ABSTRACT

DEBNATH, MRITTIKA. Soft Mechanical Treatments of Recycled Fibers Using a High-Shear Homogenizer for Hygiene and Packaging Bio Products (Under the direction of Dr. Lokendra Pal and Dr. Martin Hubbe).

The pulp and paper industry is one of the largest manufacturing sectors utilizing renewable lignocellulosic materials such as virgin and recycled fibers to produce sustainable hygiene and packaging products. Paper recycling has been practiced extensively to demonstrate good environmental stewardship practices and address energy concerns. Major problems with recycled fibers such as old corrugated containers (OCC) are their inferior properties that arise from hornification, resulting in loss of flexibility and inter-fiber bonding, which affect the strength properties. Mechanical treatments such as high-intensity refining can enhance tensile strength by increasing the surface area and fiber flexibility but at the expense of a high level of fines that reduces the dewatering of the wet paper web.

In this study, an innovative soft mechanical treatment based on homogenization was utilized to examine its effectiveness as a tool for enhancing the recycled pulp quality with respect to tissue and packaging paper production. OCC pulp was mechanically treated using an iFiber homogenizer, Double Disc Refiner (DDR), and a combination of iFiber and DDR in tandem. To evaluate the impact on tissue and packaging properties, hand sheets were prepared from unrefined, iFiber homogenized, DDR refined, and DDR refined + iFiber homogenized OCC pulp fibers. The critical properties such as bulk, softness, tensile strength, and water absorption were measured. Homogenized OCC pulp handsheets showed better bulk, water absorbance, and strength properties without adversely affecting the softness and drainage (freeness) relative to unrefined tissue sheets. Scanning electron microscopy (SEM) images, water retention (WRV) values, and hard to remove water values showed a fibrillation pattern that was consistent with observed trends of bulk,

softness, and tensile strength. It was found that homogenization helps with deflocculating the pulp stock through high-intensity homogenization without significantly affecting fiber quality and fines generation. Therefore, it can be stated that high shear homogenization treatment has the potential to improve the overall quality of recycled tissue and packaging products.

Mechanical modifications of recycled fibers using a high-shear homogenizer for sustainable hygiene and packaging solutions

by
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DEDICATION

To My Son and My family

BIOGRAPHY

Mrittika Debnath was born in a Dhaka, Bangladesh, to her parents in 1991. After completing her high school, she went to University of Dhaka to study Bachelor of Science in Applied Chemistry and Chemical Engineering. During her bachelor, she was interested in organizing cultural activities and photography. After completing her undergraduate, she did her masters from the same department. During her masters, she worked as a visiting researcher in Bangladesh Institute of Public Health. After finishing her masters, she worked as research assistant in Bangladesh University of Engineering and Technology for six months. After receiving her degree, she joined Northern Illinois University as a graduate student. There she received her Masters degree from Chemistry and Biochemistry Department. After finishing her MS there, Mrittika joined North Carolina State University in 2019 to pursue her Masters enroute Ph.D. degree under the direction of Dr. Lokendra Pal and Dr. Martin Hubbe in the Department of Forest Biomaterials. Outside of research, she likes travelling to new destination, photography, playing musical instruments. She also enjoys spending time outdoors with her friends.

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Aurko, who just came to my life like a little sunshine and yet being immensely understanding even at this age (5 months old) towards his mom’s higher study.

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CHAPTER 1. Introduction and Background

The pulp and paper industry is one of the largest industries in the world. It is an end user of the forest sector [1, 2]. Paper products fall into three main categories based on their functions: i) packaging materials for packaging purposes ii) printing and writing papers for graphics and iii) household and sanitary papers, also known as tissue and hygienic paper for wiping and absorbing [3]. Currently, both tissue paper and packaging products are the most promising sectors in the paper industry due to the constant increase in demand [4]. In case of packaging, paper manifests an environmentally friendly tag which makes it the top choice for the packaging industry. It has shown that ~ 47% of total paper and paperboard produced in the year 2000, was used for packaging applications [5]. With respect to tissue paper, it has a wide variety of applications, such as, facial tissue, toilet tissue, napkin, hand towel, kitchen towel [4]. (FAO, 2019). Most importantly, after the severe fallout of the pandemic, the demand for hygiene paper products and packaging products has increased with people's growing awareness of safety and hygiene [6].

1.1 Motivation

Both virgin and recycled pulps have been used as raw materials for making tissue paper and paper board, however, the shortage of wood fibers, high demand of raw materials in paper industry, high water and chemical consumption for recycling operations, China’s import ban on recycled waste, and a decline of forests resources due to the increasing market demand for paper products, recycled fibers are able to complements the need for virgin wood fibers [7, 8, 9, 10, 11, 12]. The main source of recycled fibers is OCC accounting for approximately 40% of the total wastepaper [13]. Therefore, the utilization of recycled instead of virgin fibers could play a significant role in meeting the raw material demand of the paper industry.

However, the major problem with OCC for the inferior properties of recycled fibers is the hornification,a phenomenon characterized by a loss of flexibility and inter-fiber bonding, which affect the strength properties [14, 15]. Although, it is known that high intensity refining can enhance tensile strength by increasing the fiber flexibility and surface area, but this happens at the expense of high level of fines that slow down dewatering of the wet paper web [16, 17].

Moreover, as refining progresses, due to external fibrillation, it causes lowering of softness which is one of the major properties of tissue paper [18]. Thus, it is important to find alternative mechanical treatments that can reverse the negative changes in the recycled fibers mainly through internal and external fibrillation without rigorously affecting fiber like fiber shortening and fines generation.

1.2 Objectives

In this study, we will be considering an innovative mechanical treatment based unit, known as iFiber that can be utilized to solve the issues with refining and OCC pulp during making tissue paper and paper board. iFiber induces both internal and external fibrillation without fiber shortening while deflocculating and separating fibers during homogenization. The fibrillation keeps the original size of the fibers and increases their ability to form strong inter-fiber bonds upon drying while dispersing effect break the flakes and separate the fibers providing better formation. iFiber can also break down big contaminants, thus enhancing the appearance of the sheets. In the second chapter, the mechanism of the unit will be discussed in detail and will be explained. This unit would offer both strength and softness along with other major properties like bulk and water absorbency without significantly affecting drainage during tissue paper manufacturing.

The third chapter will review how to achieve both strength and freeness along with other major properties, such as bulk, without significantly affecting dewatering (freeness) for packaging application. The authors hypothesize that such homogenizing has the potential to both externally and internally delaminate the fibers, thus increasing their potential to form strong inter-fiber bonds which is required for packaging paper.

1.3 Background

Tissue paper has been used for hygiene since the 1940s, and today it is one of the fastest growing industries in the world. It is a type of paper product with low grammage, dry creped in general, but some non-creped papers such as toilet paper, kitchen towels, napkins, facials, handkerchiefs, hand towels and wipes are also available. Even though tissue is recognized by its low grammage, the properties of the paper may vary substantially, depending on the requirements. For example, high wet strength is for very important for kitchen towels while it is an undesirable property in toilet papers. Softness is very important for facials and toilet papers but not required for towels [19]. Target properties for tissue products in the American market change among different applications. Softness and strength are desirable for bath tissue, on the other hand, absorbency is also important for facial tissue. Again, for napkins, strength and absorbency are target properties, while bulk is also important for towels [20]. Softness, absorbency, and brightness are very important for bath and facial tissue, while absorbency and strength are more essential for towel and napkin [21]. Table 1 presents the suggested primary properties of some AFH (Away from Home) tissue products in the Europe Market.

Table 1-1. Summary of Primary or Essential Properties of AFH Tissue Products (Adapted and edited from [22].

Product	Dry Strength	Immediate Wet Strength	Brightness	Softness	Absorption
Toilet Tissue			+	+	
Facial Tissue			+ ++	++	
Hand Towels	+	+			++
Napkins and Serviettes	+			+	

Paperboard is a very popular packaging material based on wood fibers, available in many grades, based on the type of raw materials and the fabrication process. It is like an overlapping network of cellulose fibers that self-bond to form a compact mat. Paperboard based containers are used in wide range of industries around the world such as pharmaceuticals, cosmetics, chemicals, food, bakery, tobacco, soap, textiles, hardware, toys, office products, sporting goods, electronics, and beverages. It can be classified in different ways; for example it can be based on sources which includes virgin or recyclable, based on type of machines like cylinder or Fourdrinier-produced, or based on number of ply like single-ply (known as 'solid' paperboard) or multi-ply. With respect to different shapes and forms like folding cartons, bags, sacks etc., corrugated fiberboard is the largest field where paper board is used. It is mainly used for fabricating shipping containers, but also for cushioning, mechanical protection, and pallet construction as well. The relative proportion of paper and paperboard packaging material production is shown in Figure 1-1 [23].

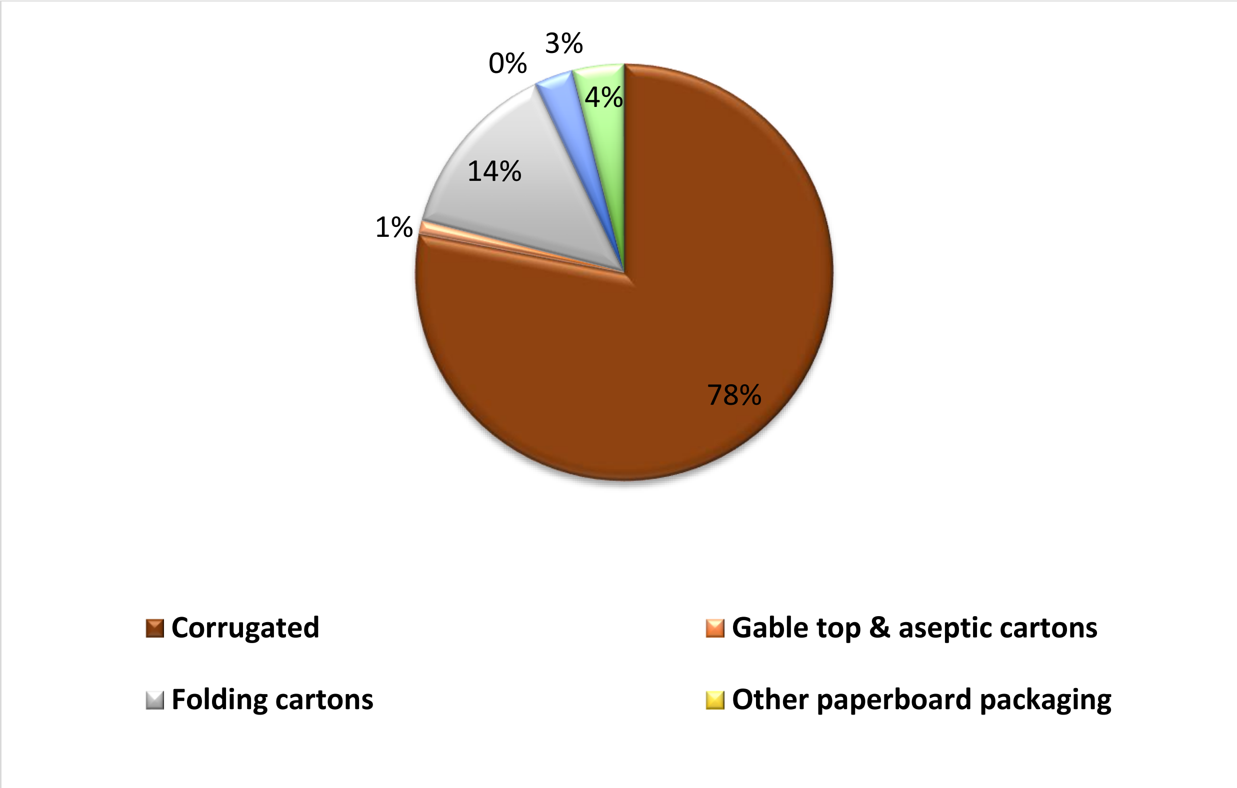


Figure 1-1. Relative proportions of paper and paperboard packaging materials in the USA [24]

Important properties of paperboard are basis weight, thickness (or caliper), density, bulk, stiffness, tensile strength etc. These properties are correlated to various degrees and largely depend on raw materials and production conditions. Basis weight has a direct relationship to the cost per unit area of a package when the board cost is based on weight. Bulk is the inverse of density and is an important determinant of stiffness as stiffness is proportional to paperboard caliper. Mechanical properties of packaging materials are important since they provides protection to the packaged foods by maintaining their integrity during handling and storage. Mechanical properties of bio-based packaging films include tensile strength, tear propagation resistance, penetration resistance, and seal strength [23, 25].

1.4 Raw Materials

Paper based products are made from cellulosic fibers, which forms a bonded random network to adhere as a mat by forming intermolecular hydrogen bonds between adjacent fibers surfaces [26, 1]. Cellulose fibers comes from wood and non-wood plants, the major global raw material in papermaking. The two main sources for wood pulps are softwood (e.g., spruce, pine) and hardwood (e.g., eucalyptus, aspen) trees, and the non-wood-based sources are crops and agriculture residues (e.g., cotton, kenaf). Tissue products are mostly made from hardwoods eucalyptus fibers with a percentage of softwood to confer strength and runnability in the paper machine process. Higher percentages of hardwood fibers improves softness and the addition of softwoods improves wet and dry strength while lowering softness [4, 27, 28, 29].

The fiber wall structure generally composed of cellulose, hemicelluloses and lignin. Cellulose is a glucose containing polysaccharide [30]. Hemicellulose is neutral polysaccharides present in the plant cell wall matrix that can be categorized as xylans, mannans, β-glucans with mixed linkages, and xyloglucans. The type of hemicellulose varies based on type of wood species. The third component of the cell wall is lignin that acts as glue and binds the different layers of cell wall. It can be removed by chemical pulping and bleaching.

The cell wall is divided into different layers: middle lamella (ML), primary cell wall, secondary cell wall and lumen (Figure 1-2). The ML consists of highest amount of lignin and acts as a cementing substance between the cells. The middle lamella surrounds the primary wall (P) which is thin and flexible. It mainly consists of hemicelluloses and lignin and a loose aggregation of microfibrils which are oriented randomly in this layer. The secondary wall is located between the P and lumen and has three distinct layers: S1, S2 and S3. Most of fiber mass belongs to the S2 layer, where the MFA (Microfibril angle) is 10–30° while the S1 layer has a high microfibril angle

(50–70°) and S3 layer is thin with fairly horizontal microfibrils (MFA is 70–90°). The last layer in the cell wall is lumen (*w*) which is the hollow core and can hold the moisture (water or water vapor) [19, 31].

Nearly 40 chains of cellulose molecules are organized into elementary fibrils, which are the narrowest fibrils (diameter 3.5 nm). Several elementary fibrils aggregate to form the microfibrils having diameters with 10 and 35 nm [32]. Finally, macrofibrillars units are formed by aggregations [33, 34]. Macrofibrils are twisted around the cell wall axis and form microfibrillar angle (MFA) which contributes to product strength [35, 36, 37].

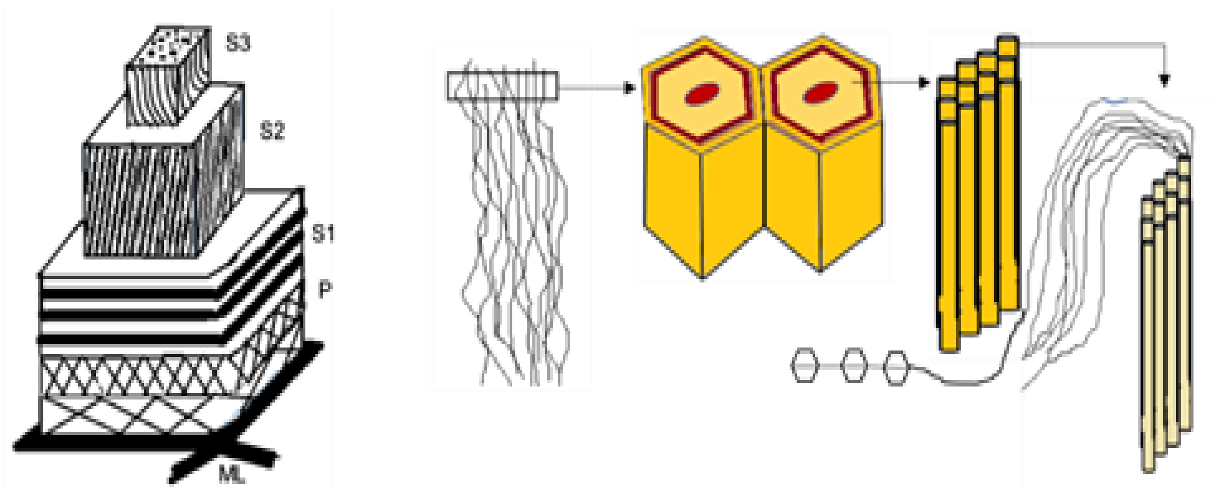


Figure 1-2. Fiber unit structure: (a) schematic of cell wall layers (middle lamella (ML), primary wall (P), secondary wall (S1, S2, S3) and lumen (W); (b) fibrillar structure of cell wall [11]

Both virgin and recycled pulps have been used as raw materials for making tissue paper and packaging paper, since the use of recycled fibers complements the need for virgin wood fibers which can reduce the exploitation of forests and save the biodiversity[7, 38]. Paper made from recycled fiber has comparatively low production cost due to less usage of water, chemicals and energy in comparison to virgin fibers [8, 39, 40]. Therefore despite having less strength, paper production in some countries is being conducted using recycled fibers [15]. The main source of recycled fibers is OCC, which includes 40% of total wastepaper as it is relatively low-priced compared with office waste paper and relatively clean source of fiber without significant fillers or plastic contaminants [13, 41]. Even though both are cellulosic fiber, the differences between recycled fibers and virgin fibers in terms of morphology and surface properties can be observed due to the effect of papermaking process such as refining, which results in inferior strength properties and semi irreversible changes of fiber [12]. The importance and issues with OCC pulp will be explained in detail in next two chapters.

1.5 Refining:

In order to improve the fiber quality of the paper product, all kind of pulp has to go through some fundamental steps, and one of the processes that is conducted in the stock preparation is called “refining” which can be described as a mechanical treatment of the pulp by using the refiner [31]. Refining is an important method to improve the properties of the final product by effecting changes to the mechanical and geometrical properties of the fibers and the fiber bonds [42]. In this process, the fibers go through several changes under compression and shear forces. Based on the initial fiber properties, pulp consistency and refiner specification, the changes in fiber result in higher bonding [43].

In the refiners, fibers are passed between two parallel grooved plates called stator and rotor (Figure 1-3). First, the fibers are accumulated and trapped between the edges of bars. Then the trapped fibers are compressed by the surfaces of the moving and stationary bars after which the fibers are subjected to the shear forces. During the bar crossing, where the fibers also hit the bars on the surface to edge and again edge to edge, two different forces act on the fibers from contact between fibers and bars and another one due to the contact between fibers. The fibers that get trapped between the stator and rotor bars, are treated as the flocs rather than single fiber due to the gap between the bars and the rotor which is as big as the thickness of several single fibers. Three types of forces get transmitted to the flocs: normal force, shear force and corner or edge force (Figure 1- 2). The normal force is due to the compression of the floc between bars, shear force is produced by the movement of bar surface on the floc and corner force appears at the bar edge [44, 45, 46].

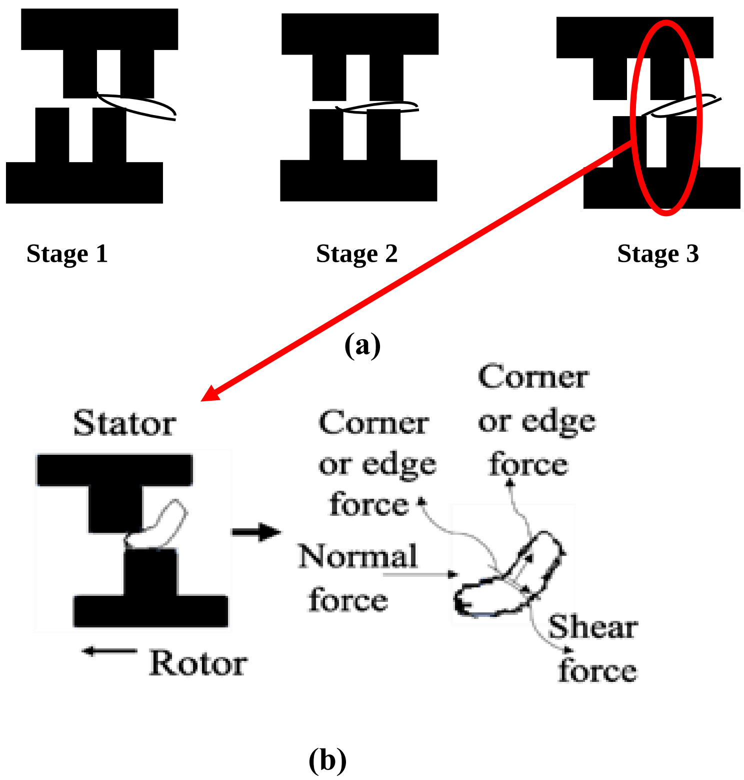


Figure 1-3. (a) Refining mechanism and (b) Forces acting during refining [11]

This whole refining is performed by repeated exposure of pulp between the gap of rotor and stator disc and subjected to repeated cyclic frictional stresses during the rotation of discs. Also, during homogenization, diluted slurries of refined cellulose fibers are pumped at high pressure and fed through a spring-loaded valve assembly and then associated with shearing and impact forces at a repeated cyclical manner as the valve opens and closes in rapid succession. The number of cycles can be 2-30 times, which can cause enormous energy consumption, such as 30,00kWh/t. The main direct effects of refining on fibers are as follows (Figure 1-4):

1.5.1 Internal fibrillation

Internal fibrillation is considered to be one of the main reasons for improving the strength properties of a paper network. It causes delamination of the P and S1 layers of the fiber wall which results in swelling the fibers as water can penetrate into the fibers' wall following the new voids that have been introduced by refining. This increase in the swelling causes better fiber conformability during paper forming and improves flexibility or, equivalently, a reduction of the bending stiffness. The increase in conformability and flexibility leads to greater opportunities for bonding, larger bonded areas and stronger bonds between the fibers [47, 48, 49, 50]. However, due to this flexibility of fiber, fibers entangle firmly together and make a web during draining water causing lowering of drainability [51].

1.5.2 External fibrillation

In external fibrillation, fibrils are peeled outward from the fiber's surface and S2 layers are exposed (while they are still attached to the fiber). The most significant effect of external fibrillation is increasing of the specific surface area of fibrils [52] enhancing the bonding between fibers through mechanisms such as mechanical interlocking and, as a result, it increases relative surface area of the fibers which leads to paper with a higher tensile strength [48, 53, 54]. However, external fibrillation may slow down dewatering because the fiber network becomes more closed; also it may create a negative effect on the tensile strength of paper, which is linked to a reduction of the fiber strength and a weakening of the inter-fiber bonds [19, 55, 56]. Occasionally the un-removed layers are sources of roughness enhancement due to external fibrillation [57].

1.5.3 Formation of fines

During refining process, small, detached fiber pieces are formed, which are generally known as fines. These are basically detached segments of fibers, fibrils, lamellae fragments, ray cells, and so on. Fines can be categorized into two groups based on their shape. The first group includes flake-like or chunky lignin-rich particles which are known as flour stuff, flakes and chunks. These have an adverse effect on the mechanical properties of the paper because they hinder the formation of fiber bonds. The second group of fines includes finer, flexible, and swellable, cellulose-rich particles with a large length to width ratio and good bonding properties. These are usually known as slime stuff, fibrils or ribbons are known to contribute to the increased strength of paper [58, 59, 60].

Also fines can be classified based on the effect of refining. Fines that are already present in the pulp before refining are called primary fines and those created during refining are known as secondary fines. Secondary fines mainly includes fibrils. Because of their small sizes, and good swelling and chemical compatibility, they improve the strength of paper by filling the gaps or openings between fibers, which helps to establish contact between adjacent fibers [50, 61]. Alternatively the formation of bridges between fiber surfaces around the points of fiber crossings have been observed through SEM and other imaging techniques [62]. However, fines creation is also considered as an unwanted effect since it has a negative influence on dewatering. As mentioned earlier, fines have a much larger specific surface area than fibers and can carry almost twice the amount of water per unit dry mass as fibers and also, due to their size, fines also block channels between fibers where water can be drained [19].

1.5.4 Shortening of fibers

Another effect of refining is the shortening of fibers. Usually fiber length is measured by using the statistical average lengths such as numerical or arithmetic, length-weighted and weight-weighted. Shortening of fiber can create negative effect for the tensile strength of paper, though it also helps to improve the formation of paper [63]. However, the shortening of the fibers leads to a better formation [64]. Occasionally, in case of paper with long fiber, it was found that short fibers help to improve the strength of the paper [65].

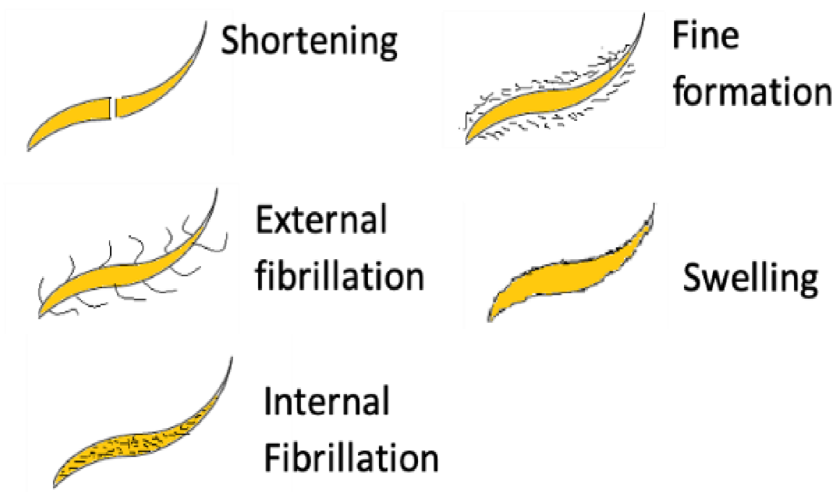


Figure 1-4. Changes in fiber properties during refining [11]

Refining can deforms the fibers by either straightening or curling. Depending on the pulp type and refining equipment, refining can cause both an increase and a decrease in the fiber curl [66]. Curl is measured by the parameter called curl index which is the relationship between the fiber "contour length" and the "projected length" (Figure 1-5) [67]. The relationship is shown below (equation 1-1):

Curl Index= (L/l)-1…… Eq. 1-1

Where L= contour length, l= projected length

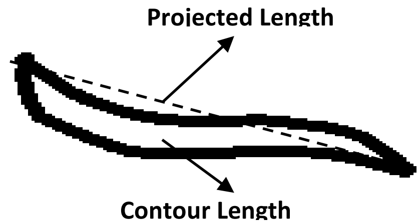


Figure 1-5. Illustration of fiber Curl Index [68]

Curl affects the drainage resistance of most pulps. A reduction in curl index results in a reduction in freeness value [67]. Fiber straightening improves the load carrying ability as well as the stress distribution in the fiber network and mostly increase the elastic modulus and tensile strength of the paper [69, 70].

1.6 Different pulp refining machines

Refiners can be categorized as laboratory and industrial refiners. The widely used laboratory refiners are PFI refiners and laboratory Hollander (Valley) beaters. PFI is a high energy and low intensity refining device in which the pulps are refined between a stainless steel roll with bars and a rotating disk. The pulp is distributed uniformly as a smooth bed in the rotating disk and both bars and disk rotate in the same direction[71]. Valley beater, consisting of the roll bars and bedplate, can refine bigger amount of samples for longer time, compared to PFI. It increases the fiber cutting and the fine formation, whereas PFI mill tends to increase the internal fibrillation and swelling [72].

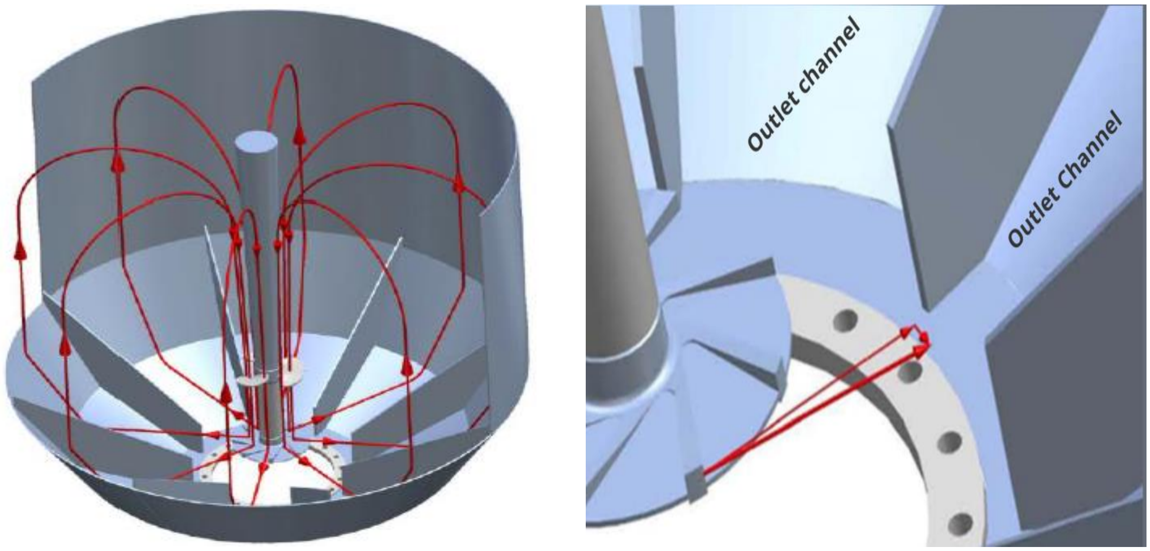
The industrial refiners are classified based on different types of geometries such as disk, conical and cylindrical. They consist of rotor and stator as mentioned earlier, which could have various patterns of plates with different grooves and bar dimensions. The disk refiner is a low cost machine that can be classified into two groups: the single disk refiner and the multiple disk refiners.

Among the multiple disk refiners, the double disk refiners are widely used in the paper mills which can improve the homogeneity and save energy more than the single disk refiners [73].

The biggest issue with a traditional refining rotor/stator system is that the fiber flocs, hanging over the bar edges and receive the first hit and most are cut, which results in shorter fiber. Also as mentioned earlier, even though refining can improve the fiber bonding, in case of OCC, as the number of cycles of fiber reuse increases, each time followed by refining, the strength of papers goes down gradually [74]. Vigorous refining causes the lowering of drainability by increasing fines [75]. Formations of fines causes fiber swelling which is reflected by linear increase of water retention value as well [18]. This is a significant issue with OCC and other reclaimed fibers.

1.7 iFiber unit

In the present work, a new technique of mild mechanical treatment has been introduced to pulp, which is a high-shear homogenizer, an IBS iFiber unit. This unit has a rotor and stator. In this approach, the stock suspension of pulp enters in the iFiber unit, and is immediately accelerated to the rotor (Figure 1-6). The rotor then forces the stock out to the static housing while the stock suspension is circulated within the unit, creating high shear forces. The shear force causes internal fibrillation and homogenization of the stock suspensions. The stator body drives the flow through the outlet channel and doesn’t let it return to the vortex. Fiber flocs are thus dissipated and debris are broken up without vigorously affecting the fiber surface properties. The main role of this unit is to disperse fiber bundles and flocs to provide a more uniform stock to the process. The benefits of the more homogenous stock are improved formation and more consistent pulp. It provides both internal and external fibrillation, but mainly external, which can be observed from the change in different physical properties like strength and fine formation pattern of the fibers due to homogenization treatment compared to that when the pulp is treated with DDR refiner. Also, its maintenance cost and energy consumption is lower than double desk refiner.



(a) (b)

Figure 1-6. **(a)** Cross section view of unit; and (**b)** Flow of pulp suspension

1.8 Conclusion

The iFiber is an innovative approach of high shear based soft mechanical treatment for pulp. Both refiner and iFiber unit accept a continuous stream of stock like a refiner. However, unlike the refiner, its main role is to disperse fiber bundles and flocs to provide uniformity. It not only improves the appearance of the paper, but also the fibrillation keeps the original size of the fibers and increases their ability to form strong inter-fiber bonds upon drying. Most importantly, the lack of fiber shortening and fine generation can be helpful for drainage issue which is a big concern during using refiner. Hence, this type of unit can be utilized for pulp like OCC for manufacturing hygiene and packaging products.

Chapter 2. Soft Mechanical Treatments of Recycled Fibers Using a High-Shear Homogenizer for Tissue and Hygiene Products

2.1 Abstract

This study introduces an innovative approach for developing high strength-high softness recycled fibers through soft mechanical treatment. Recycled fibers from old corrugated containers were treated using a homogenizer, a refiner, and in tandem. The recycled fibers and tissue paper sheets after the treatments were evaluated for the effect on critical properties such as fiber morphologies, freeness, water retention, hard-to-remove water, bulk, softness, tensile strength, and water absorption. High softness and tensile strength were achieved with mechanical treatment by utilizing a homogenizer alone or in tandem with a refiner. Overall, the homogenized recycled fibers and tissue paper sheets provided higher bulk, water absorption, and tensile strength while maintaining the softness and drainage (freeness) behavior similar to unrefined paper sheets. It was found that homogenization helps in deflocculating the recycled fibers under high-shear without negatively affecting the fiber quality, such as fines generation.

Keywords*: Sustainable tissue and towels; Refining; High shear homogenization; Softness; Strength; Absorbency*

2.2 Introduction

Paper webs or sheets are comprised of products made from low grammage, creped and some non-creped papers such as bath tissue or toilet paper, kitchen towels, napkins, facials, handkerchiefs, hand towels, and wipes [19]. These products play important roles in our society, as they contribute to improving hygiene, comfort, sustainability, convenience, and economy. In fact, per capita tissue paper consumption can give an idea about the level of development of a country [76]. Currently, because of the continual increase in demand, the tissue paper sector, along with the packaging product sector, are the most promising area for growth in the paper industry. The global market for printed tissue paper was valued at USD 657.4 million in 2018 and is anticipated to expand at a CAGR (Compound Annual Growth Rate) of 4.5% over the forecast period (2019-2025) [77]. Based on their application and performance, the tissue paper products are highly engineered to provide desired physical properties such as bulk, dry and wet strength, ultra-light weight, softness, and absorbency, *etc*. For example, in the case of kitchen towels, consumers usually expect to have good strength and water absorbency for cleaning and drying wet surfaces, whereas, for bath tissue, users expect softness and comfort [78, 79]. In the case of napkins, strength and absorbency are desired properties, while bulk is also important for tissue and towels [20, 22].

Both virgin and recycled pulps have been used as raw materials for making tissue paper, since the use of recycled fibers complements the need for virgin wood fibers to meet the demands for many reasons [7]. One of the main reasons of using recycled fibers is the decline of forest resources of the world due to the increasing demand of paper production [12] . Therefore, the utilization of recycled instead of virgin fibers could play a significant role in meeting the raw material demand of the paper industry. Recycled fiber-based paper has additional benefits, such as cost-competitive fiber production compared to virgin fibers [8]. Paper recycling operations generally use less freshwater, chemicals [9]and energy [10] compared to virgin fiber production. Moreover, China's import ban on recycled waste has affected the US due to a lack of cost-effective waste management and disrupted the logistics at recycling facilities. However, experts believe that it offers the opportunity to develop better solutions for the growing throwaway culture by recycling and reusing the wastepaper and reducing unnecessary landfilling [39, 80].

A major source of recycled fibers is old corrugated containers (OCC), which represent about 40% of total wastepaper[13, 12]. OCC is mainly composed of used unbleached kraft pulp, bleached kraft pulp, hardwood semi-chemical pulp, and non-wood pulp [79, 74]. The quality of fibers varies a lot, depending on the source. Typically, a corrugated sheet is composed of a corrugated medium from low-quality recycled fibers and a flat linerboard from high-quality virgin fibers. Considering the use of different fibers, most OCC has unique characteristics: (a) high amount of fines and short fiber length (b) slow drainage during papermaking, (c) high amount of contaminants such as inks, stickies, plastics, inorganic particles and polymeric and nonpolymeric materials; and high dissolved solid (mainly primary and secondary fines or material passing a 200 mesh screen)[81].

However, the major problem with OCC for the inferior properties of recycled fibers is the hornification, i.e., the irreversible loss in swelling capacity of the fiber wall resulting from a drying and rewetting cycle [14, 82]. The loss of inter-fiber bonding due to hornification affects the strength properties [15]. As the number of cycles of fiber-reuse increases, pulp characteristics in terms of fibers surface and chemical properties, the strength and WRV of papers goes down gradually [74, 82]. The presence of hydrophobic compounds like stickies on the surface of recycled fiber has been identified as another cause of reduced strength properties [17]. Therefore, research efforts have attempted to improve the quality and lifecycle of reclaimed paper by addressing these undesirable effects. For example, it is known that high intensity refining can enhance tensile strength by increasing the fiber flexibility and surface area, but at the expense of high level of fines that slow down dewatering of the wet paper web [16, 17]. Thus, it is important to research alternative mechanical treatments such as homogenization that can reverse the negative changes in the recycled fibers mainly through internal and external fibrillation without fiber shortening and fines generation.

In the refining process, the fibers are subjected to compression and shear forces, which cause several changes in fiber properties (Figures 2.1 (a) and (b))[83]. Wood-derived fibers mainly consist of primary wall P and secondary wall layers such as S1, S2, and S3 (Figures 2.1(c) and (d)). Refining process causes major changes in fiber, which include: internal and external fibrillation, shortening of fiber length, fines creation, and fiber deformation, such as curling or straightening of fiber, *etc*. (Figure 2.1 (b)) [83] and eventually affect the properties of final tissue paper. Internal fibrillation during refining occurs due to delamination, especially within the S2 layer, caused by the cyclic compressive forces inside the refiner [52, 70]. The breakage of inner bonds between the fibrils causes fiber swelling and thereby makes the fibers more flexible [19, 31, 84] and increases the dry tensile strength. However, high intensity refining caused by the most widely used refiners such as the double disk refiner results in fiber inflexibility as it induces more external fibrillation than internal fibrillation [31]. Flexibility allows the fibers to come into close contact during the pressing and drying processes, which enhances their internal bonding and hence the strength and bulk as well [19, 31]. External fibrillation peels off the outer layers of the fiber surface with exposure of the S2 layer, resulting in an increased relative surface area of the fibers [57]. This may increase paper strength, but after drying it may lower the water holding capacity as the network becomes more closed [19]. Moreover, as refining progresses, due to external fibrillation, it causes lowering of softness [18] .

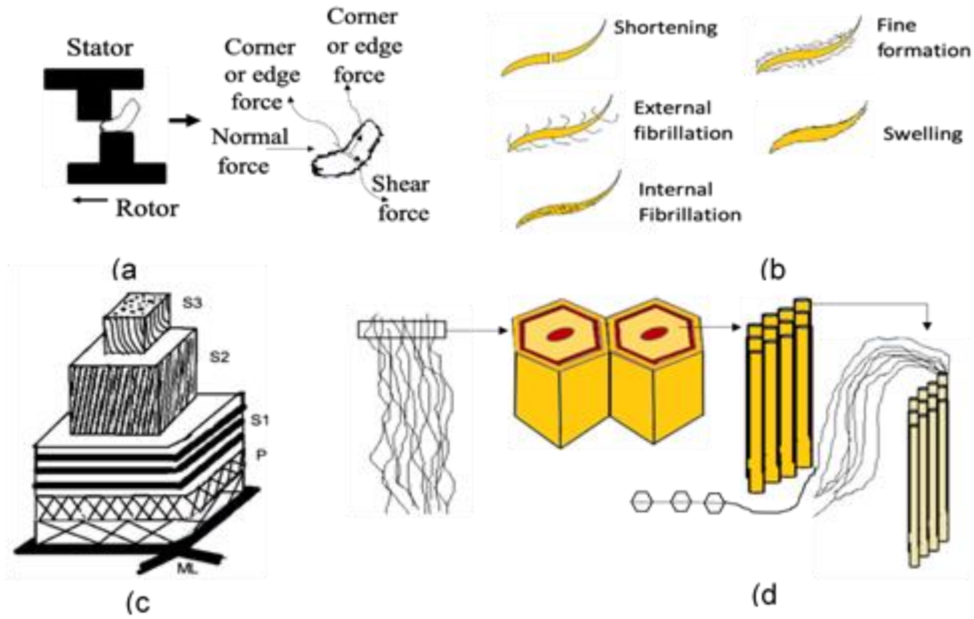


Figure 2-1. Refining process and fiber structure: (a) Forces acting during refining; (b) changes in fiber properties during refining; (c) schematic of cell wall layers (middle lamella (ML), primary wall (P), and secondary wall (S1, S2, S3)); and (d) fibrillar structure of cell wall

Thus, in the present work, a new technique was used to mildly refine the OCC fibers to impart more flexibility to increase the bulk and softness without compromising the tensile properties. This study examines an innovative approach for producing tissue paper products by using a high-shear homogenizer, an IBS iFiber unit. This method would offer both strength and softness along with other major properties like bulk and water absorbency without significantly affecting drainage during tissue paper manufacturing. Unlike a double disc refiner, this unit provides both internal and external fibrillation, but mainly external fibrillation and homogenization, which causes the fiber flocs to disseminate instead of changing the fiber characteristics extensively (Figure 2-2 (a and (b)). In this approach, the stock suspension of pulp in the iFiber unit is immediately accelerated to the rotor (Figure 2-2(c)). The rotor then forces the stock out to the static housing while the stock suspension is circulated within the unit, creating high shear forces (Figure 2-2(d). The hydrodynamic shear causes repeated flexing of individual fibers, which gives rise to internal fibrillation and homogenization of the stock suspensions. Fiber flocs are thus dissipated and debris are broken up without vigorously affecting the fiber surface properties or causing excessive fines formation through external fibrillation.

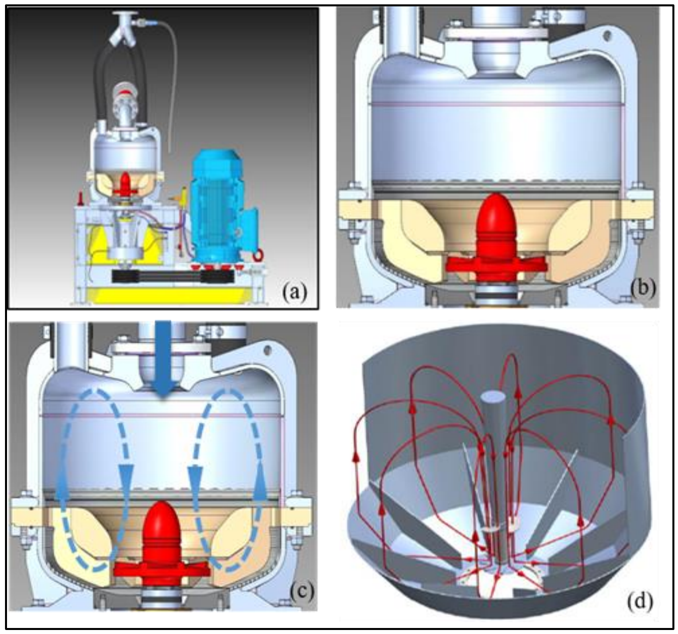


Figure 2-2. (a) iFiber unit; (b) Cross section view of unit; and (c), (d) Flow of pulp suspension

The authors hypothesize that fiber homogenization within the iFiber unit induces both internal and external fibrillation without fiber shortening while deflocculating and separating fibers during homogenization, which is visible in the SEM images (Figure. 2-4 & 2-6). With iFiber, the fibrillation keeps the original size of the fibers and increases their ability to form strong inter-fiber bonds upon drying while dispersing effect break the flakes and separate the fibers providing better formation. iFiber can also break the big contaminants enhancing the appearance of the sheets. Usually such a device has multiple styles of rotors to balance the intensity of the dispersion action and energy demand. For this work, a constant IBS standard rotor at speed ranges of 4600 rpm was used. This study analyzed the effects of iFiber homogenization on OCC fiber, and evaluated the tissue paper properties made from this fiber and compared the effects with and without using a conventional double disc refiner.

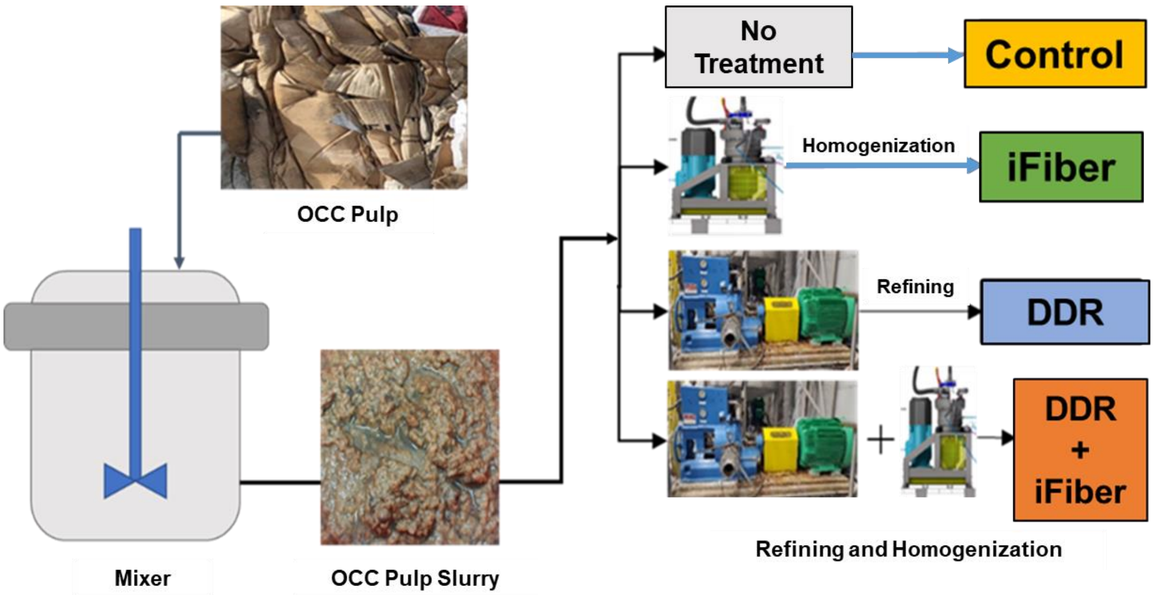
2.3 Methods and Materials

2.3.1 Materials

Old corrugated container (OCC) pulp, which is mainly unbleached softwood kraft pulp, was procured from commercial sources. The chemical composition was as follows: cellulose-60.8%, hemicellulose-21.1%, lignin-13.2%, extractive-0.7%, ash-4.2%. The properties of OCC pulp were as follows: Brightness (%ISO) 15, and pH~5.13. Other physical properties are tabulated in Table 1.

2.3.2 Refining and Homogenizing Process

The small pieces of OCC sheets were added in a hydrapulper filled with water at room temperature with a target consistency of 3.4% in a batch of 50 lbs. After 20 minutes of hydrapulping, each batch of pulp slurry was pumped into a large holding tank. The unrefined pulp was then homogenized using the iFiber homogenizer unit, denoted here as 'H' (supplied from IBS, Chesapeake VA). Homogenization was done at 4600 rpm at 40 gallons/min. The same procedure was followed for refining by double disk refiner (DDR) unit, denoted here as 'R' (Sprout-Bauer Twin-Flo Refiner) (Scheme 2-1) at 33 kWH/t. The DDR refined pulp fibers were sequentially homogenized at the same consistency and same intensity, respectively, and denoted as 'R+H.' An unrefined and unhomogenized pulp sample was used as control and is denoted as 'C.'



Scheme 2-1. Scheme for OCC pulp refining and homogenizing in DDR and iFiber units, respectively

2.3.3 Freeness, Bound water, and Hard to Remove Water Measurements

Pulp freeness testing was conducted using the Canadian standard freeness (CSF) tester as per the T 227 standard method [85]. For the bound water measurement, differential scanning calorimetry was used, where the 20 mg of each sample was placed in the pan. The sample pan was cooled to −20 °C and scanned at 1 °C/min to 20 °C. Overlapping peaks for freezing bound water and unbound water were separated by splitting the integrated areas of heat flow at the temperature of inflection between the peaks [86]. The bound water was quantified by analyzing this melting curve using the following equation –

$$\text{Bound Water, } W_B = \Delta H_{peak} / H_f * W_{dry},$$

Where, ΔH_{peak} is the change in enthalpy, H_f is the specific heat of fusion of water (334 j/g) and W_{dry} is the mass of solids (g).

Hard to remove (HR) water was determined using the thermogravimetric analysis (TGA) technique, where the samples were placed in platinum sample pans for drying. The temperature was raised to 110 °C from 30 °C at 10 °C/min to dry the samples isothermally at 110 °C for 15 min until no more drying occurred. After that, the HR water was calculated from the moisture ratio in the sample determined gravimetrically using a TGA microbalance [87].

2.3.4 Fiber Quality Analysis

The changes in physical properties of fibers were explored using a high-resolution fiber quality analyzer HiRes FQA, OpTest Equipment Inc. The fiber from the dry handsheet sample was disintegrated for the fiber quality analysis.

2.3.5 Preparation and Characterization of Tissue Handsheets

Tissue handsheets were made at a target basis weight of ~40 g/m^2 as per the TAPPI T205 method with a light weight (~ 0.15 kg) foam roller instead of a standard heavy (13 kg) brass roller, without any pressing, and dried twice on a drum dryer at 104 °C. All handsheets were conditioned at 23 °C and 50% RH before testing. The basic tissue properties such as basis weight, caliper, bulk (T258), and softness were evaluated using an Emtec TSA tissue softness analyzer, model BO 458s. The water absorption was studied based on ISO 12625-8, density by T258, and dry tensile strength as per TAPPI T494 standard methods. Morphological analysis of handsheets was carried out by using a Hitachi S3200N variable pressure scanning electron microscope (VPSEM) at an accelerating voltage of 2kV and 13 pA current. The handsheets were coated with Au-Pd before taking the images. The formation of the handsheets were evaluated using the paper perfect formation analyzer, OpTest Equipment Inc.

2.4 Results and Discussion

2.4.1 Evaluation of physical properties of fibers

The refining process resulted in the changes in fiber properties, fiber length, curl, and kink index of OCC, which were analyzed by the FQA. It was observed that the length of iFiber homogenized fiber (H) was higher than the control pulp (C), whereas, DDR refining shortened the fiber length. The difference can be attributed to the effect of homogenizing, as the dispersing action breaks up the fiber flocs, such that longer fibers were released and detected. Similarly for DDR-refined OCC fiber (R), the fiber length was lower than the control sample C as high intensity refining causes shortening of fiber [19]. Again, the DDR refined-iFiber homogenized sample R+H has the higher length than the control C, which can be the consequence of dispersion of fiber flocs caused by the iFiber homogenizer unit.

The percentage of fines formation varied due to different mechanical treatments such as the iFiber homogenization and refining, as shown in Table 2.1. The fines percentage increased when the sample was refined, which can be attributed to fiber shortening [31]. However, the fines content decreased, and fiber length increased when either the control or the refined fibers were homogenized using the iFiber unit due to deflocculation of fibers. Hence, the percentage of the fines of homogenized sample H was lower than control C and the fiber length of sample H was longer than control C. A similar trend can be observed between refined (R) and refined plus homogenized (R+H). From the percentage of fine and fiber length, it can be said that homogenization causes internal and external fibrillation without cutting the fibers and doesn't create fines compared to DDR refining.

Table 2-1. Fiber Properties of OCC pulp Used for Tissue and Hygiene Products

Pretreatment	Sample ID	Fiber Length, Ln (mm)	Fiber Length, Lw (mm)	Fines, (%)	Curl Index (%)	Kink Index (1/mm)	Freeness, CSF, ml
None	C	1.08±0.03	1.89±0.08	7.72±0.23	0.091±0.002	1.25±0.05	630±15
iFiber	H	1.01±0.05	2.01±0.12	4.39±0.18	0.085±0.001	1.02±0.025	657±13
DDR	R	0.93±0.02	1.76±0.10	6.74±0.26	0.078±0.002	1.15±0.036	558±17
DDR+iFiber	R+H	0.95±0.01	1.95±0.06	5.12±0.28	0.081±0.002	1.17±0.04	588±14

The homogenizing effect of iFiber on freeness was analyzed as well. It was found that the homogenization increased the drainability of pulp compared to that of the control and DDR refined pulp Table 2.1. This could be directly correlated with the fines content of the pulp at different conditions. Consistent with a lower proportional fines content in homogenized sample H, the freeness value was higher than the control. The freeness and drainability increased for DDR refined fibers as well when treated with iFiber homogenizer as the proportional fines content decreased due to deflocculation of the pulps.

2.4.2 Bulk and Water Absorption

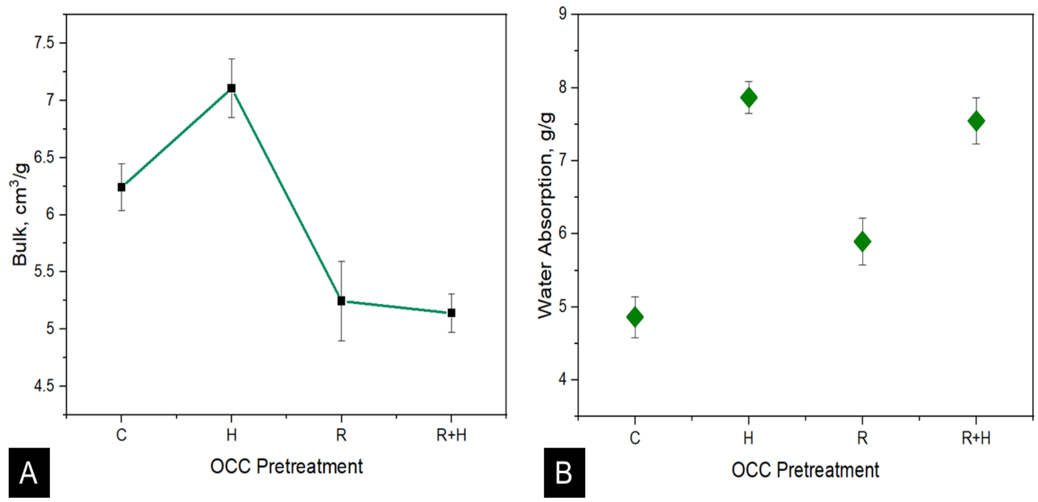


Figure 2-3. (A) Bulk and (B) Water Absorption of OCC fibers at different conditions (control-C; iFiber homogenized-H; DDR refined-R; and DDR-iFiber refined-homogenized-R+H)

Bulk is an important property of tissue paper, as it regulates the porosity, pore volume, softness, liquid absorption potential, air/gas permeability, and often also dimensional stability of the paper [88]. The thickness of paper is reduced when the pulp has undergone high refining. Hence, one of the best ways to produce paper with the highest possible bulk is by mild refining of the fibers [19]. In this study, the iFiber homogenized sample H exhibited the highest average bulk at a given basis weight, followed by the control sample C, DDR refined sample R and DDR-iFiber refined-homogenized R+H (Figure 2-3.A). The decrease in bulk of the DDR refined OCC fibers compared to the control C can be attributed to predominantly external fibrillation of fibers, which increased the surface area and consequently the fiber-fiber interactions. Thus, the fibers were closer to each other and formed more hydrogen bonds as it were dried which resulted in more fiber collapsing. The homogenization of the control increased bulk, since the iFiber unit could maintain the original size of the fibers during deflocculation as the dispersing effect break the flakes and separate the fibers providing better formation and a bulkier sheet. Thus, when dried, iFiber homogenized fibers encounter less collapsing compared to refining and would create handsheetswith bulk. The homogenization of the refined OCC pulp reduced the average bulk slightly due to cutting of flocs during refining and diminishing the effect of homogenization. However, if the standard deviation is considered along with the average bulk, it could be said that the bulk of R+H and R samples does not have any significant difference.

Water absorbency and bulk are correlated with the thickness of the paper. Usually water absorption of a paper product is the measure of its ability to absorb quantities of water at a given mass of the paper at a certain rate until it gets saturated [89, 90]. Along with external fibrillation, as mentioned earlier, refining causes internal fibrillation or breakage of internal bonds; for example bonds between cellulosic fibrils, between fibrils and hemicellulose, between cellulose and lignin, and between hemicellulose and lignin. Due to this internal breakage of bonding, the fibers will have a greater swelling potential because water can penetrate into the fibers [91]. However, with the increase of refining energy through external fibrillation, the network becomes more compact that would lower the water absorption [92]. Here, the OCC fiber after treatment with iFiber unit showed the highest water absorption value among all the four samples (Figure 2-3. B). This sample displayed 38.25% higher water absorption than the control sample C and 33.61 % higher water absorption than DDR refined sample R due to the internal fibrillation which increased the swelling by creating handsheets with higher porosity.

The SEM images as shown in Figure 2-4 depict the effect of homogenization and refining on the bulk. It can be clearly seen that the homogenized OCC pulps showed higher bulk compared to other (Figure 2-4. B). The fibers when homogenized (Figure 2-4. B and D), suffered less collapsing and had higher fiber pore openings due to internal fibrillation. The refined fibers suffered higher collapsing due to overwhelming external fibrillation due to refining. The SEM images directly correlate with the bulk value of the OCC fiber treated at different conditions and support the hypothesis we have proposed earlier.

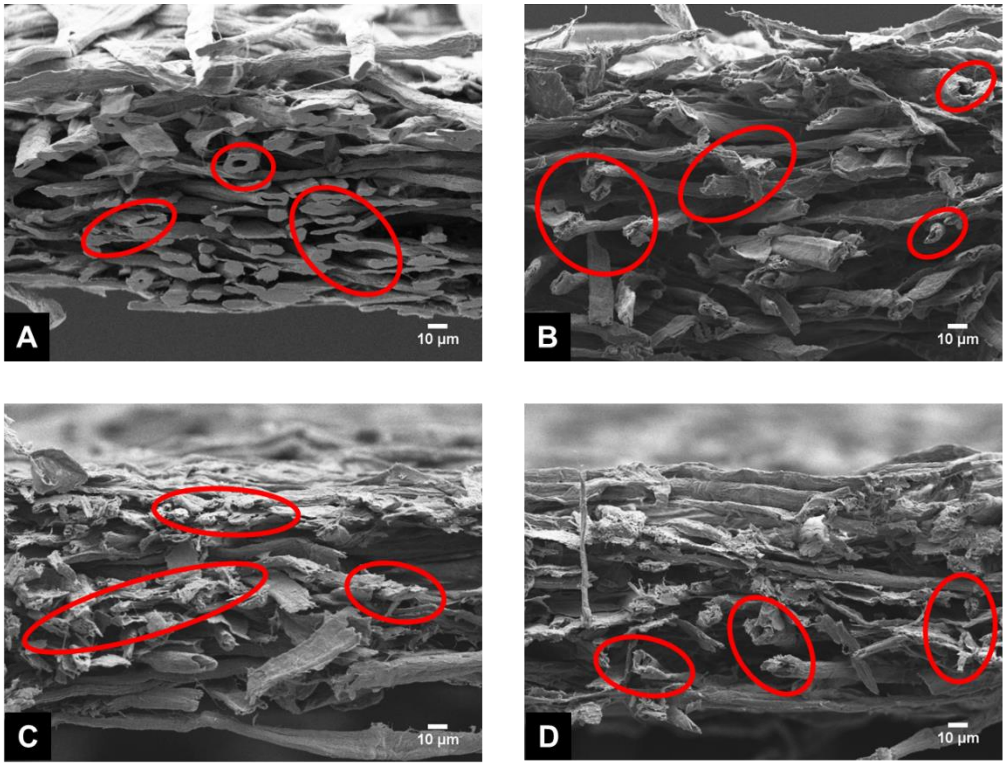


Figure 2-4. SEM of cross section images (A) control-C; (B) iFiber homogenized-H; (C) DDR refined-R; and (D) DDR-iFiber refined-homogenized-R+H OCC fibers.

2.4.3 Softness and Tensile Index

The Emtec softness of the handsheets made from different OCC pulps were measured. Here, the TS7 value increased in both the cases when the unrefined (control) and refined OCC pulps were homogenized with iFiber unit. DDR refined handsheets had the lowest softness, which might be due to the extensive external fibrillation that causes the loss of flexibility of the fibers upon drying [14]. The increase in softness can be due to increase in two types of softness, surface, and bulk softness [93]. The softness in this case increased due to increase in bulk by homogenization treatment as the perception of bulk softness is generated by the light folding and crumpling of a tissue which is related to the overall flexibility. The iFiber homogenization unit imparted flexibility through internal fibrillation which has increased the overall softness compared to the respective control and refined OCC pulp. These findings were in agreement with our earlier reported data within the range of statistical uncertainty [94]

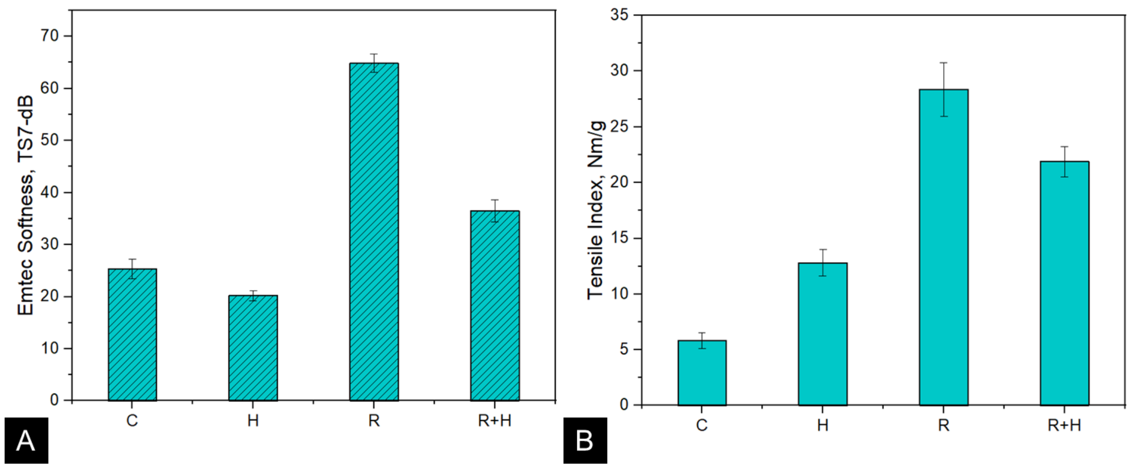


Figure 2-5. (A) Emtec TSA softness; and (B) Dry tensile indices of OCC fibers at different conditions (control-C; iFiber homogenized-H; DDR refined-R; and DDR-iFiber refined-homogenized-R+H)

Tensile strength of tissue paper is considered as the traditional vital feature to meet the market requirements [95]. Along with softness, a tissue paper requires good strength, as it experiences stresses and strains during manufacturing and by consumers as well. It can be observed that DDR-refined handsheets R showed the highest fines content due to vigorous external delamination and consequently the highest strength **(**Figure 2-5.B). The refining process increased the external fibrillation, which in turn increased the tensile strength due to enhanced fiber-fiber interactions which ultimately enhanced the bonding between fibers through mechanisms such as mechanical interlocking [31, 42]. However, due to the effect of refining, the fibers are heavily beaten which affects the softness [14]. On the contrary, iFiber-homogenized sample H showed higher strength and softness than the control sample C, which is interesting as the softness and tensile strength are inversely related to each other [96]. The higher tensile strength for homogenized samples than control was attributed to better fiber-fiber network forming due to deflocculation of the fibers by iFiber unit, which is shown by the SEM image in Figure 2-6.B and reduction in the curl and kink index, which increased the fiber-fiber bonding area and specific bond strength due to reduction of fiber curling. However, the tensile strength of the R+H sample was lower than that of refined sample, though the fiber distribution was more uniform due to homogenization (Figure 2-6.D). The higher tensile strength of the DDR refined OCC handsheets was due to overwhelming fiber-fiber interaction (Figure 2-6.C) due to extensive external fibrillation and an increase in the interacting surface area of fibers due to decrease in its length, as discussed above, and higher fines content, which facilitated stronger fiber-fiber bonding.

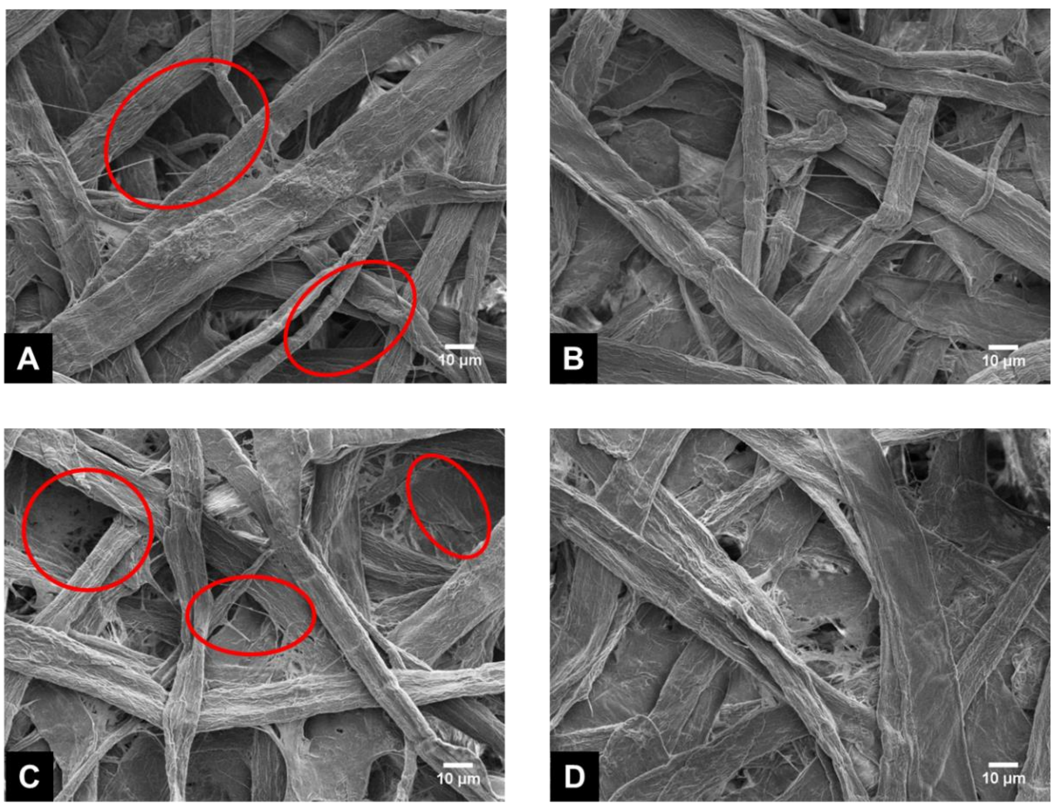


Figure 2-6. SEM of planar images (A) control-C; (B) iFiber homogenized-H; (C) DDR refined-R; and (D) DDR-iFiber refined-homogenized-R+H OCC fibers.

The red circled area in the controlled and DDR refined handsheets shows that the fibers are less homogeneously distributed compared to the respective iFiber treated handsheets.

As mentioned earlier, softness and strength typically have an inverse relationship. It can be observed from the data that the refined sample had the highest strength and the lowest softness. This was because the fibers were heavily beaten during refining, which caused them to bind tightly [97]. For homogenized and refined-homogenized samples, the softness did not decrease as much as the increase in strength when compared with the control. Along with strength and softness, better bulk and water absorption value is also vital for tissue products. Through iFiber homogenization, better strength, and softness along with higher bulk and water absorption could be obtained compared to the control and DDR refined samples.

2.4.4 Formation of Handsheets

The formation tests of the handsheets made from control and different pretreated pulps were conducted and the relative value were calculated as shown in Figure 2-7. The formation values of control handsheets were taken as reference and formation values of handsheets made from other pretreated samples were calculated relative to the control.

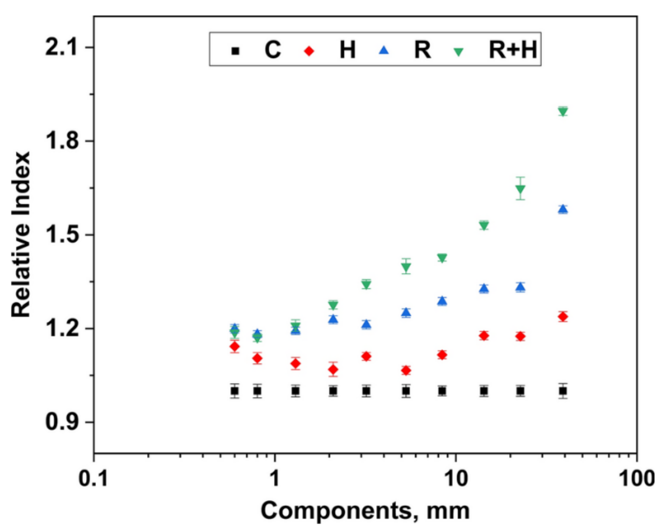


Figure 2-7. Relative formation value of the handsheets at different conditions (control-C; iFiber homogenized-H; DDR refined-R; and DDR-iFiber refined-homogenized-R+H)

It was observed that the relative formation values for each size components were greater than one, i.e., greater than the control Figure 2-7. This means the handsheets of pretreated samples had better formation than the control, since if the value is greater than one, then the tested paper is better [98]. The homogenization by iFiber unit has facilitated better formation for both the control and the refined samples (Figure 2-7). This correlates with the SEM images shown in Figure 2-6**.** The better formation of the handsheets might have resulted in higher tensile properties of the homogenized handsheets compared to control.

2.4.5 Hard to Remove and Bound Water of Fibers

Hard-to-remove (HR) water and Bound water measure the fiber-water interactions. The HR water is a very important parameter to enhance processability and performance of paper products, as the water molecules act as inter-chain bonding agents, leading to closer packing of the chains [87]. The bound water is the type of water adsorbed by the free sorption sites or by hydroxyl groups of cellulose. The combination of the bound water and trapped water during drying are identified as HR water. Thermogravimetric analysis was used to measure the HR water and differential scanning calorimetry method was used to measure the bound water following the method of Park et al. (2006) [99] and Weise et al. (1996) [86], and the results are shown in Figure 2-8**.** The HR water increased for sample R, followed by samples R+H, and control C. The homogenized sample H had the lowest HR water. The increase in HR water content can be attributed to severity of refining, where the refining process increases the surface area and creates more cellulosic hydroxyls available to water through delaminations. Thus, more hydrogen bonds are formed due to fiber-water interactions, which causes an increase in HR water. The decrease the in HR water after homogenization for both the H and R+H conditions was due to decrease in trapped water since the homogenization breaks the fiber flocs and facilitates release of trapped water. However, the bound water, as shown by Figure 2-8.B, increased due to both the homogenization and refining, since both the mechanical pretreatments open up of the submicroscopic spaces within the lamellar structure of the fiber cell wall. Thus, more water could be accommodated into the surfaces and pores of the fiber, where the water molecules reach the newer sorption sites and form a hydrogen bond with the newly exposed cellulose hydroxyls, which increases the bound water of the pulp.

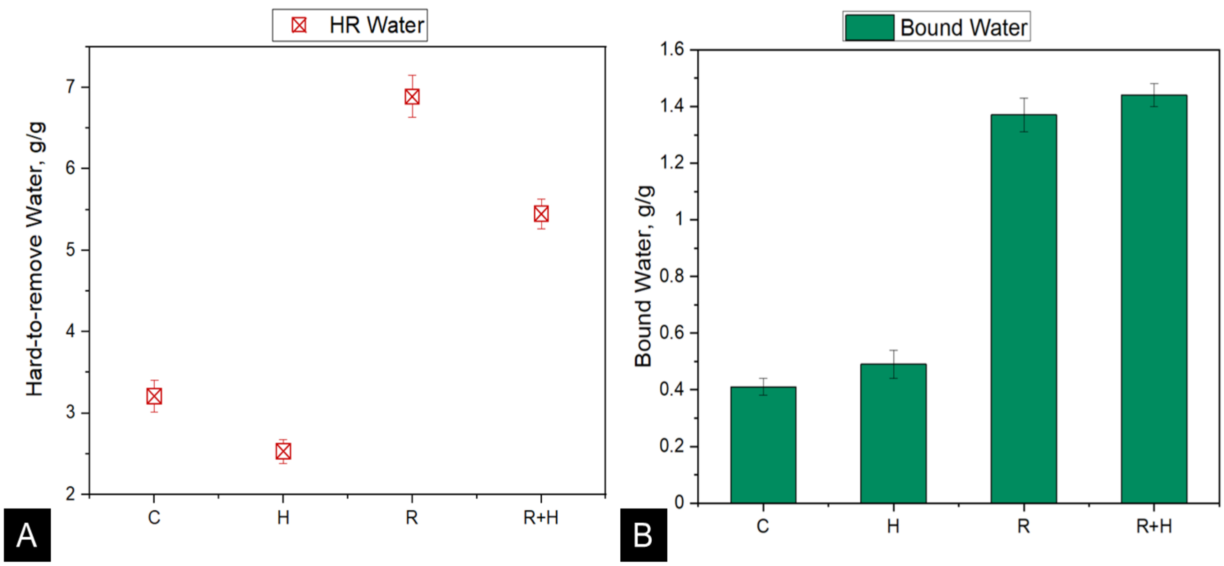


Figure 2-8. (A) Hard to remove water and (B) Bound water value of of OCC fibers at different conditions (control-C; iFiber homogenized-H; DDR refined-R; and DDR-iFiber refined-homogenized-R+H)

HR water also regulates the fiber collapsing and thus the bulk of the handsheets. The higher the HR water of a fiber, the lower will be the bulk of the handsheets made from that fiber as more and more cellulose hydroxyls forms inter-chain hydrogen bonds as the water is removed due to drying [87]. Thus, the OCC handsheets made from sample H showed higher bulk in comparison to the handsheets made from other samples, which is also supported by the SEM images as shown in Figure 2-4. HR water also affects the tensile properties as the dry tensile index of handsheets increases with increase in HR water of the respective fibers as shown in Figure 2-5. B. The tensile strength of handsheets builds up as the fiber-fiber interaction increases as water is removed from the pulp. So, fiber containing higher HR water allows more fiber-fiber interaction through hydrogen bonds as higher number of hydroxyls of fibers become open due to removal of more HR water molecules from fiber. This leads to stronger fiber-fiber interaction and increases the tensile strength. Thus, handsheets formed from samples R and R+H showed higher tensile index compared to handsheets formed from sample H. However, the homogenized H samples showed higher strength than the control though it has lower HR water which was due to better distribution of fibers within the wet-web matrix and reduction of fiber curling due to homogenization.

2.5 Conclusions

This work has examined the effectiveness of mechanical homogenization as a tool for enhancing the recycled pulp quality with respect to tissue production. It was observed that it is possible to achieve high strength in addition to high softness tissue paper from recycled fibers by using a homogenizer. The SEM images, WRV, and HR water values showed a fibrillation pattern that was consistent with the observed trends of bulk, softness, and tensile strength. Even though the strength of tissue paper obtained by using the refiner was higher than from the homogenizer, the softness was reduced significantly. By contrast, higher bulk, water absorbency, and freeness were achieved without affecting the softness of the tissue paper by using the homogenizer. Unlike the refiner, the homogenizer did not lower the freeness value of pulp, which indicated that it did not affect the drainability of pulp, which is a highly desirable result. Therefore, it can be stated that high shear homogenization treatment has the potential to improve the overall quality of recycled tissue and hygiene products.

CHAPTER 3. Innovating Recycled Fibers Using High-Shear Homogenization for Sustainable Packaging and other Bioproducts

3.1 Abstract

The use of recycled fibers from old corrugated containers (OCC) complements the need for virgin wood fibers to meet societal demands for paper and board products. However, the major problems with recycled fibers are loss of strength and physico-chemical properties due to changes in length, flexibility, bonding surface area, and contamination. This work introduces an innovative approach to recycled fibers modification through mild mechanical treatment for developing strength without adversely affecting dewatering. OCC pulp was treated using a pilot-scale high-shear homogenizer, double-disc refiner (DDR), and a combination of homogenizer and DDR. Handsheets were prepared from unrefined, homogenized, refined, and refined + homogenized pulp fibers. The fiber quality and handsheets properties such as fiber length, fines content, freeness, bulk, and strength were measured. It was found that homogenization deflocculates the recycled fibers without affecting freeness, unlike DDR. Overall, the homogenized recycled fiber-based paper provided good bulk and tensile strength without lowering the drainage (freeness).

3.2 Introduction

Packaging plays an important role of our current economy. Along with providing basic protection of goods inside, packaging helps in promoting its value, offers convenience of handling and display, and provides other useful features [101]. According to a Smithers study, the global packaging market is set to reach over $1 trillion by 2021 [102]. Most importantly, living in the era of e-commerce, as more and more consumers embrace online shopping, packaging continues to play a pivotal role in brand's reputation and the consumer's expectation [103].

Three major classifications of packaging are primary, secondary, and tertiary packaging. Primary is normally in contact with the goods, secondary includes larger packaging such as boxes that carry quantities of primary packaged goods, and tertiary is used to assist transport of large quantities of goods, such as wooden pallets and plastic wrapping [104]. However, packaging has also raised concerns over environmental sustainability. Every year, large amounts of packaging materials are wastefully disposed, where large portions of them are made of non-biodegradable and non-renewable elements. Even incineration methods exert adverse impacts on the environment. Hence, consumer preference is shifting away from petroleum-based packaging to more sustainable options, for which paper-based packaging is experiencing a compound annual growth rate of 3.41% [101, 105, 106]. Ideally such materials are 100% biodegradable, as they are based on natural cellulosic fibers and can be obtained by recovery of used paper cardboard, corrugated boards, or molded pulp products, etc. The utilization of such fibers results in a much lower adverse impact on the environment [107].

Paper is a widely used packaging material for food and also for non-food products including bags, wrappings, cups, boxes, folding cartons, composite cans, corrugated fiberboard boxes, etc. [108, 109]. By far the largest application of paperboard is corrugated fiberboard, an inexpensive, lightweight material with high strength–to–weight and stiffness–to–weight ratios which makes it the best choice for the manufacturing of packages for transportation of products [23, 110].

Considering the raw materials for paper packaging, even though both recycled and virgin pulp are used, the shortage of wood fibers, high demand of raw materials in paper industry, energy demands, water and chemical consumption for recycling, China’s import ban on recycled waste, and increasing market demand for packaging products, recycled fibers virtually mandates the need for virgin wood fibers [7, 8, 9, 10, 11]. Also it has been seen that 81% of paper packaging is recycled into new products, whereas in case of plastic, the percentage of recycling is only 18% [111]. Recycled old corrugated container (OCC) pulp has been widely used by most linerboard manufacturers to produce boards of various grades and corrugating medium of different specifications for different industrial packaging purposes [112, 113]. It is one of the main sources of recycled fibers, with a high degree of recycling for many years, accounting for approximately 40% of the total wastepaper [13]. OCC is mainly composed of used unbleached kraft pulp, bleached kraft pulp, hardwood semi-chemical pulp, and grass pulp. Due to rapid consumption around the world that are creating environmental and economic issues, recycling and utilization of OCC as secondary fibers has become important [114, 115].

Major properties of paperboard include basis weight, thickness (or caliper), density, bulk, strength, stiffness, bending, ply bonding, and glueability [31]. The manufacturing paper and paperboard can lead to difficulties maintaining specified strength and targets for caliper and drainability [116, 117, 118], especially when working with OCC pulp, due to hornification, i.e., the irreversible loss in swelling capacity of the fiber wall that affects the paper strength properties [14, 15, 82]. Moreover, the strength and water retention value (WRV) of paper goes down gradually as the number of cycles of fiber-reuse increases [74, 82]. Solutions such as high intensity refining can be considered by using refiners such as the double disk refiner. Generally, in refining, the fibers experience compression and shear forces. Such forces cause several changes in fiber properties which include internal in their wet state and external fibrillation, shortening of fiber length, fines creation, and fiber deformation such as curling or straightening of fiber, etc. [83]. Internal fibrillation makes the fibers more flexible and increases the dry tensile strength due to swelling, generated by the cyclic compressive forces inside the refiner [19, 31, 52, 70, 84]. Flexibility improves the strength and bulk of the resulting paper because the fibers come into close contact during pressing and drying [19, 31]. External fibrillation also increases the strength due to increased relative surface area [57]. Formation of fines, observed during refining, has both negative and positive effects on paper product attributes. Fines have high surface area and can carry almost twice the amount of water per unit dry mass as fiber [119, 19, 31]. However, fines formation, along with fiber swelling due to internal fibrillation reduce drainability. In case of high-intensity refining, it enhances tensile strength by increasing fiber flexibility and surface area, but at the expense of high level of fines that slow down dewatering of the wet paper web [16, 81]. Also it causes fiber inflexibility as it induces more external fibrillation than internal fibrillation [31]. Thus, it is important to find an alternative mechanical treatment that can reverse negative changes in the recycled fibers by a treatment addressing internal and external fibrillation without fiber shortening and fines generation.

In the present work, a new treatment was used to mildly refine the OCC fibers to impart flexibility without compromising pulp's freeness. This study examines an innovative approach for producing paper products using a high-shear homogenizer known as an IBS iFiber unit (Figure 2-2). The goal was to achieve increased strength along with favorable effects on other major properties such as bulk without significantly affecting drainage during packaging paper manufacturing. This mechanism was explained previously in which its mode of operation is distinct from the double disc refiner [11]. The unit was applied to make a handsheet to obtain both strength and softness along with bulk and water absorbency without significantly affecting drainage during tissue paper manufacturing. Currently, the unit was examined to achieve strength and freeness in addition to bulk, without significantly affecting freeness for packaging applications. The authors hypothesize that such homogenizing has the potential to both externally and internally delaminate the fibers, thus increasing their potential to form strong inter-fiber bonds which is required for packaging paper. In the current work, a constant IBS standard rotor at a speed of 4600 rpm was used. This study analyzed the effects of iFiber homogenization on OCC fiber, and evaluated the packaging paper properties made from this fiber and compared the effects with and without using a conventional double disc refiner.

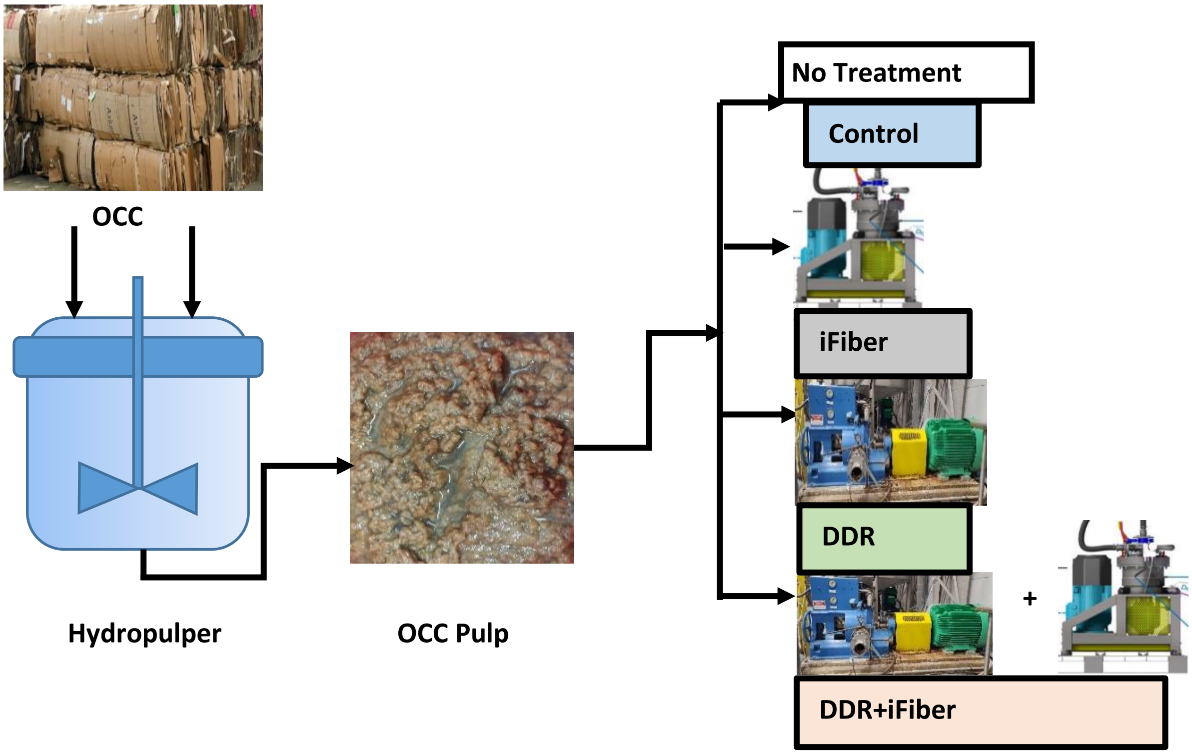
3.3 Methods and Materials:

3.3.1 Materials

Old corrugated container (OCC) pulp, mainly unbleached softwood kraft pulp, was procured from commercial sources. The properties of OCC were as follows: Brightness (%ISO) 15, and pH ~ 5.13. Old Corrugated Container (OCC) pulp was used as the raw material for making handsheets. The properties of OCC pulp were as follows: Brightness (%ISO) 15, pH ~5.0.

3.3.2 Homogenizing Pulp

The OCC pulp was added in a hydrapulper filled with water at room temperature with a target consistency of 3.4% in a batch of 50 lbs. After 20 min of hydrapulping, each batch of pulp slurry was pumped into a large holding tank. The unrefined pulp was then homogenized using the iFiber homogenizer unit, denoted here as 'H' (supplied from IBS, Chesapeake VA). Homogenization was done at 4600 rpm at 40 gallons/min. As an option, the same procedure was followed for refining by double disk refiner (DDR) unit, denoted here as 'R' (Sprout-Bauer Twin-Flo Refiner) (Scheme1) at 33 kWH/t. The DDR refined pulp fibers were sequentially homogenized at the same consistency and same intensity, respectively, and denoted as 'R + H.' An unrefined and unhomogenized pulp sample was used as control and is denoted as ‘C’.



Scheme 3-1. Scheme for pulp homogenizing in iFiber unit

3.3.3 Freeness Test, Bound Water and Hard to Remove Water Measurements

The CSF (Canadian Standard Freeness) tester as per the T 221 "Drainage Time of Pulp" was used [85]. For bound water, differential scanning calorimetry was used following the method explained previously [11, 86]. The bound water was quantified by analyzing this melting curve using the following equation

$W_B = \Delta H_{peak}/H_f * W_{dry}$,

where, ΔH_{peak} is the change in enthalpy, H_f is the specific heat of fusion of water (334 j/g) and W_{dry} is the mass of solids (g).

Hard to remove (HR) water was determined using the thermogravimetric analysis (TGA) technique [11, 87], which measures the capacity of fibers to hold water.

3.3.4 Handsheets Making and Characterization of Their Physical Properties

Handsheets were made at a target basis weight of ~60 g/m^2 as per the TAPPI T205 method. After pressing, all handsheets were conditioned at 23 °C and 50% RH before testing. The handsheets were then tested for basic paper properties such as basis weight, caliper, bulk (T258), strength properties such as dry tensile strength employing TAPPI T494 methods, STFI or SCT (Short Span Compression Test) according to TAPPI Standard 826 etc. Morphological analyses of handsheets were done by using a Hitachi S3200N variable pressure scanning electron microscope (VPSEM) at an accelerating voltage of 2 kV and 13 pA current. The handsheets were coated with Au–Pd before taking the images. To observe the uniformity, the formation of the handsheets were done using the Paper Perfect Formation Analyzer, OpTest Equipment Inc.

3.3.5 Fiber Quality Analysis

To observe the changes in physical properties of fibers, testing was carried out by using a high resolution fiber quality analyzer HiRes FQA, OpTest Equipment Inc. Before testing, the FQA was calibrated and used according to the manufacturers' specifications. The pulp for each sample was disintegrated before the measurement.

3.4. Results and Discussion

3.4.1 Evaluation of fiber properties

Changes in handsheet properties were observed after homogenizing and refining. These were caused by the changes in fiber properties, more specifically in the fiber length, fines content, curl, and kink index of OCC pulps, analyzed by the Fiber Quality Analyzer (FQA). The values of these properties are given in Table 3.1. According to the refining theory, it is known that refining causes decreases in fiber length [31]. Here, it was observed that homogenized fiber (H) had fiber length very close to that of the control pulp (C), whereas, for DDR-refined OCC fiber (R), the fiber length was lower than the control sample C. This finding is consistent with an expectation that high-intensity refining causes shortening of fiber [19]. Again, the DDR refined-iFiber homogenized sample R + H and refined sample R had very similar fiber length, a consequence a consequence of dispersion of fiber flocs caused by iFiber homogenizer unit. Previously, it was observed that the homogenized sample had higher fiber length than the control condition [11]. In the present case, a similar trend was observed in terms of fiber length, but the variation could be the nature of OCC pulp. As a recycled fiber, its sources vary a lot i.e. hardwood fiber, recycled fiber, soft wood fiber, etc. A similar trend was observed in case of curl index and kink index as well.

Coarseness is another important fiber property that is related to flexibility and formation of fiber. Due to flexibility of thin wall fiber, it is generally expected to have low coarseness at a given refining energy. This can be expected to promote tensile strength through high sheet density on account of the higher flexibility of thin-walled fibers where low coarseness implies more fibers per unit mass of pulp and a higher number of bonds per unit fiber length at a given sheet density [120]. Here, the coarseness value didn't vary much.

Table 3-1. Fiber Properties of OCC pulp Used for Packaging Products

Sample ID	**Fiber Length Ln (mm)**	**Fiber Length Lw (mm)**	**Coarseness (L = 0.07 to 10 mm)mg/m**	**Curl Index (%)**	**Kink Index (1/mm)**
C	0.86±0.03	1.64±0.09	0.030	0.095±0.002	1.52±0.05
H	0.85±0.05	1.74±0.11	0.034	0.0890±0.002	1.27±0.03
R	0.82±0.02	1.56±0.1	0.048	0.074±0.002	1.26±0.03
R+H	0.81±0.01	1.66±0.08	0.038	0.069±0.002	1.19±0.02

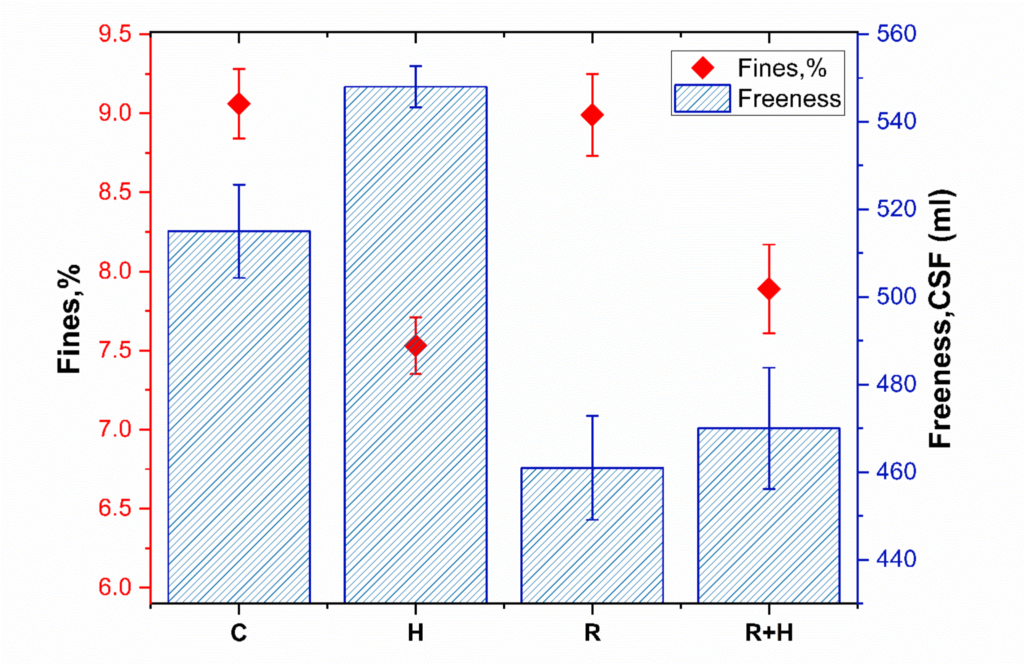
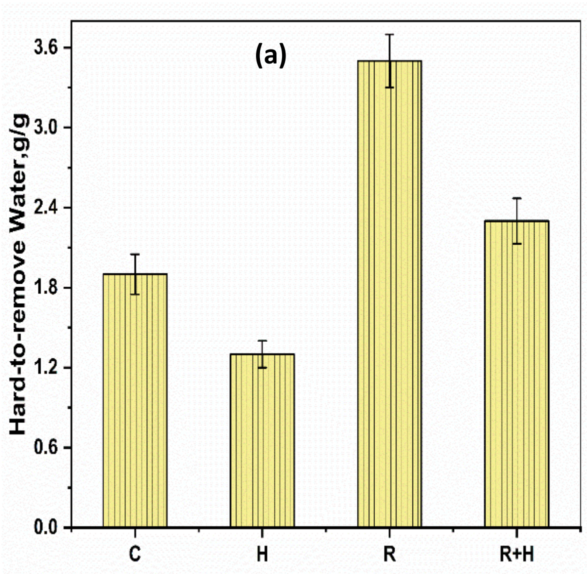


Figure 3-1. Relationship between fine and Freeness of OCC fibers at different conditions (control-C; iFiber homogenized-H; DDR refined-R; and DDR-iFiber refined-homogenized-R + H)

As mentioned earlier, refining produces fines due to shortening of fiber, which increases drainage time. Here, the relation between fines formations and freeness is shown in Figure 3-1. Sample R had a high fines content due the effect of refining. However, due to the effect of deflocculation, the fines content decreased, and fiber length increased when either the control or the refined fibers were homogenized using the iFiber unit. Hence, homogenized sample H had lower fine content but longer fiber length than the control ‘C’. This actually confirms the effect of internal and external fibrillation, which take place during homogenization in the iFiber unit without cutting the fibers or fine creation compared to DDR refining. This can be observed between refined (R) and refined plus homogenized sample (R + H). The effect of fines content has been reflected to the freeness as well. As seen in Figure 3-1, the fines content was inversely related to freeness. For example, the homogenized sample had higher freeness than the control due its lower proportional fines content. The freeness increased for DDR refined fibers when sample 'R+H', was treated as evidenced by the proportional fines content decreased due to deflocculation of the pulps. This means that unlike DDR, the iFiber unit helped to improve the freeness. This trend is consistent with previous data [11].

3.4.2 Hard to remove and bound water of fibers

Hard-to-remove (HR) water and bound water are important parameters to measure fiber-water interactions, which have a great impact on paper making process and performance of paper products [87]. HR water is a combination of trapped water and bound water (adsorbed by the free sorption sites or by hydroxyl groups of cellulose). As demonstrated previously, thermogravimetric analysis was used to measure HR water and differential scanning calorimetry was used to quantify bound water [86, 99]. Figure 3-2**.** shows that sample R had the highest HR water value due to the effect of refining, followed by samples R + H, then control C. The homogenized sample H had the lowest HR water. The refining process makes the cellulosic hydroxyls available, such that the water can create more hydrogen bond as a consequence fiber-water interactions, which causes increase in HR water. By contrast, for homogenized samples, the HR water decreases because of the decreased trapped water as the homogenization breaks the fiber flocs and facilitates release of trapped water. However, the bound water increases with the severity of mechanical treatments, as both the homogenization and refining open up of the submicroscopic spaces within the lamellar structure of the fiber cell wall. Thus, more water could be accommodated into the surfaces and pores of the fiber where the water molecules reach the newer sorption sites and form hydrogen bonds with the newly exposed cellulose hydroxyls, which increases the bound water of the pulp.



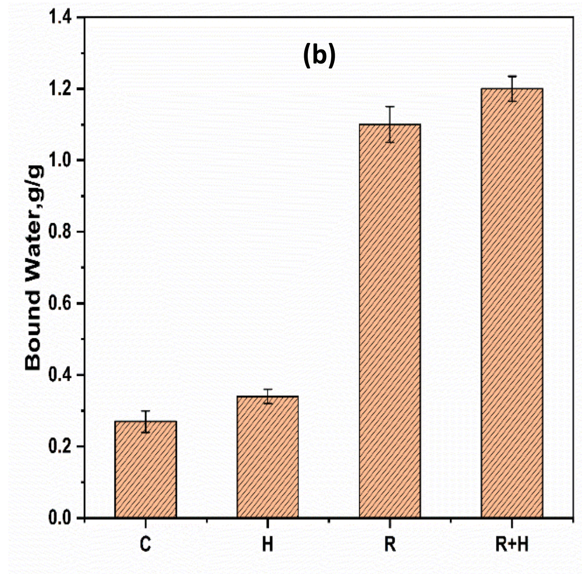


Figure 3-2. (a) Hard to remove water and (b) Bound water value of of OCC fibers at different conditions (control-C; iFiber homogenized-H; DDR refined-R; and DDR-iFiber refined-homogenized-R + H)

3.4.3 Bulk

Bulk is a necessity for many paperboard and packaging products such as carton boards because it increases porosity, pore volume, thermal insulation, sheet thickness, bending stiffness, softness, z-directional compressibility, liquid absorption potential, air/gas permeability, and often dimensional stability [88]. The thickness of paper is reduced due to high refining which causes an increase in the density. Hence, one of the best ways to produce paper with low density or high bulk is by using a milder beating of the fibers [19]. Here, for 60 gsm handsheets, the iFiber homogenized samples had higher or similar bulk to the control sample both for refined and unrefined samples.

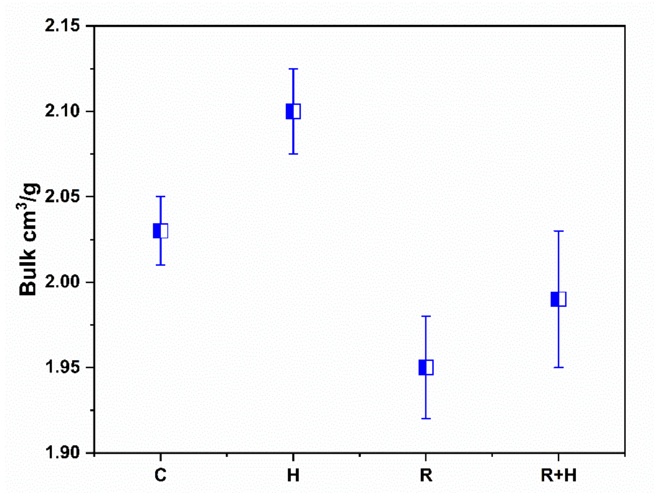


Figure 3-3. Bulk of OCC fibers at different conditions (control-C; iFiber homogenized-H; DDR refined-R; and DDR-iFiber refined-homogenized-R + H)

The SEM images in Figure 3-4 reveal the effects of homogenization and refining on bulk. The homogenized OCC pulps had higher bulk compared to other conditions (Figure 3-3). The fibers when homogenized (Figure 3-4b and d) suffered less collapsing and had higher fiber pore openings due to internal fibrillation. The refined fibers suffered higher collapsing due to overwhelming external fibrillation due to refining. The SEM images directly correlate with the bulk value of the OCC fiber treated at different conditions.

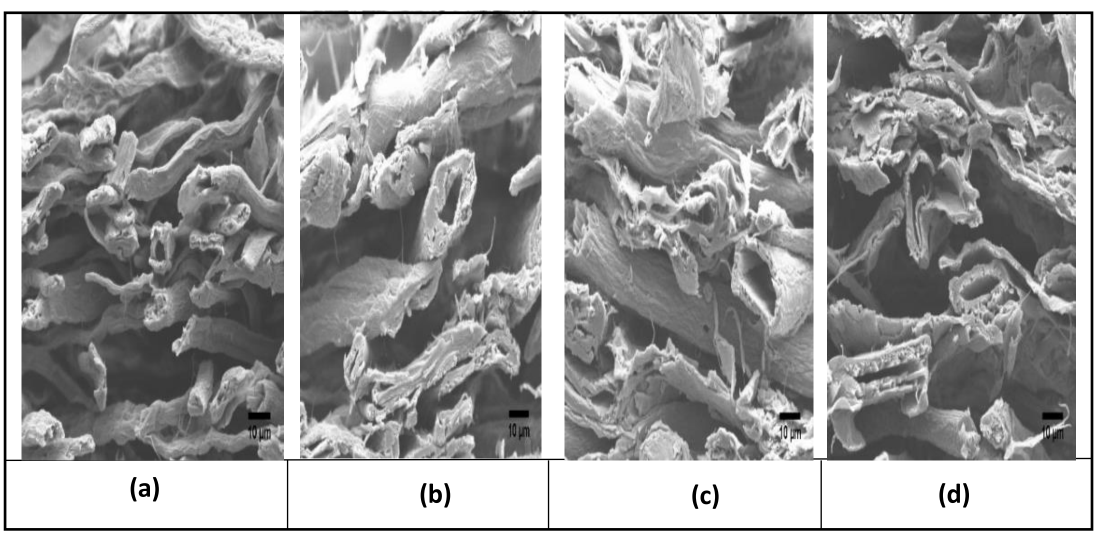


Figure 3-4. SEM of cross section images (a) control-C; (b) iFiber homogenized-H; (c) DDR refined-R; and (d) DDR-iFiber refined-homogenized-R + H OCC fibers

3.4.4 Strength Properties

Strength is one of the most important properties for packaging. To measure strength performance, two important tests were carried out: tensile strength and the STFI short-span test. Compression and tensile strengths are related to fiber modulus and the degree of fiber bonding. The failure of tensile strength is due to a combination of fiber fracture and fiber pull-out separation from the matrix, whereas compression failure consists of a combination of fiber buckling and separation of fiber layers at a visible crease, which is out-of-plane from the sheet [121]. As mentioned earlier, refining increases external and internal fibrillation. It is very effective for improving both the compression and tensile strength.

As shown in Figure 3-5. (a), the refined sample R achieved the highest tensile and compression strength, followed by refined-homogenized sample R+H and then homogenized sample H. The higher tensile and STFI value of homogenized sample than the control was attributed to better fiber–fiber network forming due to deflocculation of the fibers by the iFiber unit. This is shown by the SEM image in Figure 3-6 (b) and reduction in the curl and kink index, which increased the fiber–fiber bonding area and specific bond strength due to reduction of fiber curling. However, the tensile strength of the R + H sample was nearly the same as the refined sample, although the fiber distribution was more uniform due to homogenization (Figure 3-5 (a)). The higher tensile and compressive strength of the DDR refined OCC handsheets were attributed to higher fines content, which facilitated stronger fiber–fiber bonding both for unrefined and refined arising from very strong fiber–fiber interactions (Figure 3-5) due to extensive external fibrillation and an increase in the interacting surface area of fibers due to decrease in its length, and higher fines content.

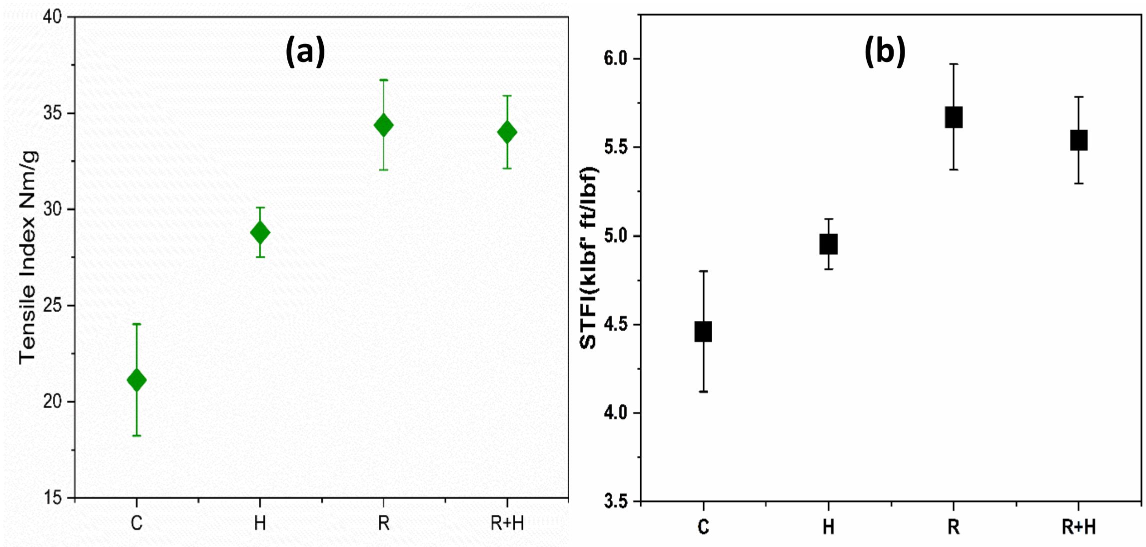


Figure 3-5. (a) Tensile Index and (b) STFI of OCC fibers at different conditions (control-C; iFiber homogenized-H; DDR refined-R; and DDR-iFiber refined-homogenized-R + H)

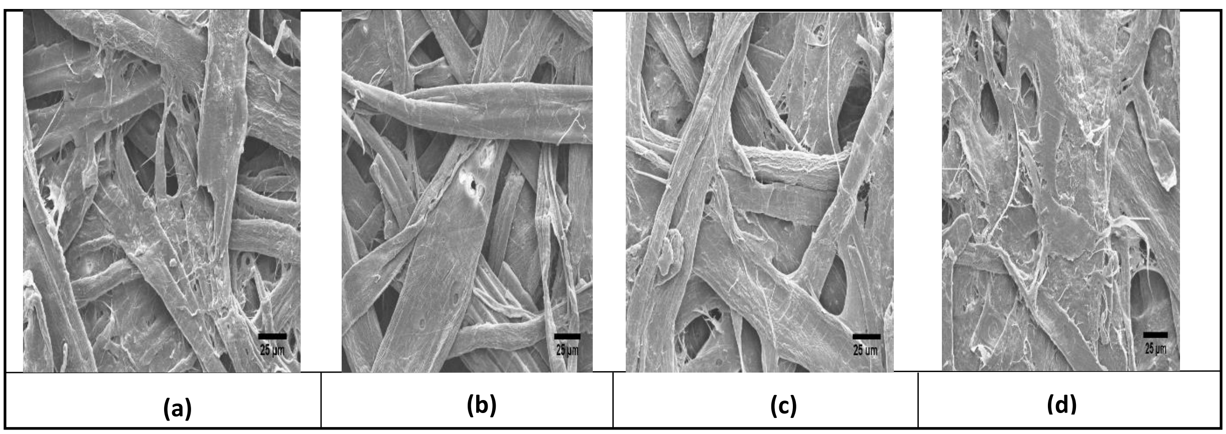


Figure 3-6. SEM of planar images a control-C; b iFiber homogenized-H; c DDR refined-R; and d DDR-iFiber refined-homogenized-R + H OCC fibers

3.5. Conclusions

This work examined the effectiveness of a homogenizing unit, the iFiber, for mechanical treatment of the recycled pulp to be used in making paperboards. It was observed that it is possible to achieve high strength with good bulk. The fibrillation pattern that has been observed in SEM images, bound water and HR water values, was consistent with the observed trends of bulk and strength properties. Unlike other refiners, iFiber didn't lower the freeness value, which indicated that it helps in maintaining the drainability of pulp. It is therefore offered that this high shear force mechanical treatment has the potential to improve the overall quality of recyceled paperboard which can be applied in the future for other pulp-based industrial production.

CHAPTER 4. SUMMARY AND FUTURE DIRECTIONS

This chapter provides a brief summary and gives major highlights to the readers in each chapter. In addition, the limitations with current work and future direction in my understanding to continue the current work is included.

4.1 Chapter 1

4.1.1 Summary

Here, importance of the pulp and paper industry, how tissue paper and paperboard packaging have impacted life, and the effect of fiber properties on the end product are described. The properties of end product largely depend on the types of raw materials, additives, and manufacturing methods. One of the most common steps during preparing raw materials is refining, which effects the fiber properties reflected on the final product and drainage system. Here, the mechanism of refining has been described with respect to the effect of harsh treatment on fiber properties, especially in case of recycled pulp and impact on the end product. Hence, an alternative mechanical treatment is required which finds a solution in the iFiber unit that was introduced for treating pulp. Here, the mechanism of this unit was explained. In case of refining, due to its harsh mechanical treatment, fibers experience fiber shortening and fines generation which negatively impact drainage. In case of the iFiber unit, fiber shortening or high percentage fine generation do not occur because of a soft mechanical treatment which is based on high shear based homogenization. It can be concluded that there is great scope for exploring this unit for pulp mechanical treatment in the pulp and paper industry

4.2 Chapter 2

4.2.1 Summary

This chapter focused on exploring this iFiber unit for recycled fiber based high strength-high softness tissue and hygiene products. Recycled fibers from old corrugated containers were treated with it, a refiner, and in tandem. With regular refiner, it is quite challenging to get tensile strength and softness at the same time for tissue papers, especially with recycled pulp. In order to observe and compare the effect of the two mechanical treatment, the treated fibers and tissue paper sheets were evaluated with respect to fiber morphologies, freeness, hard-to-remove water, bulk, softness, tensile strength, and water absorption. High softness and tensile strength were achieved with mechanical treatment by utilizing the homogenizer unit alone or in tandem with a refiner. Overall, the homogenized recycled fibers and tissue paper sheets demonstrated higher bulk, water absorption, and tensile strength while maintaining softness and freeness behavior similar to unrefined paper sheets. Overall, it was also found that homogenization helps in deflocculating the recycled fibers under high-shear without negatively affecting the fiber quality and fines generation.

4.3 Chapter 3

4.3.1 Summary

The use of recycled fibers from old corrugated containers (OCC) complements the need for virgin wood fibers to meet societal demands for paper and board products. This chapter focused on exploring this iFiber unit for paper based packaging product or paperboard. Similar treatment was carried on recycle pulp OCC using this iFiber homogenizer, a refiner, and in tandem. As mentioned previously, the major problems with recycled fibers are the loss of strength and other properties due to changes in length, flexibility, bonding, and purity. This work focuses on recycled fibers modification through mild mechanical treatment for developing strength without adversely affecting dewatering. The pulp and hadsheet properties such as fiber length, fines content, curl and kink index, hard to remove water, bound water, freeness, morphology, bulk, and strength were analyzed here to evaluate the effect of refining and homogenization. This homogenization treatment not only helps deflocculating the recycled fibers without affecting freeness, but also fibrillation pattern improves the inter fiber bond as well which was reflected on tensile strength and compression strength. Overall, the homogenized recycled fiber-based paper provided good bulk and tensile strength without lowering freeness.

4.4 Future Direction

This is a preliminary study on utilizing this homogenization based mechanical treatment at the pilot scale. This unit has shown promising data for recycle pulp to make hygiene tissue and packaging products, especially for improving drainage rate and maintaining good strength, bulk and softness for tissue. Here, we have mostly focused on treating the pulp for making handsheets. Afterwards, this unit can be utilized for various kinds of virgin pulp both at the lab and pilot scales..

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