ABSTRACT

CHACON OVALLES, LISANDRA V. Towards the Redesign of Advanced Recycled Paper-based Containers: At the Nexus of Environmental Perception and Packaging Performance (Under the direction of Dr. Richard Venditti and Dr. Nathalie Lavoine).

This research explored the development of redesigned paperboards with obvious recycled content appearance for packaging applications as a better strategy to communicate sustainability to consumers. The research aimed to (i) endow the samples with direct clues of eco-friendliness that can help consumers identify packaging with high recyclability levels and facilitate the purchase decision and (ii) decrease the amount of waste materials ending in landfills by proposing a novel valorization pathway. The strategy developed involves incorporating partially disintegrated waste from different sources into old corrugated containers (OCC) pulp to produce specimens with a recycled appearance.

The first part of this research evaluated the effect of mixed office waste (MOW) particle size on the visual appearance and performance of recycled containers. MOW particles with different apparent aspect ratios, *i.e.*, 52 (macroparticles), 72 (microparticles), and 163 (nanoparticles), were used during the study. The results showed that the incorporation of MOW particles with the lower apparent aspect ratio (larger particles) resulted in paperboards with a redesigned appearance (visible particles of recycled material on their surface). However, increasing loading amounts of large waste particles had a detrimental effect on the mechanical performance of the samples. MOW particles with the highest apparent aspect ratio (smaller particles) did not endow the paperboards with obvious clues of sustainability. Still, they improved the mechanical properties of the samples significantly. Thus, using both low and high aspect ratio MOW particles could be used as a strategy to engineer packaging with a high environmental perception that meets product strength specifications.

The second part of this dissertation further evaluated the influence of waste physicochemical properties on the performance of recycled packaging with redesigned appearance. In this study, representative groups of waste were selected to mimic the diversity found in recycling mills and studied their chemical interaction with recycled fibers. Mixed office waste (MOW), old magazines (OMG), and paper cups laminated with polylactic acid (PLA) were used in this study. MOW had a lesser effect on the mechanical performance of the paperboards due to its easy disintegration and predominant hydrophilic groups found in its chemical composition that allowed the interaction with the recycled fibers through hydrogen bonds. More hydrophobic waste, such as PLA paper cups, on the other hand, significantly affected the samples' performance. Hydrophobic particles disrupted the inter-fiber network by preventing the formation of hydrogen bonds. The utilization of well-investigated dry strength agents such as cationic starch and cellulose microfibrils (CMFs) could effectively restore the mechanical properties of the paperboards containing the less hydrophobic and easy-to-disintegrate waste (MOW and OMG). The production of multi-layer paperboards using the redesigned paperboards as an outer layer and only OCC in the inner layer enhanced the mechanical properties of all the samples regardless of the physicochemical properties of the waste. Thus, by using a multi-layer configuration, waste with different physicochemical properties can be used to redesign the appearance of recycled paper-based containers without compromising the mechanical performance of the final product.

The third study explored consumers' sustainability perception toward the redesigned paperboards (with an obvious waste content appearance from paper and agricultural waste) and the influence of the presentation format (online vs. in-person) on the packaging perception using the best-worst scaling (BWS) experiment. Interviewed consumers perceived the redesigned packaging as more environmentally friendly regardless of the type of waste compared to the traditional packaging (neat appearance). Obvious particles of waste served as signals to communicate sustainability effectively to consumers and can be used to guide their purchase decision.

Finally, **the fourth study** explored further exploitation of recycled fibers for food packaging applications by enhancing the barrier properties of the paperboards using two bio-based coatings (*i.e.*, CMF-reinforced starch (S-CMF) and epoxidized cottonseed oil cross-linked with citric acid (CEPO)). This research aimed to (i) replace the traditional poly-fluoroalkyl substances (PFAS) and petroleum-based barrier coatings that have an adverse effect on the health and environment and (ii) broaden the applications of the redesigned paperboards to food packaging by providing water and oil & grease resistance properties. Recycled paperboards coated with multiple layers of both CEPO and S-CMF showed good barrier properties against water, water vapor, and oil & grease, comparable to commercial paper-based food packaging used for fast food applications. The hydrophobic nature and the cross-linked structure developed by the CEPO coating contributed to the water resistance of the samples, whereas the hydrophilic behavior of the S-CMF and the closed structure developed by the presence of the nanocellulose significantly improved the oil & grease, and water vapor resistance of the paperboards. This multi-layer barrier approach in which the individual barrier properties of each coating amalgamated together to enhance the overall properties of the paperboard could be used as a more sustainable alternative for food packaging with high environmental perception.

Towards the Redesign of Advanced Recycled Paper-based Containers: At the Nexus of Environmental Perception and Packaging Performance

by
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DEDICATION

To my lovely husband and my adorable kids. Without any doubt, your support, patience, and love were vital to achieving this goal.

To my wonderful parents, your trust and encouragement motivated me to keep going and never give up.

BIOGRAPHY

Lisandra Chacon was born in 1982 in Merida, Venezuela. She received a B.S. in Chemical Engineering from Los Andes University in Merida, Venezuela, in 2006. During her senior year, she interned in R&D in one of the most important food industries in Venezuela. In 2008 she joined the Department of Industrial and Applied Chemistry, College of Chemical Engineering, at Los Andes University as an assistant professor and worked there for eleven years. She earned a MSc in Analytical Chemistry from the Department of Chemistry, College of Science, Los Andes University, in 2012. She continued her professional career in Mexico as a professor in the Department of Analytical Chemistry at the National Autonomous University of Mexico in 2019. In 2020 she joined the Department of Forest Biomaterials at North Carolina State University, Raleigh, North Carolina, to pursue her doctorate in Forest Biomaterials. After graduation, she will continue contributing to a more sustainable life, working with bio-based materials for multiple applications.

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Chapter 1 . Introduction

1.1. Overview of recycled fiber-based materials in the packaging industry

The growth of e-commerce has influenced and impacted the pulp and paper industry in several ways, from decreasing the demand for printing & writing paper grades to empowering the packaging sector. This growth has been exacerbated by the COVID-19 pandemic, which increased e-commerce sales in 2021 by 50% over 2019 (Goldberg, 2022). The changes in consumer behavior pair with an increasingly online retail activity, which in turn results in higher demands for packaging material for delivery than for retail in a store. The e-commerce packaging market was valued at $ 27.5 bn. in 2020 and is expected to reach $65.6 bn. by 2028due to this new trend (DataM Intelligence 4Market Research LLP, 2022).

The new e-commerce marketing trend has generated two important challenges: (i) the need for increased production of paper-based products to satisfy the higher demand and (ii) the implementation of sustainable solid waste management practices to reduce the negative impact of resulting consumer waste (Escursell et al., 2020).

Recycling paper-based products is one strategy that can address the previously mentioned challenges. Herein, we define recycling as the successive steps to produce paper-based products from post-consumer recycled fibers, meaning fibers extracted from any paper, paperboard, fibrous materials, and related products that have passed through their first intended use (Scott 2019). On one hand, recycled fibers are a valuable source of raw material for paper-based products. Old newsprint (ONP), old magazines (OMG), mixed office waste (MOW), and old corrugated containers (OCC) are examples of different paper wastes used as secondary (or recycled) fibers to generate new paper-based products. On the other hand, reusing paper waste can significantly reduce the amount of waste accumulating in landfills. Effective recycling could save energy and water resources, reduce greenhouse gas emissions (Iosip et al., 2010), and help achieve the Development Sustainable Goals set up by the United Nations that promote the (re)use of renewable resources and better waste management practices.

Recycling has a long history, but with the increasing environmental awareness of society, efforts in recycling have been accelerated. On average, 68% of paper waste is recycled in the US (EPA, 2020). Strategies to increase this global recycling rate would involve, among others, understanding better the implications of contaminants in recycled pulp, educating consumers on recycling, and increasing the number of available recycling facilities.

One of the main challenges/concerns in using recycled fibers in recycling/papermaking operations lies in the low quality and poorer performance of recycled fibers compared to their virgin counterpart (*i.e.*, non-recycled fibers) and the presence of contaminants among the recycled fibers slurry. Contaminants (components other than the fibers in the recycled furnish) can disrupt the papermaking and recycling operations and alter the performance of the end-use products. With the increasing utilization of recycled products and the growing amount of paper waste, recycling mills are toned to find strategic solutions for converting a higher amount of low-quality fibers into high-value products and addressing the increasing quantities of contaminants from these waste streams. The push from society and governmental agencies to reduce, reuse, and recycle materials also exerts additional pressure to implement quick and cost-effective solutions to the previously mentioned challenges. Thus, efforts to better understand how contaminants with different physicochemical properties and sizes interact with the recycled fibers are needed to (1) increase the use of recycled fibers as fiber-based products such as packaging, (2) reduce the impact of low-performance recycled fibers on the performance of the final product, and further (3) exploit this valuable source into sustainable advanced materials.

1.2. Visual appearance and consumers' recognition of recycled packaging

With the technological advancements in the paper industry, the recycled fibers used in packaging manufacturing look as clean as non-recycled ones and could have similar properties as paper made out of virgin fibers if treated properly (Hasanin et al., 2020; Sanchez-Salvador et al., 2020). Over the past decades, research efforts dedicated to the development and improvement of methods to remove and reduce the size of contaminants present in recycled fibers slurries have been a major focus to decrease the impact of contaminants on the appearance and performance of recycled fiber-based materials (Ballinas et al., 2020; Geng et al., 2020; Hasanin et al., 2020).

Nowadays, the efforts to utilize more recycled materials in paper-based packaging are accompanied by the need to reduce the negative impact of packaging waste accumulation on the environment and satisfy consumers' demand for more eco-friendly options. Recent studies have reported that consumers' purchase decisions are grandly influenced by the sustainability of the products and the brand's commitment to the environment (Carvalho et al., 2022; Mohan, 2019). As a result, consumers prefer the so-called eco-friendly packaging over conventional ones and are willing to pay more for these *greener* options (Carvalho et al., 2022). However, if recycled paper products look like conventional ones, how could consumers recognize them?

While industries endeavor to shift from no eco-friendly manufacturing processes and raw materials to sustainable alternatives, they are also working to communicate the environmental attributes of the packaging assertively to the consumers. Thus far, extrinsic characteristics such as labels (also referred to as ecolabelling) have been the preferred strategy to convey the message (Borgman et al., 2019; Boz et al., 2020; Herbes et al., 2020; Krah et al., 2019; Magnier et al., 2016; Magnier & Crié, 2015; Martinho et al., 2015; Steenis et al., 2018). Ecolabels provide graphical information that aims to help consumers recognize sustainable packaging. Yet ecolabels' effectiveness has been compromised due to the extensive proliferation, complexity, and technicality of the information displayed and the loss of credibility with publicly condemned dishonest practices such as greenwashing (Yokessa et al., 2020). Ecolabels can confuse consumers rather than ease their purchase decisions (Boz et al., 2020; Moon et al., 2017).

Additionally, consumers rely on implicit packaging characteristics such as the type of packaging material (glass, paper, plastics, among others) to determine whether or not the packaging is sustainable (Ischen et al., 2022). For instance, paper-based packaging is perceived as a more environmentally friendly option than plastic-based packaging (Magnier et al., 2016), although this is not necessarily true.

A better approach to communicating sustainability is thus required to convey correct and proper information to the consumers because of the highlighted uncertainty of consumers in assertively recognizing sustainable packaging.

1.3. Recycled packaging for food applications

A better understanding of recycled fibers and the role/effect of contaminants on recycled products could contribute to developing new advanced and sustainable materials for food packaging applications. Food packaging is the major contributor to the environmental impact associated with packaging waste generation and accumulation. 55% of the global production of rigid plastic packaging is used in the food industry (Figure 1-1), and only 21% is recycled (Tiseo, 2021a). The rest of the material is either landfilled (54%) or incinerated (21%), causing significant environmental issues (Tiseo, 2021b). Although petroleum-based plastics have been the preferred material in food packaging, the food industry is making massive efforts to replace them due to their lack of biodegradability and fossil fuel dependency (Singh et al., 2021),

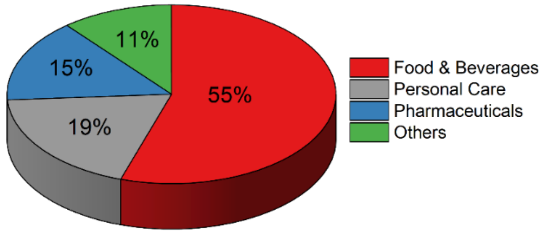

Figure 1-1. Global rigid plastic market (Adapted from Grand View Research, (2018)).

Companies are increasingly considering using fiber-based materials, such as paper & paperboard, to provide a sustainable alternative to plastic packaging. WestRock, for instance, has designed a paper-based solution called CanCollar® to replace the harmful plastic rings used in the canned beverage industry (Figure 1-2a) (WestRock, 2017). The new alternative is fully biodegradable and recyclable. Similarly, the Coca-Cola Company has developed a paper-based bottle prototype as a packaging solution for the thousands of plastic bottles ending up in the oceans yearly (Figure 1-2b) (The CoCa-Cola Company, 2020). Paper-based materials promise to replace plastic-derived packaging materials predominant in the food packaging industry.

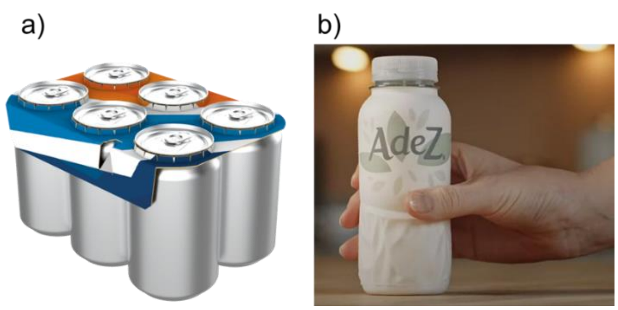

Figure 1-2. Paper-based packaging substitutes for traditional plastic options. a) Can ring developed by WestRock (WestRock, 2017), b) Coca-Cola bottles (The CoCa-Cola Company, 2020).

Herein, we propose to exploit recycled fibers as another sustainable solution to plastic-food packaging. The utilization of secondary fibers in food packaging is, however, known to be challenging because (i) recycled fibers do not necessarily provide a food-contact, food-grade approved platform (Suciu et al., 2013), and (ii) the performance of recycled fibers does not match the performance of virgin fibers or plastic materials, nor do they meet the stringent specifications of food packaging materials (Wang et al., 2022).

Recycled fibers are already used either as primary or secondary packaging for food products, but mainly for packaging dry food products such as flour, sugar, rice, and eggs. However, the main concern regarding recycled paper-based boxes is the possible migration of potentially harmful contaminants that remain in the recycled stock when used in direct contact with wet food. Recycled fibers could contain multiple contaminants that can compromise their suitability for food contact applications. Some contaminants are unsafe and could be at concentrations that jeopardize human health or the product's organoleptic properties (Jamnicki et al., 2012). Since there are current legislations and regulations in the US and EU for recycled food-contact materials (Misko, 2013), such as the Food and Drug Administration (FDA, US) and the Council of Europe resolution (CoE), additional investigations need to be conducted in this area to expand the utilization of recycled materials further.

Food packaging needs to protect the product from any damage or contamination triggered by the air, moisture, and microorganism, and these characteristics are provided essentially by the packaging materials. Paper substrates, on their own, cannot accomplish all the mentioned requirements primarily because of the lack of water-, gas- and oil & grease resistance behavior (Wang et al., 2022). That is why in the last decades, multiple studies have centered their attention on pairing the properties of paper-based packaging with the properties of the material they are replacing.

In the past, per- and polyfluoroalkyl (PFAS) substances were extensively used in paper-based packaging to provide the substrate with good barrier properties, especially grease resistance. PFAS were utilized in popcorn bags and food wrappers, among others (Hubbe & Pruszynski, 2020). However, due to the adverse effect on human health and the environment (Glenn et al., 2021), PFAS are being replaced (See Chapter 5). Therefore, there is a need to develop paper-based food packaging with outstanding performance like that provided by non-sustainable materials but, in turn, sustainable (biodegradable and recyclable) and low-cost.

1.4. Research objectives

The work aims to address the previously discussed gaps in the utilization and valorization of recycled fibers in fiber-based packaging. The overarching goals and the respective hypotheses considered are as follows:

Objective I: Redesign recycled containerboards by incorporating obvious waste particles of recycled material on the surface of the substrates so that consumers comprehensibly identify and refer to these containerboards as recycled without the need for any labels and/or claims (**Chapter 2**).

Hypotheses:

- The "environmental perception" of recycled paperboards can be enhanced by incorporating visible particles of waste (here, recycled) materials into the fiber furnish. These particles can change the aesthetic of containerboards and be clearly identified by the consumers.

  In this work, "**environmental perception**" refers to direct clues of sustainability on containerboards that help consumers recognize recycled packaging without the need for any other external signal. In this research, the clues are provided by the visible waste particles on the surface of the paperboard substrates.

- The mechanical performance of paperboards manufactured with waste particles is primarily affected by the size of the waste particles. The larger the particles and, in turn, the higher the environmental perception, the lower the strength of the paperboard.

Objective II: Understand the effect of waste physicochemical properties on the performance of redesigned recycled containerboards with high environmental perception (*i.e.*, including easily-recognized large waste particles) (**Chapter 3**).

Hypotheses:

- The size of waste particles is not the only and primary factor responsible for the decrease in the mechanical performance of containerboards designed with high environmental perception.
- The physicochemical properties (such as hydrophobicity and water absorptiveness) of waste particles can significantly affect the performance of containerboards with high environmental perceptions. For instance, the presence of hydrophobic/water-repellent compounds can prevent the formation of hydrogen bonds between the fibers decreasing the tensile and burst strength of the samples.
- The physicochemical properties of waste particles determine the effectiveness of restorative treatments applied to recover and improve the mechanical performance of containerboards with high environmental perceptions.

Objective III: Evaluate the consumer's perception of redesigned recycled containerboards with high environmental perception (**Chapter 4**).

Hypotheses:

- Consumers will perceive redesigned recycled containerboard with obvious waste content appearance as more environmentally friendly than traditional packaging (with no obvious waste particles).

- The environmental perception of packaging can significantly be altered by the shopping experience of a consumer, for instance, between an online and in-person experience.

Objective IV: Develop a bio-based multilayer coating using epoxidized cottonseed oil cross-linked with citric acid (CEPO) and CMF-reinforced starch for recycled containerboards with good water and oil & grease barrier properties for food packaging applications (**Chapter 5**).

Hypotheses:

- The first layer of epoxidized cottonseed oil cross-linked with citric acid, coated onto the recycled paperboard, renders the substrate with water and water vapor barrier properties because of the hydrophobic behavior and cross-linked structure of the coating.
- The starch matrix reinforced with bio-based nanofillers (*i.e.*, cellulose microfibrils (CMFs)) coated onto the CEPO-coated paperboards endows the paper with an oil & grease resistance behavior. The hydrophilic nature and the reduced porous surface developed by the presence of the coating prevent the penetration of the oil & grease and delay the pass of water and water vapor.

To accomplish **Objectives I, II, and III** of the research, the appearance of recycled paperboards was intentionally modified by incorporating minimally-processed waste from different sources into the recycled furnish prior to papermaking. The primary purpose was to endow recycled paperboards (made from Old Corrugated Containerboard recycled pulp) with a noticeable recycled content that could be easily identified by consumers (by the naked eye) as an innovative solution for enhanced recognition of recycled packaging.

A major part of the work was devoted to investigating the influence of the properties of added waste materials on the visual appearance and performance of recycled paperboards. Three different studies were conducted and are presented in Chapters 2, 3, and 4, as follows:

Chapter 2 discusses how the addition of mixed office waste (MOW) of different apparent aspect ratios (*i.e.*, macro-, micro-, and nano-scale waste) alters the visual appearance, mechanical properties, and air resistance of OCC-based paperboards.

Chapter 3 focuses on understanding how differences in the physicochemical properties of selected paper waste can influence the mechanical performance of recycled paperboards. Three paper wastes (*i.e.*, mixed office waste (MOW), old magazines (OMG), and paper cups laminated with polylactic acid (PLA)) were selected to simulate the large diversity of contaminants found in recycling facilities. Each waste was processed into large particles and incorporated into the paperboards to also endow the substrates with high environmental perception. This study discusses the interaction of waste particles with distinct properties (*e.g.*, hydrophilicity vs. hydrophobicity) with the OCC pulp to offset the negative impact of selected waste and propose solutions to overcome the overall loss of performance observed with the presence of large waste particles.

Chapter 4 explores consumers' perception of paper-based packaging made with a high amount of obvious waste particles for enhanced environmental perception. This study incorporated large agricultural and paper waste particles into the recycled fibers to produce specimens with obvious recycled content. Online and in-person surveys were conducted using the best-worst scaling (BWS) method to determine whether packages with direct clues instilled an enhanced sustainable recognition and promoted a better identification of the product compared to traditional containers with no agricultural and paper waste.

Chapter 5 focuses on **Objective 4** of this Ph.D. work and aims to extend the utilization of recycled fibers to food packaging applications. This research discusses strategies to primarily enhance the water, water vapor, and oil & grease barrier properties of recycled paperboard substrates using exclusively renewable resources. The goal is to develop a fully bio-based coating solution that can replace PFAS and selected petroleum-derived barrier materials. To this end, this last chapter explores the design and engineering of a multilayer system composed of both CEPO and CMF-reinforced starch as a barrier coating onto recycled paper-based materials for food packaging applications.

Chapter 2 . Valorization of Mixed Office Waste as Macro-, Micro-, and Nano-Sized Particles in Recycled Paper Containerboards for Enhanced Performance and Improved Environmental Perception[1]

2.1. Abstract

Recent surveys have shown that consumers do not know how to recognize sustainable packaging and are misled by the excessive usage of environmental cues by the packaging industry. A better approach to communicate sustainability is therefore needed to promote purchasing towards sustainable products. This study proposes to re-design recycled paper-based containers so that consumers easily recognize visually large contaminants in the paper influencing the consumer to refer to this product as recycled and perceive it as sustainable. To this end, the appearance of recycled containers from old corrugated containers was intentionally altered with the addition of processed mixed office waste (MOW) of distinct average apparent aspect ratio (AR) (length divided by width), namely 52 (macro-scale), 72 (micro-scale), and 163 (nano-scale), to produce recycled paperboards with visually noticeable recycled contents. The addition of MOW with the lowest AR resulted in visible particles on the surface of paperboards, evidencing the presence of recycled materials. The mechanical performance with this material, however, decreased. On the other hand, the addition of MOW with the highest AR improved the mechanical properties of the paperboards similar to the addition of nanocellulose but with less obvious cues of it having recycled content in the product. Thus, the combination of low and high AR

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contaminants is suggested to strategically engineer sustainable packaging with high performance and clear visual clues of recycled content and positive environmental perception.

2.2. Introduction

Waste from paper and paperboard-based substrates accounts for more than 67 million tons of municipal solid waste generated in the U.S. annually (EPA, 2020). Although paper and paperboard-based waste is a valuable source of lignocellulosic biomass, overall only 68% is recycled. The rest is either landfilled or incinerated, which negatively impacts the environment (EPA, 2020).

Old newsprint (ONP), old magazine (OMG), mixed office waste (MOW), and old corrugated containers (OCC) are all categories of paper waste that can be used as a source of (recycled) fibers to generate new paper-based products. Among them, OCC pulp has the highest utilization rate (94%), while MOW has the lowest, estimated at 13% (V. Kumar et al., 2020). OCC is mainly used to produce new corrugated boxes or paperboard for consumers' goods, whereas deinking is not required because of the brown color of the final product. MOW, however, is used to produce high-grade de-inked paper products, namely printing, writing, and tissue papers. MOW is mainly composed of recycled copy papers, envelopes, printer scraps, among others, and thus requires an intense deinking process for re-use as high-grade paper products. The difficulty in removing ink, which is the primary contaminant in MOW, makes the rate of recovery of MOW low (B. Li et al., 2011).

The packaging industry is one of the main industries that exploits and valorizes paper waste as a source of fibers. Different products can be engineered using recycled fibers, such as containerboards for shipping and transportation, paperboards for retail sales (*e.g.*, cereal and medicine boxes), and paper bags for shopping, shipping, and goods transportation (American

Forest & Paper Association, 2019). The main challenge in using recycled fibers for packaging engineering lies in the low quality of the recycled fibers (Ackermann et al., 2000; Hubbe et al., 2007). Recycled fibers commonly exhibit lower mechanical and barrier properties than never-dried virgin fibers (Kim et al., 2000). Upon pressing and drying, the pulp fibers undergo different changes commonly referred to as hornification (Howard, 1990). The fibrillar layers within pulp fibers collapse onto each other, creating a strong and rather irreversible intra-fiber bonding network (Minor & Atalla, 1992). The structural and chemical changes induce the semi-irreversible closure of the fibers' cell wall. Therefore, upon recycling, the porous structure of recycled fibers can only partially re-open with limited swelling, resulting in reduced fibers' quality compared with never-dried fibers (Stone and Scallan 1966; Welf *et al.* 2005; Hubbe *et al.* 2007). Several strategies, including refining, the addition of dry strength additives, enzymes, and cellulose nanomaterials to the recycled furnish, have been investigated to overcome the poor performance of recycled fibers (Hubbe et al., 2007; Nazhad, 2005).

However, despite numerous efforts in enhancing the performance of recycled fibers, the presence of a high amount of highly diverse contaminants in the recycled furnish, and especially stickies (*i.e.*, adhesive contaminants with the ability to deposit and adhere to fibers and processing equipment), still affect the quality of recycled pulps negatively. Contaminants such as inks, hot-melt glues, adhesives, and waxes can disrupt the papermaking and recycling operations, in addition to altering the performance of the end-use products (Ballinas et al., 2020; Blanco et al., 2007). Although numerous articles have demonstrated that the presence of contaminants negatively affects the properties of recycled products (Miranda et al., 2008; Venditti et al., 2000), to date, there is still a lack of studies that carefully evaluate how the origin, nature, and chemical composition of contaminants and stickies influence the performance of recycled products.

Recently published works are mostly dedicated to improving the methods for contaminant tracking and characterization (Licursi et al., 2016; Ossard et al., 2017) and developing new approaches for enhancing the properties of recycled fibers and products (Ballinas et al., 2020; Hasanin et al., 2020). A deeper understanding of how contaminants interfere with the performance and esthetic of recycled paper-based products could help manage their presence in the recycled material more effectively (by either offsetting their negative impact or exploiting them further for value addition) and reduce the need for energy-intensive and time-consuming cleaning processing.

A significant increase in the usage of recycled fibers and products has been observed over the past 10 years in the packaging industry, a trend that has been driven by three main societal changes. The online retail activity (i) has considerably increased, with consumers preferring purchasing online rather than in-store (Escursell et al., 2020). The COVID-19 pandemic has also drastically accelerated this new purchase trend (Guthrie et al., 2021). As a result, additional packaging materials are needed for product transportation and home delivery (Chueamuangphan et al., 2020). The second societal change (ii) relates to the Sustainable Development Goals (United Nations, 2020) that promote the use of renewable resources and better waste management practices. E-commerce brings comfort and convenience to users, but in turn, generates a significant amount of paper waste that negatively impacts the environment. Recycling is thus one strong strategy to lower the accumulation of packaging waste in landfills, save energy and water resources, and reduce greenhouse gas emissions (Chueamuangphan et al., 2020; Pickin et al., 2002). Finally, the increasing consumers' concerns on the end of life of packaging (Martinho et al., 2015) is the third societal change (iii) that has pushed the packaging industry to implement new strategies to utilize a higher amount of recycled fibers and replace harmfully and pollutant elements with environmentally friendly alternatives. The Frustration-Free Packaging (FFP) program of Amazon (launched in 2008) has, for instance, contributed to the elimination of one million tons of packaging materials (Amazon, 2021). This program helped redesign laundry detergent plastic bottles into eco-boxes made with 60% less plastic that can fulfill the functions of both primary and secondary packaging and can be recycled (Amazon, 2019).

Such initiatives are, however, not always broadly and effectively disseminated among customers (Boz et al., 2020). As a result, sustainable packaging with a conventional design may not be labeled as “sustainable” or “eco-friendly” by a customer (Magnier & Schoormans, 2015). The recycling of paper-based products is a great example illustrating the lack of communication between consumers and suppliers. Paper waste recycling commonly involves intense processing of recycled materials and thorough washing and removal of contaminants (*e.g.*, ink, adhesives) (Hubbe et al., 2007). Packaging made of recycled fibers thus looks very similar to those made of virgin fibers. Different studies have shown that consumers cannot easily differentiate sustainable packaging from non-sustainable ones, especially if they are not provided with direct cues to identify how eco-friendly the product is. Thus, inaccurate assumptions are made by consumers during the purchase decision. Scott and Vigar-Ellis (2014) showed that 30 to 45% of interviewed consumers utilize labels, images, or logos to differentiate between eco-friendly and regular packaging. Therefore, the packaging industry relies on the use of often excessive visual signals to explicitly communicate the eco-friendliness of a product (Magnier & Schoormans, 2015; Steenis et al., 2017). Yet, this approach has not proved to be effective for clearly communicating sustainability (Escursell et al., 2020; Herbes et al., 2020). Visual signals can sometimes be misunderstood, as is the Green Dot symbol used in some packaging products. Although this symbol seems to indicate sustainability because of its design and green color, its use indicates that the producers have made a financial contribution to the organization of recovery, sorting, and recycling of sales packaging (Herbes et al., 2020).

To more effectively communicate sustainability in packaging design, one strategy consists of changing the appearance (or esthetic) of the packaging to facilitate its appropriate categorization and promote its purchase (Krah et al., 2019). Magnier & Schoormans (2015) studied the influence of visual appearance in laundry detergent (conventional vs. ecological look) on consumers' affective attitudes and purchase intention regarding packaging sustainability. Molded-pulp bottles were used for the ecological look and red hard plastic bottles for the conventional ones. Consumers filled out a survey to assess their purchase intention and emotional commitment. The molded-pulp products positively influenced environmentally conscious consumers' purchases, as they were perceived as more natural, biodegradable, and recyclable than the plastic ones; yet, 100% of the proposed plastic bottles were also recyclable (Oloyede & Lignou, 2021).

To date, most of the studies that evaluated consumers' perception of sustainable packaging compared different packaging materials such as plastic, glass, and paper-based containers (Boesen et al., 2019; Orzan et al., 2018) and different visual elements such as graphics and information on labels (Borgman et al., 2019; Krah et al., 2019; Samant & Seo, 2016). No studies have intended to modify the appearance of the packaging within one class of material; studies have mainly focused on perceptions between material classes such as glass versus paper versus plastic.

Hence, this study proposes to redesign recycled paper-based containers so that consumers recognize residual contaminants from recycled materials and comprehensibly refer to this product as recycled. Changing the appearance of containers without externally added visual signals is conjectured to help consumers recognize sustainable (here, recycled) packaging from a non-sustainable one, and in turn, help promote a stronger identity of the product and brand. Moreover, avoiding the use of labels and printed claims on paper-based packaging will contribute to the reduction of contaminants upon recycling the labelled product. In this study, the appearance of recycled containers was intentionally altered with the addition of processed MOW with varying apparent aspect ratios to OCC to produce a series of recycled paperboard substrates with noticeable recycled contents. The performance of the recycled products was evaluated and the behavior of the resulting paperboard properties analyzed with respect to the macro, micro and nano scale contaminants introduced. In future research consumer perceptions of such material will be evaluated.

2.3. Experimental

2.3.1. Materials

Recycled fibers from old corrugated containerboards (OCC) were kindly provided by Greif (North Carolina, US) and used as received for the preparation of recycled paperboards. Copy paper of 75 g/m^2 (Husky® copy, Domtar, South Carolina, US) were printed using a laser printer (Bizhub 364e, Konica Minolta, Canada) with a standard image in black and white that covered 50% of the surface area of the paper and was used as MOW. The morphology and average dimensions of the fibers were assessed using a fiber quality analyzer (HiRes FQA, OpTest Equipment Inc., Ontario, Canada) as described in the TAPPI 271 om-07 2007 standard. The ash content of the OCC pulp and MOW was measured according to the TAPPI 211 Om-02 2007 standard (Sybron Thermolyne, Thermo Fisher Scientific, Asheville, USA) after incineration at 525 °C for 6 h. Compositional analysis of the OCC pulp was performed following the NREL report to determine structural carbohydrates and lignin in biomass (Sluiter et al., 2012). The data are reported in Table 2-1.

Table 2-1. Fiber properties and chemical composition of OCC pulp and MOW.

Raw Material	OCC	MOW
Fiber length[1] (mm)	1.5	0.93
Mean width(μm)	22	18
Mean Curl Index[1]	0.11	0.11
Kinks per mm	0.7	0.9
Fines Content[1] (%)	8.3	14.2
Ash Content (wt%)	1.7 ± 0.1	17.8 ± 0.2
Cellulose (wt%)	59.9 ± 1.2	-
Total hemicelluloses (wt%)	14.1 ± 0.5	-
Total lignin (wt%)	14.3 ± 0.2	-

[1]Length weighted
± is the standard deviation

2.3.2. Preparation of recycled paperboards

Figure 2-1 illustrates the different steps used to prepare the recycled furnishes and the recycled paperboard lab samples.

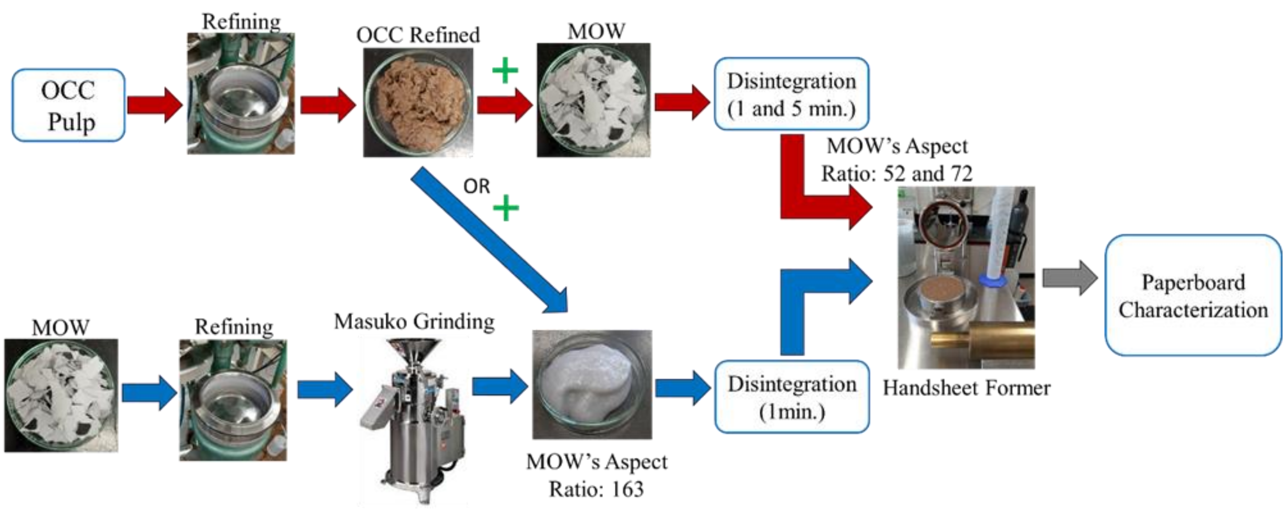


Figure 2-1. Preparation of the recycled furnishes and paperboard samples from OCC pulps and MOW of different apparent aspect ratios, 52, 72, and 163.

The OCC pulp suspension was refined using a PFI mill refiner (PFI Mill, The Norwegian Pulp and Paper Research Institute, Oslo, Norway) at 1,000 revolutions according to the TAPPI 248 Sp-00 (2000) standard. The number of refining revolutions was selected to mimic the refining process performed in a paper mill. A progression of refining was used to produce pulp that matched the strength properties of the same OCC pulp refined in a manufacturing site (Figure S1 in the supplementary information). The refined OCC pulp was then mixed with the MOW in different weight ratios using a pulp disintegrator (Testing machine Inc., New Castle-DE, USA) prior to paperboard making, as described in Table 2-2.

Varying the disintegrator time produced different apparent aspect ratios of the MOW. A disintegration time of 1 min resulted in MOW of 52 in apparent aspect ratio (length to width), whereas a disintegration time of 5 min increased further the apparent aspect ratio to 72 (Table 2-2). A higher apparent aspect ratio of MOW was obtained by processing further the MOW (before its addition to the OCC pulp) with the PFI mill refiner at 5,000 revolutions and then passing the resulting aqueous suspension at 2.5 wt% through a Supermasscolloider nano-grinder (MKZA6-5, Masuko Sangyo Co., Ltd, Saitama, Japan) at 1,200 RPM. The grinder was equipped with silicon carbide grinding stones (MKW 10-46#), and a specific energy consumption of 0.5 kWh/kg (Kriechbaum et al., 2018) of oven-dried pulp was targeted and reached after 8 passes. The resulting MOW suspension (which can be referred here as MOW cellulose micro/nanofibrils or MOW CMNFs) had an estimated apparent aspect ratio of 163. The MOW CMNF suspension was then blended with the OCC pulp at different weight ratios prior to paperboard making (Table 2-2).

The apparent aspect ratios of the three types of MOWs and OCC fibers were estimated by sedimentation, as detailed in the next section.

Table 2-2. Series of paperboard lab samples

Apparent Aspect Ratio of MOW	OCC (wt%)	MOW (wt%)	Disintegration Time (Min.)	Grammage (g/m^2)	Freeness (ml)	Bulk (cm^3/g)
87 ± 7	100	0	-	132.0 ± 2.9	395 ± 7	1.8 ± 0.04
52 ± 7	95	5	1	129.4 ± 3.8	386 ± 7	1.8 ± 0.06
	90	10	1	129.8 ± 4.9	387 ± 2	1.7 ± 0.03
	80	20	1	128.9 ± 3.3	390 ± 7	2.1 ± 0.03
	60	40	1	131.4 ± 0.4	423 ± 7	1.9 ± 0.02
72 ± 3	95	5	5	135.2 ± 1.5	368 ± 2	2.2 ± 0.02
	90	10	5	136.6 ± 1.5	370 ± 9	1.8 ± 0.01
	80	20	5	137.2 ± 1.4	368 ± 20	1.6 ± 0.03
	60	40	5	135.9 ± 1.2	391 ± 13	1.6 ± 0.02
163 ± 1	95	5	N/A	136.2 ± 0.7	276 ± 38	1.6 ± 0.03
	90	10	N/A	137.2 ± 0.3	182 ± 9	1.5 ± 0.03
	80	20	N/A	138.8 ± 0.8	105 ± 7	1.5 ± 0.07
	60	40	N/A	134.6 ± 0.3	31 ± 1	1.5 ± 0.06

± is the standard deviation
N/A: not applicable

The Canadian Standard Freeness (CSF) of the resulting slurries (OCC pulp containing the MOW) was measured according to TAPPI 227 om-09 (2009). The freeness is an indicator of the drainage rate of the fibers in a paper machine operation and thus is critical in understanding the potential rate of paper production. The freeness results are shown in Table 2-2. Paperboard specimens of 130 g/m2 were prepared using a TAPPI handsheet former (TAPPI 205 Sp-02 (2006)) from the OCC pulp slurries containing different weight ratios of MOW of varying apparent aspect ratios (0, 5, 10, 20, and 40 wt%).

2.3.3. Characterization of the MOW

2.3.3.1. Apparent aspect ratio

The sedimentation value of the macro-, micro-, and micro/nano-sized particles of MOW was estimated using the calculations for the aspect ratio by sedimentation test according to the Crowing Number theory for fibers, as reported by Martinez et al. 2001 and Varanasi et al. 2013. In brief, five aqueous MOW suspensions of solids contents of 0.5, 0.75, 1.0, 1.25, and 1.5 kg/m^3

were prepared and transferred to the same volume, and height plastic graduated cylinders for sedimentation. The initial height of the suspensions, H_0, was measured using a ruler. The particles in suspension (*i.e.,* MOW fibers and CMNFs) were then allowed to sediment for one week, after which the height of the observed sediment, H_s, was determined. The ratio between H_s and H_0 was plotted against the initial concentration of the suspensions. The gel concentration point, meaning the lowest concentration at which all flocs of fibers are interconnected, forming a self-supporting network (Martinez et al., 2001; Sanchez-Salvador et al., 2021), was determined through the linear term of the fitted quadratic curve. The sedimentation value was then estimated based on the Crowding Number theory using Eq. 2.3-1, where ϕ_g is the gel point. The analysis was performed in duplicate, and the average apparent aspect ratio values are reported in Table 2-2.

$$Sedimentation\ value = 6\sqrt{\frac{1000}{\phi_g}} \qquad \text{Eq. 2.3-1}$$

2.3.3.2. Analysis of MOW particles

The average area of the MOW particles of 52 and 72 in apparent aspect ratio was determined using the Spec*Scan™ 2000 an image analysis system (Apogee Systems Inc.) combined with a flat-bed scanner (Epson Perfection 2400 photo). The two aqueous MOW suspensions were diluted to 0.04 wt% and then filtered using black filter paper (Grade 8613, Ahlstrom). Ten filter papers per sample were scanned using a resolution of 600 dots/inch, in normal 256 grayscale, using a reverse threshold setting of 80% of the average grayscale value. This procedure was not followed with the MOW CMNFs, because of the small dimensions of the fibers and the tendency to self-aggregate. The characterization of the exact dimensions (*i.e.,* length and width) of CMNFs is well known to be challenging (Foster et al., 2018). Thus, the AR is the main parameter to refer to the CMNFs size. An SEM image is available in Supporting Information (Figure S2) to give an overview of the size difference and distribution of the fibrils; the image shows that there is significant amounts of nanofibrillar materials with diameters in the nano-size range.

The procedure was repeated to analyze the MOW-containing paperboards. The total area (in mm^2) covered by the MOW particles per square meter (m^2) of the paperboard sample and the average particle area (in mm^2) was determined using the Spec*Scan™ 2000. Five paperboard sheets per OCC/MOW weight ratio were scanned on both sides.

2.3.4. Characterization of the paperboards

Prior to testing, the paperboard samples were conditioned for 24 h at 50% relative humidity (RH) and 23 °C (TAPPI 402 sp-98, 1998). For each property described hereinafter, an average value was obtained from a minimum of 3 to 10 replicates and is used in this manuscript for further discussion.

2.3.4.1. Physical properties

The grammage and the thickness of the produced paperboards were measured using a high precision lab-scale and a micrometer (Lorentzen & Wettre, Stockholm, Sweden) according to TAPPI 410 om-08 (2008) and TAPPI 411 om-97 (1997), respectively. The bulk of the paperboards was calculated by dividing the measured thickness by the calculated grammage of each paperboard. The physical properties were measured in triplicates and are presented in Table 2-2.

2.3.4.2. Optical properties

The brightness of the paperboards was measured at 457 nm using a ColorTouch® X (Technidyne Corporation, Inc., NJ, USA) with diffuse illumination, according to TAPPI 525 om-92 (1992). Ten samples per paperboard grade were analyzed for reproducibility. The CIE L*a*b* parameters were determined using the same instrument and following the TAPPI 527 om-02 (2002) standard. The parameters measured were lightness (L*), redness (a*), and yellowness (b*).

2.3.4.3. Mechanical Properties

The tensile strength (TAPPI 494 om-96, 1996), tear strength (TAPPI 414 om-98, 1998), burst strength (TAPPI 403 om-97, 1997), and short-span compression strength or STFI (TAPPI 826 om-08, 2013) of the paperboard samples were measured on a minimum of five specimens per sample type. Strips of 15 mm wide were used for tensile strength and short-span compressive strength measurements, and the testing was carried out using a tensile tester (TMI 84-56, Testing Machines, Inc., New Castle, DE, USA) and an STFI compression strength tester (Lorentzen & Wettre, Stockholm Sweden), respectively. The tear strength was evaluated on a stack of five plies of 63-mm wide using a Lorentzen & Wettre tear instrument (Stockholm, Sweden). The burst strength was performed using a Mullen burst tester (Model A, Testing Machines, Inc., New Castle, DE, USA). The results were discussed using the Index values for each mechanical property, meaning that the average values of each mechanical property were divided by the average grammage of the corresponding tested samples.

2.3.4.4. Air Permeability

The Gurley air permeability (Testing Machines, Inc., New Castle, DE, USA) of the paperboards was measured on a minimum of ten samples per paperboard type according to the TAPPI 460 om-96 (1996).

2.3.4.5. Microscopy imaging

Imaging of the surface and cross-section of the MOW-containing paperboards and controls was performed using a Field Emission Scanning Electron Microscopy (FE-SEM, FEI Verios 460

L, OR, USA). Prior to image analysis, the surfaces and cross-sections were coated with a thin layer of gold (of ca. 7 nm thick). A working distance of 6 mm and an accelerating voltage of 2.00 kV were used at a magnitude of ×200 for the analysis of the surfaces, while a working distance of 3.5 mm, an accelerating voltage of 2.00 kV, and a magnitude of ×500 were applied for the cross-sections.

2.3.5. Statistical Analysis

The Tukeys' range test was applied to the average values reported for the mechanical and barrier properties of paperboards to determine the ones that are significantly different from each other, using the JMP software (SAS Institute). The Tukey's range test considers that the data from different groups of mean comes from a population that has a normal distribution and has the same standard deviation. *p*-values lesser than 0.05 suggest a significant statistical difference between the data, while *p*-values greater than 0.05 suggest that there is no significant statistical difference between the data. The statistical evaluation was performed with a confidence interval of 95%.

2.4. Results and Discussion

2.4.1. Recycled pulps

MOW of different apparent aspect ratios was purposely added to the recycled OCC pulp for designing and engineering recycled containerboards with obvious (*i.e.*, visible by naked eyes) recycled content. The disintegration processing time of the MOW-containing OCC pulps was varied to alter the particle size of the MOW according to Figure 2-1. As shown in Table 2-3, a disintegration time of 1 min resulted in an estimated apparent aspect ratio of 52. An extended processing time of 5 min increased by almost 50% the apparent aspect ratio of the MOW (apparent aspect ratio of 72). A nano-grinder was needed to process even further the MOW and increase its apparent aspect ratio considerably to 163, which relates to an apparent aspect ratio of micro- and nano-scale elements (Kriechbaum et al., 2018).

In the following, we will refer to the three-size ranges of MOW as follows: L-MOW in reference to the lowest MOW apparent aspect ratio (*i.e.*, 52, macro size range), M-LOW for the middle range MOW apparent aspect ratio (*i.e.*, 72, micro size range), and H-MOW for the highest MOW apparent aspect ratio (*i.e.*, 163, a combination of micro- and nano-size range) (Table 2-3).

Table 2-3. Apparent aspect ratio and area of MOW particles.

	MOW Treatment	Apparent Aspect Ratio	Average Particle Area (mm^2)
L-MOW	Disintegration (1 min.)	52 ± 7	0.33
M-MOW	Disintegration (5 min.)	72 ± 3	0.08
H-MOW	Nano-grinding (0.5 kWh/kg)	163 ± 1	N/A

The apparent aspect ratio of OCC fibers was 87 ± 7
N/A: not applicable
± is the standard deviation

With a disintegration time of 1 minute, the added MOW went through partial disintegration only, resulting in a blend of individual fibers and macro-sized fiber cluster particles with an average particle area of 0.33 mm^2 (Figure S3a). A disintegration time of 5 min further broke down the MOW into almost exclusively individual fibers with an average particle area of 0.08 mm^2. Although the 5-min processed MOW particles are still sized in the microscale range (Figure S3b), it was impossible to differentiate them from the OCC fibers by the naked eye (OCC apparent aspect ratio of 87). Grinding the MOW reduced even further the dimensions of the individual fibers to cellulose micro- and nano-fibrils (a mixture of micro/nano-sized particles), explaining the significant increase in apparent aspect ratio (from 72 to 163). The average particle area of the CMNFs could not be determined using the Spec*Scan™ due to their small size and high apparent aspect ratio, which is beyond the detection limit of the scanner used at 600 dots per inch which provided a minimum spot area detectable of 0.02 mm^2 (Figure S3).

2.4.2. Impact of MOW on the pulp drainage rate

Figure 2-2 shows the Canadian Standard Freeness (CSF) values of the MOW-containing OCC pulps and controls (*i.e.*, without MOW). The addition of L-MOW to the OCC pulp kept the freeness constant up to 20 wt% added MOW, with no significant differences observed between the values (Table S2), while further addition (40 wt% L-MOW) increased the freeness value by 7%. This is probably due to the partial replacement in the furnish of the well-separated individual OCC fibers and associated fines (Table 2-1) (which have strong water holding capacity) with undisintegrated clusters of MOW (with lower fines and water holding capacity). Additionally, due to partial disintegration of the L-MOW, the addition of a certain amount of large particles in the fiber network may have limited the number of fiber-fiber bonding, thus creating spaces between the OCC fibers through which the water drained faster.

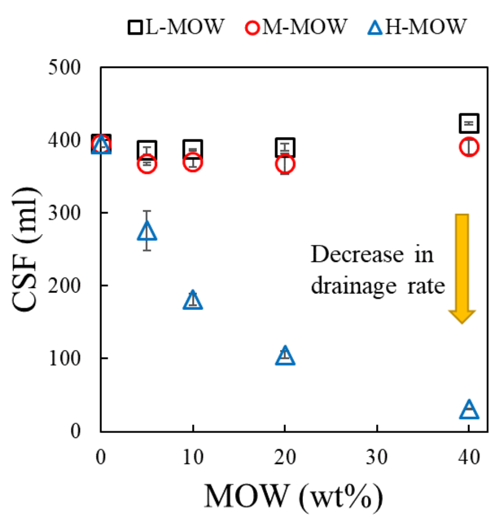


Figure 2-2. Canadian Standard Freeness (CSF, in ml) of MOW-containing OCC pulps at different MOW weight ratios for three apparent aspect ratios (low, 52; medium, 72; and high, 163). The error bars indicate the standard error.

The addition of M-MOW had a less significant change of the freeness values of the recycled slurry (Table S2). It is observed that the freeness of the M-MOW was lower for all levels relative to the L-MOW, reasonable as the M-MOW had more liberated fibers/fines than did the L-MOW which existed more as chips. No large MOW particles could be observed in the M-MOW containing recycled paperboard because of the complete disintegration of the MOW into individual fibers with a similar apparent aspect ratio to that of the OCC fibers.

The further increase in apparent aspect ratio with the addition of H-MOW drastically decreased the freeness of the recycled pulp. With the addition of 5 to 40 wt% of H-MOW, the freeness was reduced by 30% and 92%, respectively. The micro- and nano-sized scale MOW has significant water holding capacity and fills in space and voids between the micro-scale OCC fibers resulting in the considerably lower drainage rates observed. Moreover, as observed in the SEM images (Figure 2-3), some of the H-MOW accumulated at the surface of the recycled paperboard upon drainage. The large surface area and high OH-bond availability of the MOW CMNFs promoted water absorption and fiber swelling (Campano et al., 2018; Taipale et al., 2010; Zambrano et al., 2021), explaining the observed significant decrease in freeness with increasing H-MOW weight ratio in the OCC pulp. It is possible to improve the freeness of the pulp stock by cationic retention aids such as polyacrylamides or cationic starches.

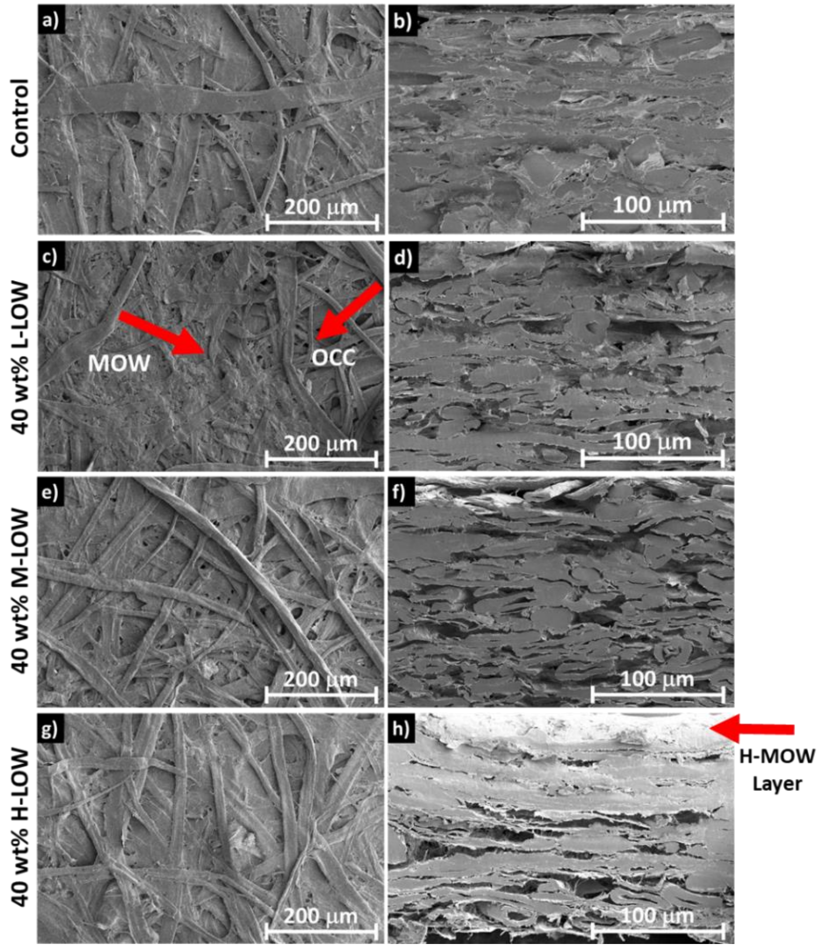


Figure 2-3. Scanning Electron Microscopy (SEM) images of the surface (*left*) and cross-section (*right*) of (a) and (b) the paperboard controls; (c) and (d) 40 wt% L-MOW containing paperboards; (e) and (f) 40 wt% M-MOW-containing paperboards, and (g) and (h) 40 wt% H-MOW containing paperboards.

2.4.3. Visual appearance and optical properties of recycled paperboards

Figure 2-4a-g show the appearance of the paperboards made with 0, 10, and 40 wt% MOW of low, medium, and high apparent aspect ratios (Figure S4 in the supplementary information shows the complete series of recycled paperboards made with the addition of the three levels of MOW apparent aspect ratio, from 0 to 40 wt%). A higher amount of MOW resulted in whiter recycled paperboards, as confirmed by the increase in L* values and decrease in a* and b* intensities (Figure S5). Beside the difference in color, addition of L-MOW changed further the appearance of the paperboards leaving visible macro particles of MOW on the surface of the substrates. The change in the appearance is noticeable when compared to the recycled paperboard without MOW (Figure 2-4a) and to the non-recycled paperboard (Figure 2-4h). With the increase in L-MOW weight ratio, the area of the recycled paperboards covered by the MOW particles, also increased (Figure 2-4i) from 1,960 mm^2/m^2 to 20,300 mm^2/m^2 for 5 wt% and 40 wt% of added MOW, respectively (Table S1). The particles detected in the control group, *i.e.,* without added MOW, totaling a value of 98.2 mm^2/m^2, came from residual contaminants already present in the OCC pulp after recycling. The presence of L-MOW particles endowed the paperboards with an obvious recycled aesthetic, which would be encouraged for the design of sustainable packaging with an ecologically friendly appearance.

Unlike the L-MOW, the MOW with either a medium or high apparent aspect ratio did not change the appearance of the paperboards relative to the recycled paperboard without MOW (Figure 2-4a) and to the non-recycled paperboard (Figure 2-4h). The high-intensity processing of the MOW (M-MOW and H-MOW) reduced the size of the particles to an extent that could not be differentiated by the eye from the OCC fibers. Only a change in the color of the paper-based material could be perceived, as shown in Figure 2-4d-g. The coverage area of the M-MOW particles increased from 97 to 406 mm^2/m^2 with the addition of 5 and 40 wt% of MOW, respectively, while no H-MOW particles at all could be detected due to the significant difference between the micro-scale OCC fibers and the micro/nano-scale MOW. The increase in drainage time that was observed during paperboard making with the addition of H-MOW resulted in the formation of an H-MOW film-like layer on the top side of the paperboard sheets, as shown in Figure 2-4g.

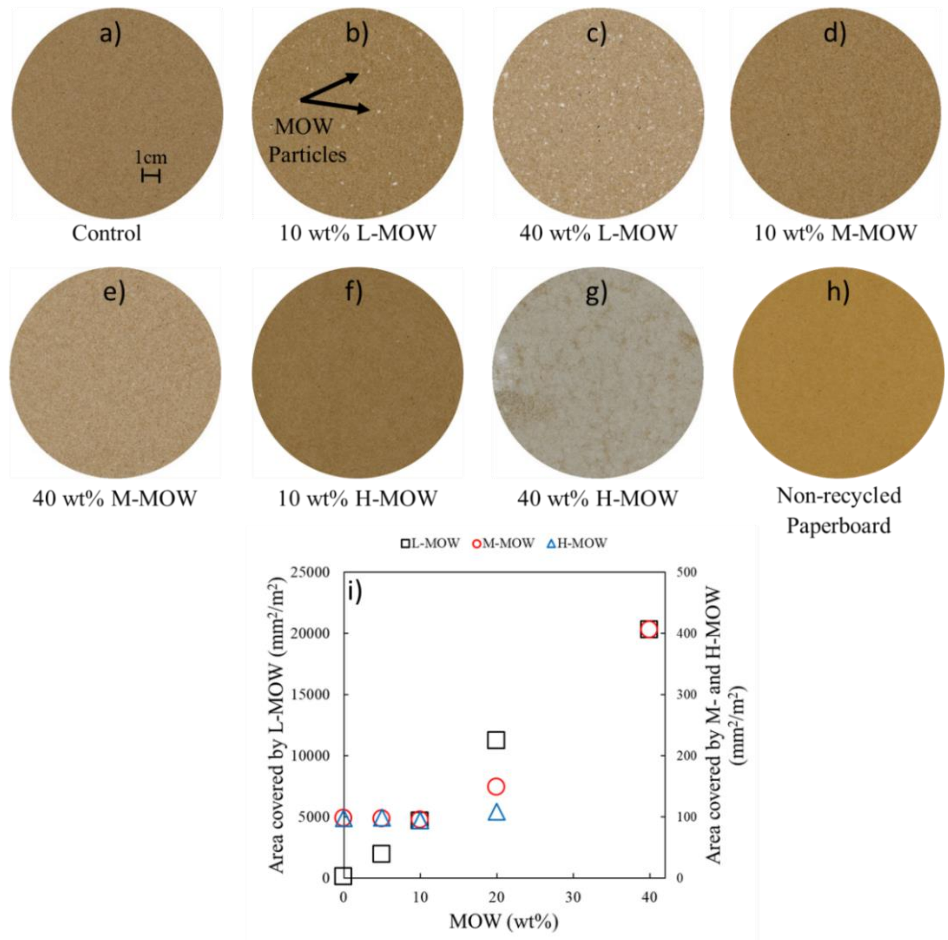


Figure 2-4. Visual appearance of the MOW-containing paperboards. Scans of the (a) control paperboard (OCC pulp only) and paperboards with 10 and 40 wt% of (b and c) L-MOW, (d and e) M-MOW, and (f and g) H-MOW, respectively, (h) non-recycle paperboard, and (i) total area covered by the MOW particles, in mm^2/m^2, as a function of the weight ratio of MOW added to the OCC pulp (left y-axis for L-MOW particles, right y-axis for M-, and H-MOW).

The brightness of the recycled paperboards also overall increased with the added MOW content for all apparent aspect ratio materials (Figure 2-5a). The MOW used in this study was mainly composed of bleached fibers and calcium carbonate that can reflect a larger portion of light than the unbleached fibers from the lignin containing OCC pulp. However, a more significant increase in brightness was observed with the L- and M-MOW.

Incorporating MOW from commercial copy paper to the OCC furnish also added a certain amount of minerals, such as calcium carbonate, to the recycled paperboards. The filler content (or ash content) in the paperboards was determined after incineration at 500 °C. As shown in Figure 2-5b, the ash content increased with the increasing amount of MOW, confirming that minerals, whose purpose is to improve opacity and/or brightness of the pulp (Hubbe & Gill, 2016), also contributed to the overall enhancement in optical properties of the MOW-containing paperboards.

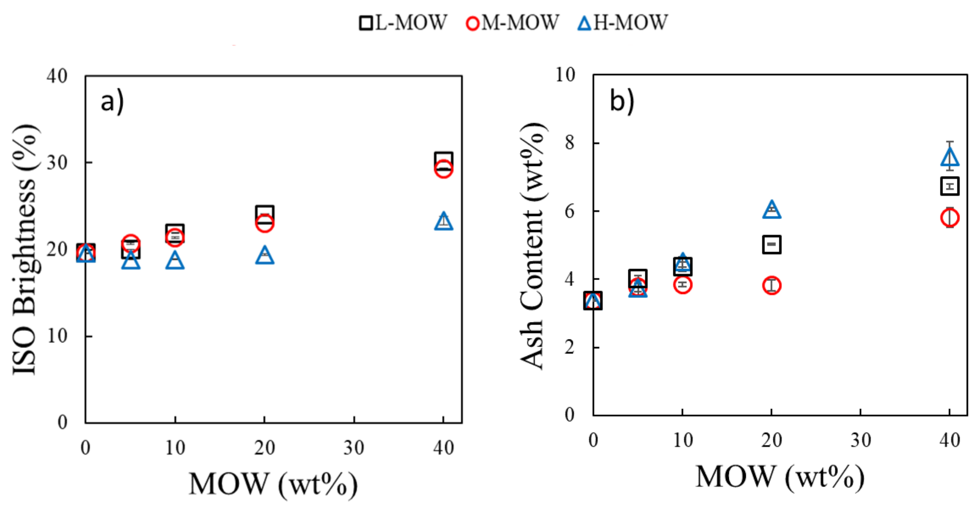


Figure 2-5. Optical properties of the MOW-containing paperboards (a) ISO brightness and ash content (b) at different MOW weight ratios and apparent aspect ratios. The error bars indicate the standard error.

2.4.4. Air permeability of recycled paperboards

Figure 2-6 depicts the effect of MOW of different apparent aspect ratios on the air permeability of the recycled paperboards. The addition of L- and M-MOW decreased the air permeability up to an addition of 20 wt%. However, the addition of 40 wt% MOW increased the air permeability to values almost similar or higher than the air permeability of the controls. With the addition of MOW at low weight percent, the voids between the OCC fibers were filled, and others were created, thus forming a more tortuous pathway through which the flow of air was resisted (*i.e.*, decrease in air permeability). The factors causing the higher air permeability at 40 wt % of L- and M-MOW have not been identified, as the bulk was the lowest at the 40 wt% level.

Air permeability of H-MOW-containing paperboards decreased drastically with an increase in the weight ratio of MOW. A reduction in paperboard porosity is expected because of the micro and nano-sized particles that promote a denser structure (see lower bulk measurements in Table 2-2) with better inter-fiber bonding (Lavoine et al., 2012). Air permeability data with H-MOW above 10 wt% could not be measured because the air permeability was so low (indicating near zero air flow) that values could not be recorded with the Gurley tester.

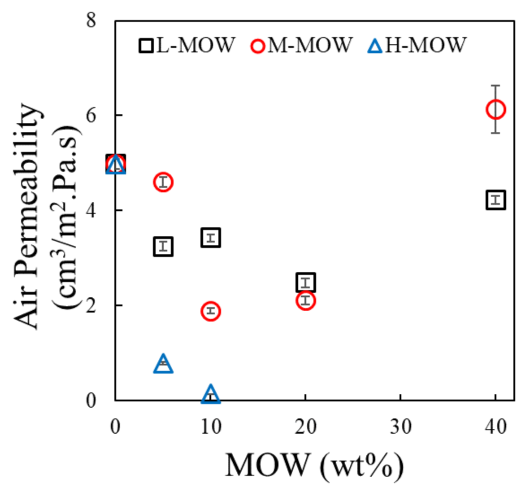


Figure 2-6. Air permeability of MOW-containing paperboards as a function of the MOW weight ratio (wt%). The error bars indicate the standard error.

2.4.5. Mechanical Properties

With the addition of an increasing amount of L-MOW to the OCC pulp, the mechanical performance of the recycled paperboards decreased with a more pronounced drop from 20 wt% of MOW (Figure 2-7). An average decrease of 20% was observed for each mechanical property tested for paperboards with 40 wt% L-MOW. The presence of MOW particles larger than the OCC fibers and the fillers disrupted the OCC inter-fiber bonding network during papermaking, creating a more porous and bulkier structure than that of the control paperboards (Table 2-2). The results were supported by the SEM image analysis of the paperboards (Figure 2-3). As shown in Figure 2-3b, the control paper showed a more compact network than the L-MOW-containing paperboards, which showed a bulkier structure with visible particles of L-MOW on their surface (Figure 2-3c and d).

The addition of more processed M-MOW (here, after 5 min disintegration time) had a lesser effect on the mechanical performance (tensile, burst, and compression) of the paperboards (Figure 2-7) with the tensile index almost independent of the wt % of M-MOW. This suggests that at high weight ratios of contaminants, the L-MOW interferes more than the M-MOW with the fiber matrix.

In contrast, with the addition of H-MOW (apparent aspect ratio of 163) the mechanical properties improved. The tensile, burst, and STFI compression indexes showed a considerable increase of 50%, 59%, and 72%, respectively, with the 40 wt% of added H-MOW, probably due to the formation of a denser and less porous structure (lower bulk, see Table 2-2). The high apparent aspect ratio and large surface area of the H-MOW enhanced the bonding area between the micro-scale OCC fibers by covering the surface of the OCC fibers and filling the space between them, in turn increasing/improving the inter-fiber contact points and the hydrogen bonding between fibers, as shown in Figure 2-3g and h. Enhanced mechanical properties were thus observed for H-MOW-containing paperboards. In contrast to the tensile, burst and compression improvements, the tear index decreased with increased H-MOW. The tear performance was decreased by 30% with 40 wt% of added H-MOW, which may result from an increased amount of fiber breakage due to increased bonding due to nanofibrils rather than fiber pull out at the tear line. Fiber pull out requires more work than does fiber breakage during a tear test (Ehman et al., 2020).

The general improvements in mechanical properties other than tear strength using H-MOW can thus be exploited to utilize lower grade recycled content in higher strength OCC pulps without a detriment to the properties. However, decreases in the drainage rate with H-MOW will be realized as a disadvantage. Future research should pursue methods to alleviate drainage issues with the presence of micro and nano-fibrillar materials.

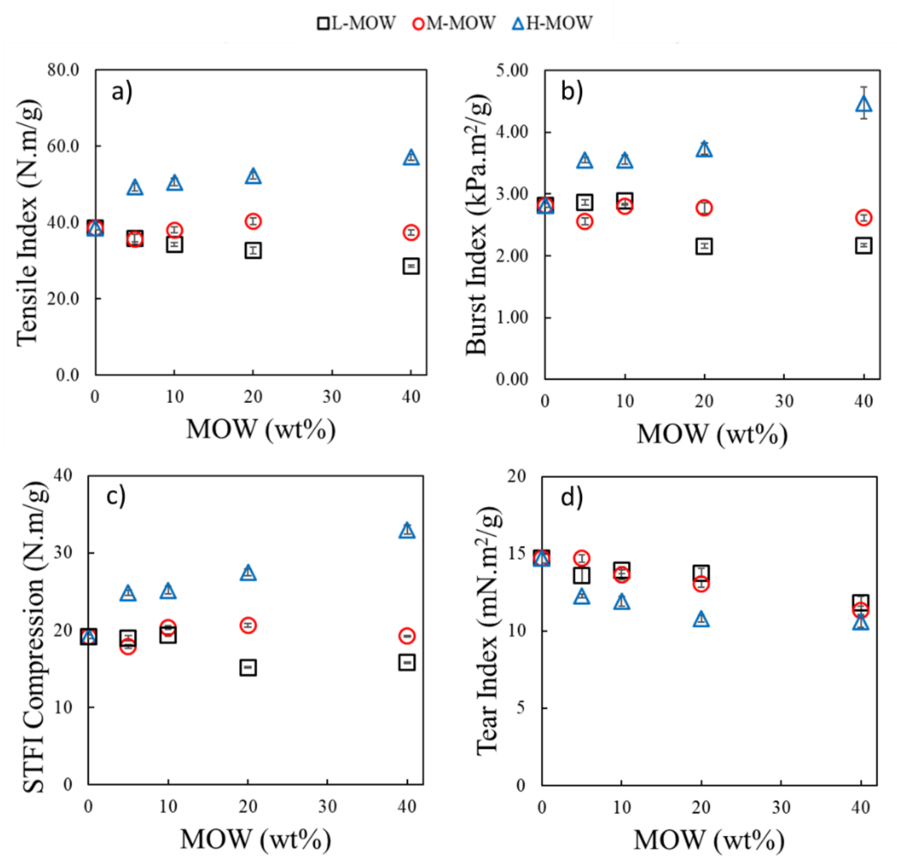


Figure 2-7. Mechanical performance of the MOW-containing paperboards for different MOW weight ratios (from 0 to 40 wt%) and MOW apparent aspect ratio. (a) Tensile index, (b) burst index, (c) short-span compression strength index (STFI compression), and (e) tear index. The error bars indicate the standard error.

In summary, we can illustrate the interactions between MOW of different apparent aspect ratios and the OCC pulp, as shown in Figure 2-8. Among the three MOW apparent aspect ratios evaluated in this study, L-MOW, and H-MOW were the MOW particles size that mainly affected the paperboard properties. The L-MOW modified the appearance of the paperboard substrates with noticeable recycled content (large particles of MOW partially disintegrated) that could be easily recognized by the consumers (Figure 2-8a). The H-MOW enhanced the mechanical and barrier properties considerably by improving the inter-fiber hydrogen bonding and filling the voids between OCC fibers (Figure 2-8c). However, the H-MOW did not present macroscopic particles that can be recognized by the naked eye as contaminants. The use of M-MOW blended in with the darker OCC fibers (Figure 2-8b) brightened the product but did not result in presenting macroscopic particles and at the same time did not greatly modify the mechanical properties. Thus, amongst the three materials, there are tradeoffs between properties of the board and the appearance to promote positive perception of the recycled packaging. It may be possible to incorporate multiple apparent aspect ratio materials into a single paperboard to allow larger weight percentages of the lower valued recovered paper and also to achieve targets in mechanical and appearance characteristics. Alternatively, dry strength agents might be required to accommodate the presence of visually obvious recycled particles that have a substantial impact on the mechanical properties. Commercially, to create high, medium or low apparent aspect ratio MOW, it might necessitate a dedicated pulper or dispersion unit to treat the waste material separately.

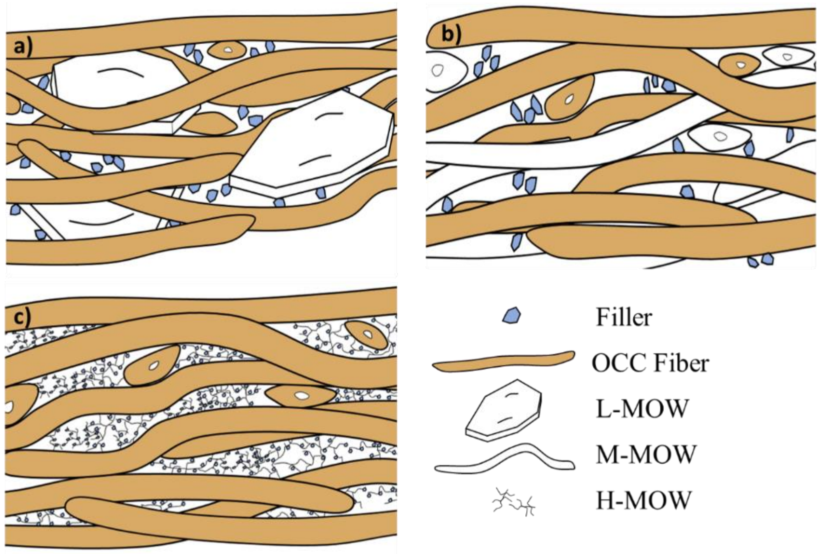


Figure 2-8. Schematic representation of the possible interactions between MOW of a) low (L-MOW, AR of 52), b) medium (M-MOW, AR of 62), and c) high apparent aspect ratio (H-MOW, AR of 163), fillers (AR estimated to 1), and OCC fibers (AR of 87).

2.5. Conclusion

In this study, mixed office waste (MOW) has been used to re-design recycled containers with enhanced environmental perception with the aim of increasing the purchase intention of consumers towards sustainable packaging. The addition of minimally processed MOW (52 in apparent aspect ratio) to recycled paperboards resulted in macroscopic and visually apparent particles on the surface of the boards, which reflect the presence of recycled materials in the container. The presence of clearly identified contaminant particles should help consumers recognize sustainable packages without any external visual aids. However, the incorporation of such large MOW particles negatively impacted the mechanical performance of the paperboards, suggesting a need for restorative treatments that can improve the performance of the products without altering their sustainable appearance. The addition of highly processed MOW (apparent aspect ratio of 72 and 163) did not endow the paperboards with a similarly high environmental perception. However, the use of micro/nano-scale MOW (with an aspect of 163) considerably enhanced the mechanical properties of the paperboards. Therefore, the combination of two sizes of MOW particles (*i.e.*, MOW with low and high apparent aspect ratios) could be a strategy to engineer sustainable packaging of high performance and environmental perception.

The previous study showed that the size of paper waste incorporated into OCC pulp plays an important role in defining the mechanical performance of recycled paperboards with high environmental perception. **But is size the only variable to consider? Or there are other aspects such as the physicochemical properties of the waste?**

The next chapter discuss the effect of waste physicochemical properties on the performance of redesigned packaging with obvious recycled content.

Chapter 3 . Redesigning the Appearance of Recycled Containers for Packaging Applications: The Effect of Paper Waste Physicochemical Properties on the Performance of Paperboards with Obvious Recycled Content[2]

3.1. Abstract

Significant efforts have been made over the past decade to facilitate the recognition of environmentally friendly packaging and promote sustainability. Yet, consumers remain confused by the excess of labels and claims used to communicate sustainability. In our previous work, we modified the appearance of recycled fiber-based packaging by incorporating visible particles of fiber-based waste on its surface. This strategy enabled consumers to better identify packages with a high recyclability level, enhancing their environmental perception towards sustainable products. However, the incorporation of such large particles of waste proved to be detrimental to the mechanical properties of the paperboards. In this study, we further investigate the influence of waste physicochemical properties on the performance of recycled packaging. Using a similar strategy to enhance the environmental perception, we herein studied the effect of mixed office waste (MOW), old magazines (OMG), and polylactic acid (PLA) paper cups. The presence of hydrophobic and difficult-to-process and -disperse waste, such as the PLA paper cups, significantly altered the mechanical performance of the paperboards, whereas more hydrophilic and easy-to-disintegrate waste (MOW and OMG) had a lesser effect regardless of the size of the particles. Well-investigated strength agents such as cationic starch (CS) and cellulose microfibrils (CMFs) successfully restored the properties of the paperboards containing MOW and OMG but

[2] The material in this chapter has been submitted to the Packaging Technology and Science Journal and is under review

were less effective for PLA paper cups. A multi-ply strategy overcame the limitations of CS and CMFs using the redesigned paperboard as an outer ply for aesthetic purposes and a 100% recycled inner ply for restoring strength.

3.2. Introduction

The exacerbated ecological crises and the spread of the COVID-19 pandemic have profoundly marked the consumers' responses to packaging design and engineering (Kitz et al., 2022). Consumer perception has become an important driver for making effective environmental decisions. Hence, understanding the attributes consumers use to identify sustainable packaging becomes essential to improving marketing strategies, promoting sustainability, and lowering packaging's environmental impact on the planet and society (Chekima et al., 2016; Magnier & Schoormans, 2015; Oloyede & Lignou, 2021; Rokka & Uusitalo, 2008). Using solely recycled pulps to convey a high environmental perception message to consumers has been shown to be ineffective, essentially because recycled packaging looks like conventional ones (Magnier & Schoormans, 2015). Consumers need clearer signals to make correct assessments and purchase decisions (Rees et al., 2019; Yokessa et al., 2020). The effect of visual signals such as labels and colors on consumers' perception has been reported quite exhaustively (Boz et al., 2020; Herbes et al., 2020; Krah et al., 2019), but only a few studies have discussed the importance of redesigning the package itself. Modifying the appearance of the package could result in enhancing the recognition of a substrate as eco-friendly (*e.g.*, from recycled resources), avoiding the increasing consumers' confusion toward the proliferated eco-labeling (Moon et al., 2017). In a previous study, we surveyed 734 consumers to understand whether recycled paper-based containers, whose aesthetics were intentionally modified with the incorporation of visible, recognizable particles of waste, were "more eco-friendly" than containers with a *conventional appearance* like the corrugated boxes from Amazon (Van Schoubroeck et al., 2023). More than 90% of the consumers surveyed understood the obvious waste content appearance as clues for judging the packaging as recycled and, in turn, a more sustainable option.

Despite the enhanced environmental perception instilled by the presence of visible recycled material in the packaging, the redesigned containers can have lower mechanical performance than their non-modified analogs. In our previous work, it was shown that the apparent size of waste particles that were incorporated in a recycled fiber matrix had a more detrimental effect on the mechanical properties of the redesigned recycled containers than their weight ratio (wt%) (between 5 and 40 wt%) (Chacon et al., 2022) Large waste particles (with an apparent aspect ratio of 52) disrupted the inter-fiber bonding network more significantly than small particles of larger apparent aspect ratios (72 and 163 in the reported study (Chacon et al., 2022)), which resulted in a 20% decrease of the paperboard strength (Chacon et al., 2022).

The influence of waste properties, specifically their physicochemical characteristics, on the performance of recycled fiber-based containers was unaddressed in our previous work. Yet, previous studies have suggested that the physicochemical properties of wastes such as hydrophobicity and repulpability could impact their chemical and physical interaction with the fiber matrix and, in turn, the properties of the recycled containers (Cao & Heise, 2005; Venditti et al., 2000). However, most of the dedicated research efforts in the field have developed or improved strategies to efficiently remove the increasing amount of non-fibrous materials (*e.g.,* stickies) from recycled furnishes in order to decrease their impact on the appearance and performance of recycled paper-based products (A. Kumar & Dutt, 2021; Kuňa et al., 2021).

Herein, we provide a deeper understanding of how variations in physicochemical properties of waste can alter the performance of recycled containers. Valorization of these large quantities of waste through packaging design is a proposed sustainable route to endow recycled containers with a higher environmentally friendly profile. The overarching goal of this study is thus twofold: (1) raise awareness of consumers on recycled containers to guide their purchase decision towards bettering the environmental impact of packaging, and (2) lower the amount of waste particles in recycling facilities by proposing a novel valorization pathway. To this end, we selected different fiber-based waste sources, namely mixed office waste (MOW), old magazines (OMG), and paper cups laminated with polylactic acid (PLA), with distinct physicochemical properties to best represent the large diversity of waste found in recycling facilities. The waste was minimally processed and added to old corrugated containers (OCC) pulp to produce a series of recycled paperboards with obvious recycled content appearance, thus with enhanced environmental perception (Van Schoubroeck et al., 2023). Three strategies to engineer OCC-based recycled packaging with both a high environmental perception and good mechanical performance were also investigated: i) addition of cationic starch, ii) incorporation of cellulose microfibrils, and iii) multi-ply paperboards.

3.3. Material and methods

3.3.1. Materials

Old Corrugated Containers (OCC) pulp was provided by Greif (Gladstone, VA, US) and used as the fiber source to produce recycled paperboards. Waste materials of different origins and physicochemical properties were selected as follows: mixed office waste (MOW) was simulated using copy papers of 75 g/m^2 (Husky® copy, Domtar, South Carolina, US) that were laser printed (Bizhub 364e, Konica Minolta, Canada) on one side with an image in black and white covering 50% of the paper surface area; color printed double-side coated magazines of 152 g/m^2 with ca. 85% coverage were selected and referred to as old magazines (OMG); unprinted disposable paper cups laminated with polylactic acid (PLA) were purchased from Papernain (China) to simulate single-use packaging waste. Cationic starch (CATO® 237, National Starch) with a degree of substitution between 0.043-0.053 % was used as received as a dry strength additive.

3.3.2. Production of paperboards with visible recycled content

Paperboard sheets containing different amounts of waste materials were prepared using a TAPPI handsheet former at a grammage of 120 g/m^2 as described in the TAPPI 205 Sp-02 (2006) standard. Prior to paperboard making, a 10 wt% OCC pulp suspension was refined at 1000 revs. using a PFI mill refiner (the Norwegian Pulp and Paper Research Institute, Oslo, Norway, TAPPI 248 Sp-00 (2000)). The refined OCC pulp was blended with 10 or 40 wt% of waste materials (*i.e.*, MOW, OMG, or PLA paper cups) using a pulp disintegrator (Testing machine Inc., New Castle-DE, USA) for one minute.

The PLA paper cups were also processed using a Wiley mill (Model No. 3, Arthur H. Thomas, Philadelphia, US) with an accepts screen of 2 mm prior to the 1-min blending with the OCC pulp using the disintegrator. This additional step aimed to produce PLA paper cup waste of similar particle size to the MOW and OMG waste.

The Canadian Standard Freeness (CSF) of the waste-containing OCC slurries was determined in compliance with TAPPI 227 om-09 (2009).

3.3.3. Upgrading the mechanical performance of redesigned paperboards with obvious recycled appearance

3.3.3.1. Utilization of Cationic starch (CS)

A solution of 1 wt% of CS was prepared by cooking the aqueous dispersion of CS under continuous magnetic stirring (200 rpm) at 90 °C for 30 min until complete gelatinization of the starch. (Hamzeh et al., 2013) A fresh starch solution was prepared and used the same day for papermaking to prevent starch retrogradation. CS was added at 0.5, 1.0, 1.5, and 2.0 wt% into the refined OCC pulp and mixed for 1 min at 3000 rpm to ensure complete dispersion of the CS. The same paperboard-making procedure described in section 2.2 was followed to produce these sets of recycled specimens.

3.3.3.2. Addition of cellulose microfibrils (CMFs) from MOW

The MOW (as described above) were shredded and suspended in tap water at an initial solids content of 10 wt% and refined using the PFI mill refiner at 5000 rvs. (TAPPI T 248 Sp-00 (2000)). The consistency of the refined pulp suspension was adjusted to 2.5 wt% prior to processing through a supermasscolloider grinder (MKZA6-5, Masuko Sangyo Co., Ltd, Saitama, Japan) for CMF production as previously reported. The grinder was equipped with silicon carbide grinding stones (MKW 10-46#) and ran at a constant rotating speed of 1200 rpm to target a specific energy consumption of 0.5 kWh/kg of oven-dried pulp.(Trovagunta et al., 2021) The apparent aspect ratio of the produced MOW CMFs was 163.(Chacon et al., 2022) The complete characterization of the thus-produced CMFs is available in Chacon et al. (2022).

The prepared CMFs were added at 1.0, 3.0, and 5 wt% to the refined OCC pulp. Complete dispersion of the additive was confirmed after 1 min disintegration of the slurry. The same paperboard-making procedure described in section 2.2 was followed to produce the paperboards reinforced with CMFs.

3.3.3.3. Production of multi-ply paperboards

2-ply paperboards were produced by pressing together (while wet) two sheets of paperboard produced following the protocol described in section 2.2. One of the plies (outer layer) consisted of 40 wt% waste-containing paperboards, whereas the other ply (inner layer) was produced using 100% OCC pulp (Figure 3-1). MOW- and PLA-paper cups of reduced size were used as the outer layer. The grammage of the single plies was varied, but the total grammage of the 2-ply paperboards was kept constant (ca. 120 g/m^2). Waste-containing paperboards with 60 g/m^2 and 40 g/m^2 and OCC paperboards with 60 g/m^2 and 80 g/m^2 were used in the 2-ply paperboard production and were denoted as 2P 60-60 and 2P 40-80, respectively (Figure 3-1). Samples were dried as described in the TAPPI 205 Sp-02 (2006) standard prior to testing.

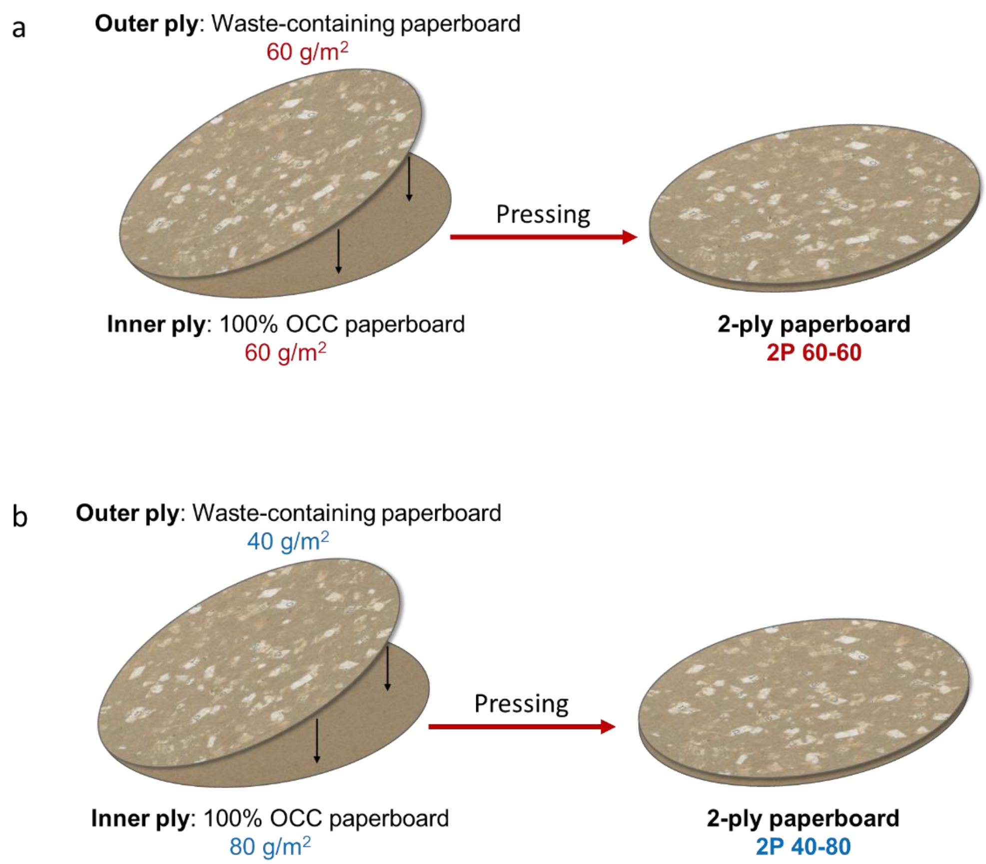


Figure 3-1. Schematic representation of 2-ply paperboard production. (a) 2 ply paperboard with an outer and inner layer with a grammage of 60 g/m^2 (2P 60-60), (b) 2 ply paperboard with an outer and inner layer with a grammage of 40 g/m^2, and 80 g/m^2, respectively (2p 40-80).

3.3.4. Characterization of the waste materials and recycled paperboards

3.3.4.1. Charge Demand Measurements

The charge demand of CS, OCC fibers, waste materials, and 40 wt% waste-containing OCC fibers with either 0 wt%, 1.0 wt%, or 2.0 wt% CS was determined using a streaming current titration analyzer (CAS-Touch, Emtec, Germany). Each specimen was prepared at 0.1 wt% consistency and titrated using either polydiallyldimethylammonium (poly-DADMAC) if negatively charged and potassium poly(vinylsulfate) (PVSK) if positively charged surfaces.

3.3.4.2. Fourier-transform Infrared spectroscopy (FTIR)

FTIR spectra of the waste materials were recorded using an attenuated total reflectance-Fourier transform infrared spectrometer (ATR-FTIR) (Frontier, PerkinElmer, Massachusetts, USA). The data was collected in the spectral range from 4000 cm^{-1} to 650 cm^{-1} with a resolution of 4 cm^{-1} for 10 scans.

3.3.4.3. Surface Wettability

The wettability of the waste materials was evaluated by water contact angle measurement using a SEO Phoenix 300 analyzer (Suwon city, Korea). A drop of about 7 µL of deionized water was placed on the paper waste surface using a micro-syringe, followed by image acquisition immediately after the first contact of the probe liquid with the surface. The average water contact angle obtained from at least five measurements per waste type is reported for discussion.

3.3.4.4. Water Absorption

Water absorptiveness of the waste materials was measured by adapting the 2-min Cobb test (TAPPI 441 om-98, 1998) to a proportionally smaller test area of 16 cm^2. The absorption of 10 mL of water by the 16 cm^2 sample area was determined by measuring the weight uptake of the wetted specimen after 120 s of contact with the water. An average value was obtained from a minimum of five replicates.

3.3.4.5. Microscopy Analysis

A benchtop scanning electron microscope (SEM) equipped with energy-dispersive X-ray spectroscopy (EDS) (JCM-6000PLUS, JEOL, Tokyo) was used for surface and cross-sectional analysis of the waste materials and waste-containing OCC paperboards. EDS analysis of the waste materials was performed at an accelerating voltage of 15.0 kV. Surface and cross-section images of the paperboards with visible recycled content were taken at an accelerating voltage of 5.0 kV and a magnitude of ×100 and ×200, respectively. The samples were coated with a thin layer of gold prior to image analysis.

3.3.4.6. Image acquisition of waste particles and paperboard substrates

The average covered area of waste particles after 1 min disintegration was determined using Spec*Scan^{TM} 2000 image analysis system (Apogee Systems Inc.) with a flat-bed scanner (Epson Perfection 2400 photo). Aqueous suspensions of disintegrated waste at 0.04 wt% were filtered using a black filter paper (Grade 8613, Ahlstrom) and then subjected to image analysis using a resolution of 600 dpi, with a 256 grayscale and a reverse threshold setting of 80% of the average grayscale value. Ten specimens per waste type were measured and averaged for discussion.

3.3.4.7. Physical and mechanical properties of the waste-containing paperboards

The waste-containing paperboards were stored for at least 24 h at 50% relative humidity (RH) and 23 °C prior to testing (TAPPI 402 sp-98 1998). The grammage, thickness, and apparent density of the specimen were determined using a high precision lab-scale and a micrometer (Lorentzen & Wettre, Sweden) (TAPPI 220 Sp-96 1996).

The mechanical performance of the specimens, specifically the tensile strength(TAPPI 494 om-96, 1996), tear strength(TAPPI 414 om-98, 1998), burst strength(TAPPI 403 om-97, 1997), and short-span compression strength (STFI compression)(TAPPI 826 om-08, 2013) were assessed and reported as an average of a minimum of five measurements per waste-containing paperboards. The tensile strength was performed using a tensile tester TMI 84-56 from Testing Machines, Inc. (New Castle, DE, USA). The tear strength was evaluated using a Lorentzen & Wettre tear instrument (Sweden). A Mullen burst tester (Model A, Testing Machines, Inc., New Castle, DE, USA) and a STFI compression strength tester (Lorentzen & Wettre) were used to determine the burst and compression strength of the paperboards, respectively.

The mechanical properties were reported as indexes, meaning that the average values were normalized by the average grammage of the samples tested.

3.3.4.8. Statistical Analysis

The values reported for the mechanical properties of the paperboard specimens were evaluated by analysis of variance (ANOVA) followed by a post hoc Tuckey test using the JMP software (SAS Institute). The statistical analyses were performed to determine the significantly different means with a confidence level of 95%.

3.4. Results and discussion

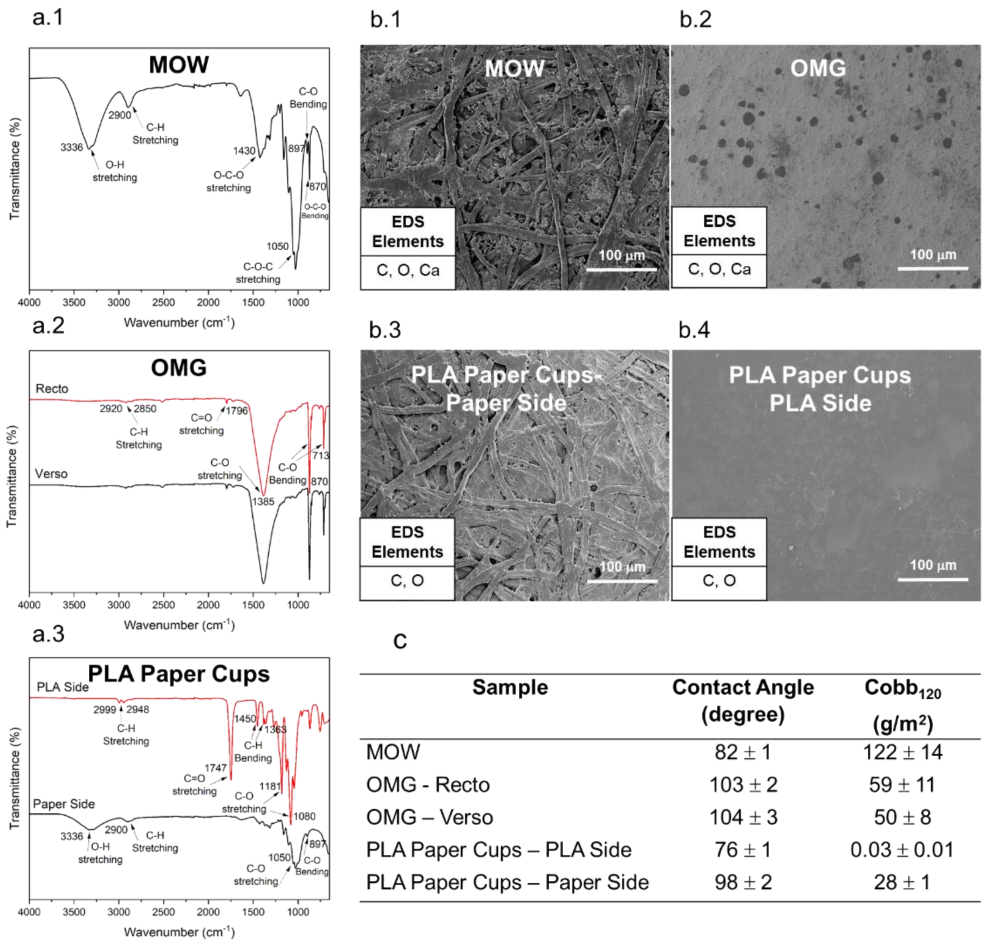
3.4.1. Identification and characterization of the waste materials

The main structural and physicochemical properties of the selected waste materials were analyzed by combining different spectroscopy techniques and image analyses. Figure 3-2a shows the IR spectra of MOW, OMG, and PLA paper cups. The recto and verso (*i.e.,* inner and outer layers of the cups) of the two latter samples were studied to confirm their manufacturing process. The MOW (Figure 3-2a.1) and the paper side (outer layer) of the PLA paper cups (Figure 3-2a.3) showed prominent absorption bands at wavenumbers characteristics to cellulose. The signals at 3336, 2900, 1050, and 897 cm^{-1} correspond to the stretching vibration of O-H, C-H, and C-O-C bonds and the C-O bending vibration of the β-glycosidic linkages, respectively (Chen et al., 2018). In addition, the MOW IR spectra displayed peaks at 1430 and 870 cm^{-1}, which can be assigned to the bending and the stretching vibrations of the O-C-O bonds, respectively, as found in calcite (Noah et al., 2017). This suggests that fillers such as calcium carbonate ($CaCO_3$) that are commonly added to bleached wood pulp to enhance opacity, brightness, and printability of copy paper (Pöykiö & Nurmesniemi, 2008; X. Wang et al., 2011) were present in the selected MOW. The results were supported by the SEM-EDS analysis, as shown in Figure 3-2b.1.

The IR analysis of the PLA-coated side of the paper cups showed characteristic peaks of polylactic acid, namely the stretching vibrations at 2999 and 2948 cm^{-1} (C-H bonds), 1747 cm^{-1} (C=O), and 1181 and 1080 cm^{-1} (C-O), as well as bending vibrations assigned to the C-H bonds at 1450 and 1363 cm^{-1} (Chieng et al., 2014).

The IR spectra conducted on the recto and verso of the OMG showed identical absorption bands (Figure 3-2a.2), suggesting that both sides of the OMG substrates were of similar chemical composition. The high-intensity bands observed at 1385, 870, and 713 cm^{-1} can be assigned to the vibration of calcite bonds, indicating the presence of calcium carbonate in the coating slurry. The presence of calcium was also confirmed by SEM-EDS analysis (Figure 3-2b.2). The weaker signals at around 2900 and 1796 cm^{-1} related to the stretching vibration of C-H and C=O bonds suggest the presence of a polymeric material that may have been used as a binder in the composition of the coating slurry. The exact nature of the binder could not be identified from the collected data, but common commercial solutions used in magazines coating could include styrene-butadiene, styrene acrylate, and polyvinyl acrylate latex.

The analysis of the surface properties of the waste materials and their interaction with water was more insightful to understand the formation and resulting properties of the waste-containing paperboards than knowing the exact composition of the waste materials. Hence, the water contact angle and water uptake of each waste material were evaluated (Figure 3-2c). The MOW showed the highest water absorption with a $Cobb_{120}$ value of 122 g/m^2 and a low wettability with a contact angle of 82° which is in accordance with values from literature (Stankovská et al., 2014). On the other hand, the OMG showed a much lower water absorption (ca. 50 g/m^2) and a higher contact angle (ca. 103°) than the MOW, suggesting that both sides of the magazines were coated (and possibly calendered) to reduce the porosity of the paper surface and decrease the hydrophilicity of the substrates, ultimately resulting in higher resistance to wetting and liquid penetration (Paltakari et al., 2009). The PLA paper cups demonstrated distinct surface and bulk properties between their PLA liner (inside the cups) and paper-based outer side. The paper-based side showed a much higher $Cobb_{120}$ value and water contact angle (98° *vs.* 76°) than the PLA-coated side, which may be explained by the presence of sizing agents such as alkenyl succinic anhydride (ASA) or alkyl ketene dimer (AKD) in the pulp. These sizing agents are traditionally added to the composition of packaging-intended pulp to reduce its liquid absorbency (Triantafillopoulos & Koukoulas, 2020). In contrast, the PLA-coated side showed a more uniform and denser surface (no pores were observed on the SEM surface images) as a result of possible lamination (Figure 3-2b.4), which would corroborate the lower contact angle values. PLA is known to have excellent resistance to water penetration showing a swelling ratio and water solubility of only 2% and 5%, respectively (Hosseini et al., 2016; Rhim & Kim, 2009).

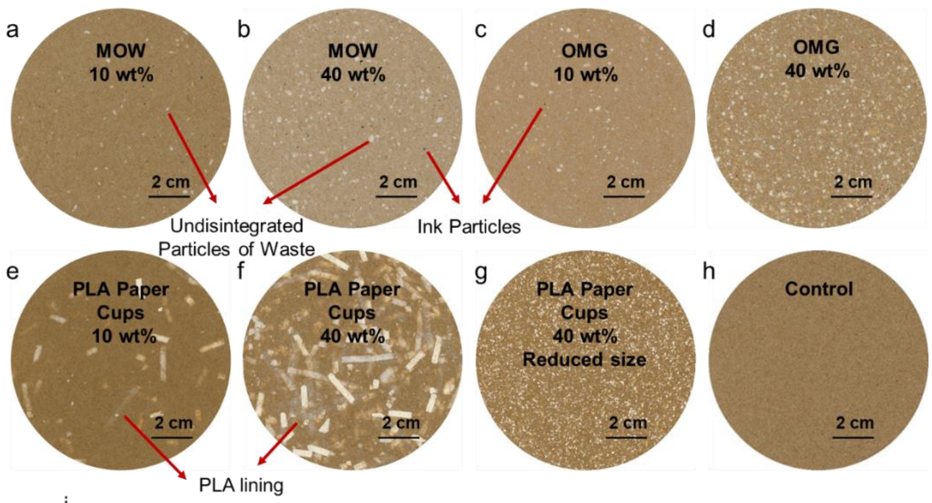


Sample	Contact Angle (degree)	$Cobb_{120}$ (g/m²)
MOW	82 ± 1	122 ± 14
OMG - Recto	103 ± 2	59 ± 11
OMG – Verso	104 ± 3	50 ± 8
PLA Paper Cups – PLA Side	76 ± 1	0.03 ± 0.01
PLA Paper Cups – Paper Side	98 ± 2	28 ± 1

Figure 3-2. Properties of selected waste materials for the production of OCC paperboards with obvious recycled content. (a) IR spectra of a.1 MOW, a.2 OMG, and a.3 PLA paper cups, (b) SEM surface images of b.1 MOW, b.2 OMG, b.3 outer paper side, and b.4 inner PLA side of PLA paper cups, and (c) water contact angle and water uptake of MOW, OMG, and PLA paper cups. The insets, b.1 to b.4, show the atoms detected in higher concentration (elemental analysis) by EDS on the respective SEM surface images.

3.4.2. Properties of waste-containing paperboards

Figure 3-3a-h shows the scanned images of the paperboards with 10 and 40 wt% of waste materials (*i.e.*, MOW, OMG, and PLA paper cups). The low-intensity processing applied to the waste/OCC pulp slurry resulted in the partial disintegration of the waste materials into a blend of fibers, individual released components such as ink or PLA lining, and large particles of undisintegrated waste (Figure 3-3a-g and Figure S1 in the supplementary information). The partially disintegrated waste materials could be easily observed on the surface of the recycled paperboards after handsheet-making endowing the substrates with a “more obvious” recycled content appearance and, in turn, guiding consumers towards identifying recycled packaging assertively (Van Schoubroeck et al., 2023).



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Paper waste	Treatment	Average Particle Area (mm^2)
MOW	1 min. disintegration	0.33
OMG	1 min. disintegration	0.26
PLA paper cups	1 min. disintegration	3.35
PLA paper cups reduced size	Grinding/ 1 min. disintegration	0.33

Figure 3-3. Scanned images of OCC paperboards with 10 and 40 wt% of (a, b) MOW, (c, d) OMG, (e, f) PLA paper cups, respectively, (g) with 40 wt% PLA paper cups processed further, *i.e.*, with reduced particle area, (h) control (*i.e.*, OCC pulp with no waste), and (i) average particle area of the waste materials after disintegration.

Each waste underwent a different break-up during disintegration due to differences in chemical composition and structure of the waste materials. OMG produced the smallest particles, while similar disintegration conditions for the PLA-paper cups resulted in the largest ones with an average particle size of 0.26 mm^2 and 3.35 mm^2, respectively (Figure 3-3i). The MOW showed an intermediate particle area of 0.33 mm^2. The particle size distribution of each waste material is plotted in Figure S2. The presence of waste particles (undisintegrated fragments of waste and released components during disintegration such as PLA lining) in the OCC fibers network created low-density paperboards with a porous and bulkier structure, as shown in the SEM images (Figure 3-4). The effect is more prominent in the presence of large pieces of hydrophobic particles of PLA paper cups, as shown in Figure 3-4g and h. Surface and cross-section images of the PLA paper cups-containing paperboards show a more disrupted network with noticeable de-bonded areas.

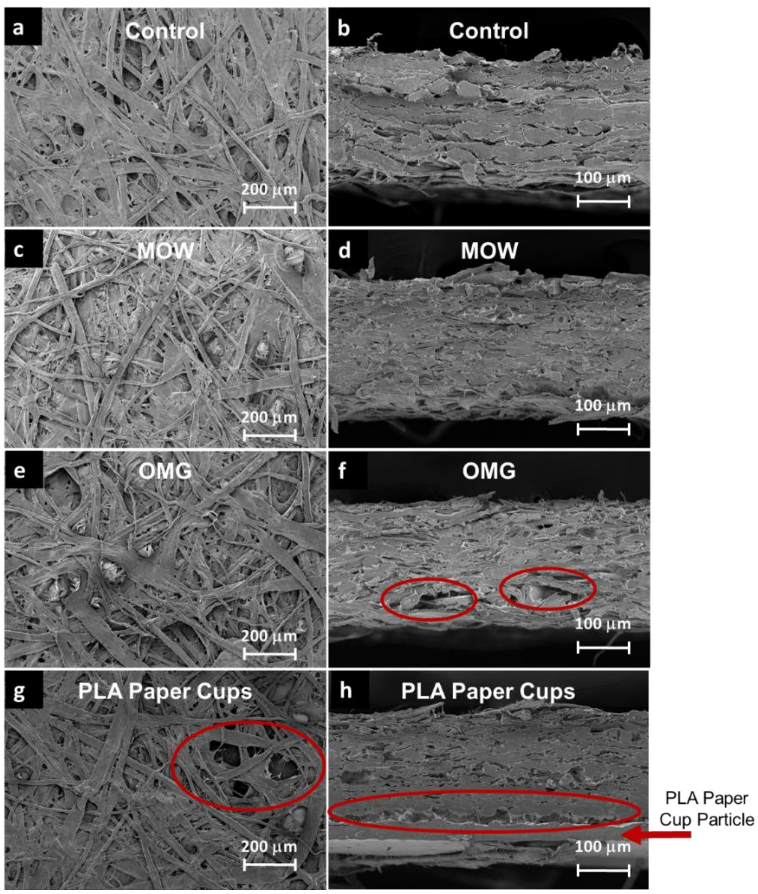
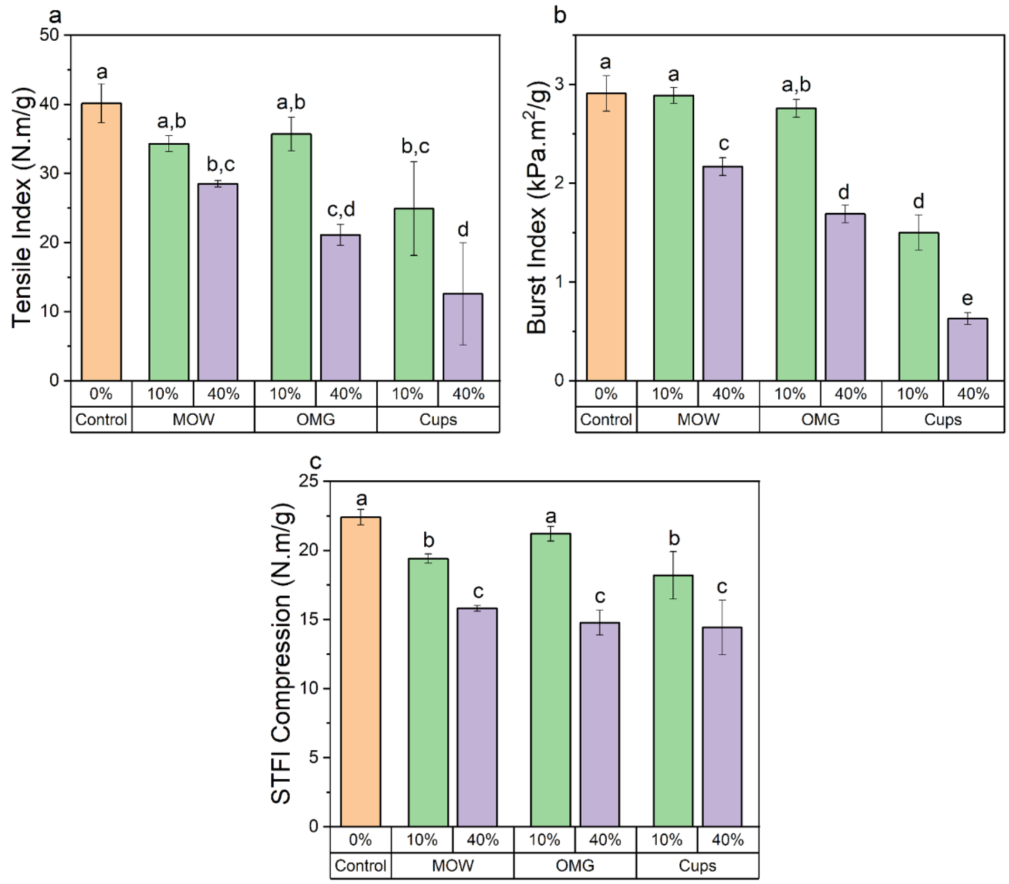


Figure 3-4. Scanning Electron Microscopy (SEM) images of the surface (left) and the cross-section (right) of (a) and (b) control paperboards, (c) and (d) MOW-containing paperboards, (e) and (f) OMG-containing paperboards, and (g) and (h) PLA paper cups-containing paperboards.

3.4.2.1. Mechanical performance of waste-containing paperboards

The mechanical performance of the waste-containing paperboards was affected by the presence of the waste particles. Tensile, and burst indexes of waste-containing paperboards are shown in Figure 3-5a, and b. The presence of waste particles (MOW, OMG, and PLA paper cups) decreased the tensile, and burst strength of the paperboards, with a stronger decrease observed with the increasing weight ratio of waste (40 wt%). The failure during the tensile strength test occurred where the large waste particles were placed, as shown in Figure S3. The presence of waste particles can reduce the bonding interaction between adjacent fibers, thus creating weaker spots where failure can easily occur (Hubbe, 2014). The decrease in the mechanical performance of the waste-containing paperboards was supported by an increase in the freeness of the pulp slurries and a decrease in the density of the paperboards (Figure 3-5d).

The more prominent drop was observed in paperboards containing PLA paper cups. The addition of 40 wt% of PLA paper cups decreased the tensile, and burst strength by 68%, and 78%, respectively. Large standard deviation values were also reported for this series of samples, thus reflecting the high heterogeneity of the samples caused by the large waste particles (ca. ten (10) times larger than the MOW and the OMG particles). In a previous study, Chacon et al. (2022) demonstrated the effect of waste particle size from MOW on the strength of OCC paperboards. Waste particles with low apparent aspect ratio, corresponding to the largest particle dimensions, disrupted the inter-fiber bonding network more significantly than smallest particles, resulting in a 20% decrease in mechanical strength. This corroborates the present findings, explaining why PLA paper cup-containing paperboards showed the lowest mechanical performance. However, the high water repellency and low water uptake of the PLA paper cups (Fig. 1c) may also suggest weaker interactions and bonding between the OCC pulp fibers and the waste particles.



d

Waste type	Control	MOW		OMG		Cups	
	Pulp and paper-waste containing paperboards						
Waste (wt%)	-	10	40	10	40	10	40
Grammage (g/m^2)	132 ± 3	130 ± 5	131 ± 0.4	132 ± 3	129 ± 6	131 ± 3	140 ± 5
Thickness (μm)	239 ± 11	220 ± 11	244 ± 7	222 ± 7	240 ± 15	248 ± 49	390 ± 102
Density (kg/m^3)	610 ± 10	592 ± 10	540 ± 7	594 ± 8	536 ± 10	527 ± 33	360 ± 22
Freeness (ml)	395 ± 7	387 ± 2	423 ± 7	413 ± 16	483 ± 7	440 ± 21	472 ± 6

Figure 3-5. Structural characteristics and mechanical properties of recycled paperboards made with different weight ratios (10 and 40 wt%) of waste materials (a) Tensile index, (b) burst index, and (c) short-span compression strength index (STFI compression). Different letters on the top of the bars indicate significant differences between the means ($p < 0.05$). The physical properties of the pulp and paperboards are shown in (d).

The short-span compression strength (STFI Compression) of the waste-containing paperboards also decreased with the increasing amount of waste materials (from 10 to 40 wt%) (Figure 3-5c) but no significant differences were observed between the three types of waste materials when added at a similar weight ratio. The results from the STFI compression test of the PLA-paper cups-containing paperboards should, however, be cautiously discussed. The STFI values may indeed reflect the compressive strength of the waste particles alone (large pieces of PLA paper cups) and not be representative of the fiber/waste materials network.

The PLA paper cups were thus further processed to reduce their apparent particle size to a size range similar to that of the MOW and OMG particles. They were reduced from 3.35 mm^2 to 0.33 mm^2 using a Wiley Mill prior to their blending with the OCC pulp (Figure 3-3g and i and Figure S1d). Figure 3-6 compares the tensile, burst, and STFI indexes of the 40wt% waste-containing paperboards. Although the three types of waste materials had a similar apparent size (ca. 0.3 mm^2), a greater decrease in the strength was still observed for the PLA-paper cups-containing paperboards, followed by the OMG-containing paperboard. The standard deviation values were considerably reduced, implying that the apparent particle size mainly affected the formation of the paperboards. The results suggest that the structural and chemical properties of the waste materials seem to also play a major role in the performance of the waste-containing paperboards (Venditti et al., 2000). The release of hydrophobic/water repellent compounds (*e.g.*, binders and sizing agents with large alkyl and vinyl groups) added to the coating of OMG or manufacturing of PLA-paper cups (an aliphatic polyester) upon pulp processing may have prevented the formation of hydrogen bonds between OCC fibers, thus negatively affecting the strength performance of the waste-containing paperboards. However, the mechanical performance of both 10 wt% OMG- and MOW-containing boards was quite similar, suggesting that there exists a threshold value (*e.g.*, >10 wt% of OMG waste) beyond which chemical incompatibility between the waste materials and the fiber network will drastically reduce the mechanical performance of the paperboards, thus limiting the potential for strength recovery (Figure 3-5a, b, and c).

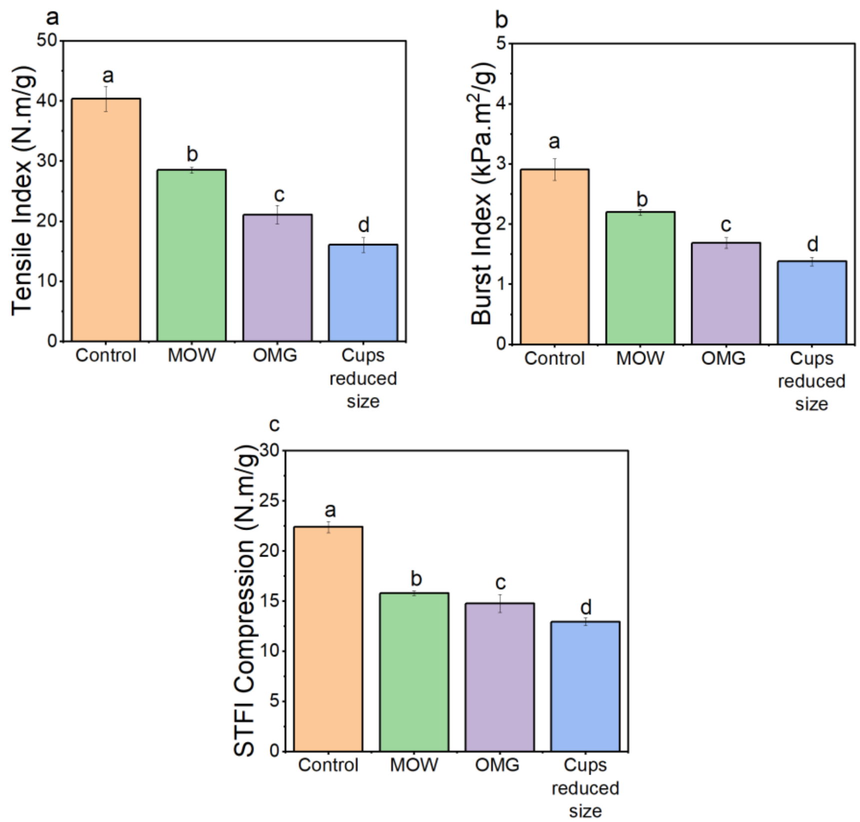


Figure 3-6. Mechanical performance of OCC paperboard with 40 wt% waste of similar apparent size (ca. 0.3 mm^2), as estimated by image analysis using Spec*Scan^{TM} system) (a) Tensile index and (b) burst index, and (c) short-span compression strength index (STFI compression). Different letters on the top of the graph bars indicate significant differences between the means ($p < 0.05$).

3.4.3. Upgrading the mechanical performance of redesigned paperboards with obvious recycled appearance

Two widely reported restorative treatments were first investigated to enhance the mechanical performance of the redesigned paperboards, (i) the addition of cationic starch

(Ghasemian et al., 2012) and (ii) the use of cellulose microfibrils (CMFs) (V. Kumar et al., 2020). CS and CMFs were chosen because of their possible interaction with the hydrophilic components of the paper waste through fiber-waste electrostatic interaction, or hydrogen bonds, respectively.

Cationic starch was added in different weight ratios (0.5 %, 1.0 %, 1.5 %, and 2.0 wt%) to the 40 wt% waste-containing OCC pulp slurries) (Figure 3-7a-c). The results showed that increasing addition of CS up to 2 wt% enhanced the tensile index by 44% and 69%, burst index by 67% and 80%, and STFI compression by 28% and 29%, of MOW- and OMG-containing paperboards, respectively. The substrates recovered almost all of their full mechanical performance (*i.e.*, their performance aligned with that of neat OCC paperboards). Improvement of recycled paper strength with CS was previously assigned to the creation of electrostatic interactions between the protonated quaternary amine groups on CS and the negatively charged sites present in the lignocellulosic substrates(H. Li et al., 2004) Such interactions can enhance fines retention (estimated at 8% for the OCC pulp used in this study) in the inter-fiber network, forming more fiber-to-fines and fiber-to-fiber hydrogen bonds and thus increasing the mechanical performance (Ghasemian et al., 2012; Liu et al., 2015). In the present study, the overall decrease in the cationic charge demand with an increasing added amount of cationic starch to the MOW- and OMG-containing OCC pulp slurries supported the previously proposed mechanism (Figure 3-7d).

Unlike MOW- and OMG-containing paperboards, the substrates containing PLA paper cups did not show any significant improvement in their mechanical properties with the addition of CS, probably due to the inability of CS to bond to the PLA-paper cup particles. The presence of the PLA paper cup waste apparently interferes with the effective charge neutralization of the system, as shown in Figure 3-7d, with only a small decrease in charge demand (13%) at the 2% CS addition level.

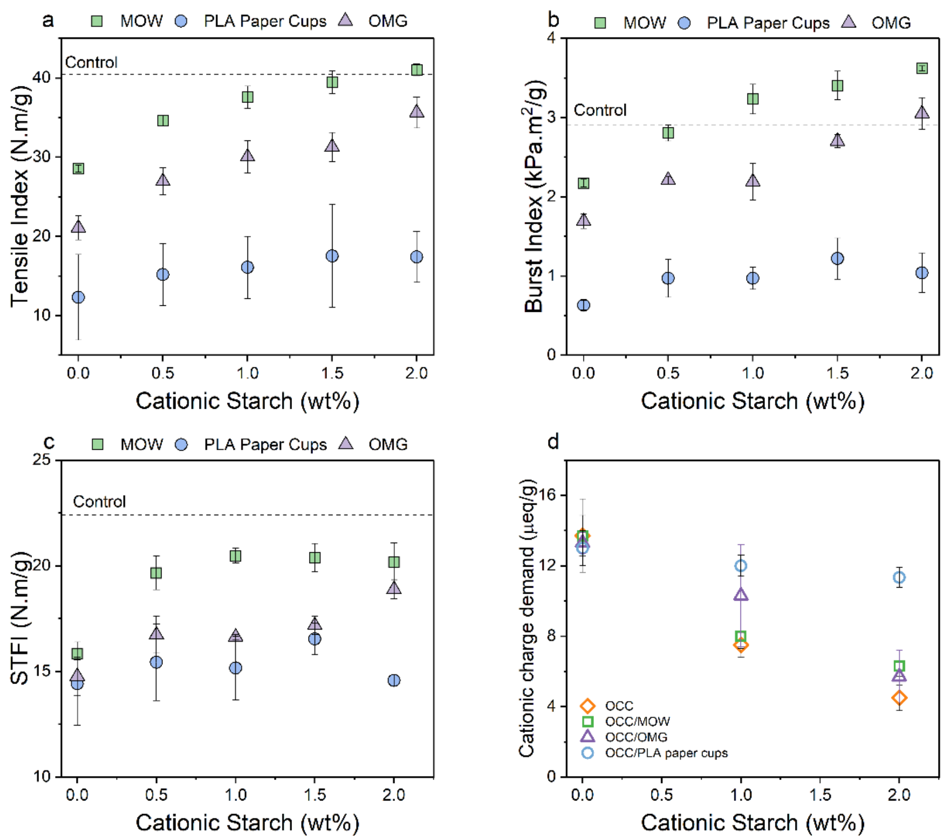


Figure 3-7. Effect of cationic starch addition on (a) tensile index, (b) burst index, and (c) STFI compression index of 40 wt% waste-containing paperboards, and (d) cationic charge demand of OCC and waste-containing OCC pulp slurries with increasing addition of cationic starch.

Similar results were obtained with the utilization of CMFs (Figure 3-8). The addition of 5 wt% CMFs increased the performance of the MOW- and OMG-containing paperboards. These results are consistent with previously published studies that assigned this enhancement effect to the high surface area and aspect ratio of the CMFs, increasing the potential for hydrogen bonding in the paper web (Ang et al., 2020; Delgado-Aguilar et al., 2015; Hu et al., 2021; Laitinen et al., 2020; Zambrano et al., 2020). The CMFs can fill pores created by the waste materials within the inter-fiber network, which in turn increases the density of the paperboard structure, as shown in Table S1. No improvement in the strength of the samples was observed with the PLA paper cups-containing paperboards. The hydrophobic components present in the PLA paper cups could not participate in forming hydrogen bonds between the CMFs and the waste particles and may have thus interfered with the bonding ability of OCC fibers.

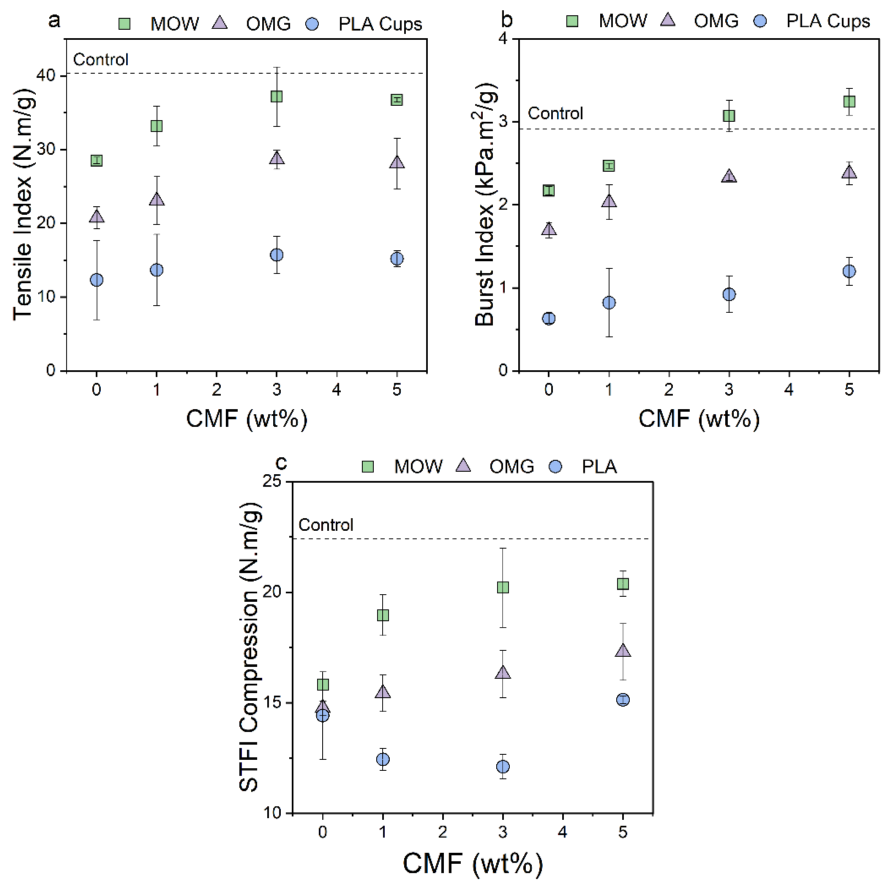


Figure 3-8. Effect of cellulose microfibrils on (a) tensile index, (b) burst index, and (c) STFI compression index of waste-containing paperboards.

The performance of multi-ply paperboards (2-plies) manufactured using the redesigned paperboards in one of the plies was also investigated to mimic the industrial production of linerboards for corrugated packaging. The first ply (outer layer) consisted of the redesigned paperboard with an obvious recycled appearance (with 40 wt% of waste), and the second ply (inner layer) contained only OCC pulp (Figure 3-1). Two configurations of the multi-ply system were evaluated, varying the grammage of each ply, i) 60 g/m^2 each ply (2P 60-60), and ii) 40 g/m^2 outer layer, 80 g/m^2 inner layer (2P 40-80).

Figure 3-9 shows the mechanical performance of the multilayer paperboards. The 2P 60-60 paperboards showed enhanced tensile, burst and STFI compression indexes compared to the one-ply paperboards (1P); for instance, the MOW-containing 2-ply paperboards exhibited similar tensile and burst strength properties to control samples (1P OCC, and 2P 60-60 OCC paperboards). The inner ply, composed of 100% OCC pulp, with no waste particles disrupting the inter-fiber network, has better strength properties than the waste-containing paperboards; hence, the OCC ply conferred better mechanical properties to the overall multi-ply system.

However, the improvement shown by 2P 60-60 PLA-paper cups-containing multi-ply paperboards was not enough to meet the properties of the control samples. Thus, the basis weight of the outer layer was reduced to 40 g/m^{2,} and the inner layer increased to 80 g/m^2 (2P 40-80). Increasing the OCC ply grammage to 80 g/m^2 improved the mechanical properties of the MOW- and PLA-containing 2-ply paperboards to an extent that some of the reported properties were superior to the control samples. Increasing the grammage of the inner ply led to the formation of a denser paperboard (Table S2) which increased the bonded area and thus the strength of the samples (Hubbe, 2014). Moreover, decreasing the grammage of the outer ply reduced the thickness of the disrupted waste-containing layer reducing the impact of the waste properties on the entire performance of the multi-ply paperboard.

Utilizing the multi-ply manufacturing process in redesigned paperboards allowed the formation of specimens with good mechanical performance regardless of the nature of the waste while keeping the recycled appearance that serves as consumers' indicators of sustainability and facilitates the purchase decision. Thus, multi-ply paperboards with redesigned appearance could be manufactured and meet the product specifications regarding the strength of the packaging.

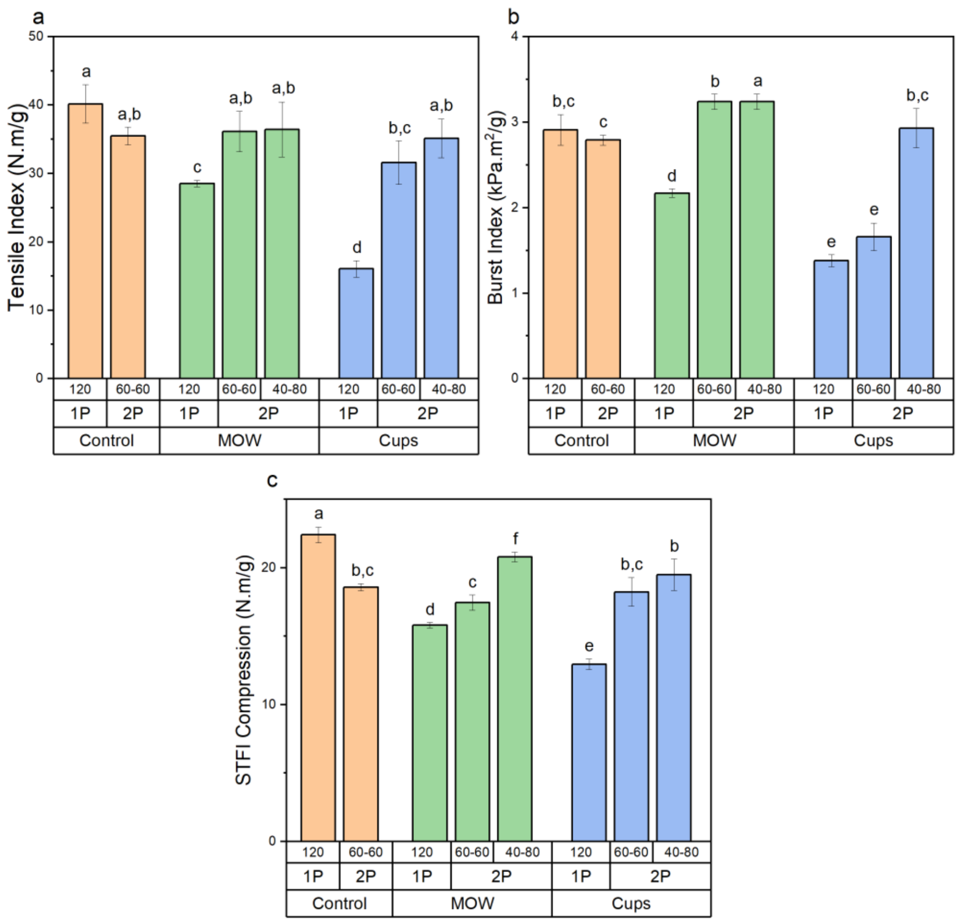


Figure 3-9. Mechanical performance of single-ply (1P) waste-containing paperboards with a grammage of 120 g/m^2, and 2-ply (2P) paperboards with different grammage (60 g/m^2 each ply (60-60), and 40 g/m^2 outer layer, 80 g/m^2 inner layer (40-80)). (a) Tensile index, (b) burst index, and (c) short-span compression strength index (STFI compression). Different letters on the top of the graph bars indicate significant differences between the means ($p < 0.05$).

3.5. Conclusion

Recycled paperboards were produced by blending partially disintegrated fiber-based waste of different physicochemical characteristics with old corrugated containers (OCC) fiber furnish to endow the resulting recycled containers with an obvious recycled appearance. The influence of the physicochemical properties of three types of waste, namely mixed office waste (MOW), old magazines (OMG), and PLA paper cups, on the mechanical performance of the redesigned recycled paperboards was studied to assess the potential of such new design to meet the performance of current corrugated boxes, in addition to guide consumers in their purchasing decisions towards sustainable packaging.

MOW was the most easily disintegrated into a blend of individual fibers and small waste particles. The presence of hydrophilic functional groups in MOW allowed the waste to interact with the OCC fibers through the formation of hydrogen bonds, resulting in recycled containers with the highest mechanical performance compared to the other waste-containing paperboards. Still, this series of redesigned recycled boards performed less than OCC paperboards.

The addition of waste materials with hydrophobic functional groups, such as those present in OMG (alkyl and vinyl groups in polymeric binders) and PLA paper cups (an aliphatic polyester), reduced more significantly the mechanical performance of the waste-containing paperboards by disrupting physically and chemically the inter-fiber network. A drastic decrease was observed with the addition of PLA paper cup waste. Due to its physicochemical properties, the re-pulping of PLA paper cups released large and non-processed hydrophobic particles that prevented the formation of hydrogen bonds between the OCC fibers.

Adding up to 2 wt% of cationic starch or 5 wt% of cellulose microfibrils to the waste-containing OCC pulps successfully restored the mechanical properties of MOW- and OMG-containing paperboards, owing to their more hydrophilic nature favoring OH-bond formation and anionic character of fiber components. However, the performance of the recycled paperboards containing PLA paper cups was not improved. The production of 2-ply paperboards with an outer layer composed of the redesigned paperboard (with obvious recycled content) and an inner layer made of 100% OCC fibers overcame the limitations of the aforementioned strength agents. Irrespective of the type and size of the added waste, the multi-ply paperboards showed similar properties to the 2-ply 100% OCC boards.

This study demonstrates that high ratios of low-quality fiber-based waste can be incorporated into recycled furnishes as a strategy to produce sustainable packages that perform similarly (or beyond) existing recycled corrugated boxes, in addition to more directly informing consumers on the recyclability and sustainability nature of such boxes. Redesigning packaging with high quantities of hydrophobic and difficult-to-process waste could significantly contribute to the bio-based circular economy by reducing the amount of waste ending in landfills through valorization of low-grade paper waste such as paper cups, which, to date are not recycled (recycle rate lower than 1%).

The previous two chapters (Chapter 2, and 3) have demonstrated that:

(i) The appearance of recycled containers can be redesigned by incorporating low-intensity processed waste into the recycled furnishes

(ii) Although the mechanical performance could be affected by the size and the physicochemical properties of the waste, using the multi-layer approach can produce specimens with good mechanical properties

But **can consumers perceive redesigned recycled containerboards as more environmentally friendly?** Chapter 4 investigates consumers' sustainability perception toward redesigned containers

Chapter 4 . Environmental Sustainability Perception Toward Obvious Recovered Waste Content in Paper-Based Packaging: An Online and In-Person Survey Best-Worst Scaling Experiment[3]

4.1. Abstract

This study explores consumers' visual sustainability impressions of paper-based packaging that has incorporated obvious waste content. Two research questions were addressed concerning (i) the environmental sustainability perception of noticeable waste content in packaging and (ii) the impact of the presentation format (i.e., online versus in-person surveys) when studying these perceptions. Best-worst scaling experiments were conducted, which made respondents choose the 'most' and 'least' environmentally friendly package. Packages were designed using paperboard substrates blending either brown linerboard or white hardwood pulp with different recovered waste materials. The results showed that consumers perceive obvious waste-containing packaging as more environmentally friendly than classical packaging (with no visual waste). Samples with a brown base and agricultural waste were perceived as more sustainable compared to white packaging and the use of paper waste. In addition, the presentation format changed respondents' perception, and should therefore be carefully considered when designing surveys.

[3] The material in this chapter has been published as:
Van Schoubroeck, S., Chacon, L., Reynolds, A., Lavoine, L., Hakovirta, M., Gonzalez, R., Van Passe, S., & Venditti, R. A. (2023). Environmental sustainability perception toward obvious recovered waste content in paper-based packaging: an online and in-person survey best-worst scaling experiment. *Resouces Conservation and Recycling, 188* (106682). https://doi.org/https://doi.org/10.1016/j.resconrec.2022.106682

4.2. Introduction

Together with increased global production and consumption, the use of packaging materials and packaging waste has grown. Many packages are intended for single-use applications and consequently disposed of by the final customer. The Environmental Protection Agency (EPA) reported that containers and packaging make up about 28 percent of total municipal solid waste (MSW) generation in the US, amounting to 82.2 million tons of containers and packaging waste generated in 2018. Half of these packaging products are paper-based, of which 6,440 thousand tons end up being landfilled (EPA, 2022). While striving for a sustainable and circular packaging industry, opportunities to further recycle and reuse (paper-based) waste for new packaging products exist and should be further explored.

The use of recovered waste content in packaging can reduce its environmental footprint. However, the question remains: is this also perceived as such by the customer or the end user? Comparisons between consumer judgements and environmental life cycle analysis (LCA) results indicated that consumers often rely on misleading and inaccurate beliefs to judge the sustainability of packaging (Steenis et al., 2017). A consumer can judge the package based on the structural (e.g., the packaging material), verbal, and graphical design. While striving toward more sustainable products, both the intrinsic attributes (e.g., manufacturing efficiency or organic ingredients) and the extrinsic attributes (e.g., the package) of the product play an important role (Magnier et al., 2016). Intrinsic sustainability can only be communicated via labels and logos, while extrinsic attributes have the opportunity to be redesigned. Steenis et al. (2017) demonstrated that consumers' sustainability evaluations are highly influenced by graphical packaging cues that have no actual sustainability consequences (Steenis et al., 2017). For example, the use of green color in the graphical design of a package is automatically associated with a higher level of sustainability (Pancer et al., 2017; Steenis et al., 2017).

Environmental sustainability performance has been perceived as a key product attribute, and therefore a source of potential differentiation and competitive advantage for companies (Porter & Van Der Linde, 1995). When consumers make product decisions, the environmental criterion is increasingly important (Peattie & Peattie, 2009). Multiple studies have been conducted analyzing the willingness-to-pay (WTP) for products with beneficial social and environmental performance characteristics. The majority of these research studies point to a higher premium for sustainable products, compared to their less-sustainable alternatives (Salazar & Oerlemans, 2016). However, customers are growing acutely aware of “greenwashing”, the practice of disclosure of false or incomplete information by an organization to present an environmentally responsible public image (Furlow, 2010). The perception of greenwashing strategies harms consumer attitudes and can lead to loss of credibility and poor purchasing decisions (Lewandowska et al., 2017; Parguel et al., 2011).

Product sustainability perception should be accounted for when developing and designing new sustainable packaging materials. Consumers should be able to easily recognize sustainable packaging based on direct cues provided by the material itself (i.e., implicit packaging cues), without the need for labels and claims (i.e., explicit packaging cues). The redesign of sustainable packaging could be achieved by adding low-intensity processed waste to the fiber furnish prior to papermaking (Chacon et al., 2022). Chacon et al. (2022) showed that by minimally processing the waste, the paper could be endowed with macroscopic and visual particles on the surface of the substrate that could communicate sustainability. It is suggested that the obvious recovered waste content within the package will help consumers to identify the product as environmentally friendly and guide consumers' purchasing decisions toward these options. Therefore, a deep understanding of consumers' attitudes toward packaging with noticeable waste content is needed. Previous research has focused on the influence of implicit packaging cues on the perception of consumers by using surveys (Granato et al., 2022). Some of these studies focused on the perception of different types of materials such plastic packaging, or a comparison between glass, plastic, and aluminum packages (De Feo et al., 2022; Weber Macena et al., 2021). However, no studies in this regard were found focusing on paper-based packaging with visually obvious waste content.

The present study focuses on both the structural and graphical design of paper-based packaging, without any information provided by labels and logos. Consumers' visual impressions on a variety of paper-based packaging materials were explored, which provides insights into consumers' beliefs on sustainable packaging. More specifically, this study aims to investigate if paper-based packaging with incorporated visually obvious paper and agricultural waste content instils positive sustainability perceptions. Consumer perception was studied by the use of stated preference (SP) methods, which rely on data that comes from consumers' responses to hypothetical questions. Previous studies have shown that the presentation format used in consumer questionnaires has a significant impact on choice (Mokas et al., 2021; Murwirapachena & Dikgang, 2021). Traditionally, SP methods rely on text descriptions or pictures of the assessed good or service. However, the evaluation of the packaging materials might change when packaging materials are shown to consumers in real life.

Two research questions are addressed in the present study: (1) Does the obvious recovered waste content in the packaging influence the perception of how environmentally sustainable the package is? And (2) Does the presentation format (online versus in-person surveys), which is used to study consumers' preferences, change the packaging perception? This study aims to gain an understanding of consumers' choices regarding sustainable packaging and to create a unique dataset comparing both online and in-person survey responses.

4.3. Materials and Methods

4.3.1. Materials

Recycled brown linerboard (RL) (S-19318, Uline, Georgia, US) and elemental chlorine-free bleached white hardwood pulp (BHW) (International Paper, US) were used as raw materials for paperboard making. Brown (unbleached) and white (bleached) pulp represent the two type of fibers used in paper-based packaging applications (Wu, 2021). Paper and agricultural waste were utilized to endow the paperboards with a visually obvious contaminant content. Copy paper of 75 g/m^2 (Husky® copy, Domtar, South Carolina, US), coupon inserts of 45 g/m^2 composed of lightweight coated (LWC) paper collected from a local grocery store (North Carolina, US), and green and pink paper of 89 g/m^2 (Astrobrights, Georgia, US) were selected as paper waste materials. The paper waste selection was based on global paper production by category. According to Tiseo (2022), printing- and writing paper is the second largest paper consumed globally and, therefore, one of the major types of paper waste produced. Switchgrass (SW) collected from a local source (North Carolina, US) and cocoa bean shells purchased from Hull Farm (Wisconsin, US) were chosen as agricultural waste. Switchgrass is an abundant grass native to North America that shows a high growth rate, even under poor soil conditions (Fan Wang et al., 2020). Cocoa bean shells, on the other hand, are a by-product of cocoa production, representing 12% of the raw material (Gómez Hoyos et al., 2020). Both agricultural wastes represent an abundant, renewable, low-cost feedstock that can be used for high-value products such as packaging

Images of the waste materials used in this study are shown in Figure S1 in the supplementary information (SI).

4.3.2. Research methodology and design

Within this research study, the best-worst scaling (BWS) method was applied to investigate the environmental sustainability perception toward obvious recovered waste content in paper-based packaging. BWS is a stated preference (SP) method, developed by Louviere and Woodworth in 1990 (J. Louviere & Woodworth, 1990). The BWS method allows respondents to evaluate all pairwise combinations of alternatives presented in several subsets leading to the modeling assumption that their "best" and "worst" choices represent the maximum difference in utility between all attributes (Parvin et al., 2016). For that reason, the BWS method is also referred to as the "maximum difference scaling" (maxdiff) method. BWS effectively avoids scaling interpretation problems of traditional rating scales (such as the Likert scale) (Finn & Louviere, 1992). Also, compared to rating a product on a 5- or 7-point scale, choosing a product from a set of alternatives is considered a more 'natural' task that consumers undertake daily, for example, when shopping at a store (Chapman & Feit, 2019).

A BWS experiment uses a balanced incomplete block design (BIBD), in which each alternative appears equally often, and co-appears equally often with the other alternatives (Jordan Louviere et al., 2013). Within this study, the alternatives consisted of eleven different packaging alternatives, which were each shown five times over eleven different subsets (Table 4-1). Within each subset, the respondents were asked to indicate the "most" and "least" environmentally friendly box.

Table 4-1. BIBDs with 11 packaging alternatives (1-11) in 11 different subsets (A-K).

Subset	Packaging alternatives				
A	1	2	7	10	8
B	10	3	9	6	7
C	9	7	5	4	1
D	4	5	10	2	6
E	2	8	3	9	4
F	6	1	8	5	3
G	5	10	11	8	9
H	11	4	1	3	10
I	8	6	4	7	11
J	7	3	2	11	5
K	9	11	6	1	2

4.3.3. Presentation formats

The paper-based boxes were evaluated via two different presentation formats. First, an online BWS experiment was launched using 3D images of the boxes. Respondents viewed the box images on their personal desktop or laptop, and were asked not to take the survey on their smartphone. The use of pictures might be representable for the graphical design, but the structural design and the packaging material are expected to be less pronounced within a picture relative to viewing the actual object in-person. Therefore, a second presentation format was added using an in-person survey in which small packaging boxes were shown to the respondents. Both the in-person and online BWS experiments used the BWS design presented in Table 4-1. In the online survey, alternatives within a subset were randomized as well as the subsets themselves. In the in-person survey, the alternatives within a subset were shown on a fixed position to every respondent, but it was ensured that this position (from 1 to 5) differed over the different subsets so that positional bias was avoided. Also, every respondent was asked to judge the subsets (from A to K) in a different order to make sure that the subsets themselves were randomized (Figure S2 in SI).

4.3.4. Preparation of the packaging samples

The boxes were produced from paperboards containing RL and BHW as base material and partially disintegrated recovered waste of different sources (i.e., copy paper, coated paper, colored paper, switchgrass, and cocoa bean shells). For paperboards-making, the RL and BHW were each fully disintegrated in water using a pulp disintegrator (Testing machine Inc., Delaware, US) at 3000 rpm for 5 minutes. The paper waste was shredded to strips of 5 mm width. The agricultural waste was mechanically ground to a particle size of 2 mm using a Wiley laboratory mill (Model No. 3, Arthur H. Thomas, Philadelphia, US) prior to paperboard making.

The waste was then mixed with the base materials (RL or BHW), as described in Table 2, using the pulp disintegrator at 3000 rpm for 30 seconds to partially disintegrate the waste. Additionally, shredded RL and the BHW were combined to prepare paperboards with BHW as a base material with visible particles of RL. The weight ratio of the waste used for the production of the paperboards was 40 wt%, except for the green and pink paper where 10 wt% of waste was used. The pulp slurry containing the blend of base material and partially disintegrated waste was utilized to produce paperboard specimens with a targeted basis weight of 240 g/m^2, adapting the TAPPI 205 Sp-02 (2006) standard to a lab-scale rectangular handsheet former. The paperboards were dried using a drum dryer (Chromalox 2110, Adirondack Machine, New York, US) at 100 °C (± 5 °C) and then stored at 50% relative humidity (RH) and 23 °C before paper testing according to TAPPI 402 sp-98 (1998) standard.

Table 4-2. Composition of waste-containing paperboard samples. RL = recycled linerboard and BHW = bleached hardwood pulp.

Sample Number	**Sample Identification**	**Base Material**	**Recovered waste**	**Waste (wt%)**	**Content***
1	RL/Control	Recycled linerboard	-	0	
2	RL/Cocoa	Recycled linerboard	Cocoa bean shell	40	
3	RL/Grass	Recycled linerboard	Switchgrass	40	
4	RL/Pink	Recycled linerboard	Pink paper	10	
5	RL/Coated	Recycled linerboard	Coated paper	40	
6	RL/Copy	Recycled linerboard	Copy paper	40	
7	RL/Green	Recycled linerboard	Green paper	10	
8	BHW/Control	Bleached hardwood pulp	-	0	
9	BHW/Grass	Bleached hardwood pulp	Switchgrass	40	
10	BHW/Copy	Bleached hardwood pulp	Copy paper	40	
11	BHW/RL	Bleached hardwood pulp	Recycled linerboard	40	

*based on oven-dry mass

The 11 paperboard samples, i.e., two control samples containing only base materials (RL and BHW) and nine paperboards containing 10 wt% or 40 wt% of paper and agricultural waste, were scanned using a flat-bed scanner (Epson Perfection 2400 photo, California, US). The digitized pictures were used for modeling 3D box images using SketchUp computer program. The box images together with an enlarged picture of the corresponding paperboard (Figure 4-1) were used to evaluate consumers' perception through an online survey.

Additionally, 11 paper-based real boxes of 7.0 x 4.5 x 5.5 cm (L x W x H) were made with the produced paperboards (Figure 4-2). The base of the boxes was sealed with glue, mainly containing acrylic polymer, and a piece of regular tape. The real samples were used for the in-person survey.

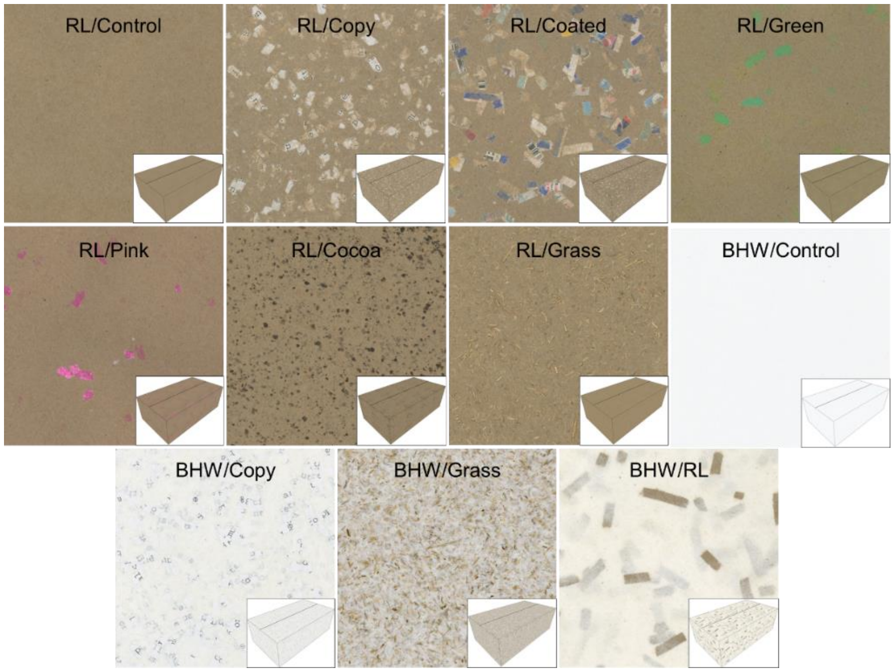


Figure 4-1. Paperboard and box images used for the online survey. The box images were created with a drawing package but included the real image of the paperboard.

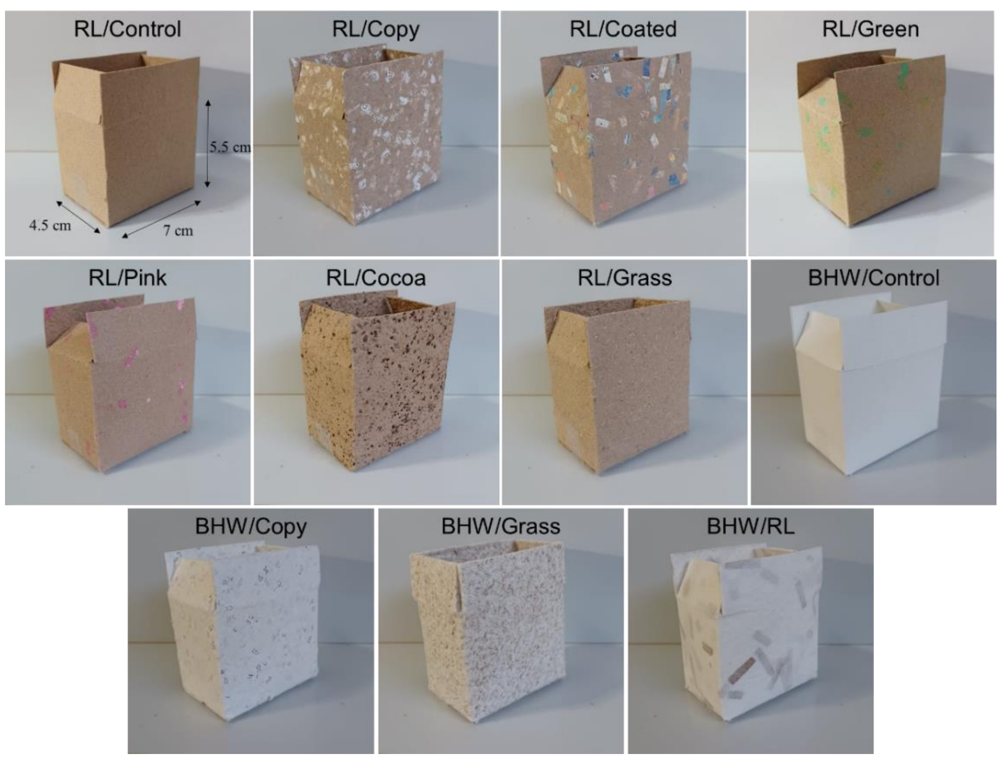


Figure 4-2. Boxes used for the in-person survey.

4.3.5. BWS data analysis

The BWS data was analyzed using a counting and a modeling approach. First, a counting approach counts the number of times each box was chosen as the most or least environmentally friendly alternative. This counting analysis results in B (best), W (worst), and BW (best-worst) scores (Flynn & Marley, 2014; Van Schoubroeck et al., 2019). The BW score of package i is calculated according to Equation (1). The calculation of BW scores enables considerable insights at the level of the individual respondent (Flynn & Marley, 2014).

$$BW_i\ score = \sum B_i - \sum W_i \quad (1)$$

Second, a conditional logit (CL) model is estimated based on respondents having a certain utility (v) (i.e., a certain value) for each package. Respondents are assumed to select the best and worst packages based on the largest difference between their utilities (i.e., the maximum difference model). This conditional logit model will estimate the average preference (Pr) for a certain package among the individuals. Under these assumptions, the probability to select package i as the best and j as the worst is expressed in Equation (2). For interpretation purposes, a share of preference (SP) for package i based on the conditional logit model choice rule was also calculated according to Equation (3) (Cohen & Neira, 2004). The share of preferences must sum to one across all packaging alternatives. The SP_i reports the importance of package i on a ratio scale, meaning that if SP_i is twice that compared to the SP of another package, it can be said that package i is twice as preferred than the other package (Lusk & Briggeman, 2009).

$$\Pr(i, j) = \frac{\exp(v_i - v_j)}{\sum_{k=1}^{m} \sum_{l=1, k \neq l}^{m} (v_k - v_l)} \quad (2)$$

With m = the amount of packages in one choice subset, equals ‘5’ in this study.

$$SP_i \frac{\exp(v_i)}{\sum_{t=1}^{T} \exp(v_t)} \quad (3)$$

4.3.6. Paperboard testing

To find possible correlations between the BWS preference results and the paperboard properties, characterization of the paperboard was performed. The basis weight of the paperboards was determined using a high precision scale and dividing the mass of each sample by the area. The thickness was measured using a micrometer (Lorentzen & Wettre, Sweden). The bulk (inverse of density) was determined by dividing the thickness of the paperboard specimens by the basis weight. Roughness was measured using the Parker Print Surf roughness tester (SE 115, Lorentzen & Wettre, Sweden) using a contact pressure of 1.0 MPa according to TAPPI 555 pm-94 (1997). Bending resistance (stiffness) was evaluated using a Taber V-5 stiffness tester (Model 150-B, Taber Instrument Corporation, North Tonawanda, N.Y., US) (TAPPI 489 om-08, 2013).

Surface topography of all the paperboard samples was performed using a confocal laser scanning microscope (Keyence VK-X1100, Osaka, Japan). Color analysis was performed using TinEye Color Extraction tool which is a free web service that extracts color palette for all the colors identified in an uploaded image (TinEye, 2022). The average particle size of paper and agricultural waste was measured from scanned paperboards (Epson Perfection 2400 photo) at a resolution of 800 dpi followed by image analysis using ImageJ software (National Institutes of Health and Laboratory for Optical and Computational Instrumentation, US).

4.4. Results and discussion

4.4.1. Data collection

The online survey was launched using a respondents' database from the North Carolina State Sensory Service Center on November 1, 2021 (survey shown in SI S3). The in-person surveys were launched at three different locations in Raleigh, North Carolina, i.e., university campus, a Harris Teeter supermarket, and the NC State Farmers' Market, throughout November of 2021. A total of 506 respondents filled out the online survey and 228 respondents the in-person survey. A total of 19 (3.75%) online respondents and 17 (7.46%) in-person respondents were identified as "bad" respondents. "Bad" respondents were defined as respondents who did not understand the BWS experiment or answered randomly probably with the sole focus to receive the final incentive (i.e., a gift card).

To identify these "bad" respondents, the time to complete the survey and their straight-lining behavior was examined. Responses were deleted based on three criteria: (1) if the survey took less than three minutes, (2) if a respondent scored package i as "most" and package j as "least" in subset x, and at the same time scored package i as "least" and package j as "most" in subset y, and (3) if a respondent scored more than three packages as both "most" and "least" during the full survey. A total sample of 487 online responses and 211 in-person responses were used for final data analysis. Socio-demographical information concerning the included participants and their environmental awareness is provided in SI (S4 and S5).

4.4.2. General packaging preferences

Table 4-3 summarizes the results of both the counting and modeling analysis of the online sample data. The B (best) and W (worst) scores show the frequency of the packages chosen as the most and least environmentally friendly alternatives. Based on the BW (best-worst) scores, the top three chosen packages consisted of the RL/Coated, followed by the RL/Copy and RL/Grass packages. The lowest BW scores were calculated for the two control packaging samples (RL/Control and BHW/Control) without contaminants. In addition to the aggregated BW scores, the BW scores per respondent per package were calculated and used in further analysis to represent the preferences per respondent. The results of the counting analysis were confirmed by the conditional logit (CL) model (Table 4-3). All the means of the packaging alternatives with obvious recovered waste content were positive and significant at $p < 0.01$. This indicates that these packaging alternatives are all more preferred than the RL/Control package, which is used as the benchmark having a coefficient of zero. The mean for BHW/Control is negatively significant at $p < 0.01$, meaning that it is considered less preferred compared to the RL/Control by the respondents. Figure 4-3 shows a preference ranking of the packages, based on the results of the share of preferences for each package, reflecting the preference of respondents toward the packages with obvious recovered waste content.

Table 4-3. Online best-worst scaling results (n = 487).

	Counting analysis			**Conditional logit model**				
	Best	**Worst**	**BW**	**Mean**	**P value**		**Standard errors**	**Share of preference**
RL/Coated	1386	120	1266	2.428	< 2e-16	***	0.0560	0.2738
RL/Copy	1034	66	968	2.104	< 2e-16	***	0.0555	0.1979
RL/Grass	851	141	710	1.799	< 2e-16	***	0.0547	0.1459
BHW/Grass	655	246	409	1.467	< 2e-16	***	0.0548	0.1047
BHW/Copy	547	375	172	1.071	< 2e-16	***	0.0533	0.0705
RL/Cocoa	386	265	121	1.034	< 2e-16	***	0.0530	0.0679
BHW/RL	170	383	-213	0.509	< 2e-16	***	0.0518	0.0439
RL/Green	122	504	-382	0.462	< 2e-16	***	0.0529	0.0383
RL/Pink	73	622	-549	0.150	3.1e-03	***	0.0508	0.0281
RL/Control	257	871	-614	0	-		-	0.0241
BHW/Control	85	1973	-1888	-1.599	< 2e-16	***	0.0556	0.0049

*** $p < 0.01$

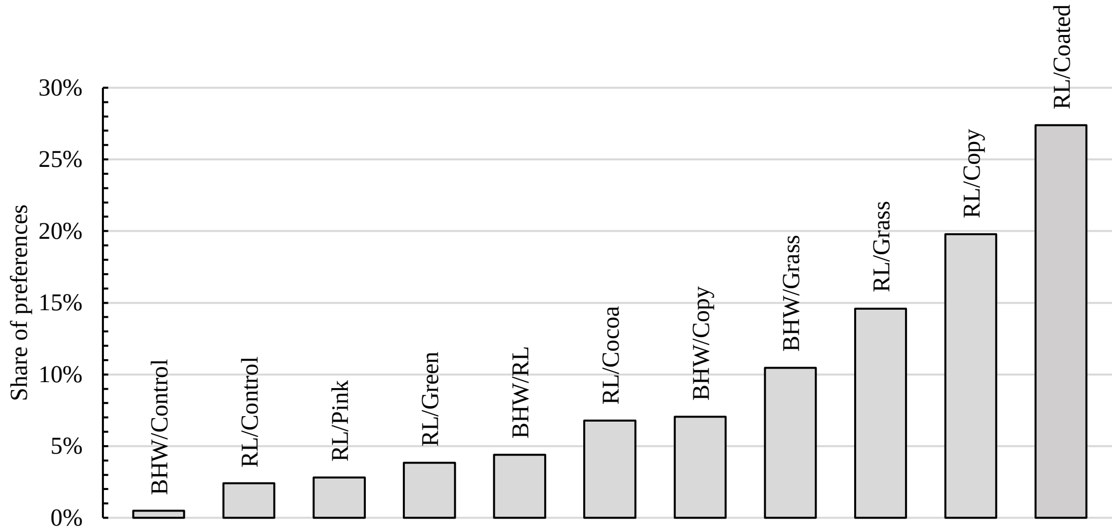


Figure 4-3. Share of preferences for the packaging materials, from least preferred (left) to most preferred (right) package. Results from the online survey based on CL model.

Table 4-4 summarizes the results of both the counting and modeling analysis of the in-person sample data. The B and W scores show the frequency of the packages chosen as the most and least environmentally friendly alternatives. Based on the BW scores, the top three packages consisted of the RL/Grass, followed by the RL/Cocoa and BHW/Grass packages. The lowest BW scores were again given to the two control samples, the RL/Control and BHW/Control. The results of the counting analysis were again confirmed by the conditional logit model (Table 4-4). All the means were significant at $p < 0.05$. Figure 4-4 shows a preference ranking of the packages, based on the results of the share of preferences for each package, visualizing the preference of respondents toward the packages with obvious recovered waste content.

Table 4-4. In-person best-worst scaling results (n = 211).

	Counting analysis			Conditional logit model				
	Best	**Worst**	**BW**	**Mean**	**P value**		**Standard errors**	**Share of preference**
RL/Grass	392	71	321	1.219	< 2e-16	***	0.0726	0.1714
RL/Cocoa	329	82	247	1.035	< 2e-16	***	0.0718	0.1426
BHW/Grass	399	181	218	0.973	< 2e-16	***	0.0720	0.1339
RL/Copy	236	60	176	0.876	< 2e-16	***	0.0724	0.1217
RL/Coated	241	100	141	0.780	< 2e-16	***	0.0719	0.1104
BHW/Copy	143	201	-58	0.298	3.1e-05	***	0.0716	0.0682
RL/Pink	79	158	-79	0.220	1.8e-03	***	0.0707	0.0631
BHW/RL	94	198	-104	0.170	1.5e-02	**	0.0700	0.0600
RL/Green	110	226	-116	0.156	2.8e-02	**	0.0710	0.0592
RL/Control	200	369	-169	0	-		-	0.0506
BHW/Control	98	675	-577	-0.995	< 2e-16	***	0.0721	0.0187

*** $p < 0.01$; ** $p < 0.05$

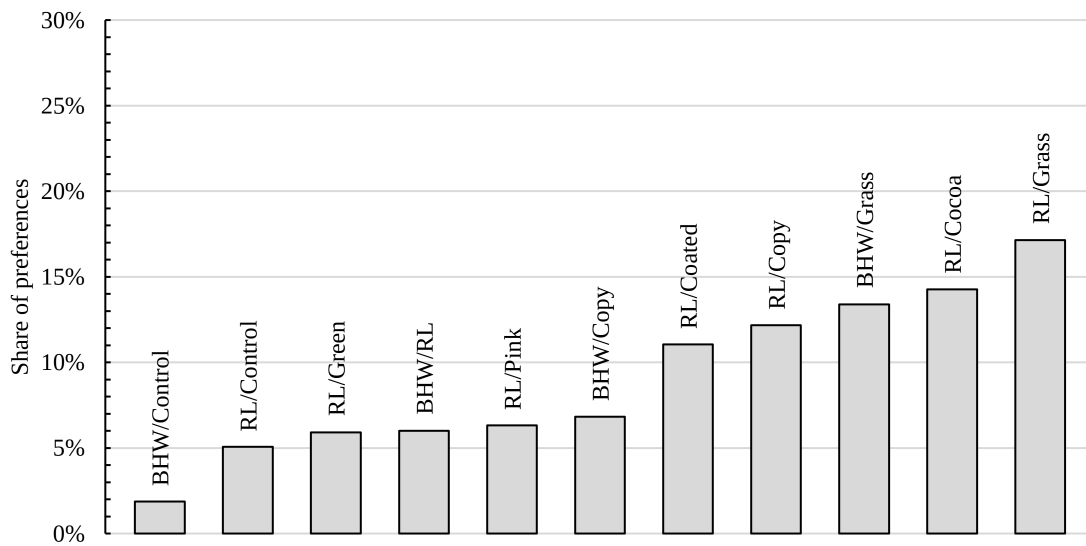


Figure 4-4. Share of preferences for the packaging materials, from least preferred (left) to most preferred (right) package. Results from the in-person survey based on CL model.

A comparison between Figure 4-3 and Figure 4-4 indicates a difference in respondents' preferences between the online and in-person sample. To check if there was a difference between the two samples, Mann-Whitney-Wilcoxon Tests were performed per package (Table 4-5). The

BW scores per respondent were used to measure the preference toward a certain package. The null hypothesis states that the BW scores of the online and the in-person sample are identical populations. When the P value is less than the 0.05 significance level, the null hypothesis is rejected. For the packages RL/Cocoa, RL/Pink, RL/Coated, RL/Cocoa, BHW/Control, BHW/Grass, and BHW/Copy, significant differences were noted between the online and in-person sample.

Table 4-5. Results of Mann-Whitney-Wilcoxon tests per package when comparing the BW scores between the online and in-person sample, and the direction of change in preference when switching from the online to the in-person presentation format (with ↑ = "preference going up", and ↓ = "preference going down").

Sample ID	P value		Direction of change in preference from online to in-person format
RL/Control	3.2e-01		
RL/Cocoa	6.1e-11	***	↑
RL/Grass	4.1e-01		
RL/Pink	3.5e-06	***	↑
RL/Coated	2.2e-16	***	↓
RL/Copy	2.2e-16	***	↓
RL/Green	5.2e-01		
BHW/Control	5.4e-10	***	↑
BHW/Grass	7.4e-02	*	↑
BHW/Copy	2.0e-04	***	↓
BHW/RL	7.0e-01		

*** $p < 0.01$; * $p < 0.1$

At the end of both the online and in-person surveys, the respondents were asked to rate the BWS experiment on a five-point Likert scale, going from 'very easy' to 'very difficult' (Figure 4-5). The respondents in the online survey generally perceived the experiment as more difficult compared to the in-person participants. Within the survey, the online participants were able to explain their rating in an open comment box. These responses were analyzed using open coding analysis, defining different keywords within the answers (Strauss & Corbin, 1998). The open question box was not provided in the in-person survey due to time constraints.

Overall, respondents that considered the survey difficult and responded with low ratings referred to certain 'knowledge gaps', which prevented them from making informed decisions. These were related to the lack of knowledge they had on the materials, processing, end-of-life, or sustainability assessment itself. Also, they often acknowledged that visual appearances can be deceiving, and they felt uncomfortable judging the products based on visual impressions alone. This could indicate the awareness of respondents toward greenwashing in marketing. In addition, some of them mentioned the need to see, feel, or smell the boxes in-person. Respondents that considered the survey easy and responded with high ratings indicated that they made the choices between the packages based on their own judgements on the perceived feedstocks, the color of the materials, processing, and the recognition of obvious recovered waste content within the boxes. An overview of the coding analysis is provided in SI (Figure S6).

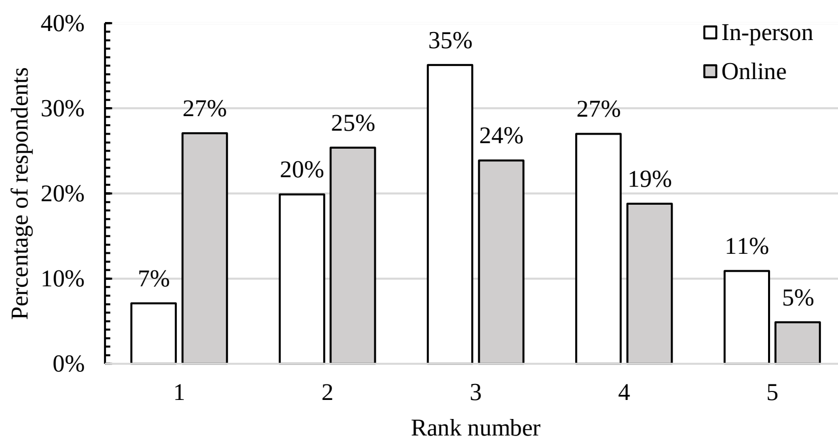


Figure 4-5. Rating the choice experiment from "1" = "very difficult" to "5" = "very easy".

4.4.3. Preferences related to respondents' characteristics

The sustainability rankings of packages, as shown in Figure 4-3 and Figure 4-4, might differ based on respondents' characteristics such as gender and age. To analyze these differences, the conditional logit model was estimated again for certain sub-groups in the sample (i.e., 'female' versus 'male', 'born in 1988 or after' versus 'born before 1988'). Table 4-6 shows the differences in rankings, based on the share of preferences, between females and males for both the online and the in-person survey. The packaging samples shaded in grey represent the ones that change ranking position when comparing both genders. For example, the BHW/Copy and RL/Cocoa packages switch ranking positions when calculating the preferences for females compared to males. However, these switches are caused by rather small differences between the preference results.

Table 4-7 shows the differences in rankings between the age groups. For the online sample, no large differences were noted between both age groups. For the in-person sample, a clear lower preference was given to RL/Control and a higher preference for BHW/grass in the age category 'born in 1988 or after'. This can be confirmed by the boxplots of the BW scores in SI (S7 in SI). However, it should be noted that the differences between the share of preferences for the age category 'born before 1988' in the in-person survey are relatively low.

Mann-Whitney-Wilcoxon tests were performed to verify if the differences between both gender and age categories were significant. The BW scores per respondent per package of both the online and in-person samples together were used as an indication of the individual's preference toward a certain package. No significant P values were found between individual preferences and gender. However, the null hypothesis (stating that the populations are the same) can be rejected when comparing both age categories for the RL/Control, RL/Pink, RL/Green, BHW/Grass, and BHW/Copy packages (S8 in SI). Additional boxplots can be consulted visualizing the BW scores for groups with different educational backgrounds (Figure S7 in SI). Significant differences were noted between these education-based groups for the RL/Control, RL/Grass, BHW/Control, BHW/Grass, and BHW/Copy packages (S8 in SI).

Table 4-6. Gender differences in preference results based on CL model (columns are sorted from highest to lowest share of preferences). Packaging samples shaded in grey represent the ones that change ranking position.

Online				**In-person**			
Female (n=286)		**Male (n=194)**		**Female (n=121)**		**Male (n=90)**	
RL/Copy	0.2726	RL/Copy	0.2722	RL/Grass	0.1654	RL/Grass	0.1793
RL/Coated	0.2063	RL/Coated	0.1858	RL/Cocoa	0.1467	BHW/Grass	0.1557
RL/Grass	0.1491	RL/Grass	0.1409	RL/Coated	0.1299	RL/Cocoa	0.1366
BHW/Grass	0.0990	BHW/Grass	0.1176	BHW/Grass	0.1195	RL/Coated	0.1107
BHW/Copy	0.0701	RL/Cocoa	0.0723	RL/Copy	0.1120	RL/Copy	0.1077
RL/Cocoa	0.0643	BHW/Copy	0.0718	BHW/Copy	0.0680	BHW/Copy	0.0680
BHW/RL	0.0435	BHW/RL	0.0440	RL/Green	0.0651	BHW/RL	0.0644
RL/Green	0.0386	RL/Green	0.0371	RL/Pink	0.0644	RL/Pink	0.0610
RL/Pink	0.0259	RL/Pink	0.0316	BHW/RL	0.0568	RL/Green	0.0516
RL/Control	0.0256	RL/Control	0.0217	RL/Control	0.0534	RL/Control	0.0467
BHW/Control	0.0050	BHW/Control	0.0049	BHW/Control	0.0188	BHW/Control	0.0183

Table 4-7. Age differences in preference results based on CL model (columns are sorted from highest to lowest share of preferences). Packaging samples shaded in grey represent the ones that change ranking position.

Online				**In-person**			
Born in 1988 or after		**Born before 1988**		**Born in 1988 or after**		**Born before 1988**	
RL/Copy	0.2700	RL/Copy	0.2787	BHW/Grass	0.2016	RL/Grass	0.1599
RL/Coated	0.1932	RL/Coated	0.2041	RL/Grass	0.1768	RL/Cocoa	0.1311
RL/Grass	0.1503	RL/Grass	0.1398	RL/Cocoa	0.1464	RL/Coated	0.1109
BHW/Grass	0.1151	BHW/Grass	0.0915	RL/Coated	0.1271	RL/Control	0.1044
BHW/Copy	0.0769	RL/Cocoa	0.0754	RL/Copy	0.1144	RL/Copy	0.0976
RL/Cocoa	0.0624	BHW/Copy	0.0622	BHW/Copy	0.0690	RL/Green	0.0857
BHW/RL	0.0428	BHW/RL	0.0450	BHW/RL	0.0567	RL/Pink	0.0837
RL/Green	0.0364	RL/Green	0.0407	RL/Pink	0.0411	BHW/Grass	0.0825
RL/Pink	0.0252	RL/Pink	0.0320	RL/Green	0.0365	BHW/Copy	0.0597
RL/Control	0.0228	RL/Control	0.0257	RL/Control	0.0211	BHW/RL	0.0541
BHW/Control	0.0049	BHW/Control	0.0049	BHW/Control	0.0092	BHW/Control	0.0303

4.4.4. Preferences related to packaging properties

Product characteristics, such as waste origin, base material, color, etc., might influence the online and in-person sustainability perception of the respondents. By visualizing and correlating the respondents' preference scores (i.e., the BW scores per respondent) with different paperboard properties, insights can be provided on the sustainability perception of product properties and the difference between online and in-person perceptions. Figure 4-6 shows four different boxplots, where the categorical variables "waste origin" ("control", "paper", and "agricultural") and "base material" ("BHW" and "RL") are mapped on the y-axis and the numeric variable "BW scores" on the x-axis. The boxplots of "waste origin" (Figure 4-6) show again a lower sustainability perception for the control boxes, and a higher preference for the boxes with obvious recovered waste content regardless of the format of the survey, i.e., online or in-person. For the online survey, it can be noted that there is almost no overlap between the BW scores of the control boxes and the boxes with the obvious recovered waste content. In addition, the correlations between the type of waste and the BW scores were quantified using Spearman's rho calculations. The control boxes were omitted from the dataset to create a direct comparison between obvious recovered paper and agricultural waste. The correlation coefficients indicate a weak, positive relationship between the use of agricultural waste and its sustainability perception. This correlation is relatively stronger for the in-person sample (i.e., a correlation of 0.307) compared to the online sample (i.e., a correlation of 0.105). Agricultural waste was composed of brown particles with an average particle size of 0.006 cm^2 while the paper waste was composed of bigger particles (c.a. 0.1 cm^2) of brighter colors such as white, blue, neon green, and neon pink (Table 4-8). Previous researched showed that consumers associate dull colors, especially brown and green, with sustainability (Herbes et al., 2020; Magnier & Crié, 2015), thus it could be inferred that the color of the agricultural waste could have influenced consumers' preferences toward the boxes containing this type of waste. Paper waste, on the other hand, with brighter colors could have been perceived as more synthetic and therefore perceived as less environmentally friendly.

The boxplots on "base material" (Figure 4-6) shows a slightly lower sustainability perception for the BHW based boxes compared to the RL based boxes. A comparison between the online and in-person survey samples shows almost no difference between the presentation formats, except for a neglectable difference in the 95% confidence interval around the median (represented by the notch). The correlations between the base material and the BW scores were quantified using Spearman's rho calculations. Both correlation coefficients indicate a weak, positive relationship between the brown RL based packaging and its environmental friendliness, with correlations of 0.208 for the online sample and 0.187 for the offline sample, with no remarkable difference between the presentation formats. However, the preference for RL based boxes becomes more apparent when a direct comparison between RL and BHW boxes containing the same waste was performed (Figure 4-7). For the control boxes, as well as the boxes containing switchgrass, and copy paper, RL was always preferred over BHW base material. Other academic studies, that focused on neat paper-based packaging with no obvious waste content appearance, have shown that eco-consciousness people prefer brown paper-based packaging over other materials (Liem et al., 2022), with this parameter being a decisive factor in their purchasing decision (Medinskaia, 2020). Brown (unbleached) paper could be perceived as more natural, less processed, and therefore more environmentally friendly which would explain the preferences of RL paperboards over the BHW ones.

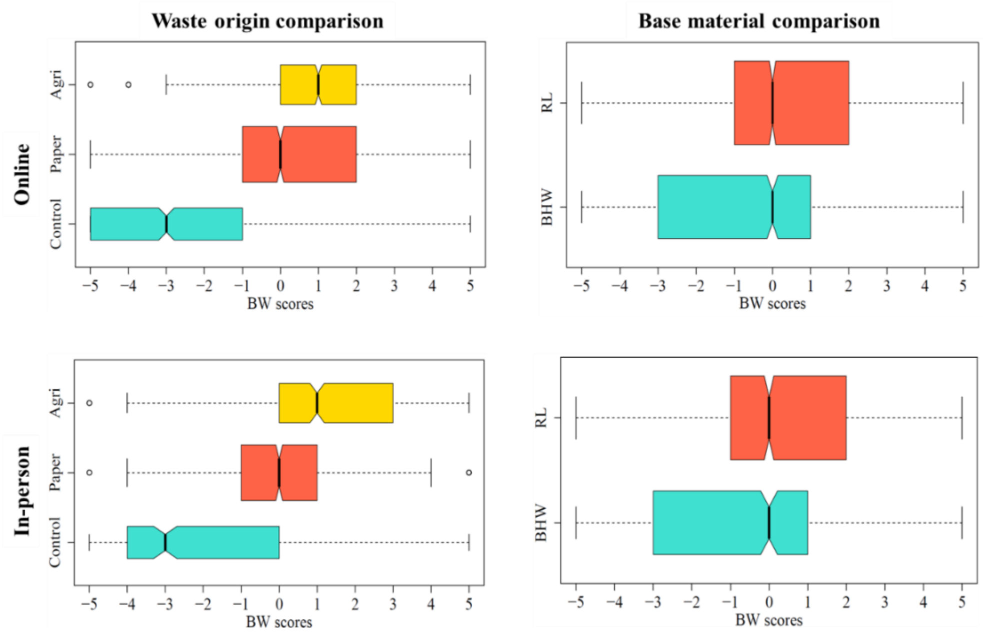


Figure 4-6. BW (best-worst) scores for both waste origin (“control”, “paper”, and “agricultural”) and base material (“BHW” and “RL”), with “○” indicating the outliers, the thin vertical lines indicating the maximum and minimum values (without outliers), the colored rectangles indicating the values of the upper quartile and the lower quartile, and the thick vertical line in the colored rectangles indicating the median. The 95% confidence interval around the median is represented by the notch in the colored rectangle.

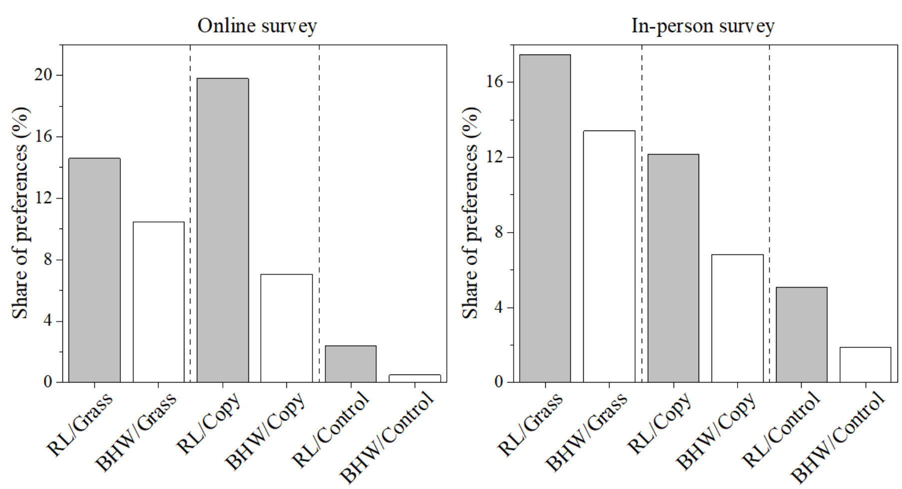


Figure 4-7. Comparison between packaging’ base materials (RL and BHW) having the same type of waste.

Table 4-8. Paper and agricultural waste characteristics.

Waste origin	Waste	Particle Size (cm^2)	Color
Paper	Copy paper	0.103 ± 0.003	65% white, 35% black
	Coated paper	0.109 ± 0.001	Multicolored (blue (59%), pink (17%), green (11%), and yellow (13%))
	Neon green paper	0.117 ± 0.023	Green
	Neon pink paper	0.119 ± 0.026	Pink
Agricultural	Switchgrass	0.006 ± 0.001	Light brown
	Cocoa bean shells	0.006 ± 0.001	Dark brown

Apart from the waste origin and the base material, other paperboard properties could have influenced consumers' preferences between the different boxes and the presentation formats. Figure 4-8 shows the thickness, bulk, roughness, and bending stiffness of the different paperboard samples, and Figure 4-9 shows the correlation between the respondents' preferences (based on the CL model) and the mentioned paperboard properties. For the in-person survey (Figure 4-9b, d, and f) there is a positive (Pearson) correlation between the share of preferences and the thickness, bulk, and roughness of the paperboard specimens, showing r values of 0.69, 0.72, and 0.91, respectively. Thicker, bulkier and especially rougher waste-containing paperboards, i.e., RL/Cocoa, RL/Grass, and BHW/Grass, were perceived as the most environmentally sustainable samples. Roughness had the strongest correlation of the aforementioned properties, and could have potentially influenced respondents' decisions.

Although respondents were not allowed to touch the samples, differences in the texture between the in-person samples were visually noticeable as can be supported by the surface topography images depicted in Figure S9 in SI. The topographic maps of the paperboards containing agricultural waste showed the most heterogeneous and roughest surfaces with non-uniform and prominent peaks and valley distributions. However, the question remains to which degree the consumers could have identified the differences in roughness without physical interaction with the samples. Also, a negative and strong correlation ($r = -0.71$) between the bending stiffness and the sustainable appearance of the boxes was observed (Figure 4-9h). Boxes showing a stiffer appearance were less preferred by the respondents. However, without physical interaction with the samples, bending stiffness might have been difficult to identify by the survey respondents. Prior to calculating the Pearson correlation coefficient, a normal distribution of the variables was verified through the Goodness-of-fit test using the Shapiro-Wilk method. Although the results from the test confirmed that the variables followed a normal distribution, the Pearson correlation coefficients calculated should be interpreted with discretion since the size of the sample was small (n=11) and both statistical tests (i.e., goodness-of-fit and Pearson correlation) are sensitive to the sample size. Results from the online survey showed a weak relationship between respondents' preferences and the physical properties of the paperboards, reporting r values of 0.27, 0.26, 0.48, and 0.43 for thickness, bulk, roughness, and bending stiffness, respectively (Figure 4-9a, c, e, and g). The r values for the online survey were all lower than the corresponding in-person values. The differences between the correlation of the paperboard properties with the sustainable perception of the boxes in the online and in-person surveys suggest that through the images shown in the online survey, consumers could not have perceived all the physical characteristics of the boxes apart from the color of the base/waste materials. Thus, it is hypothesized that the impediment to appreciate paperboard properties could complicate the recognition of the waste, with a more prominent effect on the agricultural waste. For instance, particles of cocoa bean shells in images during the online survey might have been perceived as dark spots on the surface of the material due to the impossibility to recognize the roughness and thickness of the particles. On the other hand, clues such as pieces of waste containing letters or bar codes could have helped the respondents to identify the copy and coated paper as recycled materials. Therefore, the identification of the feedstock could be an important parameter used by the respondents to judge the samples.

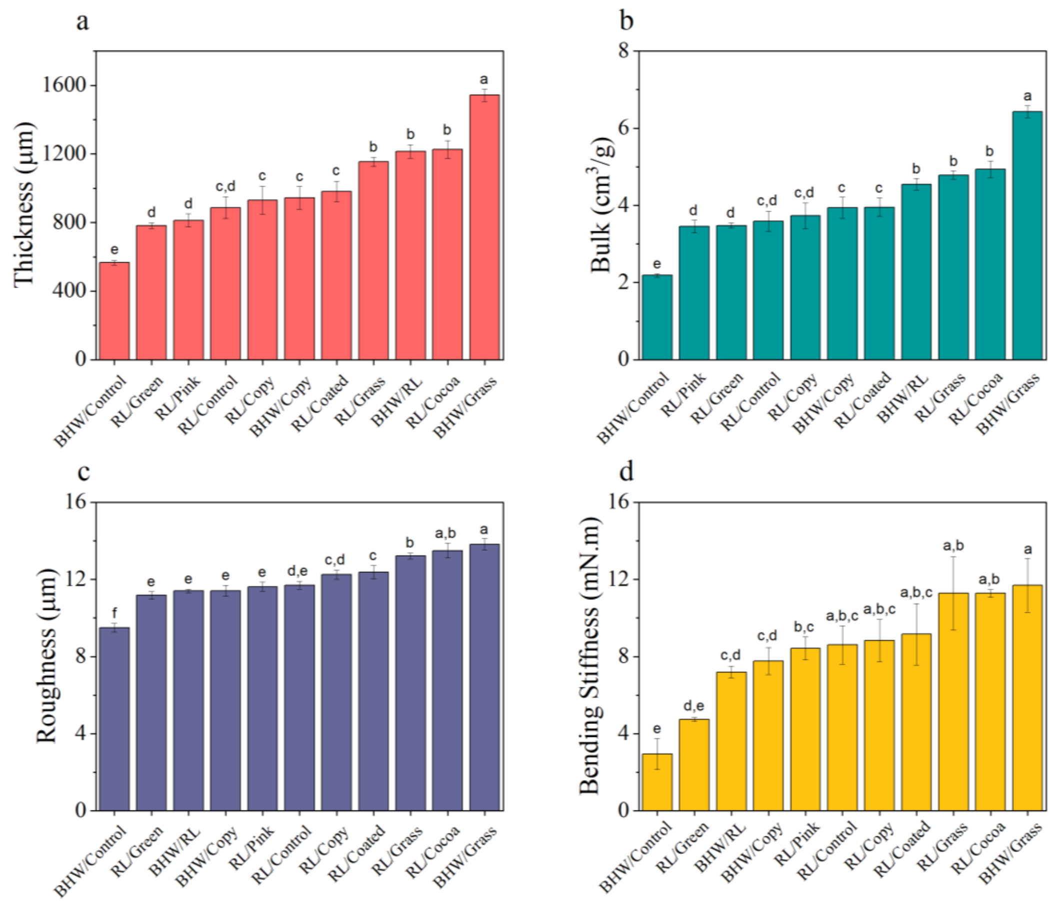


Figure 4-8. Properties of waste-containing paperboards. (a) Thickness, (b) bulk, (c) bending stiffness, and (d) roughness. Different letters at the top of the bars indicates significant differences between the means ($p < 0.05$).

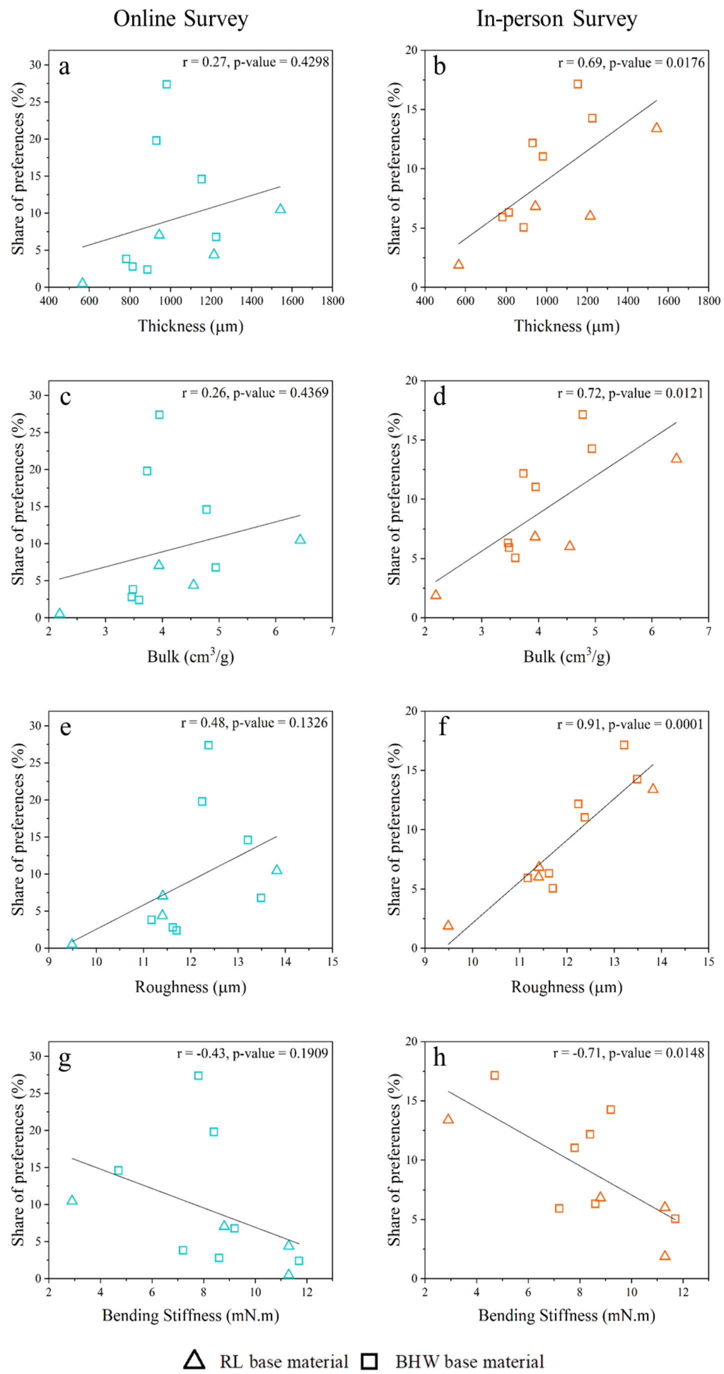


Figure 4-9. Correlations between share of preferences and paperboard physical properties for online and in person surveys. (a) and (b) thickness, (c) and (d) bulk, (e) and (f) roughness, (g) and (h) bending stiffness. The r values represents the Pearson correlation coefficients.

4.4.5. Study limitations

The BWS experiments performed in this study enabled the study to address the two main research questions on (1) the relative sustainability perception of visually obvious recovered waste content, and (2) the comparison between online and in-person survey presentation formats. However, BWS experiments only provide a relative measurement, not an absolute one. This means that it can be concluded that paper-based packaging with obvious recovered waste content was perceived as more environmentally friendly than packaging with a clean appearance, but additional information would be needed to anchor the relative scale and gather information on the absolute sustainability perception (Mueller Loose & Lockshin, 2013). In addition, the only criterion that was assessed within this study was the ‘environmental friendliness’. Future research can study additional decision criteria that can influence final consumer preferences, such as ‘perceived toxicity’ or ‘price’.

Given the aim of this study to compare multiple presentation formats (i.e., online and in-person), the geographical spread of the sample was limited to the region of Raleigh, North Carolina. Further research would make it possible to investigate different geographical regions, and focus on, for example, environmental sustainability preferences in a rural context. Also, the in-person surveys were conducted in three different outdoor locations and, as a consequence, the packaging boxes could be affected by conditions such as light intensity, shadow fall, etc. Besides that, the differences between natural lighting (in the in-person survey) and digital imaging (in the online survey) might have partially distorted the perception of color, shape, and size of the particles in the packaging. However, the results herein should give researchers additional incentive to properly select a presentation format which fits the research question, as this might have a significant influence on the study results.

Future research should investigate how the packaging perception would change if different product categories are chosen for the BWS experiment. In the present study, the product that would be inside the packaging box was not specified. Further research can compare, for instance, food and non-food products, and examine if the packaging sustainability perception would change based on what is held in the packaging container. Moreover, other value chain actors such as manufacturers, retailers etc. could be involved to further shape the acceptability and technical feasibility of paper-based packaging containing visually recovered waste.

4.5. Conclusion

This study aims to understand consumers' environmental sustainability perception toward redesigned packaging that display visually obvious recovered waste content, and the impact of the study presentation format (online versus in-person) on this packaging perception. A best-worst scaling (BWS) experiment was designed, in which a total, 698 respondents participated (i.e., 487 online responses and 211 in-person responses).

The BWS experiment showed that paper-based packaging with obvious recovered waste content, coming from paper or agricultural waste, was perceived as more environmentally friendly than packaging with a clean appearance (i.e., control samples with no waste). Particles of waste on the surface of the substrates acted as clues of sustainability and guided respondents to make certain judgements on the environmental sustainability of the boxes. From the two base materials used (brown versus white pulp) and the types of waste (paper versus agricultural waste), brown pulp and agricultural waste were perceived as being more environmentally friendly. In addition, it was found that the environmental sustainability perception of certain packages differed between age groups and respondents with a different educational background. Finally, the presentation format (online versus in-person) significantly influenced the choices made by the respondents.

Digital photographic images compared to a direct in-person presentation of the real boxes can change respondents' packaging perception, and should therefore be taken into account when designing surveys. Depending on the goal and resources of a research study, one should opt for a suitable presentation format.

The previous chapters were focused on evaluating the performance and the consumers' sustainability perception toward redesigned packaging for shipping applications. However, considering the packaging's outstanding properties, *i.e.*, high environmental perception and good mechanical performance, **why not broaden the market of redesigned recycled containers to food packaging?**

The last chapter (Chapter 5) evaluates the further exploitation and enhancement of recycled paperboard for food packaging applications by incorporating multiple layers of two bio-based coatings (cottonseed oil, and CMF-reinforced starch). Chapter 5 shows a fundamental study of the performance of the coatings with the **long-term goal of utilizing them in packaging with redesigned appearance** to extent its application to food packaging.

Chapter 5 . Enhancing the liquid and oil & grease barrier properties of recycled paperboards with epoxidized cottonseed oil and cellulose microfibrils (CMF)-reinforced starch

5.1. Abstract

Environmental and health concerns regarding the utilization of per- and poly-fluoroalkyl substances (PFAS) and petroleum-based materials in fiber-based food packaging have pushed the packaging industry to look for more sustainable options. Herein, a bio-based barrier coating for food packaging applications using a multi-component coating containing epoxidized cottonseed oil cross-linked with citric acid (CEPO) and cellulose microfibrils reinforced starch (S-CMF) was developed. The water, oil & grease absorption, and water vapor diffusion of the coated paperboards decreased significantly in the presence of the multi-component coating. The incorporation of epoxidized cottonseed oil provided the water repellency behavior owing to the non-polar long alkyl chains in its chemical structure and the less intermolecular space created due to the cross-linking reaction. The oleophobic nature of the S-CMF coating and the less porous paper surface developed by the presence of the coating opposed the penetration of oil and grease, endowing the system with an oil repellency. Thus, both bio-based coatings (CEPO and S-CMF) with opposite behaviors could be utilized within the same substrate to amalgamate their barrier properties as a sustainable material for water, water vapor, and grease & oil resistant food packaging.

5.2. Introduction

For many decades per- and poly-fluoroalkyl substances (PFAS), primarily used as a barrier coating for greaseproof products, were considered harmless and used extensively in food packaging and many other industrial applications. Nevertheless, after years of research, it was found that PFAS, especially long-chain PFAS, have adverse effects on health and the environment (Hubbe & Pruszynski, 2020). C-F bonds in PFAS are strong and stable and have the ability to migrate and accumulate in a living organism (Buck et al., 2011; Glenn et al., 2021), originating several health issues such as thyroid disease, increase in cholesterol levels, liver damage, kidney cancer, testicular cancer, and problems with the development of a fetus in a pregnant woman, among others (Fenton et al., 2021). The leading manufacturers of PFAS, thus, voluntarily stopped the production of long-chain PFAS by 2002, and some regulations were established in countries such as the U.S. to ban their production by 2015. Therefore the development of PFAS-free systems has gained increased global attention in the last decade.

Different materials have been studied or are under consideration to effectively substitute PFAS in food packaging. Among them, petroleum-based polymers such as polyethylene (PE), poly(ethylene-co-vinyl alcohol) (EVOH), and polyethylene terephthalate (PET) have been widely used (Glenn et al., 2021). A thin film of petroleum-based plastics laminated onto a fiber-based substrate can endow the substrate with outstanding characteristics, such as good thermomechanical and barrier properties, flexibility, toughness, lightweight, and low cost (Glenn et al., 2021; Silva et al., 2020). However, the major drawback of synthetic polymers is that most of them are not biodegradable, and their recovery and recycling rates are still low, even in countries with developed waste management systems (Shen & Worrel, 2014). As a result, synthetic polymers accumulate in landfills generating severe health and environmental issues (Moore, 2008; Proshad et al., 2017). Utilizing bio-based polymers, therefore, has become a more sustainable option than the current plastics used in food packaging.

Bio-based polymers offer several environmental advantages over synthetic options, such as biodegradability, recyclability, compostability, and renewability (Tyagi et al., 2021). A wide range of polysaccharides has been investigated to be used as biopolymers for food packaging, such as starch, cellulose, chitosan, and alginates. Still, starch and cellulose are the most studied among them (García-Guzmán et al., 2022). Starch is a low-cost and fully biodegradable biopolymer referred to as "the future green polymer" to replace PFAS and synthetic polymers in food packaging (Bangar & Whiteside, 2021; Vaezi et al., 2019). Starch is broadly available and non-toxic, which can be used to produce transparent coatings and films with good oil and grease barrier properties under low and ambient moisture conditions (Glenn et al., 2021). However, starch tends to swell easily under high moisture conditions (>75% RH) (Othman et al., 2019) due to the presence of abundant hydroxyl groups, which in turn negatively affect its oil & grease barrier properties. Starch also shows poor processability and low thermal and mechanical properties. These disadvantages have limited its widespread utilization in food packaging applications (Maniglia et al., 2021).

As a convenient option from an economic and environmental point of view, starch is under continuous research and has shown promising results when combined with other materials. For instance, the addition of nanofillers (particles with at least one dimension at the nanometer scale) has been a strategy recently evaluated to overcome the disadvantages of starch. Owing to their high aspect ratio and large surface area, nanofillers act as reinforcement agents that produce nanocomposites with superior mechanical and barrier properties compared to the neat biopolymer. Starch matrices reinforced with cellulose nanocrystals (CNCs) and cellulose microfibrils (CMFs) have shown lower oil permeability, oxygen transmission rate, and higher tensile strength compared to the neat starch biopolymer (Fazeli et al., 2018; M. Li et al., 2018; Vaezi et al., 2019). The water absorption and water vapor permeability of reinforced starch films with up to 50 wt% of CMFs have also been reported to decrease; however, the values obtained were still too high (*e.g.*, Cobb values 90 % higher than PVA films) to compete with commercial petroleum-based films used for food packaging applications (Carvalho do Lago et al., 2021).

Hydrophobization could be addressed by adding another biopolymer layer with less hydrophilic behavior to the system (Cherpinski et al., 2018; Pasquier et al., 2022) to amalgamate the exceptional properties of the different materials and create a product with outstanding performance (Anukiruthika et al., 2020). Epoxidized vegetable oil is a material that has gained increasing attention in the last decade due to its hydrophobic nature and biodegradable properties (Cai et al., 2018). For instance, several investigations have reported the advantages of using epoxidized soybean oil in films and paper coating (Meng et al., 2022; Miao et al., 2015; Tian et al., 2022) as a barrier layer to water and water vapor. Epoxidized cottonseed oil could be another potential candidate for water-resistant materials. Cottonseed oil is a byproduct of cotton processing, with a global production of 1.83 million metric tons in 2021 (Shahbandeh, 2022). This highly available oil possesses multiple unsaturated bonds (Iodine value of 107 g I_2/100 g) available to be epoxidized, similar to the soybean oil (Iodine value of 127.6 g I_2/100 g) (Dominguez-Candela et al., 2022). The high reactivity of the epoxy groups allows reactions with other chemicals to modify the functionalities of the oil, such as improving the interfacial adhesion with other materials (Yang et al., 2021).

In the last years, epoxidized cottonseed oil has mainly been proposed as a bio-based plasticizer to enhance the ductile properties of other polymeric materials (Carbonell-Verdu et al., 2020; Narute et al., 2015); however, it could also be further exploited for paper-based packaging applications because it could endow the substrates with water and water vapor resistance.

In this research, the utilization of two bio-based coatings, with opposite properties, in recycled paperboards to develop packaging with both water and grease & oil barrier properties was investigated. A hydrophobic coating composed of cross-linked epoxidized cottonseed oil with citric acid (CEPO) and a more hydrophilic coating containing CMF-reinforced starch (S-CMF) were evaluated. Using both bio-based coatings with different properties (*e.g.,* hydrophilicity vs. hydrophobicity) was conjectured to amalgamate their barrier properties, enhancing the final barrier properties of the packaging (without compromising their individual contributions). This study proposes a novel application for cottonseed oil and a sustainable option to replace the current PFAS and polymeric materials with renewable and biodegradable materials.

5.3. Experimental Section

5.3.1. Materials

Kraft paperboard (230 g/m^2) made of recycled content was purchased from Goefun, US. Pregelatinized wheat starch was provided by ADM (Decatur, Illinois). CMFs were produced from bleached hardwood kraft market pulp with a moisture content of ca. 5 wt%. Ethanol (95%) was purchased from Sigma-Aldrich (US), and citric acid anhydrous and calcium chloride anhydrous from Fisher Scientific (US). Commercially available 100% pure corn oil (Mazola, TN, US) was obtained from a local store (Target, US). Benchmark paper-based packaging used for fast-food applications was obtained from a local food chain (Bojangles, US) to compare the performance of the designed coated paperboards.

Epoxidized cottonseed oil (EPO) with an iodine value of 110.8 mg I_2/g, an oxirane content of 5.75%, and an epoxy equivalent weight of 278 g/eq was synthesized as described by Wijayapala et al. (2019). A complete description of the epoxidation process and characterization of the epoxidized cottonseed oil is available in the supplementary information (SI).

5.3.2. Preparation of cellulose microfibrils (CMFs)

Bleached hardwood fibers were suspended in tap water at a solids content of 10 wt% and refined at 5000 revolutions using a PFI mill refiner (the Norwegian Pulp and Paper Research Institute, Oslo, Norway) following the TAPPI 248 Sp-00, (2000) standard. The suspension was then diluted to 2.5 wt% and passed through a supermasscolloider grinder (MKZA6-5, Masuko Sangyo Co., Ltd, Saitama, Japan) for CMFs production at a constant rotating speed of 1200 rpm to targeted specific energy consumption of 0.5 kWh/kg of oven-dried pulp (Trovagunta et al., 2021). The grinder was equipped with silicon carbide stones (MKW 10-46#). The apparent aspect ratio (length to width ratio) of the CMFs produced was estimated to be 148 ± 8.

5.3.3. Coating of paperboard sheets

An 8 wt % solid content suspension was prepared using either starch (S) or CMF-reinforced starch (S-CMF). A suitable amount of starch, or starch (80 wt%) and CMFs (20wt%) was dispersed in deionized water and homogenized using a high-shear homogenizer (Ultra Turrax, IKA® T-25, Germany) at 7000 rpm for 3 min.

CEPO coating was formulated by cross-linking the epoxidized cottonseed oil with citric acid (CA), as described by Yang et al., (2021). In brief, the citric acid was dissolved in distilled water at a weight ratio of 3:1 at 90 °C. After complete dissolution, the CA was blended with the epoxidized oil (EPO) at a weight ratio of 1:5 in a 50 ml round flask. The mixture was constantly stirred, and the reaction was conducted at 90 °C for 30 min. Finally, the resulting mix of CA-EPO (CEPO) was dissolved in ethanol (40 ml of ethanol for each 3 g of oil used) and stirred with a magnetic stir bar for 3 hrs.

Different sets of coated paperboards were produced using a single-component coating (paperboards coated with S, S-CMF, or CEPO) or multi-component coating using both CEPO and S-CMF as described in Figure 5-1, Table 5-1, and Table 5-2. S and S-CMF coatings were applied onto the paperboards using a lab rod coater with a Mayer rod number 16. CEPO coating was applied using a micrometer film applicator (Microm II, Gardco®, Japan) with a blade gap of 200 µm. The paperboards were coated with 1, 2, 5, and 10 successive layers of either S or S-CMF and 1 and 2 layers of CEPO (Table 5-1 and Table 5-2); each layer was allowed to dry using compressed air flow (approximately for one minute) before applying the next one. CEPO-coated boards were cured at 105 °C for 2 h (Yang et al., 2021) in a speed dryer (Emerson, Model 140) with the coated side down on the heated metal surface; the other samples (without CEPO) were dried at 105 °C for 3 min. No deposition was observed on heated metal surface from the dried coatings.

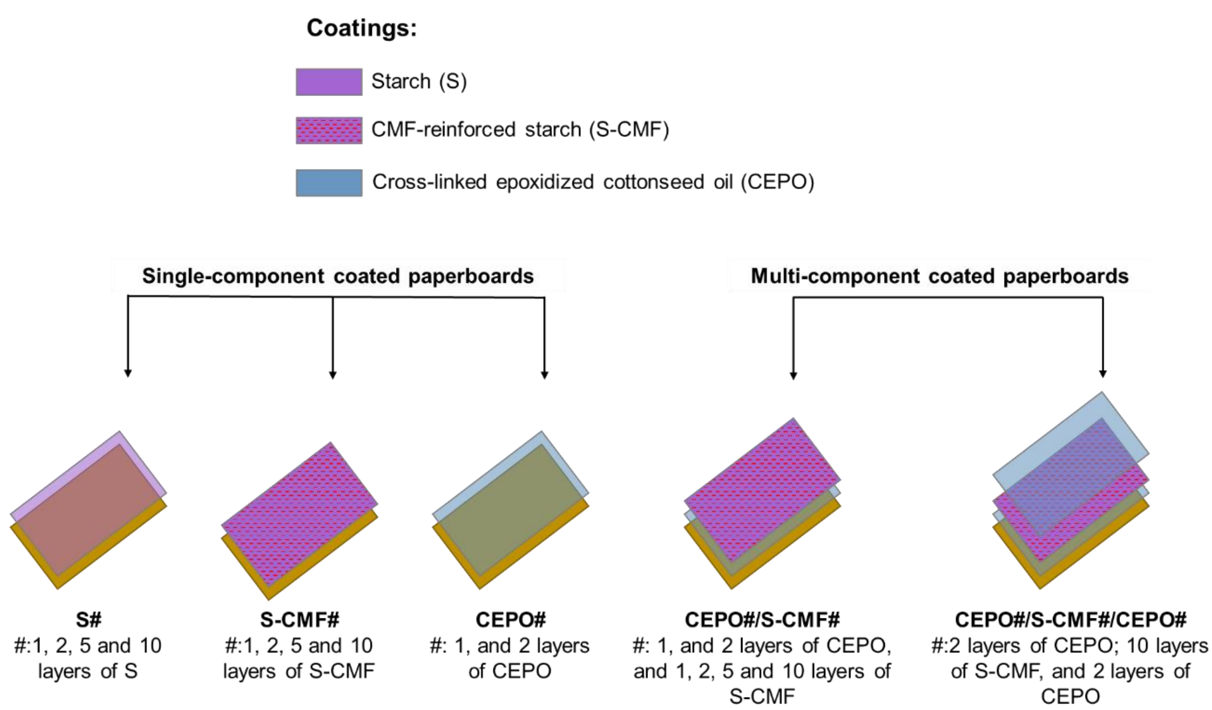


Figure 5-1. Schematic representation of single- and multi-component coated paperboards.

Table 5-1. Single-component coating composition

Sample ID	Coating composition	Layers
S1	100% starch	1
S2		2
S5		5
S10		10
S-CMF1	80% starch, 20% CMFs	1
S-CMF2		2
S-CMF5		5
S-CMF10		10
CEPO1	100% CEPO	1
CEPO2		2

S: starch, S-CMF: CMF-reinforced starch, CEPO: epoxidized cottonseed oil cross-linked with citric acid.

Table 5-2. Coating composition in multi-component coated paperboards.

Sample ID	Layers
CEPO2/S-CMF1	2 layers CEPO, 1 layer CMF-reinforced starch
CEPO2/S-CMF2	2 layers CEPO, 2 layers CMF-reinforced starch
CEPO2/S-CMF5	2 layers CEPO, 5 layers CMF-reinforced starch
CEPO2/S-CMF10	2 layers CEPO, 10 layers CMF-reinforced starch
CEPO2/S-CMF10/CEPO2	2 layers CEPO, 10 layers CMF-reinforced starch, 2 layers CEPO

S: starch, S-CMF: CMF-reinforced starch, CEPO: epoxidized cottonseed oil cross-linked with citric acid.

Single-component coated paperboards were denoted as S, S-CMF, and CEPO for the starch, CMF-reinforced starch, and citric acid-epoxidized oil coatings, respectively (Table 5-1 and Figure 5-1). Multi-component coated paperboards were denoted as CEPO/S-CMF or CEPO/S-CMF/CEPO (Table 5-2 and Figure 5-1). The number next to each sample ID indicates the number of layers applied of each coating. The coated paperboards were conditioned at 23 °C and 50% RH for at least 24 h prior to testing.

5.3.4. Characterization of the coated paperboards

The grammage of the coated paperboards and respective controls was determined by measuring the average mass per unit area of at least five conditioned paperboards cut into squares of 10 × 10 cm^2. The coat weight was calculated by the difference between the grammage of the coated and uncoated paperboards. The thickness was measured using a micrometer (Lorentzen and Wettre, Sweden) and expressed as the average of at least five measurements.

Images of the surface of the samples were taken using a benchtop scanning electron microscope (SEM) (JCM-6000PLUS, JEOL, Tokyo). The paperboards were sputtered with a thin layer (8 nm) of gold prior to imaging and were analyzed at an accelerating voltage of 5.0 kV. An attenuated total reflectance-Fourier transform infrared spectrometer (ATR-FTIR) (Frontier, PerkinElmer, Massachusetts, USA) was used to get the spectra of the samples and identify the functional groups responsible for molecular interactions between the components of the layers. The data was collected in the spectral range from 4000 cm^{-1} to 650 cm^{-1} with a resolution of 4 cm^{-1} for 10 scans. The tensile strength of coated paperboards was evaluated with a tensile tester TMI 84–56 (Testing Machines, Inc., DE, US) using an initial gap between the clamps of 80 mm (TAPPI 494 om-96, 1996). At least five measurements were performed for each sample.

The water absorptiveness of coated paperboards was determined following the 1-min Cobb test according to the TAPPI 441 om-98 (1998) but using a smaller test area (16 cm^2) and 10 ml of deionized water. The wettability was assessed by water contact angle (WCA) using 7 µL water drops (SEO Phoenix 300 analyzer, Suwon city, Korea). The WCA was recorded immediately after the first contact of the probe liquid with the surface of the paperboards. The water vapor transmission rate (WVTR) was determined according to ASTM E96/E96M-16 using ca. 5 g of $CaCl_2$ as a desiccant. The weight gain of the paperboards was monitored every hour for 8 hrs, and an average of three measurements per sample was reported.

Oil penetration was measured following the Cobb test for water absorptiveness, replacing the water with vegetable oil. Triplicates were performed for each sample. Surface repellency to oil and grease was assessed according to the Kit test (TAPPI 559 cm-02, 2002).

5.4. Results and Discussion

5.4.1. Single-component coated paperboards performance

The properties of single-component coated paperboards (*i.e.,* coated only with S, S-CMF, or CEPO) were first evaluated to understand their interactions with the fiber-based substrates and their contribution to the barrier properties.

Paperboards coated with a single layer of starch showed a coating weight of about 7 g/m^2 (Table 5-3) and a non-uniform coating layer with several pinholes visible on the surface of the paperboards (Figure 5-2b). Several layers of starch were applied (2, 5, and 10 layers) to ensure a uniform and complete coverage of the surface of the paperboard. From 5 layers and onwards, the starch coating completely covered the surface, and no pinholes were observed on the surface of the substrates (Figure 5-2d and e). However, fractures occurred in the coating, as evidenced by the presence of cracks in the SEM images (Figure 5-2d and e). Starch has poor mechanical properties (Kumari et al., 2022; Maniglia et al., 2021), and its brittle nature became more evident when increasing the weight and thickness of the coating, which affected the integrity of the coating. This was also confirmed by an observed decrease in the tensile strength of the coated paperboards with an increasing number of layers (Figure 5-3).

To overcome the shortcomings of starch, the biopolymer matrix was reinforced with 20 wt% of CMFs prior to coating onto the paperboards as reported in other studies in which films of starch reinforced with nanocellulose were studied (Carvalho do Lago et al., 2021; M. Li et al., 2018). The results of the CMF-reinforced starch coating are shown in Figure 5-2f and g. The incorporation of CMFs led to the formation of a more uniform coated layer with no pinholes or cracking observed on the surface of the paperboards by SEM. CMFs may have acted as nanofillers that, by filling the voids of the starch matrix and forming strong hydrogen bonds, improved the performance of the starch (Figure 5-3) (Freitas et al., 2021; Kumari et al., 2022; Maniglia et al., 2021). SEM images of the complete set of S-CMF coated paperboards can be found in Figure S3.

Table 5-3. Physical properties of single- and multi-component coated paperboards.

Sample ID	**Thickness (μm)**	**Coating weight (g/m²)**
Control	320 ± 1	-
Ref.	350 ± 3	-
Single-component coated paperboards		
S1	328 ± 7	7.3 ± 0.4
S2	328 ± 6	8.2 ± 1.3
S5	339 ± 6	14.6 ± 3.0
S10	343 ± 2	29.0 ± 0.1
S-CMF1	325 ± 3	3.6 ± 2.1
S-CMF2	333 ± 3	6.2 ± 3.5
S-CMF5	335 ± 3	12.1 ± 1.8
S-CMF10	346 ± 4	24.8 ± 1.1
CEPO1	319 ± 7	1.1 ± 0.1
CEPO2	321 ± 7	2.1 ± 0.2
Multi-component coated paperboards		
CEPO2/S-CMF1	321 ± 6	3.8 ± 1.8
CEPO2/S-CMF2	332 ± 4	8.7 ± 1.6
CEPO2/S-CMF5	330 ± 6	10.5 ± 1.8
CEPO2/S-CMF10	342 ± 6	21.5 ± 0.9
CCEPO2/S-CMF10/CEPO2	347 ± 8	27.3 ± 3.1

Ref.: commercial coated paper-based packaging.

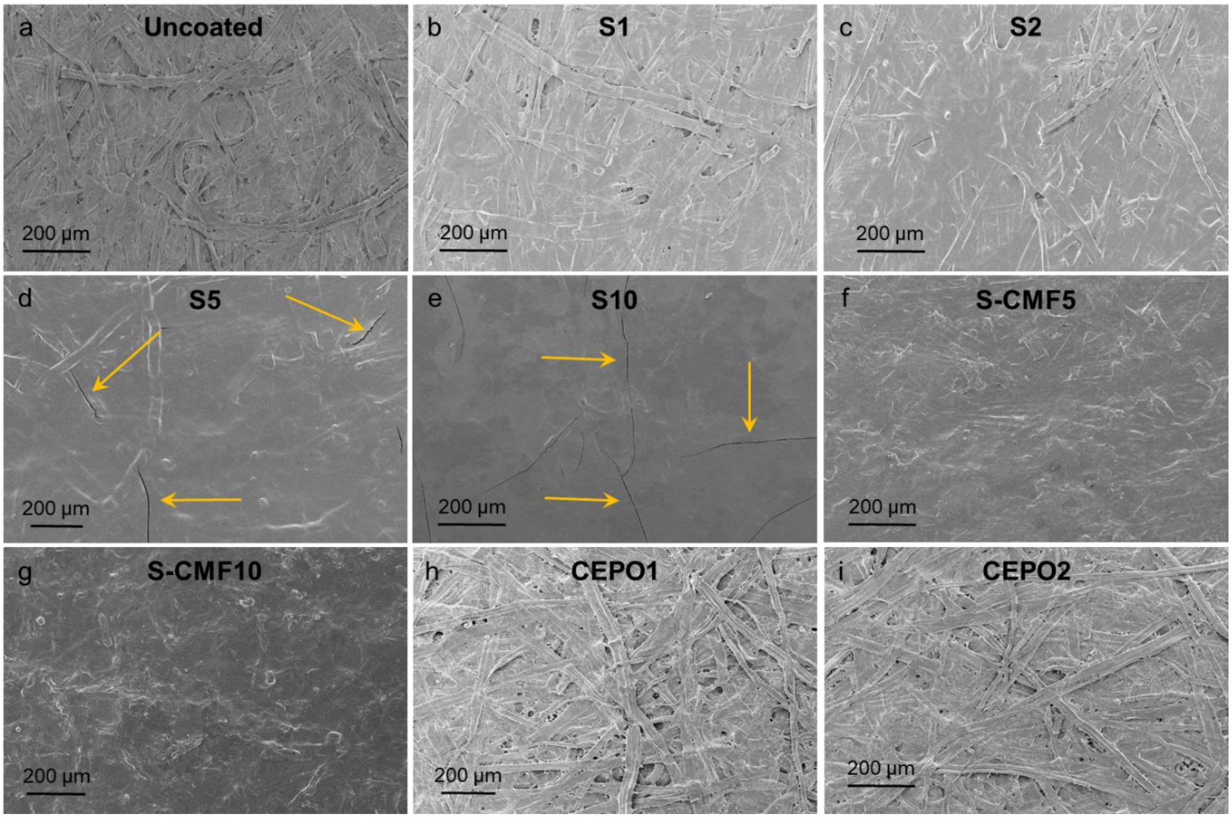


Figure 5-2. Scanning Electron Microscopy (SEM) images of the surface of uncoated and single-component coated paperboards. (a) Uncoated paperboard, (b) 1 layer (S1), (c) 2 layers (S2), (d) 5 layers (S5), and (e) 10 layers (S10) of starch; (f) 5 layers (S-CMF5) and (g) 10 layers (S-CMF10) of CMF-reinforced starch, and (h) 1 layer (CEPO1) and (i) 2 layers (CEPO2) of citric acid-epoxidized cottonseed oil. The number next to the sample ID refers to the number of layers of the coating. Arrows indicate cracks in the starch coating after drying.

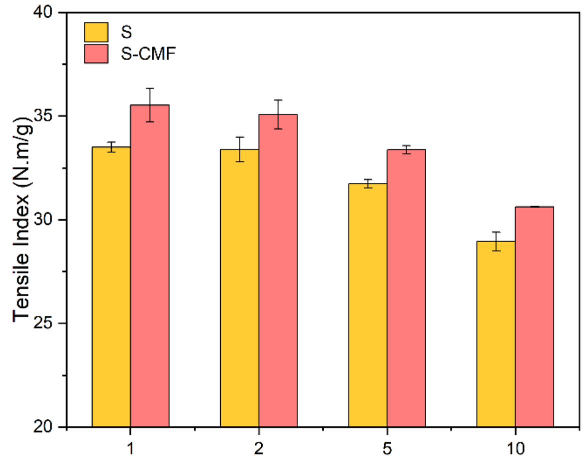


Figure 5-3. Tensile index of paperboards coated with 1 (S1), 2 (S2), 5 (S5), and 10 (S10) layers of starch.

Figure 5-4 depicts the barrier properties of the S-CMF coated paperboards. The water uptake of the samples, measured by the Cobb test, decreased with the addition of 5 layers of S-CMF coating when compared to the uncoated board (Figure 5-4a). A continuous layer of a starch coating reinforced with CMFs reduced the porosity of the samples (Figure 5-2f and g), restricting the passage of water (M. Li et al., 2018). Moreover, the strong inter-fiber network created between the starch and CMFs enhanced the cohesiveness of the matrix, reducing the interaction between the water and the coated samples (M. Li et al., 2018; Vaezi et al., 2019). Yet, the Cobb values reported for 1, 2, and 5 layers of S-CMF coated paperboards were not comparable with those reported for the commercial paper-based packaging currently used in the food industry (denoted as “Ref.” in Figure 5-4).

The water contact angle (WCA) values of the uncoated and coated paperboards with S-CMF are shown in Figure 5-4a. The WCA values decreased with the increasing number of S-CMF layers. 10 layers of S-CMF coated onto paperboards decreased the WCA by 46 wt%. The OH groups present in the starch and CMFs coating could increase the surface energy of the paperboards leading to a more hydrophilic and, in turn, wettable paper surface (Tyagi et al., 2019).

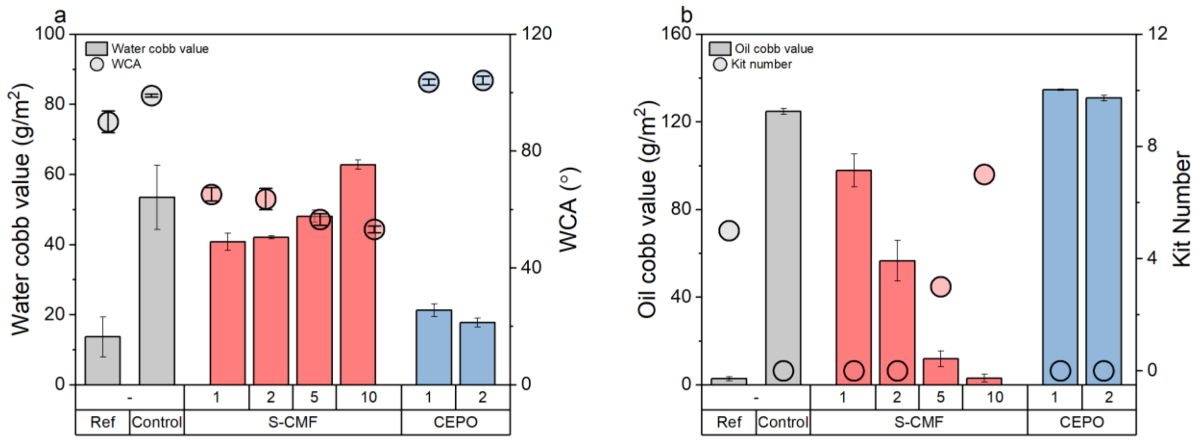


Figure 5-4. Water, oil & grease resistance of uncoated (control) and single-component coated paperboards (coated with either CMF-reinforced starch (S-CMF) or citric acid-epoxidized cottonseed oil (CEPO)). (a) Water Cobb value and contact angle (WCA), and (b) oil Cobb value and Kit number. *Ref.: commercial coated paper-based food packaging.*

Oil Cobb values and Kit number of coated paperboards with different layers of CMF-reinforced starch are shown in Figure 5-4b. A significant reduction in oil permeability (98%) can be observed in a 10 layers configuration, which is comparable to the commercial reference packaging. The Kit number increased from 0 (no resistance toward oil) to 7, which exceeds the Kit number of the reference sample (Kit number 5). The oleophobic nature of the S-CMF coating, conferred by the presence of abundant hydroxyl groups (Kansal et al., 2020), and the closed and less porous structure (Lavoine et al., 2014) promoted by the presence of the coating on the surface of the paperboards, render the samples with and oil and grease resistance behavior which is a desired property in food packaging applications.

The water vapor transmission rate (WVTR) of the paperboards coated with 5 and 10 layers of S-CMF decreased by 27% and 40%, respectively (Figure 5-5). The WVTR values obtained were lower than those reported for commercially available packaging (Ref. in Figure 5-5). The tortuosity of the paperboards developed by the presence of the S-CMF coating layer slows down significantly the diffusion of water vapor through the samples, enhancing the WVTR (Bangar & Whiteside, 2021; Nazrin et al., 2020).

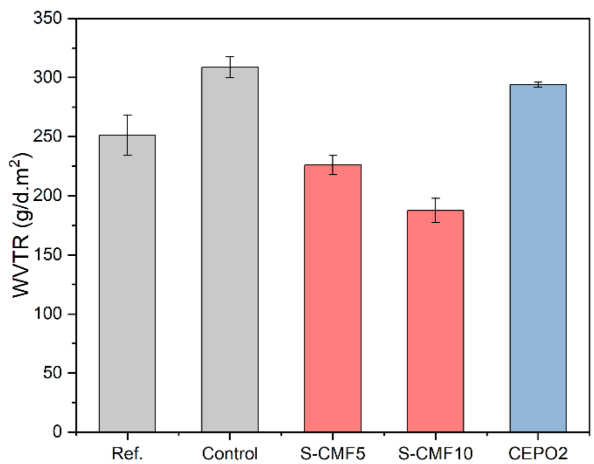


Figure 5-5. Water vapor transmission rate (WVTR) of single-component coated paperboards. *Ref.: commercial coated paper-based food packaging.*

The epoxidized cottonseed oil was treated with citric acid prior to being used as a coating. The hydrophobic nature of the epoxidized cottonseed oil, owing to the presence of long alkyl groups, makes the coating not compatible with the cellulosic substrates and the S-CMF coating, which are both mainly hydrophilic. Thus, a compatibilizer such as citric acid needs to be added to bridge the components through cross-linking (Yang et al., 2021). A schematic illustration of the cross-linking reaction is depicted in Figure 5-6a, and the complete mechanism of the reaction can be found in Altuna et al., (2013). In brief, in the first step, the protons produced during the dissociation of the CA protonate the epoxide groups in the cottonseed oil. Then, the protonated epoxide is attacked by the carboxylate anion from the CA, which promotes the ring opening and forms β-hydroxyesters, as shown in Figure 5-6a.

The crosslinking reaction was confirmed by the changes in the FTIR spectra of the epoxidized oil before (EPO) and after cross-linking (CEPO) with citric acid (Figure 5-7). Both spectra showed strong absorption bands at 2924 cm^{-1} and 2852 cm^{-1}, belonging to the symmetric and asymmetric stretching vibration of C-H bonds of the CH_2 groups present in the epoxidized oil (Miao et al., 2015). Signals at 1739 cm^{-1} and ~1188 cm^{-1} can be assigned to the stretching vibration of C=O and C-O-C bonds of ester groups already present in the epoxidized oil and the new ester groups formed during cross-linking (Pawar et al., 2016), but it was not possible to quantify the contribution of each component to the ester absorption bands. However, the most important change in the CEPO spectra is the disappearance of the peaks at 823 cm^{-1} and 843 cm^{-1} associated with the epoxy groups, suggesting the complete reaction of the epoxy groups with the citric acid (Yang et al., 2022).

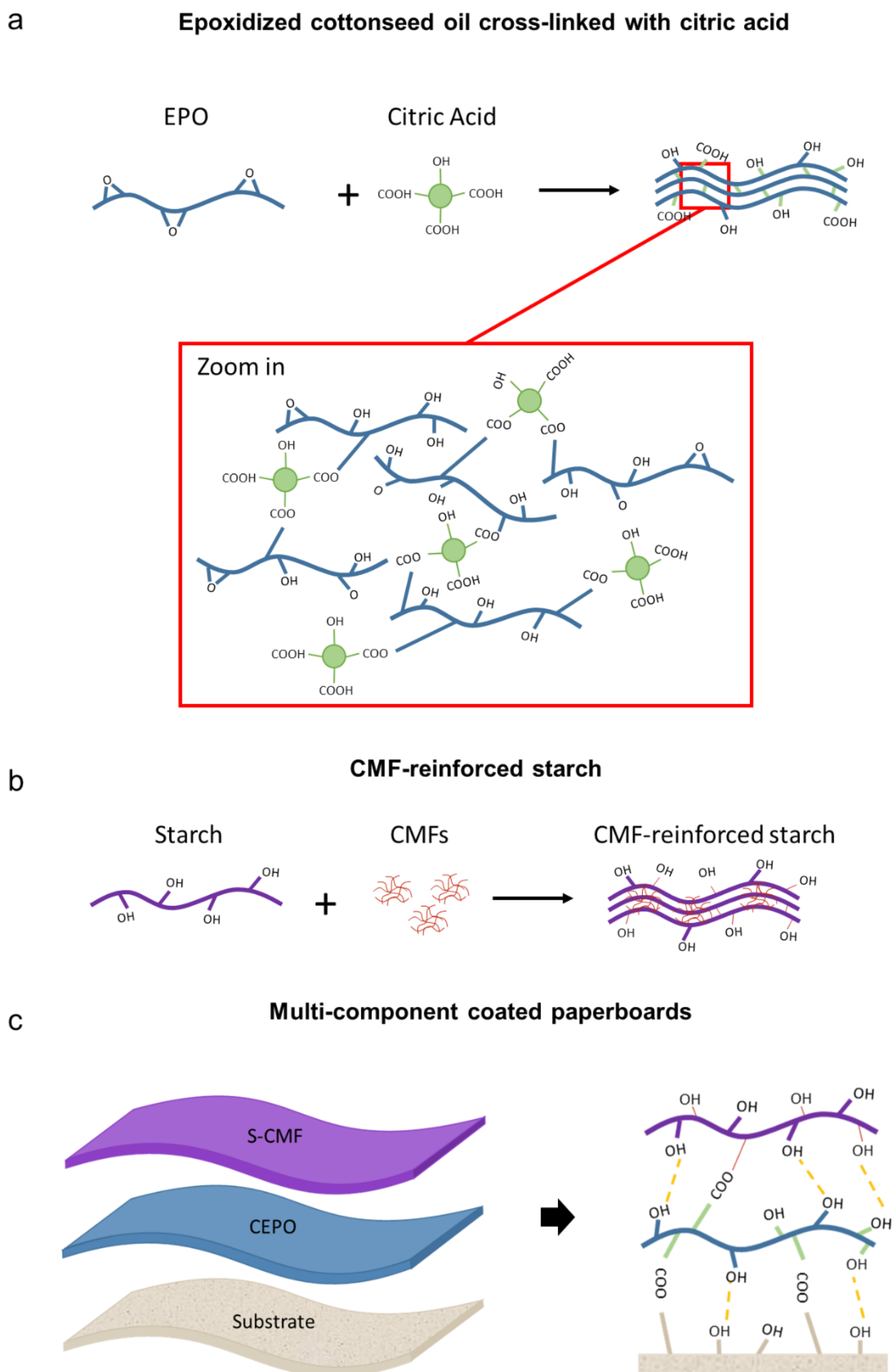


Figure 5-6. Schematic representation of coating components reactions. (a) epoxidized cottonseed oil cross-linked with citric acid, (b) starch reinforcement with CMFs, and (c) paperboard substrates coated with CEPO and S-CMF.

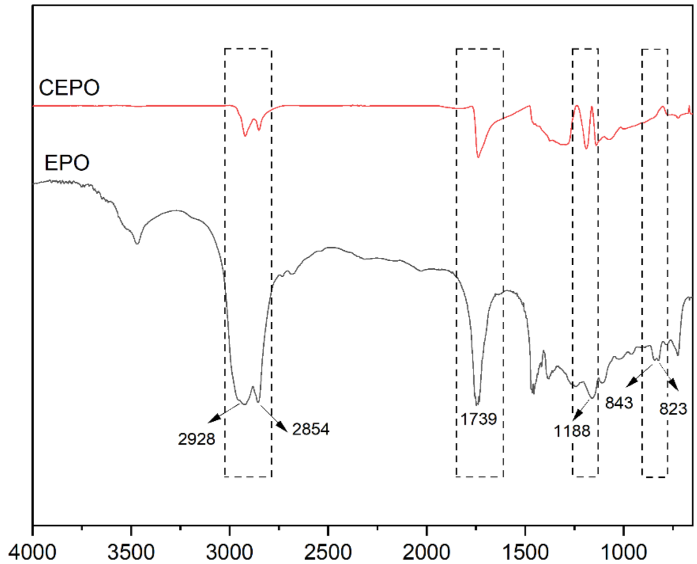


Figure 5-7. FTIR spectra of epoxidized oil before (EPO) and after cross-linking reaction with citric acid (CEPO).

After the reaction with the citric acid, the CEPO was used as a coating for paperboard substrates (1 and 2 layers of CEPO) to evaluate the barrier performance of the single-component coated paperboards. As seen in Figure 4, the paperboards coated with CEPO showed an opposite trend than the S-CMF coated samples. The water absorptiveness significantly decreased from 53.5 g/m^2 to 17.8 g/m^2, and the water wettability increased from 99° to 104° when coated with 2 layers of CEPO. The hydrophobic nature of the epoxidized cottonseed oil coating enhanced the barrier properties to water; in addition to the less intermolecular space created due to the cross-linking reaction between the epoxidized oil, citric acid, and cellulosic fibers (Lei et al., 2018; Zhao et al., 2021), as evidenced in the SEM images (Figure 5-2h and i), could also contribute to the improvement in water repellency. During curing, besides the reaction already mentioned between the EPO and the CA, the unreacted COOH groups in the CA could also react with the hydroxyl groups in the paperboard to form ester groups (Yang et al., 2021) (Figure 5-6c), thus promoting the cross-linking between the coating and the substrate.

The oil repellency of the coated paperboards, however, was affected by the presence of the CEPO coating, as can be noticed by the increase in oil permeability to 131 g/m^2 and the decrease of the Kit number to 0 (Figure 5-4b). The results agree with the lipophilic nature of the coating that facilitates the penetration of the oil.

There were no significant differences in the WVTR of CEPO coated paperboards and the control samples (Figure 5-5). The thin CEPO layer on the paperboards could not significantly seal the pores of the samples, as was done by the presence of the S-CMF coating. Consequently, the water vapor could transit through the porous sample as if no coating was present.

Thus, considering that the S-CMF coating favors the oil and grease resistance of the paperboards and contributes to the reduction of water vapor permeability, and the CEPO coating benefits the water repellency, the utilization of both coatings in paper-based substrates was proposed in this study as a strategy to build bio-based coated paperboards with good barrier properties for food packaging applications.

5.4.2. Multi-component coated paperboards performance

Taking advantage of the properties of the individual coatings (*i.e.*, S-CMF and CEPO), multi-component coated paperboards were produced using both S-CMF and CEPO to engineer samples with multiple barrier properties within the same paperboard (Pasquier et al., 2022). 2 layers of CEPO were first coated onto the paperboard substrates, followed by 1, 2, 5, or 10 layers of S-CMF. The CEPO2/S-CMF# (where # refers to 1, 2, 5, or 10 layers) coated paperboards (Figure 5-8) showed better resistance to water penetration than the single-component S-CMF coated paperboards (Figure 5-4a) but still too high when compared to the reference samples

(Figure 5-8a). The hydrophilic character of the outer coating prevailed over the water repellency of the inner coating (CEPO), avoiding the significant improvement of the water Cobb values and decreasing the WCA. An additional layer of CEPO2 was also applied to the CEPO2/S-CMF10 configuration to protect the S-CMF layer from the water attack (CEPO2/S-CMF10/CEPO2 in Figure 5-8a). An outer layer of CEPO enhanced the paperboards' water repellency, decreasing the water Cobb values to 24.2 g/m^2 and increasing the contact angle to 82°.

Multi-component coated samples also showed good oil and grease resistance properties with a kit number of 9 and an oil permeability of 3.2 g/m^2, which are comparable (and even greater in the case of the Kit number) to the values reported for the commercial samples (Figure 5-8b). The outer layer, composed of CMF-reinforced starch, contributed significantly to the samples' oil & grease barrier properties. CEPO2/S-CMF10 samples coated with an additional coating of CEPO2 increased the oil permeability to 9.7 g/m^2 and decreased the kit number to 4.

The WVTR of the multi-component coated paperboards is presented in Figure 5-9. The WVTR decreased in CEPO/S-CMF coated paperboards with increasing layers of S-CMF when compared to the control samples. The CEPO2/S-CMF5 samples showed no significant differences with the commercial reference packaging, and the CEPO2/S-CMF10 paperboards showed a decrease of 22%. An extra coating of CEPO on top of the CEPO2/S-CMF10 coated paperboards decreased even further the WVTR of the samples to a value of 177.64 g/m^2. The complementary effect of the S-CMF coating that reduces the porous structure of the paperboards, and the CEPO with hydrophobic nature, avoided the interaction and penetration of water vapor through the samples creating a substrate with outstanding barrier properties against gases for paper-based packaging applications.

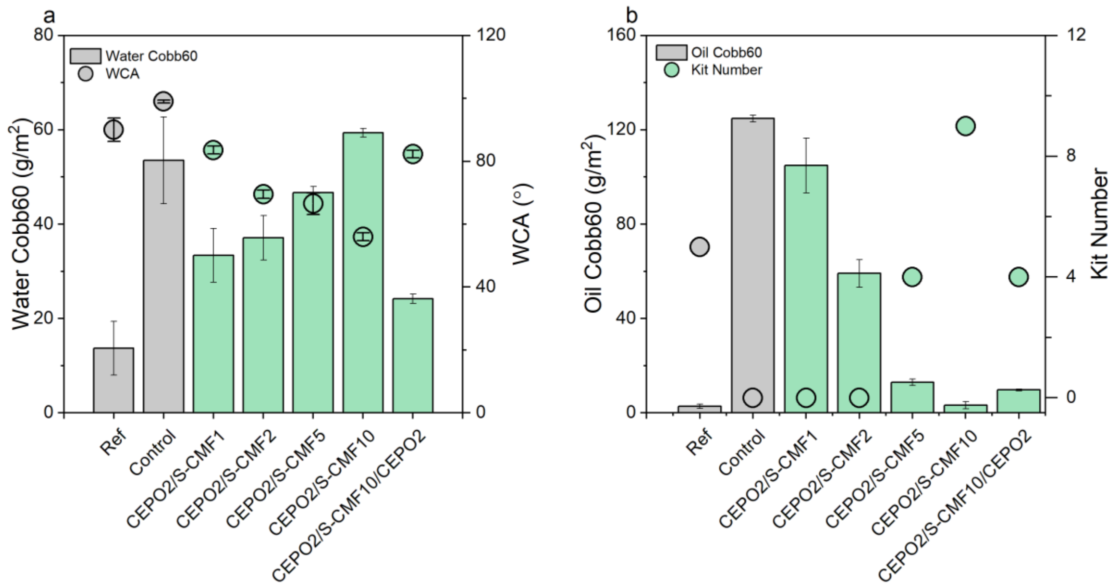


Figure 5-8. Water and oil & grease resistance of multi-component coated paperboards (coated with cross-linked epoxidized cottonseed oil (CEPO) followed by 1, 2, 5**,** and 10 layers of CMF-reinforced starch (S-CMF)). (a) Water Cobb value and contact angle (WCA), (b) oil Cobb value and kit number. *Ref.: commercial coated paper-based food packaging.*

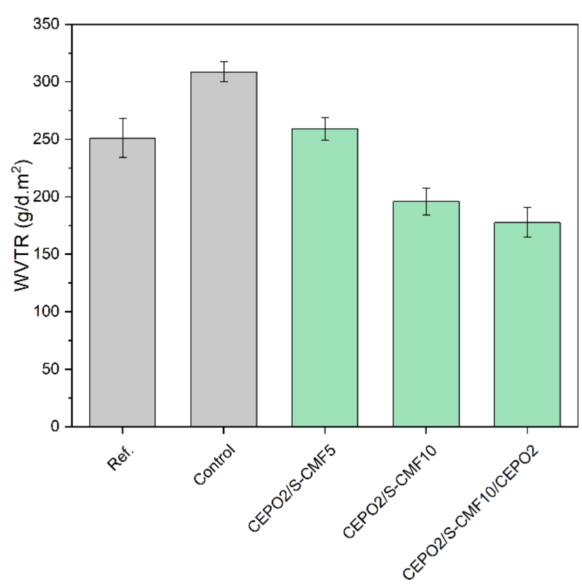


Figure 5-9. Water vapor transmission rate (WVTR) of multi-component coated paperboards. *Ref.: commercial coated paper-based packaging.*

5.5. Conclusion

In this study, we have developed a multi-component bio-based coating for paper-based packaging with good barrier properties for food applications. The amalgamate effect of CEPO and CMF-reinforced starch as coating layers onto recycled paperboards allowed the formation of samples with enhanced repellency toward the water, water vapor, and oil & grease. The epoxidized oil provided the hydrophobic character by incorporating a layer of non-polar material with less intermolecular space structure due to the cross-linking reaction when using citric acid as a compatibilizer.

The CMF-reinforced starch, on the other hand, renders the paperboards with an oil & grease barrier thanks to the oleophobic nature of the S-CMF and the less porous surface of the paper developed by the presence of the coating. S-CMF also contributed to decreasing water vapor diffusion through the samples. The multi-component coated paperboards performed similarly to commercially available paper-based packaging used in food applications. Thus, the CEPO and S-CMF could be potentially used as a more sustainable coating for paper-based packaging to replace the harmful and not biodegradable PFAS and petroleum-based materials. On the one hand, CEPO and S-CMF come from renewable resources; on the other hand, both have been reported individually as biodegradable materials, which can contribute towards lowering the environmental impact of the packaging.

Chapter 6 . Conclusions and Future Work

The scientific findings of the five chapters presented in this dissertation demonstrated the feasibility of incorporating high ratios of minimally processed low-quality waste into recycled furnishes to produce sustainable packaging with clear clues of recyclability. The produced samples meet the goal of facilitating the recognition of the packaging as more environmentally friendly when compared with the packaging with a traditional appearance (neat appearance) and, at the same time, meet the product specifications in terms of mechanical and barrier properties. The mechanical performance can be guaranteed by adding some of the well-investigated dry strength agents, such as cationic starch or cellulose microfibrils, or utilizing the currently industrial manufacturing process for linerboards, *i.e.*, building multi-layer paperboards. The barrier properties, on the other hand, can be provided by coating the paperboards with bio-based slurries such as epoxidized cottonseed oil and starch reinforced with nanocellulose to produce samples with repellency against water, water vapor, and oil & grease.

The research approach proposes a novel pathway that aligns with global efforts for a more sustainable future in several ways.

i. The replacement of a percentage (up to 40 wt%) of the pulp slurry composed of fully disintegrated recycled fibers with partially disintegrated waste could decrease the energy consumption of the pulping process. Moreover, since none of the non-fibers materials is being removed from the waste, the energy consumed in the cleaning steps during recycling (*e.g.*, screening, centrifugal cleaning, and dispersion and kneading, among others) will be saved.

ii. The valorization of low-grade waste could contribute significantly to the circular economy by reducing the amount of waste ending in landfills. The utilization of difficult-to-recycle waste, such as paper cups, could have a positive impact on the environment. To date, less than 1% of this waste is recycled.

iii. Endowing the packaging with clues of recyclability (sustainability) could raise consumers' awareness of recycled containers guiding their purchase decision towards bettering the environmental impact of packaging. This approach could inform better and educate consumers on the sustainability of the products.

iv. The replacement of PFAS and petroleum-based coatings in fiber-based packaging with bio-based materials could greatly reduce their environmental and health adverse effects.

The redesigned packaging with an obvious recycled appearance show potential to be used in different applications such as shipping and food packaging. However, future work is suggested to complement this investigation.

Consumers' perception towards redesigned containers for *primary* food packaging should be assessed. The study presented in Chapter 4 evaluated consumers' perception of the redesigned containers when used as *secondary* packaging (not in direct contact with the product). However, the direct contact of the waste with the product (*e.g.,* food) could change the perception of the packaging as well as the desirability of the product.

Since recycled fibers could contain harmful contaminants that can migrate to the food when used in primary packaging for food applications, and there are strict legislations and regulations in the US and EU for recycled food-contact materials (Misko, 2013), a migration study needs to be performed. The bio-based coating could act as a barrier to avoid direct contact of the food with the paper-based material, possibly preventing the migration of the contaminants.

Additionally, the biodegradability and recyclability of the coated paperboards with CEPO and CMF-reinforced starch should be evaluated to prove the sustainability of the proposed bio-coating.

The perception of redesigned packaging with an obvious waste content appearance from the companies' perspectives should also be investigated. Packaging companies should be interviewed to explore the advantages and challenges they find in using such boxes to instill positive product/company recognition.

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APPENDICES

Appendix A: Supporting information for Chapter 2

1. Materials and Methods

1.1. Refining level of the OCC pulp

The level of refining to be applied to the OCC pulp was selected to mimic the refining process performed in the paper mill. To this end, the mechanical properties of paperboards prepared after subjecting the OCC pulp to different refining levels (i.e., 1,000, 3,000, and 5,000 revolutions) were compared to the properties of paperboards made with OCC pulp refined at the paper mill. The OCC pulp suspension was refined using a PFI mill refiner (PFI Mill, The Norwegian Pulp and Paper Research Institute, Oslo, Norway) according to the TAPPI 248 Sp-00 (2000) standard. Paperboard specimens of 130 g/m^2 were prepared using a TAPPI handsheet former (TAPPI 205 Sp-02 (2006)). The tensile strength (TAPPI 494 om-96, 1996), tear strength (TAPPI 414 om-98, 1998), and short-span compression strength or STFI (TAPPI 826 om-08, 2013) of the paperboard samples were measured on a minimum of five specimens per sample type. Strips of 15 mm wide were used for tensile strength and short-span compressive strength measurements, and the testing was carried out using a tensile tester (TMI 84-56, Testing Machines, Inc., New Castle, DE, USA) and an STFI compression strength tester (Lorentzen & Wettre, Stockholm Sweden), respectively. The tear strength was evaluated on a stack of five plies of 63-mm wide using a Lorentzen & Wettre tear instrument (Stockholm, Sweden).

1.2.Characterization of high aspect ratio MOW suspension

Imaging of the MOW with high aspect ratio (MOW CMNFs) was performed using a field-emission scanning electron microscopy (FE-SEM, FEI Verios 460 L, Hillsboro, OR, USA). The aqueous CMNF suspension was diluted to 0.001 wt% and dispersed using a digital homogenizer (T 25 Ultra-turrax, IKA, USA) for 1 min at 10,000 rpm. A drop of the suspension was spread out onto a stub covered by carbon tape and let dry for 24 h at room temperature. Prior to imaging, the sample was coated with a thin layer of gold (approx. 7 nm). The working distance used for this analysis was 6.2 mm for an accelerating voltage of 2.00 kV and a magnitude of ×2500.

1.3. Analysis of MOW particles of different aspect ratios in suspension and paperboard substrates

The average area of the MOW particles of 52 and 72 in aspect ratio was determined using the image analysis system Spec*Scan^{TM} 2000 (Apogee Systems Inc.) combined with a flat-bed scanner (Epson Perfection 2400 photo). The aqueous MOW suspensions were diluted to 0.04 wt% and then filtered using black filter paper (Grade 8613, Ahlstrom). Ten filter papers per sample were scanned using a resolution of 600 dots/inch, in normal 256 grayscale, using a reverse threshold setting of 80% of the average grayscale value. This procedure was not feasible for the MOW CMNFs, because of the small dimensions of the fibers (not detected by the equipment) and the tendency to self-aggregate.

The procedure was repeated to analyze the MOW-containing paperboards. The total area (in mm^2) covered by the MOW particles per square meter (m^2) of paperboard sample was determined. Five paperboard sheets per OCC/MOW weight ratio were scanned on both sides.

1.4. Visual appearance of recycled MOW-containing paperboards

A flat-bed scanner (Epson Perfection 2400 photo) was used to take pictures of the MOW-containing paperboards substrates.

1.5. Color analysis of recycled MOW-containing paperboards (CIELAB parameters)

The color appearance of paperboard substrates was determined using a ColorTouch® X (Technidyne Corporation, Inc., NJ, USA) with diffuse illumination, according to (TAPPI 527 om-

02, 2002). Ten samples per paperboard grade were analyzed for reproducibility. The parameters measured were lightness (L*), redness (a*), and yellowness (b*).

1.6. Statistical Analysis

The Tukeys' range test was applied to the average values reported for the physical properties of paperboards to determine the ones that are significantly different from each other, using the JMP software (SAS Institute). The Tukey's range test considers that the data from different groups of mean comes from a population that has a normal distribution and has the same standard deviation. *p*-values lesser than 0.05 suggest a significant statistical difference between the data, while *p*-values greater than 0.05 suggest that there is no significant statistical difference between the data. The statistical evaluation was performed with a confidence interval of 95%.

2. Results and Discussion

2.1. Refining level of the OCC pulp

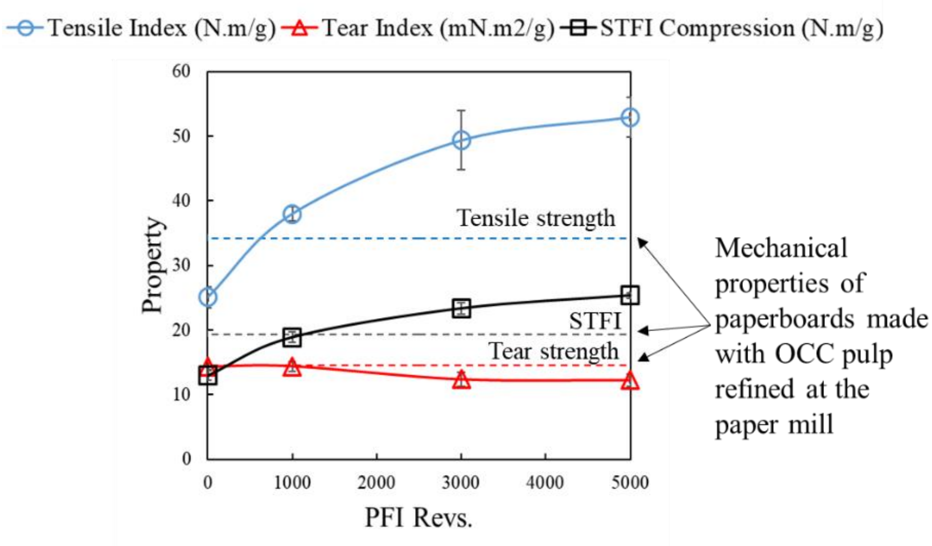


Fig. S1. Mechanical properties of OCC refined at different refining degrees. Dashed lines represent the tensile strength, tear strength, and STFI compression properties of paperboards prepared with OCC pulp refined at the paper mill.

2.2.Characterization of high aspect ratio MOW suspension

Figure S2 shows the FE-SEM image and the visual appearance of the CMNF suspension. The suspension showed a homogeneous gel-like structure with a tight entanglement network typical of cellulose micro/nano-fibrils processed by mechanical shearing (Zambrano et al., 2021). When non-functionalized CMFs are dispersed in water, strong hydrogen bonds are created between the fibrils that cause the formation of bundles (Foster et al., 2018). The aspect ratio of the CMF suspension measured by sedimentation was 162.9 (±1.2).

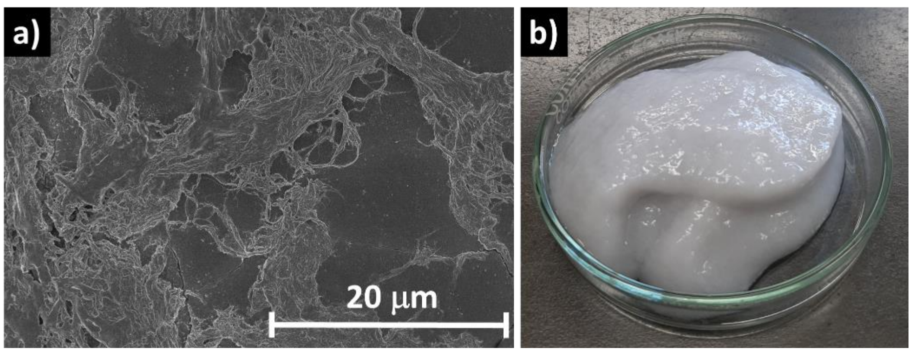


Fig. S2. MOW CMF suspension of high aspect ratio (AR=163). a) FE-SEM image of the dried suspension at 0.001 wt%, b) Picture of the aqueous suspension of MOW CMFs at 2 wt% consistency.

2.3. Analysis of MOW particles of different aspect ratios in suspension and paperboard substrates

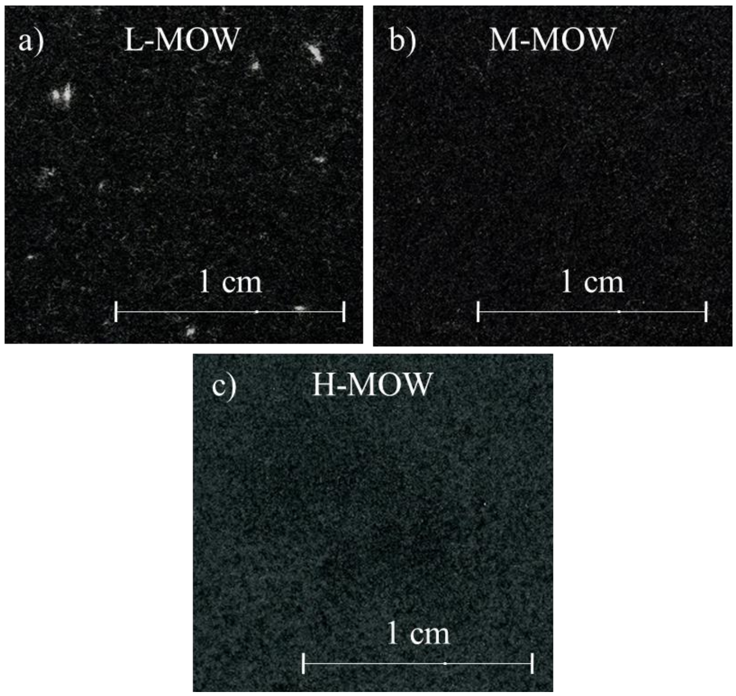


Fig. S3. MOW particles image in suspension at different aspect ratios (a) low aspect ratio or L-MOW (52), (b) medium aspect ratio or M-MOW (72), (c) high aspect ratio or H-MOW (163).

Table S1. Total area covered by the MOW particles per square meter (m^2) of paperboard sample.

Paperboard Sample	Area covered by MOW particles (mm^2/m^2)				
MOW (wt%)	**0**	**5**	**10**	**20**	**40**
L-MOW	98	1962	4669	11230	20300
M-MOW	98	97	95	149	406
H-MOW	98	99	94	109	-

2.4. Visual appearance of recycled MOW-containing paperboards

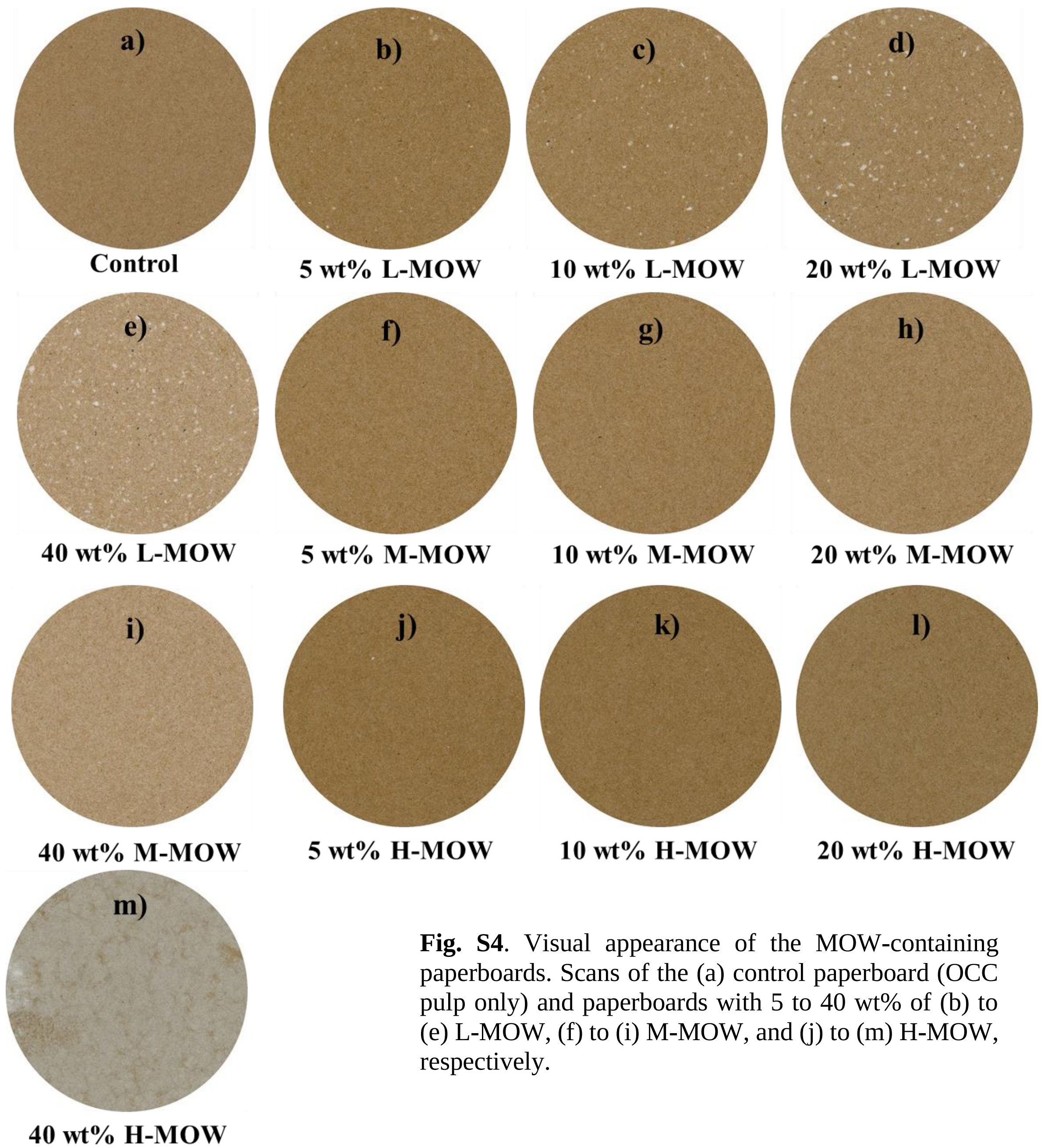


Fig. S4. Visual appearance of the MOW-containing paperboards. Scans of the (a) control paperboard (OCC pulp only) and paperboards with 5 to 40 wt% of (b) to (e) L-MOW, (f) to (i) M-MOW, and (j) to (m) H-MOW, respectively.

2.5. Color analysis of recycled MOW-containing paperboards (CIELAB parameters)

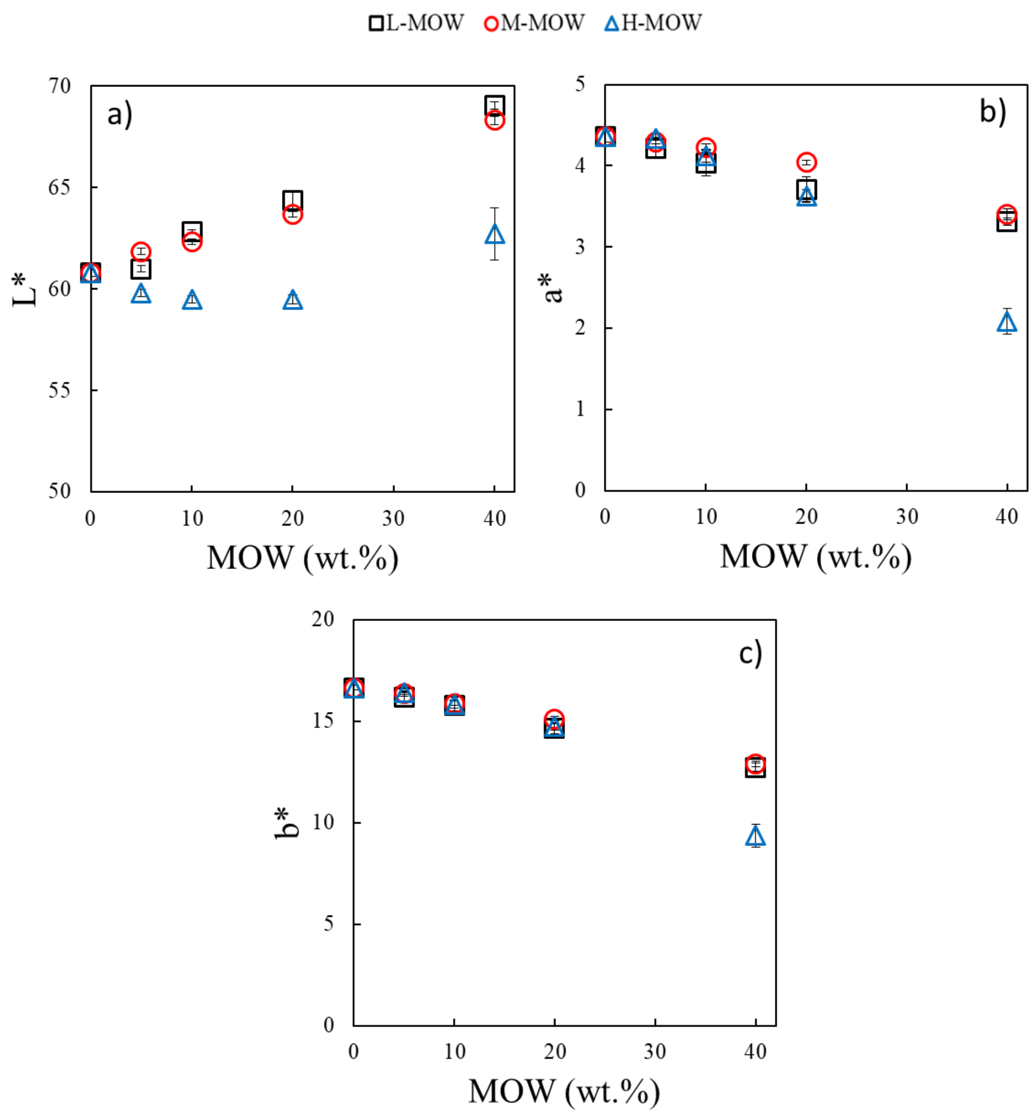


Fig. S5 CIELAB parameters of the recycled MOW-containing paperboards, (a) lightness (L*), (b) redness (a*), (c) yellowness (b*) at different MOW weight ratios and aspect ratios.

2.6. Statistical analysis of MOW-containing paperboards properties

Table S2. Statistical analysis of paperboard properties.

		L-MOW		M-MOW		H-MOW	
Property	MOW (wt%)	Mean	Connecting Letters	Mean	Connecting Letters	Mean	Connecting Letters
Freeness (ml)	0	395	a	395	a	395	a
	5	386	a	368	a	276	b
	10	387	a	370	a	182	c
	20	390	a	368	a	105	d
	40	423	b	391	a	31	e
Bulk (cm^3/g)	0	1.8	b,c	1.8	b	1.8	b
	5	1.8	b	2.2	c	1.6	a
	10	1.7	a	1.8	b	1.5	a
	20	2.1	d	1.6	a	1.5	a
	40	1.9	c	1.6	a	1.5	a
Tensile Index (N.m/g)	0	38.6	a	38.6	a,b	38.6	c
	5	35.9	a,b	35.6	b	49.3	b
	10	34.3	b	38.0	a,b	50.6	b
	20	32.7	b	40.3	a	52.2	b
	40	28.5	c	37.4	a,b	57.1	a
Burst Index ($kPa.m^2/g$)	0	2.8	a	2.8	a	2.8	c
	5	2.9	a	2.6	b	3.6	b
	10	2.9	a	2.8	a	3.6	b
	20	2.2	b	2.8	a,b	3.7	b
	40	2.2	b	2.6	a,b	4.5	a
Tear Index ($mN.m^2/g$)	0	14.7	a	14.7	a	14.7	a
	5	13.6	a	14.7	a	12.3	b
	10	13.9	a	13.7	a,b	11.9	b,c
	20	13.8	a	13.0	b	10.8	c,d
	40	11.8	b	11.3	c	10.6	d
STFI (N.m/g)	0	19.4	a	19.2	b	19.2	d
	5	19.2	a	17.9	c	24.8	c
	10	19.0	a	20.4	a	25.2	c
	20	15.2	b	20.6	a	27.5	b
	40	15.8	b	19.2	b	33.0	a

Connecting letters in Table S2 shows the significant difference between the means. If two or more data points share the same letters, there are no significant differences between the values.

Appendix B: Supporting information for Chapter 3

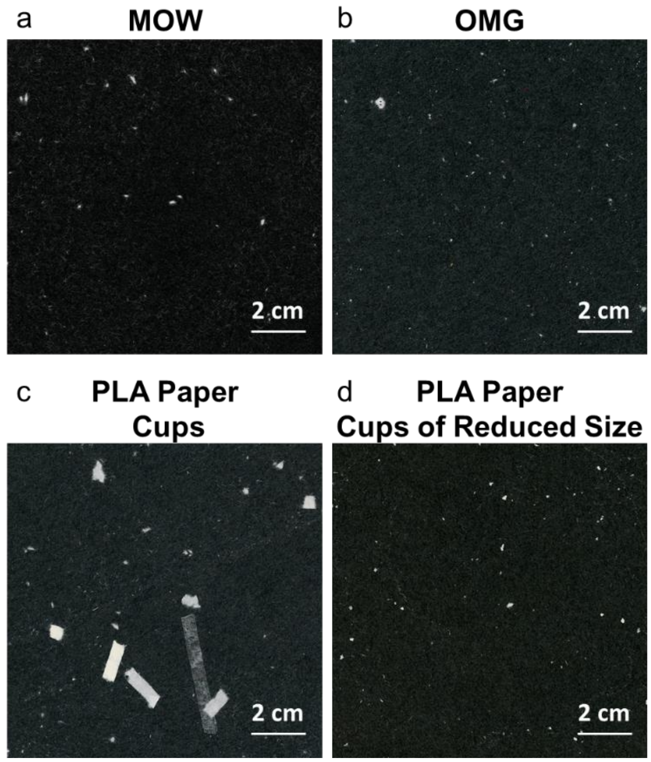


Fig. S1. Flat-bed scanner images (600 dots per inch scanner resolution) of the waste particles after 1 min disintegration. (a) MOW, (b) OMG, (c) PLA paper cups, (c) PLA paper cup of reduced size (Wiley Mill ground before disintegration). MOW: mixed office waste, OMG: old magazines, PLA: polylactic acid lining.

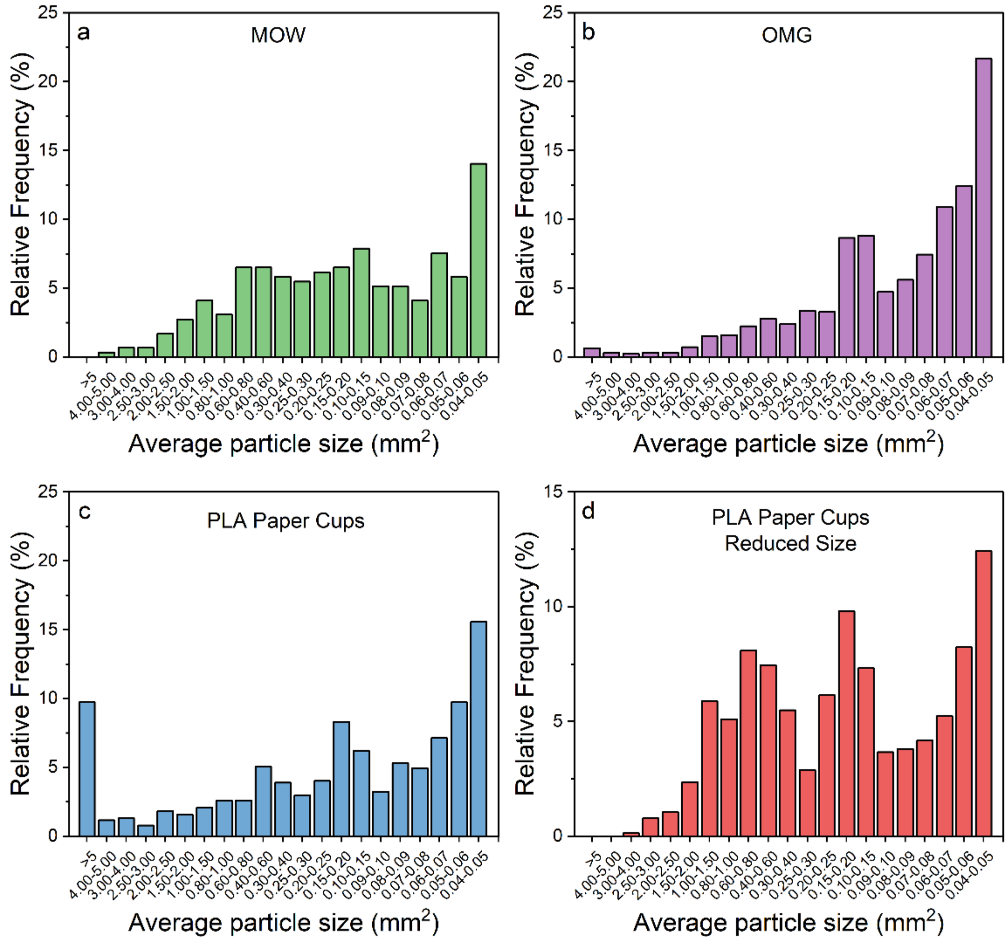


Fig. S2. Waste particle size distribution. (a) MOW, (b) OMG, (c) PLA paper cups, (d) PLA paper cups of reduced size (Wiley Mill ground before disintegration). Relative frequency: ratio of the frequency of each class in the statistical data set to the overall total of all classes.

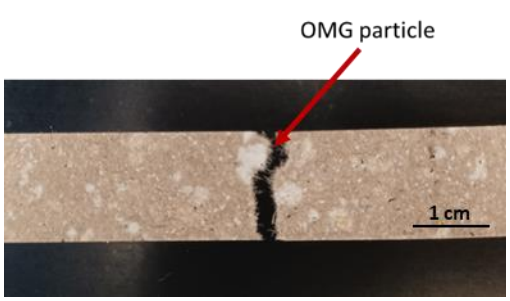


Fig. S3. OMG-containing paperboard strip failure during tensile test.

Table S1. Physical properties of waste-containing paperboards after addition of cationic starch (CS) and cellulose microfibrils (CMFs).

	Density (kg/m^3)			Freeness (ml)		
CS (wt%)	**MOW**	**OMG**	**PLA Paper Cups**	**MOW**	**OMG**	**PLA Paper Cups**
0 (Control)	539 ± 7	536 ± 10	360 ± 22	423 ± 7	483 ± 7	472 ± 6
0.5	607 ± 8	588 ± 14	449 ± 86	484 ± 2	481 ± 7	505 ± 31
1.0	623 ± 8	570 ± 10	389 ± 35	460 ± 14	491 ± 6	439 ± 6
1.5	614 ± 7	593 ± 17	371 ± 71	431 ± 6	454 ± 3	430 ± 7
2.0	627 ± 8	636 ± 13	288 ± 47	397 ± 3	416 ± 6	368 ± 6
CMFs (wt%)	**MOW**	**OMG**	**PLA Paper Cups**	**MOW**	**OMG**	**PLA Paper Cups**
1	606 ± 13	593 ± 22	292 ± 33	413 ± 6	401 ± 7	356 ± 7
3	619 ± 11	565 ± 6	288 ± 3	372 ± 7	385 ± 3	294 ± 6
5	640 ± 6	573 ± 36	286 ± 25	304 ± 6	311 ± 6	236 ± 10

±: Standard deviation

Table S2. Physical properties of 2-ply paperboards.

	Density (kg/m^3)	
Sample	**MOW**	**PLA Paper Cups**
2P 60-60	559 ± 10	334 ± 36
2P 40-80	612 ± 4	523±20

±: Standard deviation

Appendix C: Supporting information for Chapter 4

S1. Waste materials used for paperboard production with obvious recovered content.

S2. Subsets developed for the in-person survey (picture taken from set-up at university campus).

S3. Online survey.

This survey makes use of pictures. For quality reasons, this survey should be opened **only on a desktop, a laptop, or a tablet**. Please do not participate in this survey on your phone.

Q1 Are you participating in this survey using your desktop/laptop/tablet?

o Yes

o No

Hello, good day.

We'd like to invite you to participate in this survey. This way, you can contribute to academic research on **environmentally** sustainable packaging.
The survey takes about **10 minutes**. The answers you provide will be kept **confidential** and will only be used for academic purposes.
If you have questions or comments about this survey, please feel free to contact us.
Lisandra Chacon Ovalles; PhD researcher Sustainable Packaging; North Carolina State University (USA); lvchaco2@ncsu.edu
Sophie Van Schoubroeck; Postdoctoral Researcher Environmental Economics; North Carolina State University (USA); University of Antwerp (Belgium)

Consent form

You are being asked to complete a survey for research purposes. Completing this survey is voluntary and you can stop at any time by closing the questionnaire. You must be **18 years of age or older and reside in North Carolina** to participate in this study. You will receive **an Amazon gift card of $5** if you complete the survey. In order to receive the gift card, you must provide your name and e-mail address at the end of the questionnaire. Based on your demographic information, which will be requested in the first part of the survey, it can be decided that you do not qualify to participate in this survey. If that is the case, you cannot complete the survey and will not receive an Amazon gift card.

If you have questions about your rights as a participant or are concerned with your treatment throughout the research process, please contact the NC State University IRB Director at IRB-Director@ncsu.edu, 919-515-8754, or fill out a confidential form online at https://research.ncsu.edu/administration/participant-concern-and-complaint-form/

Q2 **If you consent to complete this survey, please indicate "yes":**

o Yes

o No

Q3 What is your **year of birth** (XXXX)? ________________

Q4 What is your **gender**?

- o Male
- o Female
- o Not listed

Q5 What is your 5-digit **zip code**? ________________

Q6 What is the highest level of **education** you have completed?

- o Secondary education (high school)
- o Undergraduate education (e.g., bachelor's degree, associate degree,…)
- o Master's degree
- o Doctoral degree
- o Other (Please specify): ________________

Q7 What is currently your total yearly household **gross income** (before taxes)?

- o Under 24,999 U.S. dollars
- o Between 25,000 U.S. dollars and 49,999 U.S. dollars
- o Between 50,000 U.S. dollars and 74,999 U.S. dollars
- o Between 75,000 U.S. dollars and 99,999 U.S. dollars
- o Between 100,000 U.S. dollars and 149,999 U.S. dollars
- o Above 150,000 U.S. dollars

Q8 Including yourself, how many people currently live in your **household**?

- o 1
- o 2
- o 3
- o 4
- o More than 4

Q9 Choose one or more **ethnicities** that you consider yourself to be:

▢ White

▢ Black or African American

▢ American Indian or Alaska Native

▢ Asian

▢ Native Hawaiian or Pacific Islander

▢ Hispanic or Latino

▢ Other ____

Q10 How much do you **agree/disagree** with the following **statement: "I am concerned about the environment"**

o Strongly agree

o Agree

o Somewhat agree

o Neither agree or disagree

o Somewhat disagree

o Disagree

o Strongly disagree

Q11 How much do you **agree/disagree** with the following **statement: "I try to buy environmentally friendly sustainable goods and services"**

o Strongly agree

o Agree

o Somewhat agree

o Neither agree or disagree

o Somewhat disagree

o Disagree

o Strongly disagree

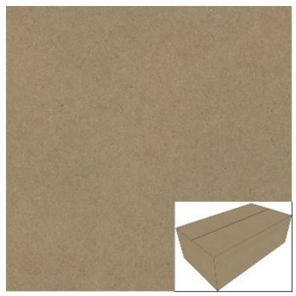
Please read carefully

Imagine you are buying products online, which will be shipped to you in a box (a secondary package, not touching the product directly). These boxes can be made out of different packaging materials, some of them being more environmentally friendly than others.

We are looking to understand **your visual impressions** regarding the **environmental sustainability** of the different packages.

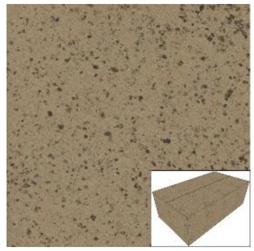
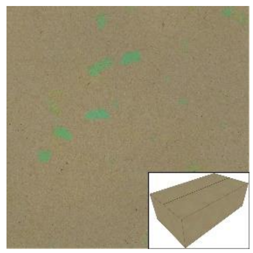
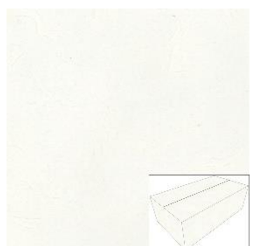
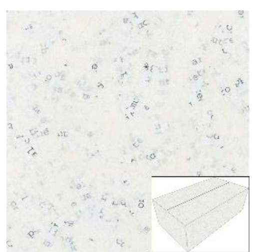
During this survey you will receive 11 different questions. Each question contains 5 packages which you can choose from. For each question you will need to compare the packages and choose **one package** which you perceive as **the most environmentally friendly**, and **one package** which you perceive as **the least environmentally friendly**. You will notice we repeat the pictures over the different questions.

Below you can find an example of a possible picture of a package. The picture shows a close-up of the packaging material. In the bottom right-corner you can find a small image of what the packaging box would look like.



Q12 Which of the following **packages** do you perceive as the **most** and the **least environmentally friendly** package? If necessary, you can enlarge the pictures by clicking on them.

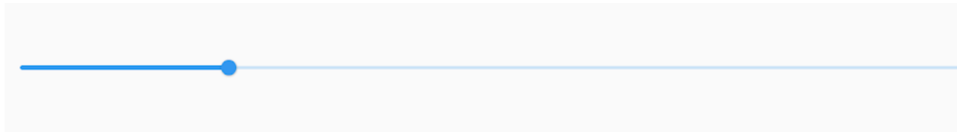
Most environmentally friendly		**Least** environmentally friendly

o		o
o		o
o		o
o		o
o		o

Q13-Q22: Q12 is repeated eleven times according to the design presented in Table 1 presented in the main paper.

Q23 On a scale of 1 to 5, **how easy** was it to make the choices **based on the visual appearance** of the packages?

	Very difficult			Very easy
1	2	3	4	5



Q24 Please **briefly explain** why it was difficult or easy to make the choices based on the visual appearance of the packages:

____

____

Q25 What is your **name** (first name/ last name)?

____

Your name will only be used to send you the **$5 Amazon gift card**. Your name will be kept strictly confidential and never associated with your answers in this survey.

Q26 What is your **e-mail** address?

____

This e-mail will only be used to send you the **$5 Amazon gift card**. Your e-mail address will be kept strictly confidential and never associated with your answers in this survey.

Thank you for participating in this survey. You will receive your Amazon gift card within 15 business days.

If you have any additional questions or comments, please **feel free to contact us**. If you have questions about your rights as a participant or are concerned with your treatment throughout the research process, please contact the NC State University IRB Director at IRB-Director@ncsu.edu, 919-515-8754, or fill out a confidential form online at https://research.ncsu.edu/administration/participant-concern-and-complaint-form/

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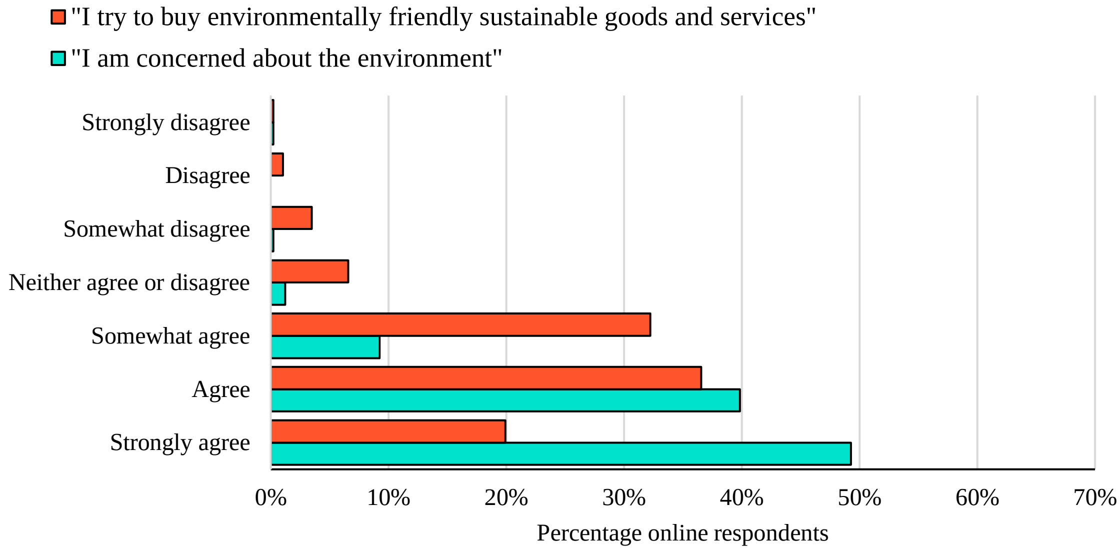
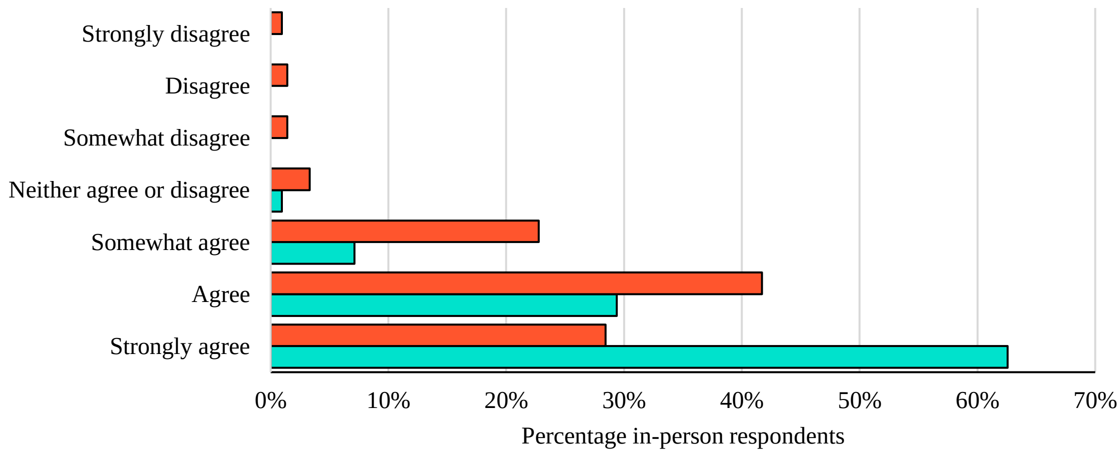
Sophie Van Schoubroeck
Postdoctoral Researcher
North Carolina State University; University

S4. Demographics from online and in-person survey.

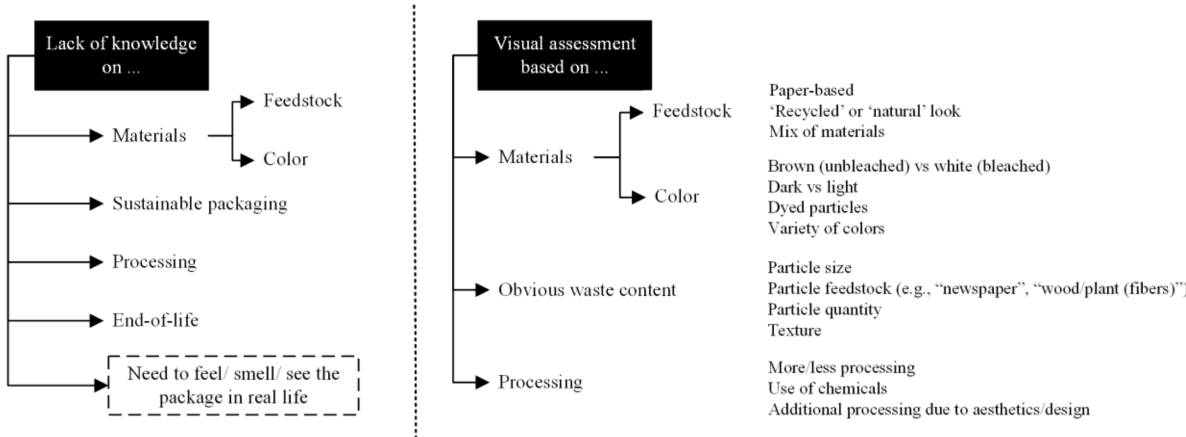
	Online	In-person
Gender		
Male	194	90
Female	286	121
Not listed	7	0
Age		
2003-1994	202	85
1993-1984	124	41
1983-1974	81	13
1973-1964	54	17
< 1964	26	55
Ethnicity		
White	75.05%	65.77%
Asian	12.28%	9.46%
Black or African American	5.65%	16.67%
Hispanic or Latino	5.65%	5.86%
Other	0.78%	0.45%
American Indian or Alaska Native	0.39%	1.80%
Native Hawaiian or Pacific Islander	0.19%	0
Education		
Secondary education	84	50
Undergraduate education	231	104
Master's degree	137	46
Doctoral degree	35	11
Household income		
< $25k	58	50
$25k - $50k	99	35
$50k - $75k	76	33
$75k - $100k	98	30
$100k - $150k	95	34
> $150k	61	29
Household size		
1	95	44
2	162	73
3	85	38
4	98	40
More than 4	47	16
Residence		
North Carolina	477	192
Outside of North Carolina	10	19

S5. Respondents' environmental awareness.



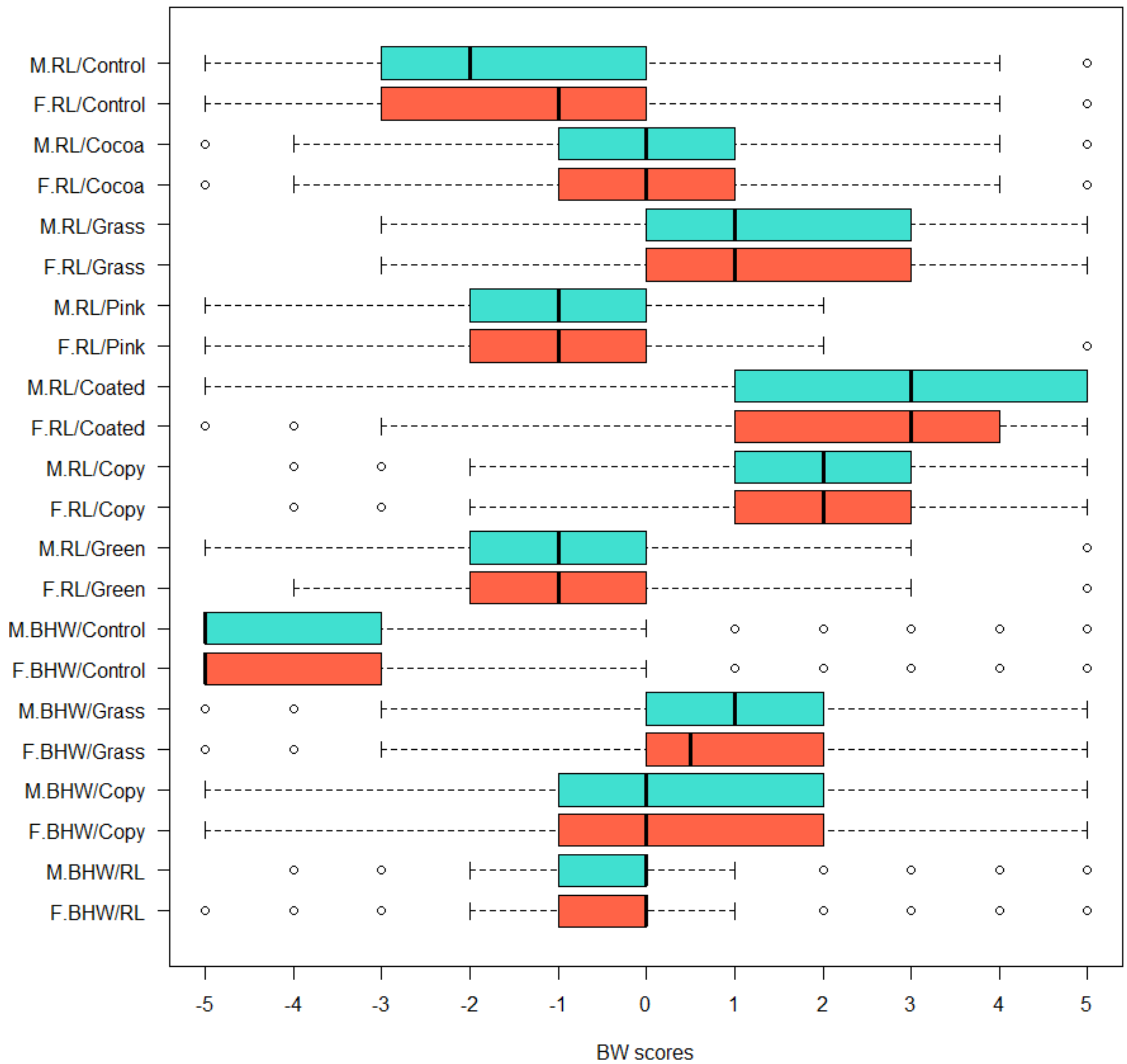


S6. Rating the choice experiment: overview of coding analysis.

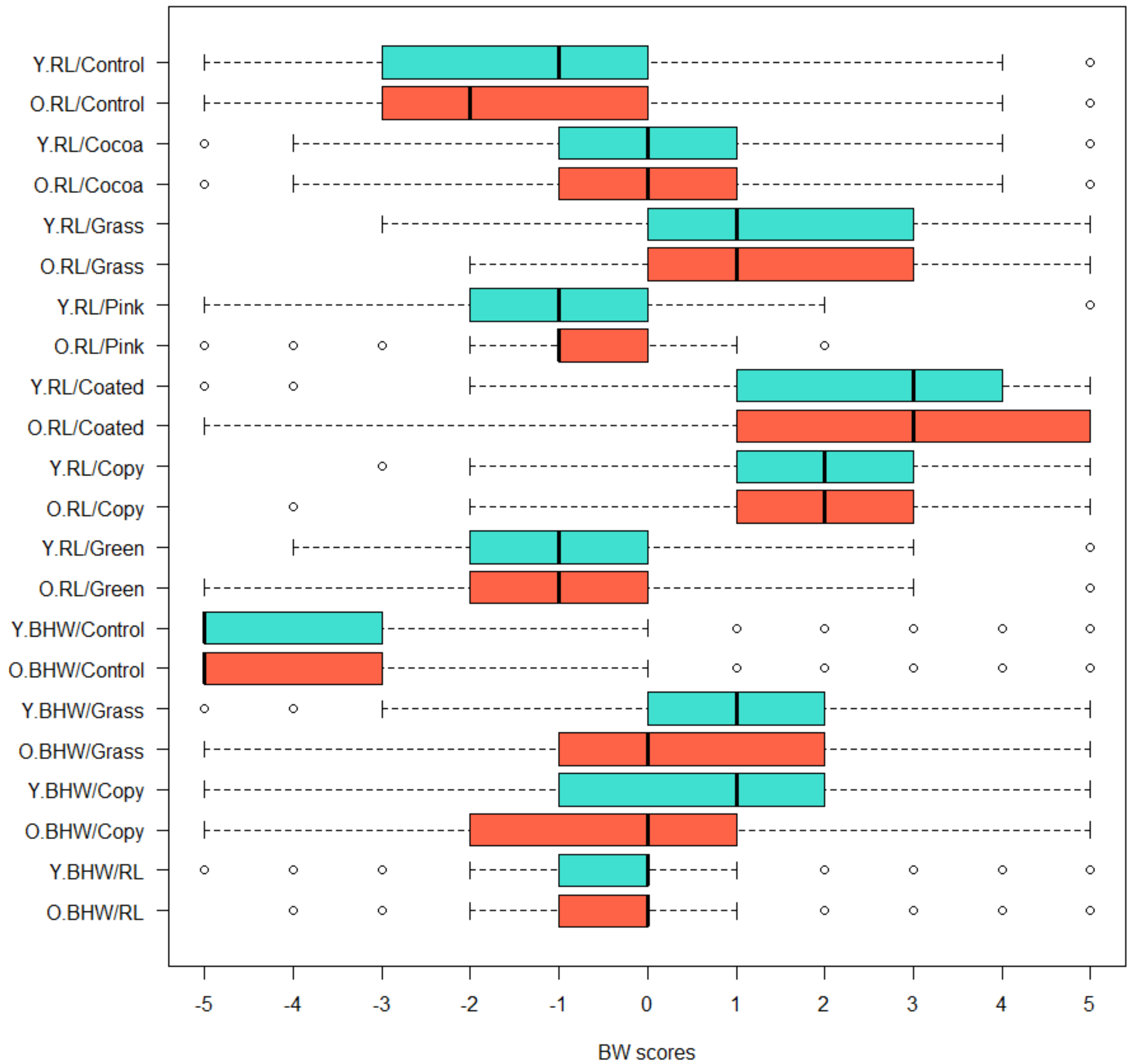


S7. BW scores and demographics.

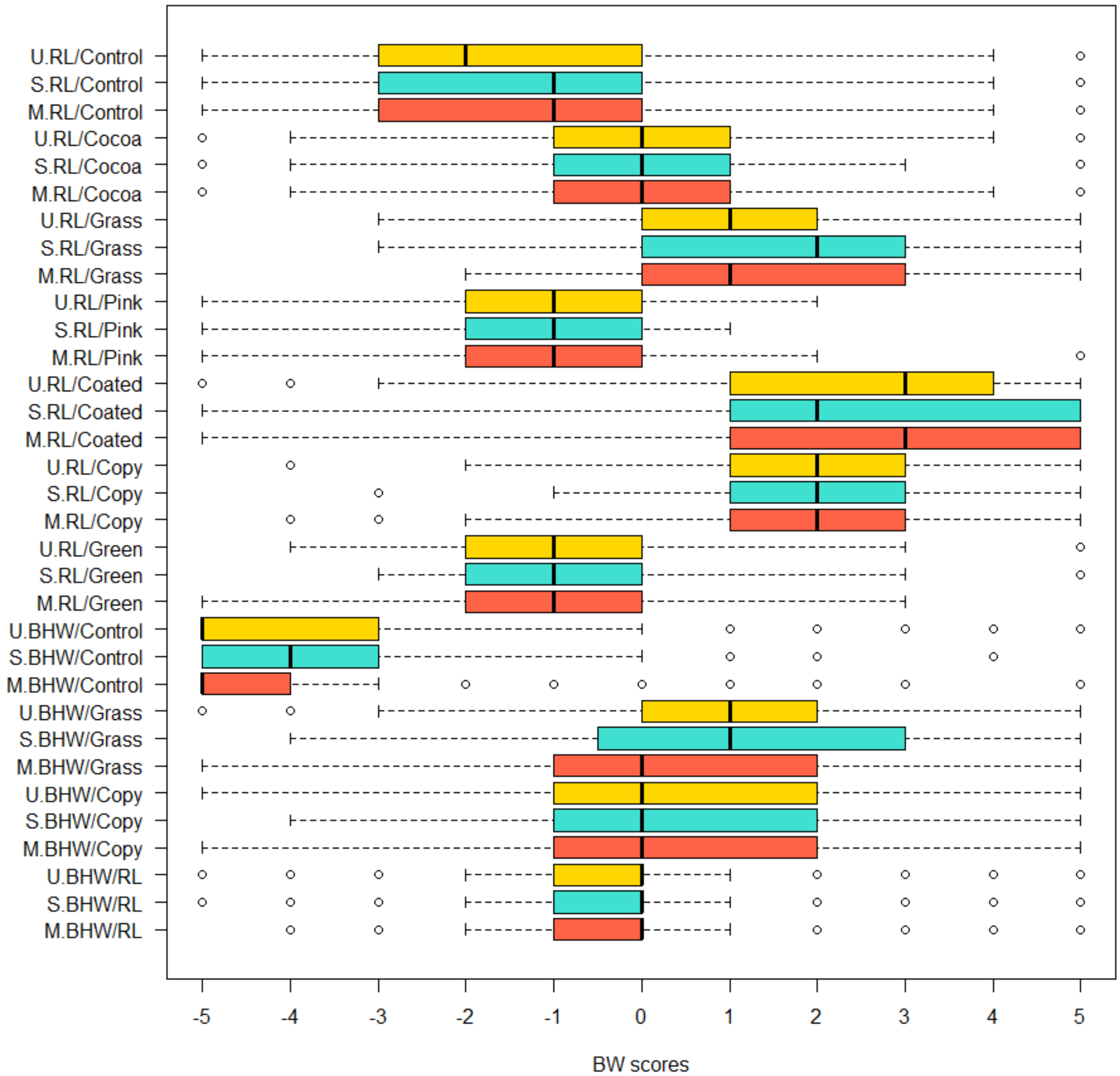
- Boxplots of online BW scores for females, labeled "F" (in red) (n=286) and males, labeled 'M' (in turquoise) (n=194).



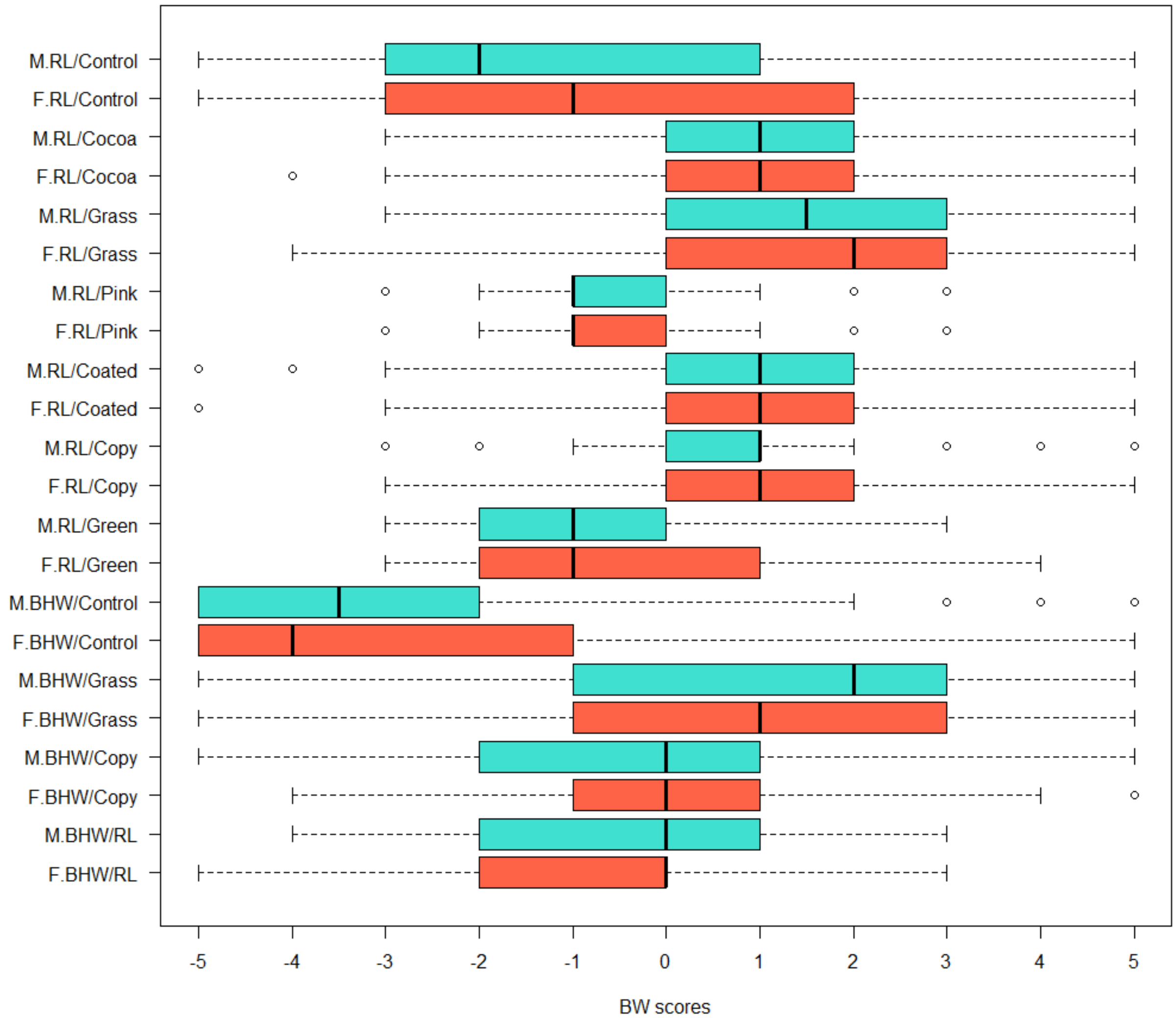
- Boxplots of online BW scores for respondents born before 1988, labeled “O” (in red) (n=206) and born in 1988 or after, labeled ‘Y’ (in turquoise) (n=281).



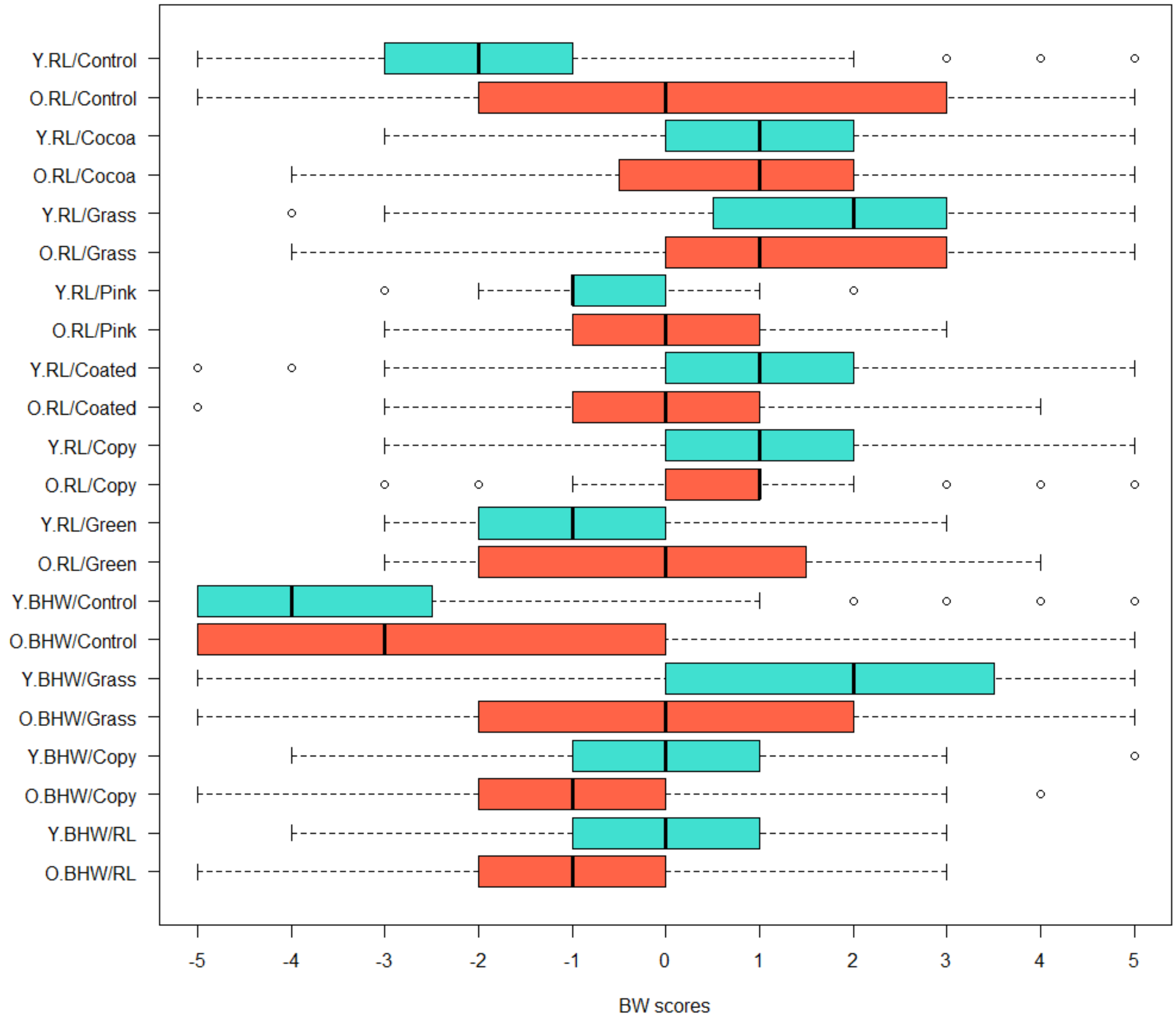
- Boxplots of online BW scores for respondents with only secondary education, labeled "S" (in turquoise) (n=84), an undergraduate degree, labeled "U" (in yellow) (n=231), and a master/doctoral degree, labeled "M" (in red) (n=172).



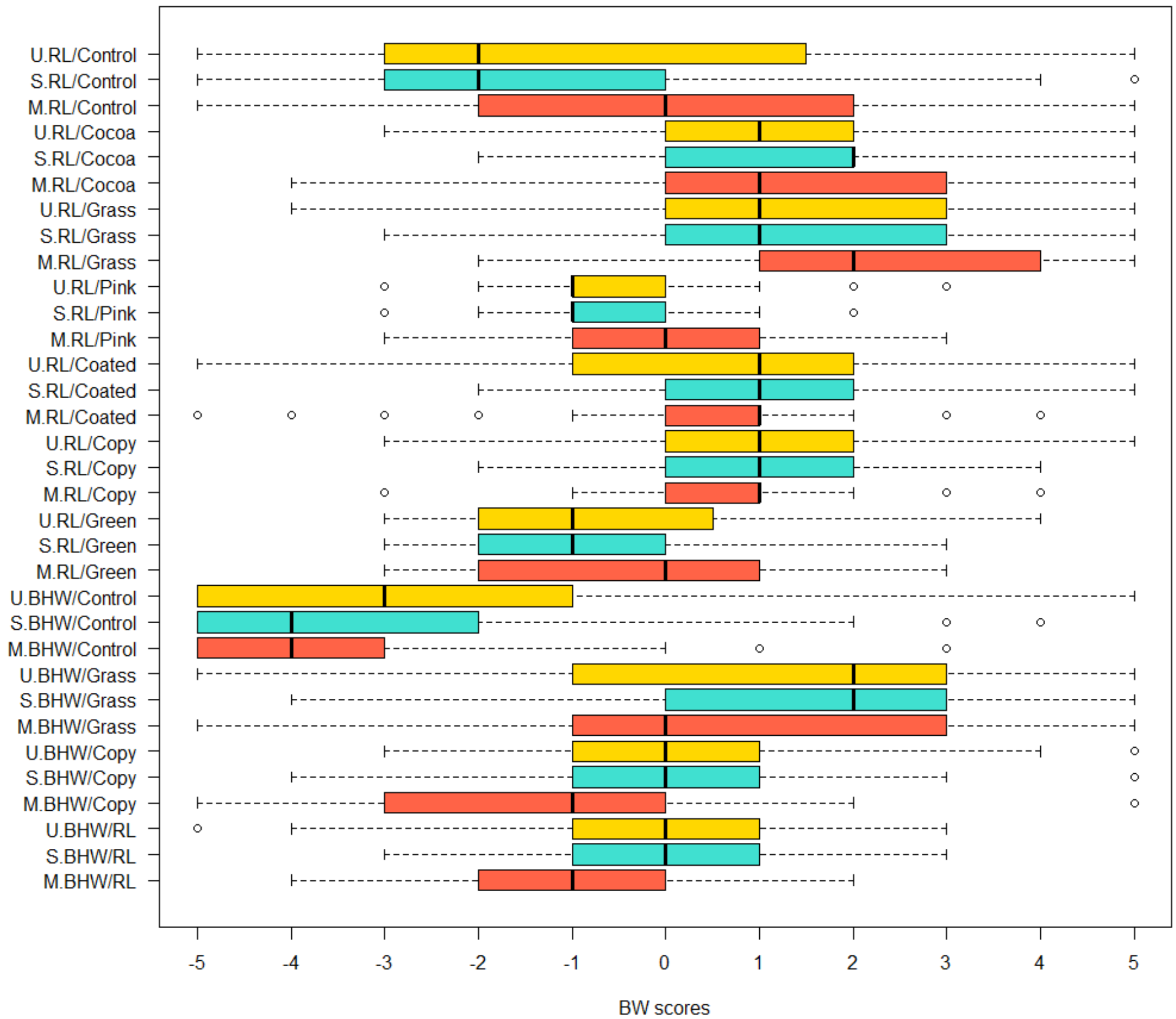
- Boxplots of in-person BW scores for females, labeled “F” (in red) (n=121) and males, labeled “M” (in turquoise) (n=90).



- Boxplots of in-person BW scores for respondents born before 1988, labeled “O” (in red) (n=95) and born in 1988 or after, labeled “Y” (in turquoise) (n=116).



- Boxplots of in-person BW scores for respondents with only secondary education, labeled "S" (in turquoise) (n=50), an undergraduate degree, labeled "U" (in yellow) (n=104), and a master/doctoral degree, labeled "M" (in red) (n=57).

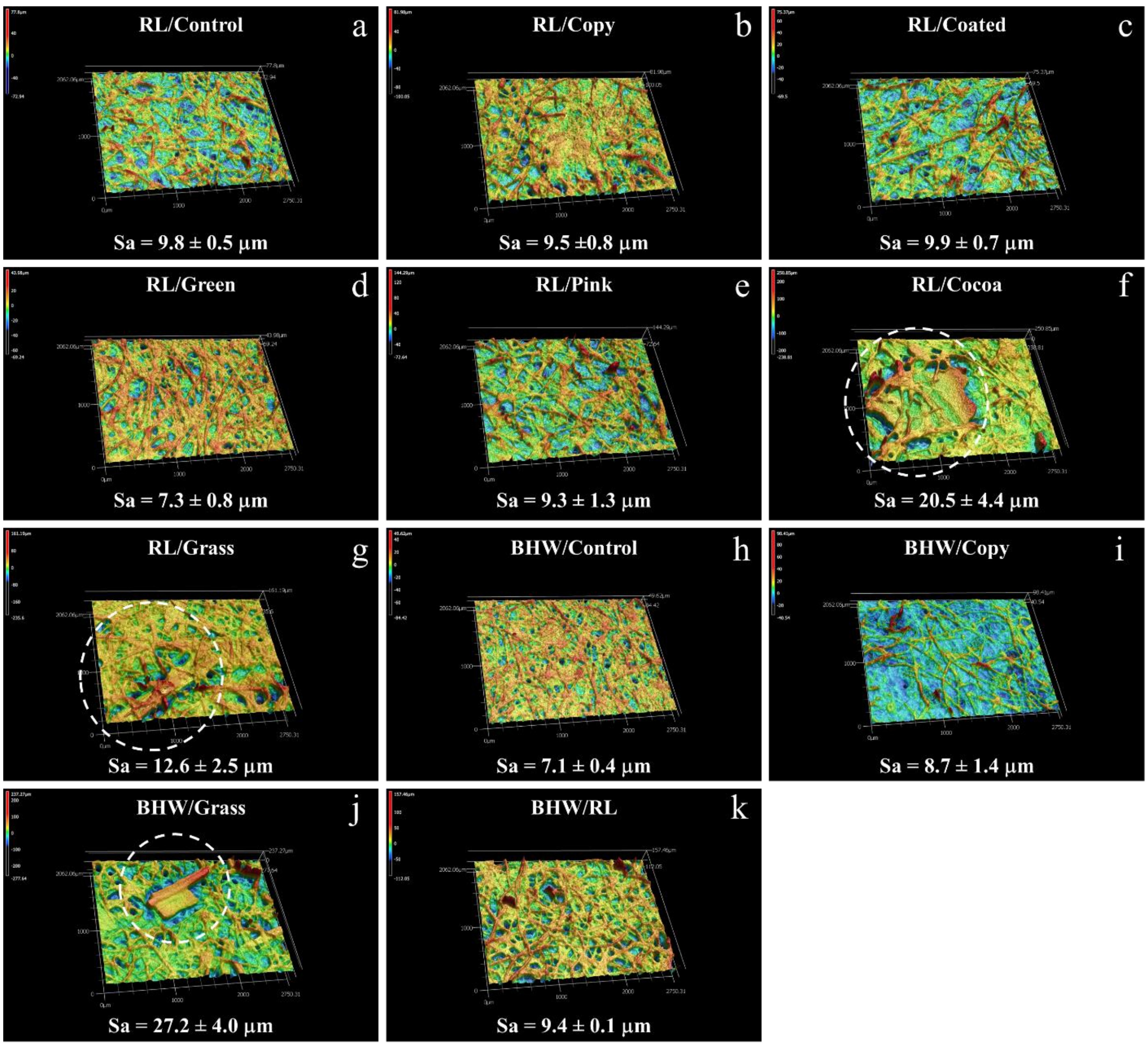


S8. Difference between gender, age, and education-based groups, using the BW scores per respondent, calculated with the Mann-Whitney-Wilcoxon (for two groups) and Kruskal-Wallis rank sum (for more than two groups) tests.

	Gender *(Male or Female)*[a]	**Age** *(Born before 1988 or Born in 1988 or after)*		**Education** *(Secondary education, Undergraduate degree, or Master/Doctoral degree)*	
Sample ID	**P value**	**P value**		**P value**	
RL/Control	1e-01	1e-03	***	4e-02	**
RL/Cocoa	2e-01	6e-01		6e-01	
RL/Grass	9e01	2e-01		2e-03	***
RL/Pink	1e-01	5e-05	***	4e-01	
RL/Coated	1e-01	7e-01		1e-01	
RL/Copy	9e-01	4e-01		3e-01	
RL/Green	1e-01	4e-05	***	1e-01	
BHW/Control	6e-01	5e-01		1e-02	**
BHW/Grass	1e-01	8e-07	***	9e-02	*
BHW/Copy	1e-01	3e-04	***	2e-02	**
BHW/RL	2e-01	4e-02	**	1e-01	

*** $p < 0.01$; ** $p < 0.05$; * $p < 0.1$
[a] "Not listed" category omitted (n=7).

S9. 3D confocal laser microscopy images of the waste-containing paperboards. (a) Recycled linerboard (control), (b) recycled linerboard with copy paper, (c) recycled linerboard with coated paper, (d) recycled linerboard with green paper, (e) recycled linerboard with pink paper, (f) recycled linerboard with cocoa bean shells, (g) recycled linerboard with Switchgrass, (h) bleached hardwood (control), (i) bleached hardwood with copy paper, (j) bleached hardwood with Switchgrass, and (k) bleached hardwood with recycled linerboard. Sa = Roughness average (arithmetic mean of the absolute value of the height above and below the mean plane within the sample area).



Appendix D: Supporting information for Chapter 5

1. Epoxidation of Cottonseed Oil

1.1. Materials

Cottonseed oil with a density of 0.92 g/cm^3, sodium bisulfite (99.7 wt%), potassium acid phthalate (≥99.95 wt%), starch solution indicator (1 wt%), and hydrogen peroxide (30 wt%) was supplied by Sigma-Aldrich (St. Louis, MO). Glacial acetic acid (99.7 wt%) and sulfuric acid (96 wt%) were purchased from Fisher Scientific (Ward Hill, MA). Cyclohexane (99wt%), potassium iodide (99 wt%), crystal violet, hydrogen bromide (33 wt%), and Wijs solution (0.1 mol ICl/l) were obtained from Thermo Scientific (Waltham, MA). Sodium thiosulfate was provided by Ricca (Arlington, TX).

1.2. Epoxidation Process

Epoxidation of cottonseed oil was performed using peroxy acid following Wijayapala et al., (2019). In brief, 94 g of cottonseed oil was heated up to 55 °C in a three-neck round-bottom flask (250mL) with a reflux condenser and a mechanical stirrer (~500 rpm). Glacial acetic acid (9.8mL) and sulfuric acid (0.8mL) were added, and the temperature was maintained at 55°C for another 10 min. Hydrogen peroxide (58mL) was added drop by drop until complete addition of the chemical. The temperature was then increased to 70°C and maintained as such for 8 hours. The resultant non-polar phase, consisting of the epoxidized oil and remaining unreacted oil, was extracted from the biphasic mixture using a separating funnel. The extracted phase was washed with deionized water twice, followed by two washings with sodium bisulfite 5%wt solution. The product was centrifuged at 3000 rpm for 10 min.

1.3. Characterization

The degree of unsaturation (double bonds) in the cottonseed oil before and after epoxidation was determined by measuring the iodine value in accordance with ASTM D5554-15, (2021) standard. The number of epoxy groups in the epoxidized oil was confirmed using the AOCS Official Method Cd 9-57, (1997) titrating the oxygen directly with hydrogen bromide in acetic acid. Both analyses were performed by duplicate.

The substitution of the double bonds in the cottonseed oil by the epoxy groups after epoxidation was tracked using an attenuated total reflectance-Fourier transform infrared spectrometer (ATR-FTIR) (Frontier, PerkinElmer, Massachusetts, USA). The data was collected in the spectral range from 4000 cm^{-1} to 650 cm^{-1} with a resolution of 4 cm^{-1} for 10 scans.

Proton nuclear magnetic resonance (^{1}H NMR) was used to confirm the chemical structure of both un-epoxidized and epoxidized oil using a Bruker Avance III 500 MHz spectrometer with 64 scans and a relaxation delay of 2 s. Samples were dissolved with deuterated chloroform before the analysis.

1.4. Results and discussion

FTIR and NMR spectroscopy was used to verify the epoxidation of the cottonseed oil. **Figure S1** shows the FTIR spectra of cottonseed oil before and after the epoxidation. The absorption bands at 3010 cm^{-1} and 1650 cm^{-1}, characteristics of stretching vibration of C=C-H and C=C double bonds, respectively, completely disappeared in the epoxidized oil spectra (Dominguez-Candela et al., 2022). This suggests the double bonds' disappearance in the oil structure after epoxidation. The FTIR findings were confirmed by the significant decrease in the amount of unsaturation expressed through the iodine value from 111.5 to 0.6 mg I_2/g (**Table S1**). New signals at 843 cm^{-1} and 823 cm^{-1} appeared after the epoxidation reaction and are attributed to the stretching vibration of C-O-C in oxirane groups, which confirms the epoxidation reaction (Ghasemi Rad et al., 2019). The percentage of oxirane oxygen in the epoxidized oil was 5.75% (**Table S1**).

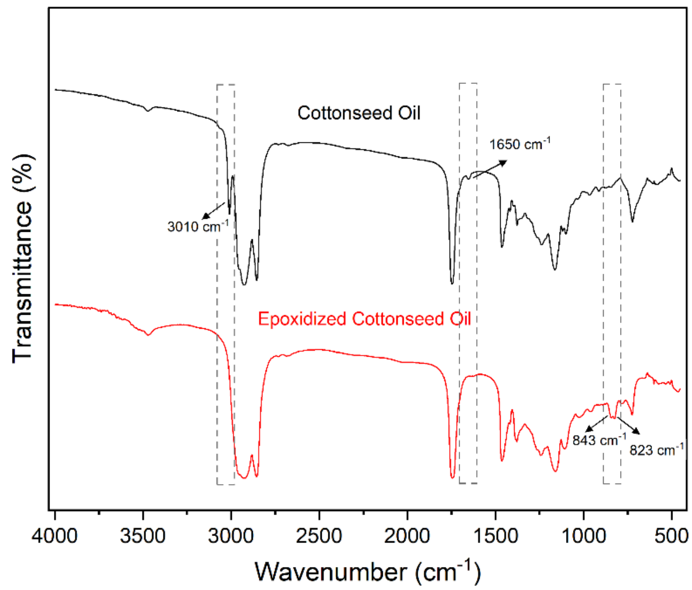


Figure S1. FTIR spectra of cottonseed oil and epoxidized cottonseed oil.

Table S1. Iodine value and oxirane content of cottonseed oil and epoxidized cottonseed oil.

Property	Cottonseed Oil	Epoxidized cottonseed oil
Iodine value (mg I_2/g)	110.8 ± 1.1	0.6 ± 0.02
Oxirane content (%)	-	5.75

The epoxidation of the cottonseed oil was further confirmed by NMR spectroscopy (**Figure S2**). The peaks at 5.5 ppm (vinyl hydrogen in double bonds) **(1)**, 2.8 ppm (hydrogens adjacent to double bonds) **(2)**, and 2.02 ppm (allyl hydrogen in double bonds) **(3)** in the cottonseed oil spectra (**Figure S2a**) almost disappeared after the epoxidation reaction (**Figure S2b**). New signals associated with the presence of epoxide groups can be observed for: methylene hydrogens adjacent to epoxy groups at 15. ppm **(4)**, hydrogens on the epoxy group at ~3 ppm **(5)**, and methylene hydrogens between two epoxy groups at 3.1 ppm **(6)** (Cogliano et al., 2022; Dominguez-Candela et al., 2022; Téllez, 2009).

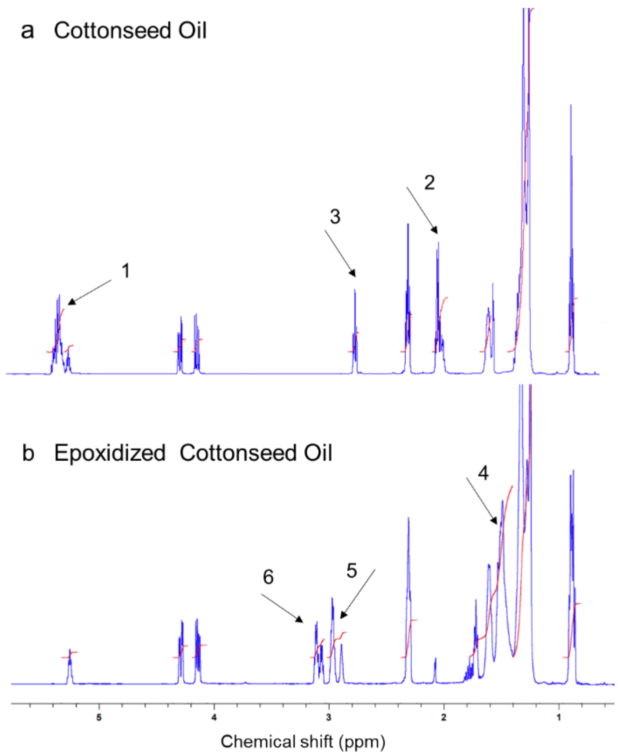


Figure S2. ^{1}H NMR spectra of (a) cottonseed oil and (b) epoxidized cottonseed oil.

2. SEM Images of S-CMF coated paperboards

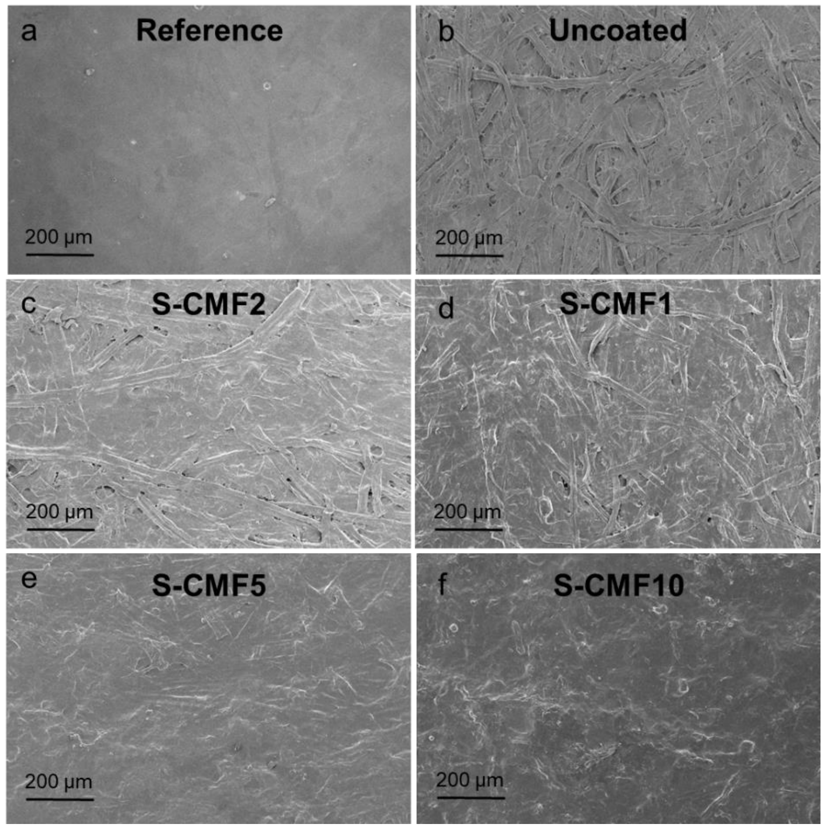


Figure S3. Scanning Electron Microscopy (SEM) images of the surface of (a) a benchmark paper-based food packaging, (b) uncoated paperboards, and S-CMF coated paperboards with (c) 1 layer, (d) 2 layers, (e) 5 layers, and (f) 10 layers.