminar flow (the estimated $N_{GrRe} < 1$, for all the cases) in the annulus of power law fluid the pressure drop across the annulus of the heat exchanger was estimated for all the examined surfaces. The results presented that under laminar flow conditions, the most important factor in the pressure drop in the annulus is the ratio $r_2/r_1$. The surface samples (untreated, the Aculon and Sprayon LU206 treated stainless steel samples) with the lowest outside radius of the inner tube had the lowest pressure drop at 42846.9 Pa (~6.2 psia). As the outside radius of the inner tube increased, the pressure drop across the annular passage also increased. Silicone sheets surface was the sample with the highest pressure drop across the annulus at 49932.5 Pa (~7.2 psia). While the rest of the samples had an intermediate pressure drop across the annulus, around 43600-45800 Pa (~6.5 ± 0.3 psia).
Table 6.1. Inner tube radius ($r_1$), volumetric flow rate (Q) and pressure drop across the annulus ($\Delta p_{HX}$) and the heat exchanger system ($\Delta p_S$) for the different examined food-contact surfaces.

<table>
<thead>
<tr>
<th>Food-contact surface</th>
<th>$r_1$ (m)</th>
<th>Q (m$^3$/s)</th>
<th>$\Delta p_{HX}$ (Pa)</th>
<th>$\Delta p_S$ (Pa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Untreated stainless steel</td>
<td>0.0255</td>
<td>0.000063</td>
<td>42846.9</td>
<td>55158.1</td>
</tr>
<tr>
<td>Rain-X</td>
<td>0.0255</td>
<td>0.000063</td>
<td>42846.9</td>
<td>55158.1</td>
</tr>
<tr>
<td>Aculon</td>
<td>0.0255</td>
<td>0.000063</td>
<td>42846.9</td>
<td>48263.3</td>
</tr>
<tr>
<td>PTFE</td>
<td>0.0263</td>
<td>0.000058</td>
<td>47042.5</td>
<td>48263.3</td>
</tr>
<tr>
<td>Sprayon LU206</td>
<td>0.0255</td>
<td>0.000063</td>
<td>42846.9</td>
<td>48263.3</td>
</tr>
<tr>
<td>Silicone sheets</td>
<td>0.0269</td>
<td>0.000051</td>
<td>49932.5</td>
<td>75842.4</td>
</tr>
<tr>
<td>Silicone fiberglass sheets</td>
<td>0.0263</td>
<td>0.000051</td>
<td>44873.6</td>
<td>48263.3</td>
</tr>
<tr>
<td>Fluoropolymer coated polyester knitted textiles</td>
<td>0.0262</td>
<td>0.000056</td>
<td>45804.7</td>
<td>48263.3</td>
</tr>
<tr>
<td>Untreated polyester knitted textiles</td>
<td>0.0262</td>
<td>0.000052</td>
<td>44834.3</td>
<td>55158.1</td>
</tr>
<tr>
<td>Fluoropolymer coated polyester woven textiles</td>
<td>0.0263</td>
<td>0.000055</td>
<td>43692.5</td>
<td>48263.3</td>
</tr>
<tr>
<td>Untreated coated polyester woven textiles</td>
<td>0.0263</td>
<td>0.000050</td>
<td>44505.5</td>
<td>75842.4</td>
</tr>
</tbody>
</table>

6.3.2. Velocity profile

The effects of the different examined food-contact materials on the velocity profile of 1.5% CMC, flowing within the annulus side of the heat exchanger, was studied using PTV video microscopy as the testing method. PTV video microscopy was used to capture the flow
of fluorescent tracer particles. The resulted frames from video microscopy were processed using the Matlab algorithm to locate and calculate the msd of each tracer particle. Knowing the msd for each particle, the particle velocity was estimated by Eq. 6.8. Knowing the velocity, and the location of each tracing particle, velocity (u) versus the distance from the outer surface of the inner tube was plotted to get the velocity profile of 1.5% CMC flowing in the annulus, for each examined surface sample.

The PTV results on the velocity profile of 1.5% CMC, presented a parabolic velocity profile for almost all the examined cases, with the maximum velocity of the fluid streams to be close to the center of the annulus area, and the slowest fluid streams close to the inner and outer tube wall (Fig. 6.4-6.14). Similar parabolic velocity profile and lower particle velocity values, lower compared to the other case were observed for the untreated and the hydrophilic chemically treated stainless steel surface samples (Fig. 6.4; Fig. 6.5). The samples with the smaller cross-sectional flow surface area (PTFE, silicone sheets, silicone fiberglass sheets) presented the highest particle velocity at the center of the annulus passage, with values higher than 0.05 m/s (Fig. 6.7; Fig. 6.9; Fig. 6.10). The maximum velocity, at 0.058 m/s, was observed for the case of silicone sheets surface, the sample with the smallest flow cross sectional area. Furthermore, the lubricated smooth stainless steel surface, treated with Sprayon LU206, was another case with the particles flowing close to the center of the annular passage with a velocity higher than 0.050 m/s (Fig. 6.8). The woven textile samples presented similar velocity profile, with the particles velocities to be higher for the treated sample (Fig. 6.13; Fig. 6.14). For the case of the hydrophobic chemically treated with Aculon stainless steel surface and the superhydrophobic treated knitted textile, Most of the tracer particles to be concentrated in specific areas away for the inner tube wall. For the case of Aculon treated stainless steel the
tracing particles were spread in the area of the annular passage of the heat exchanger, with the most of the particles to be close to the outer tube wall (Fig 6.6). While for the case of the superhydrophobic knitted textile, the tracing particles were concentrated in a narrow area of the center of the annulus, with similar velocities (0.032-0.046 m/s) (Fig. 6.11). Finally, for the case of the untreated knitted textile surface, very slow fluid streams were observed close to the inner tube pipe wall, with the fastest particles to be located in the area close to the outer tube wall (Fig. 6.12).

Finally, based on the particle velocity data, not a clear conclusion can be made regarding the slip and slip flow condition close to the inner tube wall surface. The particle closest to the surface of the inner tube wall (~0.004 m) was observed for the case of the superhydrophobic treated woven textile, with the particle having a velocity lower than 0.005 m/s. Moreover, based on the velocity profile results (particles spread in the annulus) and the slowest flow streams, untreated and Rain-X treated stainless steel samples are expected to have the most efficient and uniform thermal treatment of the product across the annulus.
Figure 6.4. Velocity profile of 1.5% CMC, flowing within the annulus of the heat exchanger, with untreated stainless steel to be the examined outside surface of the inner tube. Note the points on the graph shows the velocity magnitude and location in the annulus of the heat exchanger of the tracing particles.

Figure 6.5. Velocity profile of 1.5% CMC, flowing within the annulus of the heat exchanger, with Rain-X treated stainless steel to be the examined outside surface of the inner tube.
Figure 6.6. Velocity profile of 1.5% CMC, flowing within the annulus of the heat exchanger, with Aculon treated stainless steel to be the examined outside surface of the inner tube.

Figure 6.7. Velocity profile of 1.5% CMC, flowing within the annulus of the heat exchanger, with PTFE to be the examined outside surface of the inner tube.
Figure 6.8. Velocity profile of 1.5% CMC, flowing within the annulus of the heat exchanger, with Sprayon LU206 treated stainless steel to be the examined outside surface of the inner tube.

Figure 6.9. Velocity profile of 1.5% CMC, flowing within the annulus of the heat exchanger, with silicone sheets to be the examined outside surface of the inner tube.
Figure 6.10. Velocity profile of 1.5% CMC, flowing within the annulus of the heat exchanger, with silicone fiberglass sheets to be the examined outside surface of the inner tube.

Figure 6.11. Velocity profile of 1.5% CMC, flowing within the annulus of the heat exchanger, with fluoropolymer coated polyester knitted textiles to be the examined outside surface of the inner tube.
Figure 6.12. Velocity profile of 1.5% CMC, flowing within the annulus of the heat exchanger, with untreated polyester knitted textiles to be the examined outside surface of the inner tube.

Figure 6.13. Velocity profile of 1.5% CMC, flowing within the annulus of the heat exchanger, with fluoropolymer coated polyester woven textiles to be the examined outside surface of the inner tube.
Figure 6.14. Velocity profile of 1.5% CMC, flowing within the annulus of the heat exchanger, with untreated polyester woven textiles to be the examined outside surface of the inner tube.

6.4. Conclusions

This research investigated the effects of different surface characteristics such as wettability, roughness, and topography on adhesive and flow behavior of Carboxymethyl Cellulose (CMC) solution (conc. 1.5%). Data of pressure drop and velocity profile were collected and used as the testing methods to compare the effects of the different surface samples on adhesive and flow behavior of highly viscous 1.5% CMC solution, flowing within the annular passage of a tube in tube heat exchanger. The outer surface of the inner tube was used as the tested food-contact surface, applying on it all the different examined materials.

A variety of different food-contact surface samples were studied including untreated and chemically and lubricant treated stainless steel samples (Rain-X, Aculon, Sprayon LU206), fabrics (knitted and woven structures, untreated and treated with a commercial
fluorocarbon-based resin), polytetrafluoroethylene (PTFE) sheets, silicone sheets and fiberglass reinforced silicone polymer sheets. The examined surface samples covered a wide range of different surface characteristics such as surface wettability, roughness, and topography. The selected surface materials tested had different surface wettability properties, varying from hydrophilic (0-90°) (Rain-X treated stainless steel), low hydrophobic scale (90-100°) (untreated stainless steel, Sprayon LU206 treated stainless steel), hydrophobic (Aculon treated stainless steel, PTFE, silicone sheets, fiberglass silicone sheets, untreated textiles) up to superhydrophobic (150°) (fluorocarbon treated textiles) behavior. Surface roughness was the second parameter to differentiate the examined surface samples, having samples with a very smooth surface such as Aculon and Sprayon LU206 treated stainless steel samples and with the highly rough surface such as silicone sheets. The rest of the samples had an intermediate surface roughness but higher than the untreated stainless steel used as the reference sample for this study. Finally, two different surface structure topography were studied using untreated and treated with a commercial fluorocarbon-based resin knitted and woven fabrics surface samples.

Pressure drop was used as an indirect measurement of the presence of product deposit at the pipe wall, to study the effect of the different examined surface materials on the adhesive behavior of 1.5% CMC. The pressure drop across the heat exchanger system was estimated for the collected pressure data at the inlet and outlet of the heat exchanger system, during the experiments with the examined food-contact surface samples. Silicone sheets and untreated woven textile samples, with the characteristic highly rough hydrophobic surfaces, exhibited the highest pressure drop across the system with a value of 75842.4 Pa (12.0 psia). The rest of the examined materials had a pressure drop value around 48263.3-55158.1 Pa (7-8 psia), with
the lowest value to be observed for the samples with the smooth surface (Aculon and Sparyon treated stainless steel) and superhydrophobic (treated knitted and woven textile samples) characteristics. The pressure drop across the concentric annulus of the heat exchanger was estimated based on the collected data of volumetric flow rate (0.000050-0.000063 m³/s (0.8-1.0 gal/min)) and the rheological parameters of 1.5% CMC (flow behavior index of 0.361 ± 0.003 and a flow consistency index of 28.7 ± 0.2 Pa s). The results presented that under laminar flow conditions, the most important factor in the pressure drop in the annulus is the ratio between the outside radius of the inner tube (r₁) and the inside radius of the outer tube (r₂). The lowest pressure drop at 42846.9 Pa (~6.2 psia) was observed for the untreated and treated stainless steel materials, the samples with the lowest r₁. While silicone sheets surface, the sample with the highest r₁, exhibited the highest pressure drop across the annulus at 49932.5 Pa (~7.2 psia). The rest of the samples had an intermediate pressure drop across the annulus, around 43600-45800 Pa (~6.5 ± 0.3 psia).

Moreover, the velocity profile of 1.5% CMC was studied, using particle tracking velocimetry (PTV). Video microscopy was used to capture the movement of fluorescent tracer particles, introduced at the inlet of the heat exchanger, at three different locations of the annulus flow (close to the wall of the inner tube, at the center of the annulus cross-sectional area and close to the outer tube pipe wall). The video recorded frames were analyzed with a particle tracking Matlab algorithm. The Matlab algorithm identified the position of the particles in each examined frame and created particles trajectories as a function of time, based on the linked particles from the sequence of the examined frames. Finally, the algorithm processed the position of the trajectories as a function of a time step to calculate the mean-square displacement (msd) for each particle. The estimated results of msd data as a function of time,
were statistically processed with the position data from the particle trajectories to link the data with the tracer particles and calculate the velocity for each particle. Knowing the velocity and the location of each tracing particle, the velocity profile and particle velocity magnitude was compared for 1.5% CMC flowing in the annulus, for each examined food-contact surface sample. The PTV results on the velocity profile of 1.5% CMC, showed a parabolic velocity profile for almost all the examined cases, with the fastest particle be close to the center of the annulus flow. The fastest flowing particles with a velocity magnitude between 0.050- 0.058 m/s, were observed for the sample with the smallest flow cross sectional area (silicone sheets, silicone fiberglass sheets, PTFE and textile samples) and for the smooth stainless steel surface, treated with Sprayon LU206. For the samples with characteristic hydrophobic (Aculon treated stainless steel surface) and superhydrophobic (treated knitted textile) behavior, the most of the tracing particles were concentrated away for the inner tube wall, near to the outer tube wall and at the center of the annulus, respectively.

In conclusion, the results of this study on pressure drop indicate that food-contact materials with smooth and hydrophobic/superhydrophobic characteristics resulted in lower product deposition at the pipe wall. While based on the PTV results, wetting surfaces (hydrophilic behavior) with smooth characteristics are the anticipating materials with the most efficient and uniform thermal treatment of the product across the annulus.

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Nomenclature

Latin letters

AFM Atomic Force Microscopy
CMC Carboxymethyl Cellulose solution

$D_h$ Hydraulic diameter (m)

$f$ Friction factor (dimensionless)

$r_1$ Outside radius of the inner tube (m)

$r_2$ Inside radius of outer tube (m)

FPS Frames per Second

I.D. Inner tube Diameter (m)

$K$ Flow consistency index ($\text{Pa} \cdot \text{s}^n$)

$L$ Length (m)

$\text{msd}$ Mean-square displacement (pixels$^2$)

MSD Real displacement (m)

$n$ Flow behavior index (dimensionless)

$N_{\text{GR}e}$ Generalized Reynolds number for non-Newtonian fluids

O.D. Outer tube Diameter (m)

$Q$ Volumetric flow rate ($\text{m}^3/\text{s}$)

PTFE Polytetrafluoroethylene

PTV Particle Tracking Velocimetry

$T$ Temperature ($^\circ\text{C}$)

$u$ Average velocity of fluid (m/s)

$u_p$ Magnitude of particle velocity (m/s)
**Greek letters**

- \( \gamma \) Shear rate \((s^{-1})\)
- \( \Delta p \) Pressure drop \((Pa)\)
- \( \Delta p_s \) Pressure drop across the heat exchanger system \((Pa)\)
- \( \Delta T \) Temperature difference \((^\circ C)\)
- \( \Delta t \) Lag time \((frames or s)\)
- \( \zeta \) Correction factor for the hydraulic diameter \((dimensionless)\)
- \( \theta \) Incline angle of the consistometer units \((^\circ)\)
- \( \sigma \) Shear stress \((Pa)\)

**Subscripts**

- MAX refers to the maximum temperature difference between the 3 cross-sectional points
- GRe refers to generalized Reynolds number for non-Newtonian fluids
- HX refers to the temperature differences between the average temperature at the inlet and at the outlet of the heat exchanger
- p refers to particle
- s refers to the pressure drop across the heat exchanger system
References


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CHAPTER 7
Enhancement of Continuous Flow Cooling of Viscous Foods by Application of Engineered Textiles

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This study investigated the effect of two different food-contact materials; untreated stainless steel (series 300) and engineered food-grade polytetrafluoroethylene (PTFE) textile on continuous flow cooling of viscous foods (sweet potato puree, banana puree, and cheese sauce). The PTFE antifouling surface was constructed by taking advantage of the stability and partial non-wetting and hydrophobic properties of PTFE and the fabrication methods of knitted textile to modify the surface as desired, resulting in smooth and knit characteristics. Cooling studies for both surfaces examined were conducted under identical steady-state operating conditions (flow rates, inlet temperatures), with the tested foods flowing within the annular passage of a tubular heat exchanger. The outside surface of the inner tube of the heat exchanger was the examined food-contact surface, applying on it the PTFE textile. Cooling performance between the two different examined materials was assessed by comparing the temperature differences between the average temperatures of the processed materials at the inlet against the average temperature at the outlet of the heat exchanger ($\Delta T_{HX}$), for both cases. The amount of product deposition at the pipe wall was significantly lower when using the PTFE textile, compared to that of untreated stainless steel surface, an indication of lower product accumulation at the heat exchanger pipe wall. This resulted in more efficient cooling for all foods tested, with PTFE textile covered tube having an average $\Delta T_{HX}$ of $15 \pm 2 \, ^{\circ}C$, compared to a $\Delta T_{HX}$ of $12 \pm 2 \, ^{\circ}C$ for the untreated stainless steel tube surface. The results of cooling studies showed an improvement of 25% in the cooling of viscous foods for the case of the inner tube of the heat exchanger, covered with the engineered PTFE textile.

**Key words:** Continuous flow cooling, antifouling surface modification, PTFE textile, viscous foods
7.1. Introduction

New developments in thermal processing of viscous and multiphase food products, such as dairy products, fruit and vegetable purees, and soups using advanced thermal technologies, such as continuous flow microwave (MW) processing, ohmic heating, and radio frequency heating systems, provide uniform and rapid volumetric heating resulting in reduction of come up time, reduction of fouling deposition, and minimization of product quality losses (Coronel et al., 2005; Steed et al., 2008; Cullen et al., 2012). However, cooling of viscous products is still implemented using inefficient conventional cooling methods. During conventional cooling of viscous food products, laminar flow, low thermal conductivity, characteristic for these materials, lead to a non-uniform, slow cooling process, increase in operating cost, and degradation of final food quality (Stoforos et al., 2016; Stoforos et al., 2017a). Thermal mixing (cross-sectional temperature equalization) has been proposed as an efficient method to enhance heat transfer and the overall continuous flow cooling process of highly viscous food products (Metcalfe and Lester, 2009; Stoforos et al., 2016). However, the efficiency of thermal mixing and cooling of viscous food products, such as fruit and vegetable purees, is affected by the formation and build-up of fouled material on the surface of processing equipment (heat exchangers, mixing units) (Stoforos et al., 2016).

Fouling and accumulation of unwanted material, such as proteins, starches, and minerals, on the surface of processing equipment, a problem mainly observed during conventional thermal processing of viscous products, reduces processing efficiency and increases the operation cost. The formation and build-up of fouling layers on food processing equipment increases the resistance to heat transfer, thermal mixing and pressure drop across the processing equipment, resulting in degradation of final food product quality and increasing
the overall processing and cleaning operation time and cost (Balasubramanian and Puri, 2008; Balasubramanian and Puri, 2009; Barish and Goddard, 2013; Barish and Goddard, 2014; Stoforos et al., 2016; Stoforos et al., 2017a). Furthermore, accumulation of fouling layers on food processing surfaces is prone to increase the formation of biofilms, resulting in excessive use of chemicals and sanitizers, and frequent food shutdowns for sanitation and cleaning (Sandu and Singh, 1991; Kananeh et al., 2010; Mérian and Goddard, 2012; Barish and Goddard, 2014).

The degree of fouling and adhesion of unwanted food material on the processing equipment surface depends on several parameters, such as operating conditions (temperature and flow rate), food properties (viscosity and food composition) and surface characteristics (surface wettability and topography) (Santos et al., 2004; Awad, 2011; Mérian and Goddard, 2012). In the food industry, different types of fouling can occur, based on different physical and chemical processes involved. The types of fouling in food and dairy processing can be categorized as: 1) particulate or sedimentation fouling (deposition of suspended particles), 2) crystallization or precipitation fouling (dissolved salts from a saturated solution), 3) chemical reaction fouling (cooking, polymerization, etc.), 4) corrosion fouling (electrochemical reactions), 5) biological fouling or biofouling (attachment and growth of microorganisms), and 6) solidification or freezing fouling (formation of a solid fouling deposit onto a subcooled surface, from fluid components with a high melting point) (Bansal and Chen, 2006; Awad, 2011, Goode et al., 2013).

Unfortunately, the most commonly used material for food processing equipment - stainless steel, has surface characteristics, such as partial wetting properties and high surface energy, that are prone to fouling and adhesive behavior of viscous food materials (Goode et
Recent studies on fouling reduction have involved the use of polymer and nanoparticle coatings (polyethylene glycol, silica, fluorocarbon and PTFE), physico-chemical surface modification techniques such as plasma-enhanced chemical vapor deposition and electro-chemical and ion implantations, in order to modify the surface characteristics of stainless steel, minimize fouling and improve hygienic properties of food-contact surfaces (Santos et al., 2004; Mérian and Goddard, 2012). Surfaces characteristics, such as surface wettability (contact angle, spreadability), and morphology (roughness and topography), have been modified to produce functional surfaces that mimic the self-cleaning and repellent characteristics of natural antifouling materials such as the lotus leaf (*Nelumbo nucifera*) (Santos et al., 2004; Rosmaninho et al., 2007; Balasubramanian and Puri, 2009; Kananeh et al., 2010; Barish and Goddard, 2013). Antifouling materials have low surface energy (weak attractive forces) and specific surface structure (roughness, topography). Low surface free energy, associated with partial non-wetting surfaces (hydrophobic and superhydrophobic characteristics) such as polymer and ceramic coatings, have longer induction periods than partial wetting surfaces (high free surface energy, hydrophilic characteristics), resulting in lower amount of product deposition. On the other hand, surfaces with high roughness, increase the contact area of the surface, providing more suitable sites for nucleation, which are more favorable for initiation of fouling (Awad, 2011, Goode et al., 2013). Antifouling materials and surface modification techniques, with characteristic low surface free energy and smooth surface characteristics, have successfully been used in food and dairy research, improving the operation and cleaning efficiency, while reducing the bacterial adhesion (Beuf et al., 2003; Santos et al., 2004; Bansal and Chen, 2006; Premathilaka et al., 2006; Rosmaninho et al., 2007; Balasubramanian and Puri, 2009; Kananeh et al., 2010; Mérian
and Goddard, 2012; Barish and Goddard, 2013; Goddard et al., 2013; Cheng et al., 2015; Stoforos et al., 2017a).

Furthermore, antifouling surface properties, of low free surface energy hydrophobic surface, have been examined on cooling of highly viscous products, such as sweet potato puree, banana puree, and cheese sauce with significant enhancement of cooling and cleaning processes for most of the materials tested (Stoforos, et al., 2017a). However, antifouling surface modification techniques have not extensively been used for commercial application for food and dairy processing due to the cost and complexity of fabrication, and in some cases the incompatibility with food grade materials, additionally with the reported short-term stability of antifouling properties of the modified surfaces (Barish and Goddard, 2013). Based on these observations, there is a need for development of new food surface modifications techniques with lower cost and improved simplicity of fabrication.

The main objective of this research was to improve the continuous flow cooling process of viscous and poorly conductive foods, minimizing the cooling-related quality losses of the final food product, and potentially improving the sanitation conditions by applying surface modification techniques. For the purpose of this study, new in the food and dairy research area, low-cost and easy to implement surface modification techniques, based on textile fabrication methods were introduced. Anticipating that the new engineered antifouling materials, with the desired surface properties, will enhance cooling and accelerate cleaning of viscous foods while introducing a new concept of surface modification for future research studies and potential use in advanced thermal processing applications (heat exchangers, mixing units, holding tubes, spray dryers and food packaging).
7.2. Materials and Methods

7.2.1. Test materials

The test food products selected for this study were highly viscous food products, such as sweet potato puree (Yamco LLC, Snow Hill, NC, USA), banana puree (Aseptia/Wright Foods, Troy, NC, USA), and cheese sauce (Advanced Food Products, New Holland, PA, USA). These materials were chosen because they exhibit good behavior during research and commercial applications of advanced thermal technologies, particularly continuous flow MW heating (Coronel et al., 2005; Steed et al., 2008), while product deposition and accumulation at the pipe wall during cooling and mixing process have also been reported (Stoforos et al., 2016; Stoforos et al., 2017a).

7.2.2. PTFE textile surface samples

The engineered examined surface has been designed for reduction of product accumulation and fouling during cooling of viscous foods. The examined material was designed based on the results of quantitative and statistical studies on the adhesive behavior of viscous foods on different tested materials (Stoforos et al., 2017b; Stoforos et al., 2017c). Results of these studies showed that partial non-wetting surfaces, with hydrophobic and smooth surface characteristics, and knitted surface structure, exhibited the best repellent and non-adhesive behavior with the tested fruit and vegetable food samples. Furthermore, untreated stainless steel (series 300), and PTFE samples presented long-term stability of surface characteristics compared to the adhesive behavior of the foods tested. Based on these observations, knitted PTFE yarn based fabric was used as the examined sample, a material with characteristic low surface free energy, non-wetting behavior with hydrophobic
characteristics, excellent chemical resistance and great stability within a wide range of
temperature conditions (-80 to 260 °C).

PTFE yarn samples (Toray, Fluorofibers (America), INC, Decatur, AL, U.S.A.) were
used to fabricate the engineered surface with the smooth and knitted surface structure, using
knit construction methods (West, 2016) and units (SWG061N2 (Shima Seiki USA Inc.,
Monroe Township, NJ, U.S.A.)). The final engineered PTFE textile samples were a lightweight
(0.091 kg) with tubular knitting (25 stitches knitting pattern) characteristics, with a total length
of 1.2 m, an inner diameter of 0.0508 m (2 in), and a thickness of 0.002 m (0.08 in).

7.2.3. Experimental methods

The effects of surface modification on continuous flow cooling of highly viscous
products were examined by comparing the efficiency of cooling between an untreated stainless
steel heat exchanger against the performance of the heat exchanger where the food contact
surfaces of the inner tube have been covered with a food-grade engineered PTFE textile. To
study and understand the effects of surface modification on continuous flow cooling of viscous
foods two different series of experiments were conducted. The first series of experiments
involved the study of the surface properties, such as surface wettability and surface roughness
on the untreated stainless steel (300 series) and the engineered PTFE textile surface samples.
More comprehensive studies on continuous flow cooling of viscous foods were conducted, by
comparing the efficiency of cooling of viscous foods using the untreated and treated heat
exchangers. Approximately 5 gallons of food product sample was sufficient to fill the
experimental system and allow for recirculation of the test samples at a flow rate of
6.3 × 10^{-5} m^3/s (≈1 gal/min), flowing within the annular passage of the heat exchanger.
Municipal supply water was used as the cooling medium, flowing within the inner tube of the heat exchanger in a counter-current flow configuration, with an inlet temperature between 25-29 °C and a flow rate of $56.8 \times 10^{-5} \text{ m}^3/\text{s}$ (9 gal/min).

**7.2.4. Contact angle measurements**

Surface wettability was one of the parameters differentiating the examined surfaces. Surface wettability studies involve the measurement of contact angles (CA) as the primary data, which indicates the degree of wetting at a solid-liquid interaction. Surface wettability characteristics distinguish the surfaces to wetting and non-wetting surfaces, corresponding to small contact angles (<90°) and large contact angles (>90°), respectively (Korhonen et al., 2013). The measurement of water and oil contact angles is a common method to evaluate the surface wettability (Korhonen et al., 2013). The equilibrium static contact angles (the Sessile drop method) of water and oil (n-dodecane) on the samples tested, were measured at room temperature (18-20 °C), using OCA 15 (Future Scientific Corp, Garden City, NY, U.S.A.), an optical contact angle goniometer, and via SCA20, its software interface. Using a 500 microliter gastight syringe (Hamilton Company, Reno, NV, U.S.A), controlled 3-5 μL volume droplets of water and/or oil were deposited on different points (3 points) across the tested surfaces to measure the contact angle and determine the homogeneity of the treated surfaces.

**7.2.5. Surface roughness measurements**

Surface topography was another parameter that differentiated the food-contact surfaces examined. Surface topography differences were quantified through surface roughness studies. A multi-mode atomic force microscope (AFM) (MFP-3D, Asylum Research
Corporation, Raleigh, NC, U.S.A.) was used to measure the surface roughness of the examined materials. AFM was used to map the surface of tested samples in tapping mode using a high frequency (~300 kHz) soft tapping tip probe (Olympus AC240, Asylum Research Corporation, Raleigh, NC, U.S.A.), having a nominal spring constant of 40 N/m. The tip of the probe with a diameter of less than 10 nm, scanned an area of 5 × 5 μm with a tip velocity of 10 μm/s. Images were collected with 512 pixels per line and 256 lines of data. AFM software (MFP-3D, Asylum Research Corporation, Raleigh, NC, U.S.A.) was used to analyze the digital image from AFM and to determine surface roughness of the examined surface samples, by comparing the height (nm) variations in the surface. Finally, arithmetic average roughness (Ra (nm)) of the absolute values of the roughness profile ordinates, was used to quantify and compare the differences of surface structure between the examined samples. It is worth mentioning that AFM is not the recommended method for surface roughness measurements of such uneven textiles substrates. However, for the purposes of this study and to maintain testing consistency, the surface roughness of fabric sample was also quantified using AFM, using the same small localized scanned area of 5 × 5 μm for all the samples tested.

**7.2.6. Continuous flow cooling: experimental set up**

To study and compare the different examined food-contact surfaces during continuous flow cooling process of viscous food products, a thermal processing system in recirculation configuration was assembled, consisting of three basic sections: heating, cooling, and mixing. The thermal processing system consisted of a food grade progressive cavity pump (Seepex MD-012 12, Fluid Engineering Inc., Birmingham, AL, USA) and a continuous flow modular microwave heating system consisting of a total of 13 bench-top Panasonic Inverter MW ovens,
with a total power output of 16.350 kW, at the MW frequency of 2450 MHz. At the exit of MW system, a helical stainless steel static mixer, with a total length of 0.3337 m (13.1 in) was installed, to connect the MW heating section with the cooling unit. The static mixer was used to equalize the product temperature at the inlet of the cooling unit.

The cooling section consisted of a horizontal concentric tube in tube heat exchanger of a total length of 1.11 m. The concentric tube in tube heat exchanger consisted of an inner tube, removable and separate for each surface treatment examined and an outer tube assembly. The outside surface of the inner tube was used as the tested food-contact surface, by the application of the engineered PTFE textile sample. A stainless steel (series 300) tube, with an outside diameter (O.D.) of 0.0508 m (2.0 in) was used as the inner tube. The O.D. of the inner tube was slightly increased, being 0.0533 m (2.1 in) for the tested tube coated with PTFE textile sample. The outer tube assembly was built using 3 inches O.D. sight glass tubes and sanitary stainless steel pipe fittings. The sight glass tubes were used as a part of the heat exchanger, for visual comparison of cleaning performance between the two examined surfaces. The two different inner tubes of the heat exchanger, the one with the untreated stainless steel, and the other with the engineered PTFE textile cover, were used separately and independently for different series of experiments. For both cases, the annulus of the heat exchangers was used as the flow passage for the tested foods. Having the food product flowing within the annulus, provided an easier access to the food contact surface for the application of the PTFE textile sample and, additionally, the cooling efficiency could be improved by increasing the thermal losses to the environment (Stoforos and Simunovic, 2016).

Finally, at the exit of cooling section, the flow of the product continued within a mixing unit, which was used to minimize the temperature distribution within the product tested. The
food-contact surfaces of the mixing unit remained untreated during the studies for both the examined cases. The mixing unit, consisted of a 180° bend tube (formed from two silicone rubber-lined, reinforced flexible tube segments, each 0.53 m long), and a stainless steel 180° elbow, mounted at the top surface of a low sonic frequency tactile audio transducer Buttkicker LFE (The Guitammer Company, Westerville, OH, USA) (Stoforos et al., 2016; Stoforos et al., 2017a). A computer frequency generator software (FreqGen 1.13, Digital River Inc., Minnetonka, MN, U.S.A.) was used to control the frequency and duration of vibration of the tactile audio transducer. The amplitude of vibration was controlled via a power amplifier type BKA1000-N (The Guitammer Company, Westerville, OH, USA). An accelerometer data logger device (SlamStick C, Mide Inc., Medford, MA, USA) and a computer program (Slamstick viewer, Mide Inc., Medford, MA, USA), were used to measure, record, and process the vibration parameters (frequency, duration, and amplitude of vibration).

The temperature of the tested foods was measured using T-type thermocouples, located at the inlet and outlet of the MW heating system, cooling system, and at the entrance and exit of the mixing unit. Specially customized triple-point T-type thermocouple probes (Keyhole Multipoint probes, Windridge Sensors LLC, Holly Springs, NC) were used to measure the temperature profiles of test products at the inlet and the outlets of the tube in tube heat exchanger and the mixing unit. The multi-point probes measured the temperature of the product at three different cross-sectional locations, at the center of the pipe, close to the pipe wall and at an intermediate point equidistant between the pipe wall and pipe center. Time-temperature data were recorded once per second using a 32-Channel temperature data acquisition system (IOTEch TempScan 1100, IOtech, Inc. 25971 Cannon Road, Cleveland, OH 44146-1833) (Fig. 7.1).
Figure 7.1. a) Schematic diagram and pictures of the closed-loop, recirculating experimental system for continuous flow thermal processing, was consisted from b) MW-Heating Section, c) outer tube assembly of the heat exchanger, d) the two examined samples of untreated stainless steel (top), and PTFE textile covered inner tubes of heat exchanger and d) mixing section.
7.2.7. Continuous flow cooling: experimental methods

The cooling performance of the two different heat exchangers was compared for all the food products tested. Two different sets of duplicate experiments were conducted, initially using the untreated stainless steel tubular heat exchanger, followed by the second set of experiments with the modified heat exchanger, under the identical operational conditions (temperature and flow rate). Moreover, the modular MW heating system was used to heat all the tested food samples up to the same temperature range, with the average outlet product temperature being approximately 92-102 °C, which was used as the initial inlet temperature for all the cooling experiments. When the target temperature was reached from the MW heating, cooling studies were started by recirculating the product throughout the system and continued operation of only 6 out of 13 MW module units, to achieve and maintain steady state conditions (approximately constant inlet temperature) for a period of about 400 s.

During cooling, thermal equalization of cross-sectional temperature at the exit of the cooler was studied by imposing acoustic/mechanical vibrations for a duration of 120 s at the resonance frequency of the mixing unit and at the maximum volume level of the used audio amplifier (Stoforos et al., 2016). The resonance frequency of the mixing unit was determined, through scanning acoustic frequencies in the low sonic range (10–100 Hz), by applying 10s of vibration at the tested frequency, and the same fixed volume level of amplifier, followed by 5 s of silence (0 Hz), with incrementally increasing frequency steps of 10 Hz (Stoforos et al., 2016). The SlamStick C accelerometer was used to record the data of acceleration and frequency of vibration over the time. The resonance frequency of the mixing unit was determined from the collected data, as the frequency with the highest magnitude of acceleration (Stoforos et al., 2016).
Continuous flow cooling studies were conducted in the temperature range of 105-60 °C. Cooling studies were not performed at temperatures below 60 °C, where more uniform temperature distribution within the product is established and less product quality degradation occurs (Stoforos et al., 2016). Cooling studies were immediately followed by evaluation of cleaning efficiency. Cleaning trials were conducted via washing-down the system (modular MW heating, cooling and mixing sections) with hot water (~60 °C), flowing with a flow rate of $56.9 \times 10^{-5}$ m$^3$/s (9 gal/min), for 600 s. At the end of wash-down, photos of the outer surface of the inner tube of both heat exchanges were taken for evaluation and comparisons of cleaning efficiency.

7.3. Results and discussion

7.3.1. Contact angle measurements

The first set of experiments for this study was performed to determine the differences in surface characteristics between the untreated stainless steel and the engineered PTFE textile surfaces. Contact angle was used as the parameter to define the differences between the untreated and the treated stainless steel surfaces, as a measure to quantify the wettability properties of the surface. Using the video analysis-based OCA 15 goniometer, the results on the sessile drop static water contact angle at different points on the tested surfaces showed a homogeneous behavior for all the different samples. The contact angle for the water-stainless steel interface was at $84 \pm 3^\circ$, while for water-PTFE textile interface was at $111 \pm 3^\circ$ (Fig. 7.2). The results of the contact angle measurements with water indicated that PTFE textiles presented a partial non-wetting surface with hydrophobic characteristics, while partial wetting properties were observed for the stainless steel sample.
Furthermore, the sessile static contact angle measurements with oil (n-dodecane) showed an oleophilic behavior for both surfaces, with the contact angles of approximately $19 \pm 1^\circ$ and $31 \pm 1^\circ$, for the stainless steel and the PTFE textile samples, respectively (Fig. 7.3).

Figure 7.2. Picture of sessile static contact angle measurements with water: a) PTFE textile and b) Untreated stainless steel samples.
7.3.2. Surface roughness measurements

Surface roughness was examined as the second parameter to quantify the differences between the examined food-contact surfaces samples. Analysis of the AFM micrographs was used to determine the surface roughness ($Ra \text{ (nm)}$) of the examined food-contact surface
samples. PTFE textile sample exhibited the smoothest characteristics with a value for $R_a$ of 3.9 nm, compared with a $R_a$ of 7.8 nm for the untreated stainless steel sample (Fig. 7.4).

Figure 7.4. AFM surface topography of: a) PTFE textile and b) Untreated stainless steel samples. The shadings correspond to the $Z$ height variation. Darker shades represent lower $Z$ value and brighter regions represent higher $Z$ value.
7.3.3. Resonance Frequency

The second part of this research studied cooling and thermal mixing performance during experiments with highly viscous foods flowing within the annular passage of the examined heat exchangers. The first set of tests was to determine the resonance frequency, the frequency with the highest magnitude of the acceleration (g) of vibration. Analyzing the collected data of acceleration, frequency (Hz) and time (s), from the accelerometer, similar results were observed for all the tested products, with the resonance frequency for the mixing system at approximately 20 Hz, as can be observed from the following acceleration data for cheese sauce (Fig. 7.5). The frequency with the highest magnitude of acceleration at 20 Hz, was used during the studies for all the tested foods.

Figure 7.5. Effect of acoustic frequency on the acceleration (expressed as unit Gs) magnitude of vibration at different tested frequencies.
7.4. Continuous flow cooling studies

To study the cooling and thermal mixing performance of the examined food-contact surfaces, time-temperature data at the inlet and the outlet of the heat exchangers and the mixing unit, at 3 different cross-sectional points within the pipe (center, wall and an intermediate point), were collected. Similar to a previous study (Stoforos et al., 2017a), the effects on the cooling efficiency of untreated stainless steel and the modified/treated heat exchangers were examined by comparing the temperature differences ($\Delta T_{HX}$) between the average temperatures at the inlet against the average temperature at the outlet of the heat exchanger. Thermal mixing efficiency was evaluated by comparing the maximum temperature difference between the product temperature at the three cross-sectional points at the inlet ($\Delta T_{MAX}$) and at the outlet of the mixing unit ($\Delta T_{MAX,M}$) (Stoforos et al., 2017a).

Finally, pictures of the internal heat exchanger tube outer surfaces after experimental runs and washing-down process of the tested foods were compared to understand the effects of the engineered PTFE textile surface treatment on the cleaning process.

7.4.1. Sweet potato puree

Studies on cooling and thermal mixing were performed during the cooling stage of sweet potato puree processing, within the temperature range of 95-70 °C. Duplicate experiments, under steady state, were conducted, with sweet potato puree flowing within the annulus of the heat exchangers at a flow rate of $6.3 \times 10^{-5}$ m$^3$/s and with an inlet temperature in the range of 90-95 °C. Water was used as the coolant, flowing in counter-current mode, with a flow rate of $63.1 \times 10^{-5}$ m$^3$/s and an inlet temperature of 28 ± 1 °C. Comparing the time-temperature profiles, slightly better performance was observed for the case of PTFE textile,
with an average $\Delta T_{HX}$ at 14 $\pm$ 2 °C during the period of the steady state conditions (400 s), compared to the other case which exhibited an average $\Delta T_{HX}$ of 12 $\pm$ 2 °C (Fig. 7.6).

Temperature distribution within sweet potato puree flow cross section was more uniform during the cooling process with the untreated stainless steel heat exchanger, during all the tests. Moreover, for the study of sweet potato puree with the untreated heat exchanger, a $\Delta T_{\text{MAX}}$ of 2-4 °C was reported, at the exit of the cooler, while at the exit of mixing unit the $\Delta T_{\text{MAX,M}}$ was reduced to 0-2 °C (Fig. 7.7). For the heat exchanger with the inner tube covered with the PTFE textile, the $\Delta T_{\text{MAX}}$ was at 8-11 °C at the exit of the cooler and reduced at a $\Delta T_{\text{MAX,M}}$ of 2-4 °C, at the exit of the mixing unit with the influence of vibration at 20 Hz (Fig. 7.8). The wider temperature distribution within the product during cooling with the PTFE textile cover on the surface of the inner tube can possibly be explained by the better cooling efficiency for this case, resulting in lower temperatures for the product in that area and hence wider temperature distribution within the product.
Figure 7.6. Comparison of the average $\Delta T_{HX}$, during two sets of experiments a) and b), on cooling of sweet potato puree, for the heat exchanger with the untreated stainless steel inner tube and with the inner tube cover with the engineered PTFE textile.
Figure 7.7. Comparison of temperature distribution within sweet potato puree (at center, wall, and an intermediate point) at the inlet (a) and the outlet (b) of the mixing unit for sweet potato puree after cooling in the untreated stainless steel heat exchanger for a given product average temperature at the inlet of the heat exchanger. The vertical dashed lines represent the beginning and the vertical solid lines the end of vibration of 120 s at 20 Hz. Note that the average time for the fluid particle to move from the inlet to the outlet of the heat exchanger and mixing unit was 55 s and 24 s, respectively.
Figure 7.8. Comparison of the temperature distribution within sweet potato puree (at center, wall, and an intermediate point) at the inlet (a) and the outlet (b) of the mixing unit for sweet potato puree after cooling in the heat exchanger with the inner tube covered with the engineered PTFE textile, for a given product average temperature at the inlet of the heat exchanger.
The end of cooling experiments was followed by washing-down the thermal processing system, with hot water for a period of 600 s and visual observation of the cleaning efficiency. Comparing the photographs of the internal heat exchanger tube outer surfaces, that were taken at the end of wash-down process and after disassembly the system (Fig. 7.9), both examined surfaces exhibited a significant efficient cleaning, with no residual amount of sweet potato puree to be visible on the outer surface of the inner tube of the heat exchanger.

Figure 7.9. Comparison of a) untreated stainless steel and b) PTFE textile covered internal heat exchanger tube outer surfaces, after cooling of sweet potato puree. Picture shows the disassembled tubes after wash-down with hot water.
7.4.2. Banana puree

Cooling and thermal mixing experiments were conducted during the cooling process of banana puree, within the temperature range of 99-75 °C, and by employing vibration of 120 s duration at 20 Hz, for mixing. Duplicate experiments, under steady state conditions, were performed, with banana puree flowing within the annular passage with an inlet temperature in the range of 91-99 °C, and a flow rate of $6.3 \times 10^{-5}$ m$^3$/s. Water flowing in counter-current mode, with a flow rate of $63.1 \times 10^{-5}$ m$^3$/s and with an inlet temperature of $28 \pm 1$ °C, was used as the cooling medium. From the time-temperature data of banana puree during the steady state cooling period (for ~400 s), cooling was more efficient using the heat exchanger with the inner tube covered with the engineered PTFE textile, with a $\Delta T_{HX}$ of 14 ± 2 °C. On the other hand, a $\Delta T_{HX}$ of 9 ± 2 °C was observed, for the untreated stainless steel heat exchanger (Fig. 7.10).

Temperature distribution within the product at the exit of cooling unit was approximately the same for both examined cases, with a $\Delta T_{MAX}$ at 10-12 °C. After application of vibration at 20 Hz, $\Delta T_{MAX,M}$ was reduced to 0-3 °C, for both cases (Fig. 7.11; Fig. 7.12).
Figure 7.10. Comparison of the average $\Delta T_{HX}$, during two sets of experiments a) and b), on cooling of banana puree, for the heat exchanger with the untreated stainless steel inner tube and with the inner tube cover with the engineered PTFE textile.
Figure 7.11. Comparison of the temperature distribution within banana puree (at center, wall, and an intermediate point) at the inlet (a) and the outlet (b) of the mixing unit after cooling in the untreated stainless steel heat exchanger for a given product average temperature at the inlet of the heat exchanger.
Figure 7.12. Comparison of the temperature distribution within banana puree (at center, wall, and an intermediate point) at the inlet (a) and the outlet (b) of the mixing unit after cooling in the heat exchanger with the inner tube covered with the engineered PTFE textile, for a given product average temperature at the inlet of the heat exchanger.
Cooling experiments were followed by the washing-down process, by cleaning the heat exchanger with hot water for 600 s. By comparing the pictures of the internal heat exchanger tube outer surfaces, that were taken at the end of the wash-down process and after disassembling the system, a significantly more efficient cleaning was observed for the hydrophobic surface of the PTFE textile cover (Fig. 7.13). For the case of the untreated heat exchanger, the surface of the tube was fully covered with banana puree, residual product accumulated during cooling. The existence of a thick layer of product deposition at the pipe wall had a negative impact on heat transfer during cooling, resulting in lower cooling efficiency for the case of the untreated stainless steel heat exchanger.

Figure 7.13. Comparison of a) untreated stainless steel and b) PTFE textile covered internal heat exchanger tube outer surfaces, after cooling of banana puree. Picture shows the disassembled tubes after wash-down with hot water.
7.4.3. Cheese sauce

The final experiments on cooling and thermal mixing were conducted with cheese sauce as the examined material. Steady state operational conditions were achieved with cheese sauce flowing at $6.3 \times 10^{-5} \text{ m}^3/\text{s}$, within the annulus passage of the heat exchanger, with an inlet temperature of about 95-102 °C. Water was used as coolant, flowing in a counter-current mode, with a flow rate of $63.1 \times 10^{-5} \text{ m}^3/\text{s}$ and an inlet temperature of $28 \pm 1 °C$. Time-temperature profiles for cheese sauce presented a significantly more efficient cooling using the heat exchanger with the inner tube covered with the engineered PTFE textile, with a $\Delta T_{\text{HX}}$ of $16 \pm 2$, compared to a $\Delta T_{\text{HX}}$ of $11 \pm 2 °C$ for the case of the untreated stainless steel heat exchanger (Fig. 7.14).

A slightly more uniform temperature distribution within the food was observed with the heat exchanger with the inner tube covered with the engineered PTFE textile with a $\Delta T_{\text{MAX}}$ of 5-8 °C, while for the untreated case the $\Delta T_{\text{MAX}}$ was equal to 8-10 °C. Thermal mixing processing of cheese sauce via application of vibration at 20 Hz reduced the temperature difference within the product for the case of the untreated stainless steel heat exchanger, with a $\Delta T_{\text{MAX,M}}$ of 1-4 °C (Fig. 7.15). On the other hand, for the case of the heat exchanger with the PTFE textile cover, only a slight improvement was observed on temperature distribution assisted by vibration with a $\Delta T_{\text{MAX,M}}$ of 3-6 °C (Fig. 7.16).
Figure 7.14. Comparison of the average $\Delta T_{HX}$, during two sets of experiments a) and b), on cooling of cheese sauce, for the heat exchanger with the untreated stainless steel inner tube and with the inner tube cover with the engineered PTFE textile.
Figure 7.15. Comparison of the temperature distribution within cheese sauce (at center, wall, and an intermediate point) at the inlet (a) and the outlet (b) of the mixing unit after cooling in the untreated stainless steel heat exchanger for a given product average temperature at the inlet of the heat exchanger.
Figure 7.16. Comparison of the temperature distribution within cheese sauce (at center, wall, and an intermediate point) at the inlet (a) and the outlet (b) of the mixing unit after cooling in the heat exchanger with the inner tube covered with the engineered PTFE textile, for a given product average temperature at the inlet of the heat exchanger.
Unlike the previously tested foods, for the case of cheese sauce both the examined surfaces had a residual amount of product on the outside surface of the inner tube, as can be seen from the pictures of the internal heat exchanger tube outer surfaces that were taken at the end of the wash-down process and after disassembling the system (Fig. 7.17). For the case of the untreated stainless steel heat exchanger, the surface of the tube was fully covered with cheese sauce, a residual product accumulated during cooling, resulting in low cooling. However, the amount of area covered with cheese sauce was also substantial for the case of the engineered PTFE textile covered tube. Furthermore, similar to the observations of the previous products tested, the outside stainless steel surface of the inner tube, covered by the PTFE textile, remained clean at the end of the cooling experiments (Fig. 7.18).

Figure 7.17. Comparison of a) untreated stainless steel and b) PTFE textile covered internal heat exchanger tube outer surfaces, after cooling of cheese sauce. Pictures show the disassembled tubes after wash-down with hot water.
The results of this study showed that a combination of a good conductive material, such as stainless steel, the inner tube's material, with a food-contact surface with antifouling surface characteristics, can significantly enhance cooling, and potentially improve the sanitation conditions during the processing of viscous foods, such as sweet potato puree, banana puree, and cheese. A combination that can be achieved by reinforcement of antifouling materials, such as engineered PTFE textiles, on the food-contact surface of stainless steel wires and hoses (Rubber Fab, Sparta, NJ, U.S.A.) Furthermore, new developments in laser-etching techniques on the surface of stainless steel (Vorobyev and Guo, 2015), to create a hydrophobic material, can potentially be used as a more expensive option for the same purposes.

Moreover, a significant difference in cooling and cleaning efficiency was observed with the different foods tested. Cooling and cleaning results for banana puree and cheese sauce indicate the importance of food product surface characteristics during thermal processing of these products. A partial non-wetting surface, with hydrophobic smooth surface
characteristics, reduced the product accumulation and solidification, resulting in improvement of cooling and cleaning efficiency for these types of foods, as shown from the results of this study and the observations of previous studies (Stoforos et al., 2017a). However, the adhesive behavior of sweet potato puree is independent of the surface characteristics and behaves similarly for any surface tested in this research or in previous studies (Stoforos et al., 2017a; Stoforos et al., 2017b; Stoforos et al., 2017c). An observation that probably can be explained due to the viscoelastic behavior of sweet potato puree based foods (Ahmed and Ramaswamy, 2006). Liquid, solid or semi-solid foods, with viscoelastic properties tend to adhere less and are more easily rinsed off from the food-contact surface (Goode et al., 2013). This observation indicates the need for more focused studies on the dynamic rheological parameters of the food samples (such as viscoelastic properties and glass transition temperatures) for a better understanding of the interactions between the examined surface materials, and the flowing food materials.

7.5. Conclusions

During this work, cooling of viscous foods was studied by comparing cooling efficiency for two different food-contact materials, namely untreated stainless steel (series 300), used as the reference sample, and food-grade polytetrafluoroethylene (PTFE) textile. PTFE was chosen due to the long-term stability of anti-fouling characteristics and the stability in a wide range of temperature conditions. The fabrication method of knitted textiles was used to fabricate the examined sample because it is an easy and simple method to modify the surface topography (roughness, structure), with low production cost.
The differences between the surface characteristics of the two examined materials were quantified, conducting measurements on surface wettability and surface roughness, using the testing methods of contact angle (CA) measurements and atomic force microscopy, respectively. Measurements of contact angle were conducted using water and oil (n-dodecane) as the liquid phase. The results of contact angle measurements of water showed that PTFE textiles exhibited a partial non-wetting behavior with hydrophobic characteristics (CA=111 ± 3°), compared to a partial wetting behavior for stainless steel with hydrophilic characteristics (CA=84 ± 3°). The contact angle measurements with oil showed an oleophilic behavior for both surfaces with a CA of 19 ± 1° and 31 ± 1°, for the stainless steel and the PTFE textile samples, respectively. The surface roughness of the examined materials was compared based on the AFM measurements, with PTFE sample to exhibit a smoother surface compared to stainless steel, with average surface roughness values of 3.9 nm and 7.8 nm, respectively. PTFE textile samples were specifically engineered with anti-fouling and partial non-wetting properties, such as low surface free energy, hydrophobic and smooth surface characteristics, and knitted surface structure in order to reduce product accumulation and fouling on the surface of cooling unit, resulting in enhancement of cooling and cleaning efficiency of viscous foods.

Cooling of different viscous foods, such as sweet potato, banana puree, and cheese sauce, was studied using a tube in tube heat exchanger as the cooler. The outside surface of the inner tube of the heat exchanger was used as the tested food-contact surface, applying on it the engineered PTFE textile sample. Cooling studies for both examined materials were conducted under steady state and identical operating conditions (flow rate, inlet temperature), with the tested food products flowing within the annular passage of the heat exchanger and the coolant
(water) flowing in the inner tube, in counter-current mode, with an inlet temperature of 90-102 °C and 28 ± 1 °C, respectively. Cooling performance between two different examined materials was examined by comparing the temperature differences (ΔT_{HX}) between the average temperatures at the inlet against the average temperature at the outlet of the heat exchanger, for both cases. Furthermore, uniformity of temperature distribution within the product was compared at the exit of the heat exchanger and after application of vibration mixing at the resonance frequency of the mixing unit, 20 Hz for both cases. Temperature distribution uniformity and thermal mixing efficiency were evaluated by comparing the maximum temperature difference between the product temperature at the three cross-sectional points at the outlet of the heat exchanger (ΔT_{MAX}) and at the outlet of the mixing unit (ΔT_{MAX,M}), respectively. Finally, cleaning efficiency of the tested materials was examined by visual comparisons of the outer surface of the inner tube for both cases, at the end of a short period of washing-down the heat exchanger with hot water.

The results for all the tested foods showed a total improvement of 25% of cooling efficiency when using the inner tube of the heat exchanger covered with the PTFE textile. Cooling of cheese sauce was significantly improved for the case of PTFE textile as the food-contact surface, with a ΔT_{HX} of 16 ± 2, compared to a ΔT_{HX} of 11 ± 2 °C for the case of the untreated stainless steel heat exchanger. Similar to the cheese sauce, cooling performance for banana puree was better with the PTFE textile, with a ΔT_{HX} of 14 ± 2 °C, compared with a ΔT_{HX} of 9 ± 2 °C for the other case. For the cooling of sweet potato puree, a slight improvement of ΔT_{HX} was observed for the heat exchanger with the PTFE textile with a value of 14 ± 2 °C, while a ΔT_{HX} of 12 ± 2 °C was observed with the untreated stainless steel heat exchanger.
The results of cross sectional temperature distribution of the flowing product at the exit of the heat exchanger were different for the products examined. Moreover, more uniform temperature distribution during cooling of sweet potato puree was observed for the case of the untreated stainless steel with a $\Delta T_{\text{MAX}}$ of 2-4 °C compared to a $\Delta T_{\text{MAX}}$ of 8-11 °C, for the PTFE textile case. The opposite results were observed during cooling of cheese sauce with a $\Delta T_{\text{MAX}}$ of 5-8 °C and 8-10 °C, for PTFE textile and untreated stainless steel surfaces, respectively. Finally, no difference was observed between the examined materials for the case of banana puree with a $\Delta T_{\text{MAX}}$ of 10-12 °C, for both cases. Product temperature distribution was significantly improved by employing vibration, observing uniform temperature cross sectional distribution for all the materials, with $\Delta T_{\text{MAX}}$ of 0-3 °C.

Comparing the efficiency of cleaning of the examined materials with sweet potato, no significant difference was observed, with no residual amount of product to be visible on the outer surface of the inner tube of the heat exchanger, probably due to the viscoelastic behavior of sweet potato puree and the non-adhesive behavior exhibited by such products. However, cleaning efficiency of the heat exchanger was significantly improved using the inner tube covered with the engineered PTFE textile for both banana puree and cheese sauce. Using the PTFE textile the amount of product deposition at the pipe wall, after washing down with hot water, was significantly lower, compared to the untreated heat exchanger, an indication of lower product accumulation at the heat exchanger pipe wall, and hence a reduction of the resistance to heat transfer due to the fouling layer deposition.

Finally, this research showed that a combination of a good conductive material, such as stainless steel, with a food-contact surface with antifouling surface characteristics can significantly enhance cooling and potentially improve the sanitation conditions during the
processing of viscous foods. Antifouling food-contact materials for the purpose of this research were fabricated using the knitted textile construction methods, a newly introduced low-cost modification technique with a great potential for future research studies on advanced thermal processing applications (heat exchangers, mixing units, holding tubes, spray dryers, food packaging).

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Nomenclature

Latin letters

AFM Atomic Force Microscope (AFM)
CA Contact Angle (°)
MW Microwave system
PTFE Polytetrafluoroethylene

Ra Average roughness (nm)
T Temperature (°C)

Greek letters

ΔT Temperature difference (°C)
Subscripts

MAX refers to the maximum temperature difference between the 3 cross-sectional points at the exit of heat exchanger

MAX_M refers to the maximum temperature difference between the 3 cross-sectional points at the exit of mixing unit

HX refers to the temperature difference between the average temperature at the inlet and outlet of the heat exchanger
References


CHAPTER 8

Research summary and future outlook

8.1. Research summary

During continuous flow cooling of viscous food products, laminar flow and low thermal conductivity, characteristic for these materials, compounded by product accumulation and fouling formation at pipe walls of processing equipment (heat exchangers, mixing units), lead to non-uniform, slow cooling processes, increasing the operating costs and degradation of quality. To address these problems, enhancement of cooling of viscous products, such as applesauce, sweet potato puree, banana puree, and cheese sauce was examined numerically and experimentally, using computer simulation studies and surface modified heat exchangers, respectively. Numerical studies examined different flow configurations and heat exchanger designs to determine the most efficient operational conditions for cooling of applesauce and sweet potato puree. Experimental studies with sweet potato puree, banana puree, and cheese sauce were conducted to compare the efficiency of an untreated stainless steel (series 300) and several surface-modified heat exchangers on cooling, thermal mixing, and cleaning performance respective to the foods tested.

Numerical analyses were conducted using computer simulations with Multiphysics software system Comsol 5.2, in order to study continuous flow cooling of viscous food products, such as applesauce and sweet potato puree. Computer simulation modeling using Computational Fluid Dynamics (CFD) and heat transfer software modules, was used to compare two different flow configurations within the same tubular heat exchanger: i) cooling process of the food, while flowing within the inner tube of the heat exchanger and ii) cooling process of the food, while flowing within the annular passage of the heat exchanger (outer
Numerical studies calculated and compared the product bulk temperature ($T_{\text{bulk}}$) at the exit of the heat exchanger, temperature distribution, and velocity profile of the product during cooling studies for both examined flow configurations. Moreover, the potential improvement in the cooling efficiency, resulting from the change of operating conditions due to the movement of process material flow from the internal tube into the external annulus could be 14% and 23%, for applesauce and sweet potato puree, respectively.

The improvement of cooling for the tested foods flowing in the annulus was a result of the heat lost from the product to the environment. Energy balance calculations showed the additional energy losses from the product flowing in the annulus to the environment was in the range of 174-699 W and 91-369 W, for sweet potato puree and applesauce, respectively. The most efficient cooling of sweet potato puree was observed with the product flowing within the annular passage of a heat exchanger, with the equal cross-sectional area (the outside diameter of the inner tube at 0.0558 m (2.2 in)) for both inner tube and annular product flow, while for the case of applesauce, design characteristics of the heat exchanger (using the equal cross-sectional area or hydraulic diameter) did not have the same impact. Comparing the temperature and velocity data for both applesauce and sweet potato puree, more efficient cooling was observed during cooling of sweet potato puree flowing in the annular flow configuration, resulting in a parabolic velocity flow profile type. Comparatively less efficient cooling was observed with applesauce flowing in the annulus, with a slower flattened velocity profile (plug flow type velocity profile). The slower and flattened velocity profile of applesauce resulted in a less efficient cooling, compared with sweet potato puree, due to the lower product heat loss to the environment.
In conclusion, based on the results of numerical studies of cooling of highly viscous materials, the switch of the product flow location to the annulus of the heat exchanger could yield potential benefits of enhancement of conventional continuous flow cooling, while improving the sanitation conditions in the processing plant. However, prior to the broader industrial applications of this cooling method, additional experiments and computational studies still need to be conducted for better understanding and addressing the problem of static or "dead" spots observed in the transition area of the product flow into the annular passage, considering the potential cleaning and sanitation complications.

Using the heat exchanger design and flow configuration improvements observed from the computer simulations, a preliminary study was conducted to examine the effects of surface modifications on cooling and cleaning processes of viscous foods, such as sweet potato puree, banana puree, and cheese sauce. In this study cooling, thermal mixing, and cleaning performance were compared between two identical horizontal stainless steel tube in tube heat exchangers, one with untreated food-contact surface and one with the food-contact surfaces chemically treated with a non-food grade, commercially available hydrophobic solution (Aculon,). Cooling efficiency for the products tested was compared under identical operating conditions (flow rate, inlet temperature), under steady state regime, with the tested food products flowing within the annular passage and with the coolant (water) flowing through the inner tube in counter-current mode.

Cleaning studies shown that for the case of the treated heat exchanger, the amount of product deposition at the pipe wall was significantly reduced, compared with the untreated heat exchanger. Reduction of product accumulation and fouling formation at the pipe wall of the heat exchanger, resulted in a significant improvement of cooling performance for banana puree
and cheese sauce, using the treated heat exchanger, with the average outlet product temperature reduced by 4-6 °C more, compared with the results for the untreated heat exchanger. However, for sweet potato puree, cooling using the untreated heat exchanger was slightly better, with the average outlet product temperature 2-4 °C lower, compared to the treated case. Furthermore, cross sectional temperature distribution within the flowing product during cooling and thermal mixing for all the tested materials was more uniform with the treated heat exchanger. The maximum recorded temperature difference among the three points, at the exit of the heat exchanger (ΔT_{MAX}), was lower by 6-7 °C, 5-6 °C, and 2 °C, for sweet potato puree, banana puree, and cheese sauce, respectively, for the treated heat exchanger compared to the untreated one. Thermal mixing studies, via application of vibration at the resonance frequency of the mixing unit at 20 Hz, showed good results for all the experiments with sweet potato puree and banana puree. Application of vibration improved the temperature uniformity within the product, with a maximum recorded temperature difference at the exit of the mixing unit (ΔT_{MAX}), in the range of 2-4 °C, for sweet potato puree and banana puree. However, for cheese sauce, no significant improvement in temperature uniformity was observed via application of vibration, with the ΔT_{MAX} in the range of 8-10 °C.

Results of these preliminary experiments indicated that hydrophobic surface characteristics of the food contact surfaces resulted in enhancement of cooling of viscous foods and also in the potential reduction of the cooling-related food quality losses while improving the sanitation processing conditions. However, results obtained for sweet potato puree indicate the need for more studies in order to better understand the effects of surface characteristics on cooling of products with different composition.
In continuation of the previous preliminary studies, the effects of different surface characteristics such as wettability, roughness, and topography on the adhesive behavior of sweet potato puree, banana puree, and cheese sauce, and the flow behavior of Carboxymethyl Cellulose (CMC) solution (conc. 1.5%) were examined for potential applications in continuous flow cooling and mixing processes. Effects of the surface characteristics on the adhesive behavior and flow behavior of tested viscous products were examined by testing different engineered food-contact surface samples, including stainless steel samples, untreated and modified using chemical and lubricant surface treatments (Rain-X, Aculon, Sprayon LU206), fabrics (knitted and woven structures, untreated and treated with a commercial fluorocarbon-based resin), polytetrafluoroethylene (PTFE) sheets, silicone sheets and fiberglass-reinforced silicone polymer sheets. Surface wettability was one of the parameters used to differentiate the examined surfaces, varying from hydrophilic (0-90°) (Rain-X treated stainless steel), low hydrophobic (90-100°) (untreated stainless steel, Sprayon LU206 treated stainless steel), hydrophobic (Aculon treated stainless steel, PTFE, silicone sheets, fiberglass silicone sheets, untreated textiles) up to superhydrophobic (150°) (fluorocarbon treated textiles) characteristics. Surface roughness was the second parameter used to quantify the differences between the surface samples, represented by samples with very smooth surfaces such as Aculon and Sprayon LU206 treated stainless steel samples and at the other end of scale with the highly rough surface of silicone sheets. The rest of the samples had an intermediate surface roughness but higher than the untreated stainless steel surface used as the reference sample for this study.

Adhesive behaviors of tested foods and repellent behaviors of the engineered surfaces were examined by measuring the residual product mass which remained attached to the testing surface, during the standard methods of product depletion and centrifuge adhesion tests. Both
of these two testing methods were performed under two different temperature conditions, at room temperature (20–23 °C) and at cooling process stage temperature conditions with the initial temperature of the examined food samples at 95-100 °C. During the experiment at cooling temperature conditions, the initial temperature of the food-contact surface samples was at 5 °C for product depletion tests, while the surface temperature was maintained within the temperature range of 17–20 °C during the centrifugal adhesive tests.

Results from the product depletion test showed that the hydrophobic surfaces with smooth characteristics (chemically treated stainless steel samples), PTFE, and treated knitted fabrics retained the lowest amounts of the tested food products. The average residual mass for all the products on these surfaces was approximately at 35–45% at room temperature, and 45–65% at cooling temperature conditions.

Moreover, under cooling conditions, time-temperature data of the highest surface temperature was recorded during the product depletion tests. Comparing the temperature differences (ΔT), between the initial and the final temperature, for each of the tested products and examined food-contact surfaces, the untreated and chemically treated stainless steel samples, additionally with PTFE presented the highest ΔT values for banana puree and sweet potato puree at 35–38 °C. Cheese sauce was the food product with the lowest ΔT values for all the food-contact surfaces used, with only the hydrophobic smooth Aculon treated stainless steel sample exhibiting a ΔT higher than 35 °C. Product solidification observed for cheese sauce on the surface of the chilled/cooled examined samples, had a negative impact on product depletion, and also resulted in lower ΔT values.

Adhesive behavior of the tested foods and repellent behavior of the examined food-contact surfaces were examined using the centrifugal adhesive test. Under two different
temperature conditions, three different rotating speeds at 44 rpm, 66 rpm, and 88 rpm were tested for each food sample and contact surface material. The applied centrifugal force has increased as the speed of rotation increased, resulting in a lower amount of retained product on the surfaces of the rotating surface samples. Independent from the speed of rotation and the examined food-contact surface, sweet potato puree exhibited the least adhesive behavior, with the residual product at 0-10% at room temperature and 10-20% at cooling conditions. For a rotational speed of 66 rpm or higher, the best repellent behavior with banana puree was observed for the hydrophobic surfaces treated with Aculon stainless steel and the knitted fabric samples, with a residual amount of product at 0-4% and 15-32%, at room and cooling temperature conditions, respectively. Finally, for cheese sauce, only knitted fabric samples (superhydrophobic and hydrophobic) reduced the amount of the residual product on the surface of the rotating sample, under all temperature conditions. The best repellent behavior with cheese sauce was observed with the superhydrophobic knitted fabric samples, at the maximum speed of rotation, with a residual amount of product at 11% and 39%, at room temperature and cooling temperature conditions, respectively.

Moreover, the velocity profile of 1.5% CMC was studied using particle tracking velocimetry (PTV) as the testing method. Video microscopy was used to capture the movement of fluorescent tracer particles, introduced at the inlet of the heat exchanger. Recorded video frames were analyzed using a particle tracking Matlab algorithm. The Matlab algorithm was used to identify the position of the particles in each examined frame and created particles trajectories as a function of time, based on the linked particles from the sequence of the examined frames. The Matlab algorithm processed the position of the trajectories as a function of a time step to calculate the mean-square displacement (msd) of each particle. The estimated
results of msd data as a function of time were statistically processed with the position data from the particle trajectories to link the data with the tracer particles and calculate the velocity of each particle. Velocity profiles and particle velocity magnitudes were compared for 1.5% CMC flowing in the annulus, for each examined food-contact surface sample. The PTV results of the velocity profile of 1.5% CMC, showed a parabolic velocity profile for almost all the examined cases. The flowing particle was located close to the center of the annulus flow, with a velocity magnitude between 0.050-0.058 m/s, for samples with the smallest flow cross sectional area (silicone sheets, silicone fiberglass sheets, PTFE and textile samples) and for the lubricated smooth stainless steel surface, treated with Sprayon LU206. For samples with the characteristic hydrophobic (Aculon treated stainless steel surface) and superhydrophobic (treated knitted textile) behavior, most of the tracing particles were concentrated away from the inner tube wall, near to the outer tube wall and at the center of the annulus, respectively.

The results of the adhesive and flow behavior of viscous products against different examined surfaces showed that hydrophobic surfaces, with partial non-wetting and smooth characteristics, are the ideal surfaces for cooling of viscous foods. On the other hand, more rough surfaces, with more complex topography, and hydrophobic/superhydrophobic behavior appear to be the preferred surface types for mixing application.

The final study of this research was to fabricate a surface with hydrophobic, partial non-wetting and smooth characteristics, food-grade and with long term stability for cooling applications of viscous foods. Using the method of knit textile construction, an easy and simple method to modify the surface topography (roughness, structure) and with low production cost, food-grade PTFE textile samples were fabricated as the examined surface for cooling experiments. PTFE was chosen due to the hydrophobic and partial non-wetting behavior, the
long-term stability of anti-fouling characteristics and the great stability in a wide range of temperature conditions.

Similar to the preliminary studies with the hydrophobic chemical treated heat exchanger, cooling, thermal mixing and cleaning efficiency of sweet potato, banana puree, and cheese sauce were examined by comparing two different food-contact materials, namely untreated stainless steel (series 300), and the food-grade PTFE textile surface sample. The outside surface of the inner tube of a tube in tube heat exchanger was used as the tested food-contact surface, applying on it the engineered PTFE textile sample. Cooling experiments for both examined materials were conducted under steady state and identical operating conditions (flow rate, inlet temperature), with the tested food products flowing within the annular passage of the heat exchanger and the coolant (water) flowing in the inner tube, in counter-current mode, with the inlet temperatures of 90-102 °C and 28 ± 1 °C, respectively.

Results for all the tested foods showed a total improvement of cooling efficiency between 15-25% when using the inner tube of the heat exchanger covered with the PTFE textile. Cooling of cheese sauce and banana puree were significantly improved for the case of PTFE textile as the food-contact surface, with the average outlet product temperature reduced by 6-9 °C, compared with the results for the case of the untreated stainless steel heat exchanger. Cooling of sweet potato puree was slightly improved with the PTFE textile, with the average outlet product temperature been 2-4 °C lower, compared to the untreated stainless steel case.

Results of temperature cross sectional distribution of the flowing products at the exit of the heat exchanger were different for the products examined. More uniform temperature distribution during cooling was observed for sweet potato puree for the case of the untreated stainless steel with a $\Delta T_{\text{MAX}}$ of 2-4 °C, compared to a $\Delta T_{\text{MAX}}$ of 8-11 °C, for the PTFE textile
case. The opposite results were observed during cooling of cheese sauce with a $\Delta T_{\text{MAX}}$ of 5-8 °C for PTFE textile, while a $\Delta T_{\text{MAX}}$ of 8-10 °C was observed for the untreated stainless steel surface. Finally, no difference was observed between the examined materials for the case of banana puree with a $\Delta T_{\text{MAX}}$ of 10-12 °C, for both cases. Product temperature distribution was significantly improved by employing vibration, observing uniform temperature cross sectional distribution for all the materials, with $\Delta T_{\text{MAX}}$ of 0-3 °C.

Furthermore, cleaning efficiency of the heat exchanger was significantly improved using the inner tube covered with the engineered PTFE textile for both banana puree and cheese sauce. The amount of product deposition at the pipe wall, after washing down with hot water, was significantly lower with the PTFE, an indication of lower product accumulation at the heat exchanger pipe wall, and hence a reduction of the resistance to heat transfer due to the fouling layer deposition. Finally, no difference was observed in cleaning efficiency of the examined materials with sweet potato, probably due to the viscoelastic behavior of sweet potato puree and the non-adhesive behavior exhibited by such products.

In conclusion, based on the results of this research, having the food product in the annulus of a surface-modified heat exchanger, with good conductive and antifouling surface characteristics, can significantly enhance cooling and cleaning processes of viscous foods, by reducing product accumulation and fouling formation, and hence reduce the resistance to heat transfer. Furthermore, antifouling food-contact materials for the purpose of this research were fabricated using the knitted textile construction methods, a newly introduced low-cost modification technique with a great potential for future research studies on advanced thermal processing applications (heat exchangers, mixing units, holding tubes, spray dryers, food packaging).
8.2. Future outlook

The presented research demonstrated several methods to improve cooling of viscous foods. The following list notes some recommendations for future work in the field of cooling and surface modification for potential studies and applications in advanced thermal processing:

- Numerical studies showed that having the food product flowing within the annular passage of the heat exchanger resulted in enhancements of cooling process while maintaining the sanitation conditions in the processing plant. However, before broader industrial applications of this cooling method are implemented, proposal for future work, would be to conduct additional experiments and computational studies for better understanding and addressing the issue of static or "dead" spots observed in the transitional area of product flow to the annular passage, regarding the potential cleaning and sanitation difficulties.

- During this study, a significant difference in cooling and cleaning efficiency was observed with different foods tested. Cooling and cleaning results for banana puree and cheese sauce indicate the importance of food product surface characteristics during thermal processing of these products. However, the adhesive behavior of sweet potato puree appeared to be independent of the tested surface characteristics and behaved similarly for all surfaces tested in this research. This is an observation that probably can be explained due to the viscoelastic behavior of sweet potato puree based foods. This observation further indicates the need for future studies, more focused on the dynamic rheological parameters of the food samples, such as viscoelastic properties, glass transition temperatures, and cloud point of lipids, for a better understanding of interactions between the examined surface materials and flowing food materials. This information can be very useful for adjustments of operating conditions and surface characteristics to reduce fouling formation.
- For this study, antifouling food-contact materials were fabricated using the knitted textile construction methods, a newly introduced low-cost modification technique, with a great potential for future research studies to advanced thermal processing applications (heat exchangers, mixing units, holding tubes, spray dryers) and food packaging.

- This study showed that using surface modified heat exchangers, by combining good thermal conductivity (stainless steel) and antifouling surface characteristics (engineered PTFE textile), can significantly enhance cooling, and potentially improve the sanitation conditions during the processing of viscous foods. For future recommendations, regarding broader commercial applications in the food industry, surfaces with these characteristics can be achieved by reinforcement of antifouling materials, such as the engineered PTFE textiles, on the food-contact surface of stainless steel wires and hoses. Furthermore, new developments in laser-etching techniques on the surface of stainless steel, to create hydrophobic food contact surfaces, could potentially be used as a more expensive option for the same purposes.

- Finally, this study presented additional improvements of cooling of viscous foods, using independently, the methods of surface modification of heat exchangers and thermal mixing. Based on these observations the next step on cooling studies and developments could be the combination of these methods. Potential advanced cooling methods, using surface modified heat exchangers, with the product flowing in the annulus, while rotating both the inner and outer tube of the heat exchanger, could potentially minimize cooling time and cooling-related product quality losses, while improving the sanitation processing conditions and reducting both time and chemical usage required for the post-process clean in place treatments.
REFERENCES


APPENDIX A:

Computer Simulation of Cooling of Applesauce under Processing Conditions

For the case of applesauce an additional computer simulation study was conducted, comparing the efficiency of cooling of food in a counter-current flow mode with cold water (4 °C), for the same two scenarios: i) with the product flowing within the annulus and ii) within the inner tube. For this study case, the initial temperature of the food was at 100 °C, a more representative temperature regarding the commercial thermal processing of applesauce, characteristic high acid food product (applesauce pH<4.6). Moreover, the results of this simulation presented a slightly better performance for the case with applesauce flowing within the annulus passage with a bulk temperature at the outlet of the cooler at 69.3 °C, compared with a temperature of 70.9 °C, for the other case (Fig. A.1).
Figure A.1. Simulation results of temperature distribution for cooling of applesauce, flowing in a counter-current flow mode: a) within the annulus side and b) within the inner tube. Note the inlet temperature at 100 °C.