

## Prospects for Replacement of Some Plastics in Packaging with Lignocellulose Materials: A Brief Review

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There has been increasing concern regarding environmental problems arising from the widespread use of petroleum-based plastic materials for packaging. Many efforts have been made to develop sustainable and biodegradable packaging materials to replace plastic products. The current review summarizes recent research progress in developing cellulose packaging materials to replace plastics used for cushioning and barrier packaging functions based on pulp fibers, cellulose nanofibers, and regenerated cellulose films to benefit from their renewability, sustainability and biodegradability. The cushioning packaging materials include molded pulp products and bio-based foams. Advanced cellulose films and paper can be good barriers for oxygen and carbon dioxide gases, as well as for water vapor. Several cellulose fiber-based packaging products have been commercialized in areas that used to be occupied solely by plastic products.

*Keywords:* Natural fiber; Cellulose nanofiber; Regenerated cellulose film; Cushioning packaging; Barrier performance

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### INTRODUCTION

In our modern economy, packaging is playing a viable and catalytic role. Not only does it protect the contents, but it also contributes to the convenient transportation, storage, and display of products (Gutta *et al.* 2013). In that context, packaging is one of the fastest growing markets. The global packaging market was valued at \$799 billion in 2012. It continues to grow at 4% per year, and it is expected to reach \$1 trillion by 2018 (PIRA 2017).

Table 1 shows the packaging market trends. It is evident that plastic packaging has been a strongly growing product segment from 2011 to 2016, which is particularly true for flexible packaging. The growth of plastics is mainly due to its low cost, convenience to use with chemical resistance, processing possibility, transparency, strength, and so on (Khosravi-Darani and Bucci 2015; Nurul Fazita *et al.* 2016). The properties of the synthetic polymers used in plastic materials generally are favorable with respect to the processing of films, hot sealing, printing, and integration with other materials in manufacturing operations (Bharimalla *et al.* 2017).

**Table 1.** Packaging Market Trends (Source: Pira 2011)

Materials	Market share (%)		Compound annual growth rate (%)
	2011	2016	
Flexible packaging	20.4	21.6	4.8
Flexible plastic	13.6	14.9	5.4
Flexible foil	1.7	1.7	3.3
Flexible paper	5.1	5	3.4
Rigid plastic packaging	22.1	24.4	5.7
Board packaging	30.6	29.4	2.9
Metal packaging	15.6	14.3	1.9
Glass packaging	6.7	6.4	2.7
Other packaging	4.7	3.9	-0.4

The wide usage of plastic packaging has caused concerns about environmental problems in the world (Greene and Tonjes 2014; Rafieian *et al.* 2014; Campbell *et al.* 2015; Attaran *et al.* 2017; Kuswandi 2017). Plastic packaging materials include PP (polypropylene), PET (polyester), polyethylene, polystyrene, and other petroleum-based polymers. Although most of these plastics are recyclable, in many countries post-consumer plastic packaging waste is rarely recycled because of technical and economic constraints. There are about 1 million tons of plastic waste generated annually in China, but only around 20% of it is recycled (He and Nie 2011). A major proportion of the used plastic materials end up as waste, which is either mainly deposited in landfills or contributes to litter on our roadsides, public spaces, and waterways (Gómez and Michel Jr 2013; Jambeck *et al.* 2015). The strain and stress of environmental balance imposed by plastic packaging materials is a driving force to develop sustainable packaging materials (Peelman *et al.* 2014; Scarfato *et al.* 2015). Natural fibers obtained from forestry and agricultural residues are renewable, completely or partially recyclable, and biodegradable (Johansson *et al.* 2012; Rohit and Dixit 2016). Their relatively abundant availability, absence of associated health hazards, toughness, good thermal stability, and easy surface modification, as well as satisfactory mechanical properties make them an attractive alternative for some plastic packaging materials (Azlan and David 2011; Asokan *et al.* 2012; Zaman *et al.* 2012; Zhu *et al.* 2014; Pan *et al.* 2016; Rohit and Dixit 2016). Packaging based on cellulosic fibers plays an essential role in the storage and transport of goods. In such applications, the cellulosic fibers often contribute greatly to the strength and structural stability of a package. The fibers can be used as self-standing thin films, as filler in composites, and as coating to provide high barrier properties (Ferrer *et al.* 2017). In addition, the fibers can be used to fabricate cushioning packaging materials, and they can serve as reinforcements in biodegradable foam (Bénézet *et al.* 2012; Kaisangsri *et al.* 2012; Ago *et al.* 2016) and as molded pulp products (Didone *et al.* 2017).

In current practices, materials based on fossil resources, mainly petroleum, hold a dominant position with respect to the preparation of barrier layers to resist the permeation of oxygen gas, water vapor, and other compounds that may affect the quality of a product (Ferrer *et al.* 2017). Typical materials for barrier polymer films with thickness between about 10  $\mu\text{m}$  and 250  $\mu\text{m}$  are ethylenevinyl alcohol (EVOH), polyvinylidene chloride (PVDC), and polyamide (PA) (Barlow and Morgan 2013). The most widely used plastic cushioning packaging products are EPS (expanded

polystyrene) and EPE (expanded polyethylene). As post-consumer waste, those polymers are difficult to reuse and recycle because of technical and economic constraints, and they can easily produce chemical contaminants during their disposal by incineration or landfilling (Greene and Tonjes 2014). The ideal solutions entail substituting for them with degradable and sustainable materials.

This review aims to give an overview of natural fibers replacing plastics for cushioning and barrier packaging as well as to provide state-of-the-art examples of cushioning and barrier packaging based on natural fibers to make clear the future prospects, challenges and research and development needs.

## NATURAL FIBERS AND THEIR KEY CHARACTERISTICS

Natural fibers can be obtained from plants including wood, agriculture crops grown mainly to obtain fibers (such as flax, hemp, and sisal), and as the by-products of some crops that have other primary uses (such as wheat, corn, rice, and sugar, *etc.*) using appropriate physical and chemical treatments. The plants mostly consist of cellulose, hemicellulose, and lignin. Pectin, ash, and extractives can be found in lower quantities. The properties of fibers are different depending on the processing technologies, which affect the chemical composition and morphology of natural fibers. Therefore, the applications in packaging materials are different. The key characteristics of natural fibers have been introduced in terms of general cellulosic fibers (pulp), cellulose nanofiber, and regenerated cellulose fibers for packaging (Fig. 1).

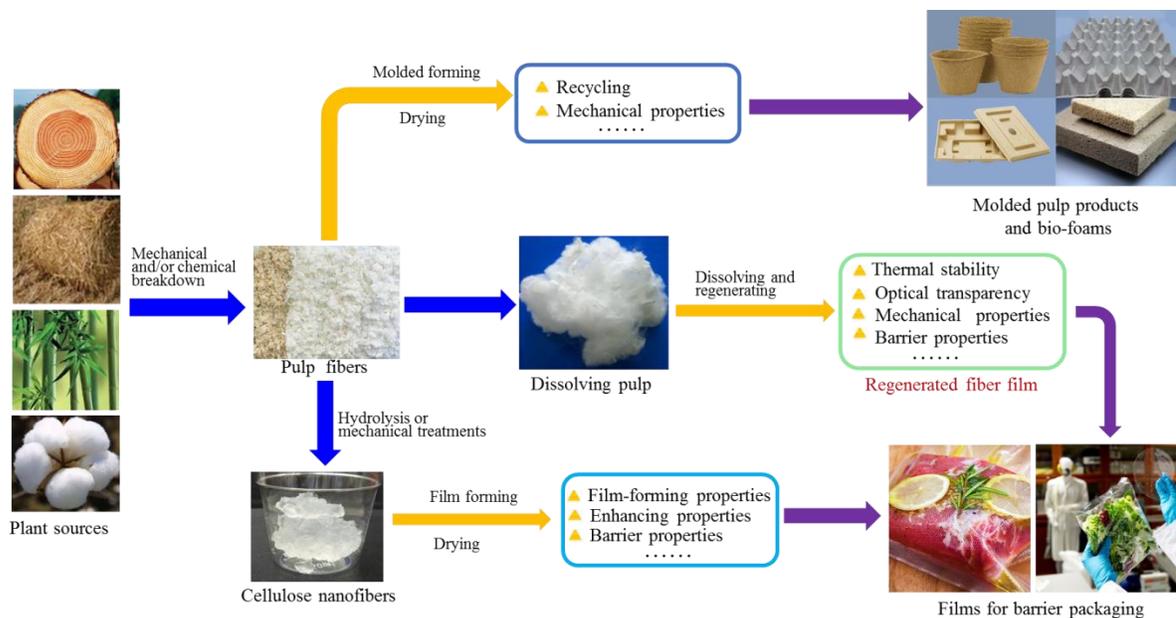


Fig. 1. Molded pulp products/ bio-foams and cellulose- based films as packaging products

### Paper Grade Pulp Fibers

Cellulosic fibers are obtained from plants by mechanical and/or chemical defiberization. They can be mechanical pulp, chemical pulp, and semi-chemical pulp, depending on different pulping processes (Laftah and Wan Abdul Rahman 2016).

Mechanical pulps are manufactured using different mechanical processes, such as refining and grinding, which preserve most of the lignin in the raw materials. Chemical pulps can be almost free of lignin, as the chemical reactions in the pulping and bleaching processes degrade and dissolve lignin to purify the cellulose fibers. The semi-chemical pulp is obtained by means of chemical digestion/pretreatment, followed by a post-refining, which retains most of the hemicellulose and part of the lignin. As the main component of pulp, cellulose can be described as a highly crystalline chain of D-anhydroglucose ( $C_6H_{10}O_5$ ) repeating units that are connected by 1-4  $\beta$ -D-glycosidic linkages at the C1 and C4 positions. The hydrogen bonds are of critical importance in forming cellulose fiber networks, in particular in determining the mechanical properties of the cellulose products.

### Dissolving Pulp

Dissolving pulp is a specialty grade of cellulose pulp that has a high alpha cellulose content (>90%). About 90% of global dissolving pulp production is isolated from wood pulp, and only about 10% comes from cotton linters (Kumar and Christopher 2017). Dissolving pulp from wood is produced chemically by the acid sulfite (AS) process or the prehydrolysis kraft (PHK) process (Duan *et al.* 2015). By means of the AS process, lignin, hemicellulose, and various minor components are separated from the wood material. This is achieved in the acid sulfite cooking and bleaching processes. In the PHK process, the hemicelluloses are degraded and removed in an acid prehydrolysis step. Subsequently, kraft pulping removes most of the lignin, and the required purity for dissolving pulp is achieved after a bleaching/purification step. In comparison to pulp made for paper manufacturing, dissolving pulp requires higher purity, higher brightness, more uniform molecular weight distribution, and higher cellulose reactivity (Chen *et al.* 2016).

Dissolving pulp can be used to manufacture regenerated cellulose films (RC film) with good thermal stability, optical transparency, good tensile strength, and high oxygen barrier properties under dry conditions for packaging (Wang *et al.* 2016). The degree of polymerization (DP) and alpha cellulose content of dissolving pulp can affect the mechanical properties and thermal stability of RC film. Cheng *et al.* (2017) found that less cellulose degradation can provide better mechanical properties and thermal stability of RC film. Pang *et al.* (2015) compared cellulose films prepared from four kinds of cellulose (pine, bamboo cellulose, cotton, and microcrystalline cellulose) with the ionic liquid 1-ethyl-3-methylimidazolium acetate (EmimAc) as solvent. Their results showed that the thermal stability of RC films increased with increasing DP of dissolving pulp, and the tensile stress of the cellulose films from pine cellulose was the highest due to its higher DP and degree of crystallinity.

### Nanocellulose

Nanocellulose materials have a high specific surface area with rich hydroxyl groups and nanoscale morphology. This contributes to properties such as high strength, low density, transparency, barrier properties, and low thermal expansion, which make them ideally suited for packaging films (Bharimalla *et al.* 2017; Mondal 2017). Nanocellulose can be isolated from various cellulosic pulp fibers through mechanical and chemical operations. Nanocellulose can be classified as cellulose nanocrystals (CNCs) and cellulose nanofibrils (CNFs) (Klemm *et al.* 2011). In some publications (Moon *et al.* 2011; Lavoine *et al.* 2012; An *et al.* 2016), the terms NFC (nanofibrillated

cellulose) and MFC (microfibrillated cellulose) are sometimes used interchangeably with CNFs, because the only thing that differs between the designations is higher input of mechanical energy, sometimes supplemented by chemical and enzymatic treatments, in the case of NFC. Thus, products that are called NFC typically have finer fibril size and a generally lower length.

CNCs are often prepared by treating cellulosic fiber with sulfuric acid or hydrochloric acid, which may be followed by a mechanical process, typically sonication. Strongly acidic conditions result in aggressive hydrolysis to attack the non-crystalline portion of cellulose fibers (Islam *et al.* 2014). CNCs are rod-like cellulose crystals with width of 5 to 70 nm and length between 100 nm and several micrometers, depending on the raw materials and processing methods (Ferrer *et al.* 2017). CNCs have excellent mechanical properties, optical properties, high aspect ratio, and well-defined dimensions (Dufresne and Castaño 2017; George and Sabapathi 2015; Scaffaro *et al.* 2017). These properties render CNCs as potential reinforcements in packaging films (Scaffaro *et al.* 2017). Because CNCs have a highly crystalline nature, the oxygen barrier properties of packaging materials can be improved when they are used as filler in composite films by increasing both crystallinity and the pathway for the diffusion of gas molecules (Miao and Hamad 2013; Rhim *et al.* 2013).

CNFs are fabricated by treating cellulosic fibers in mechanical processes. The commonly used apparatus for mechanical treatment are homogenizers, microfluidizers, and microgrinders (Lavoine *et al.* 2012). All of these methods require a large amount of energy input, and there also can be a serious loss of both fibril length and yield. In order to overcome these problems, enzymatic pretreatments and introduction of negatively charged groups (*e.g.*, carboxyl or carboxyl-methyl) on cellulosic fibers have been applied to improve delamination of the nanofibrils (Aulin *et al.* 2009; Saito *et al.* 2009; Siddiqui *et al.* 2011). The width and length of CNFs from mechanical operations are 5 to 100 nm and several micrometers, respectively (Ferrer *et al.* 2017). CNFs have interesting intrinsic characteristics that make them attractive for packaging applications. They exhibit a particularly high specific surface area, high aspect ratio, flexibility, and contain a high amount of hydroxyl groups (Abdul Khalil *et al.* 2016; Mondal 2017; Nechporchuk *et al.* 2016). Unlike CNCs, fibrillated CNFs contain both crystalline and disordered regions. Accordingly, the CNFs have lower crystallinity and higher aspect ratio (Guo and Catchmark 2012; Nair *et al.* 2014a). It is this difference that provides the pure CNFs films excellent oxygen barrier properties due to the increasing tortuosity and entanglements (Nair *et al.* 2014b). The higher aspect ratio together with high surface area leads to a denser network of CNFs film with more entanglements. As a result, the pathway for the diffusion of gas molecules increases, and the film shows better barrier properties. In addition, CNFs can be used as filler to obtain nanocomposites to increase the oxygen and grease barrier properties by making films that have fewer and smaller pores to prevent grease penetration, and are tough enough to resist the development of various defects, such as cracks (Dai *et al.* 2017; Sirvio *et al.* 2014).

Even though cellulose nanofibers can provide excellent gas barrier properties in dry conditions, as well as grease barrier properties and reinforcing performance for packaging materials, their bulk applications are often hindered due to their low resistance to water vapor permeation and poor dispersibility in hydrophobic matrices. The low water vapor barrier properties are derived from the high affinity existing between water and nanofibers (Spence *et al.* 2010).

**Table 2.** Chemical Modifications of Nanocellulose

Techniques	Function		Assessment	References
Non-covalent surface modification	Good compatibility and dispersion in hydrophobic matrix	Adsorption of surfactants; oppositely charged entities or polyelectrolytes.	Environmentally friendly processing; aqueous solution	(Huang <i>et al.</i> 2016a; Pan <i>et al.</i> 2016; Sato <i>et al.</i> 2016; Sobhana <i>et al.</i> 2017; Yan <i>et al.</i> 2016)
Esterification	Pretreatment for mechanical process; hydrophobization	Acetylation, modifying the hydroxyl groups present at the surface of the NFC through the esterification; esterification of NFC with organic fatty acid	Ease and straightforwardness; Involved the catalyst (initiator) and organic solvents	(Kellersztein and Dotan 2016; Singh <i>et al.</i> 2016)
<b>Covalent surface modification</b>				
Silylation	Improving the dispersion in non-polar solvents and matrix; hydrophobization	Silylating the hydroxyl groups present at the surface of NFC	Resulting in the disintegration of the crystal;	(Julkapli and Bagheri 2017; Vartiainen and Malm 2016)
Urethanization	Enhancing the hydrophobicity;	Proceeding with the reaction of an isocyanate with hydroxyl groups on nanocellulose surfaces, giving urethane bonds	Solvent reaction, catalyst for improving the reaction	(Follain <i>et al.</i> 2013; Siqueira <i>et al.</i> 2013)
Amidation	Enhancing the hydrophobicity;	The covalent attachment of amine derivatives on the surface of nanocelluloses with carboxylic functions on oxidized biomass	Reaction in water or DMF	(Guigo <i>et al.</i> 2014)
<b>Polymer grafting</b>				
Grafting onto approaches	Improving the dispersion, compatibility and hydrophobicity	Attachment of previously made polymer chains having reactive end groups, onto cellulose at the surface	Easy to control the properties of the polymer before grafting onto; Reduction of surface grafting density	(Ahmadi <i>et al.</i> 2017; Huang <i>et al.</i> 2016b; Madrid and Abad 2015; Roeder <i>et al.</i> 2016; Wei and McDonald 2016)
Grafting from approaches	Improving mechanical properties and dispersion; providing special functions such as conductivity and thermos-responsive	Polymer is grown <i>in situ</i> from the nanocelluloses with surface hydroxyl groups as initiating sites for ring opening polymerization (ROP)	Increasing the grafting density	(Peltzer <i>et al.</i> 2014; Wei and McDonald 2016)

Cellulose nanofibers as filler have a tendency to aggregate because of the inter- and intra-molecular hydrogen bonding of cellulose, and this results in poor dispersion of the filler in nonpolar polymer matrices (Mondal 2017). In order to improve the water vapor barrier properties and compatibility with nonpolar polymer matrices, various hydrophobic modification methods for cellulose nanofibers have been explored. Table 2 shows some chemical modifications for the functionalization of nanofibers. Such chemical modifications change the interfacial characteristics of the nanocelluloses, often making them more or less hydrophobic or hydrophilic. This can extend their applications as packaging materials.

## CUSHIONING PACKAGING

Cushioning packaging, as a form of single-use foam, can serve the role of protecting and preserving the product in the course of distribution and handling. The most popular cushioning materials include expanded polyethylene (EPE), expanded polypropylene (EPP), expanded polystyrene (EPS), and expanded polyurethane (EPU), due to their strength, light weight per unit volume, and low cost (Razza *et al.* 2015). For example, EPS is a white hard foam that is used extensively in single-use packages for packing commercial and electronic goods. Approximately 95 to 98% of the overall content of EPS is made up of air, and it is lightweight and durable. It is also resistant to chemicals, including some corrosive agents (Tan and Khoo 2005). All those properties make EPS an outstanding cushioning material. However, the post-consumer petroleum-based products are difficult to recycle due to time-intensity and cost, and the fact that they are also non-biodegradable. Concerns arise about the environmental impact of their manufacture and disposal. Developing alternative cushioning materials from renewable and degradable resources meets the need. Starch and cellulose can serve as more eco-friendly alternatives for making foams.

### Molded Pulp Packaging Materials

Molded pulp, as a kind of bio-based single-use packaging material, is also named moulded pulp or molded fibers (IMFA 2017). Starting in 1930s, molded pulp has been employed to make containers, trays and other packages. There was a market decline in the 1970s, coincident with the implementation of plastic foam packaging. Recently the molded pulp market has been recovering (Stratasys 2017). Molded pulp is an excellent protective packaging medium that can be used for electronics, consumer products, retail, industrial products, health and beauty, medical, and scientific items (UFP 2017). The pulp can be molded into whatever shapes may be sought to cushion and immobilize the contents in a package. Compared with EPS and corrugated paperboard, molded pulp has its own properties for cushioning packaging (Table 3) (EnviroPAK 2017).

Molded pulp products are manufactured with waste paper or other natural fibers, which are recyclable, biodegradable, and compostable. They can also be incinerated with less environmental impact. The International Molded Fiber Association (IMFA) has identified four main types of molded pulp. These have been categorized based on the manufacturing process: type 1) thick wall; type 2) transfer; type 3) thermoformed (thin-wall); and type 4) processed (IMFA 2017). The characteristics of the four types are shown in Table 4 (Didone *et al.* 2017; IMFA 2017).

**Table 3.** Molded Pulp vs. Alternates

	Molded Pulp	EPS	Corrugated paperboard
Material	Plant fiber	Petroleum	Plant fiber
Sustainability	Recyclable, biodegradable & compostable	Non-biodegradable	Recyclable, biodegradable & compostable
Cushioning	Excellent vibration & cushioning properties	Good vibration & cushioning properties	Inconsistent vibration & cushioning properties
Shipping & Storage	Easily nests	Does not nest	Requires labor and assembly
Climate Tolerance	Unaffected by extreme temperature or humidity	Temperature affects brittleness	Humidity affects performances
Static	Neutral	Requires treatment with antistatic agents	Neutral
Protection	Geometry	Density	Complicated origami

**Table 4.** The Characteristics of Four Types of Molded Pulp Products

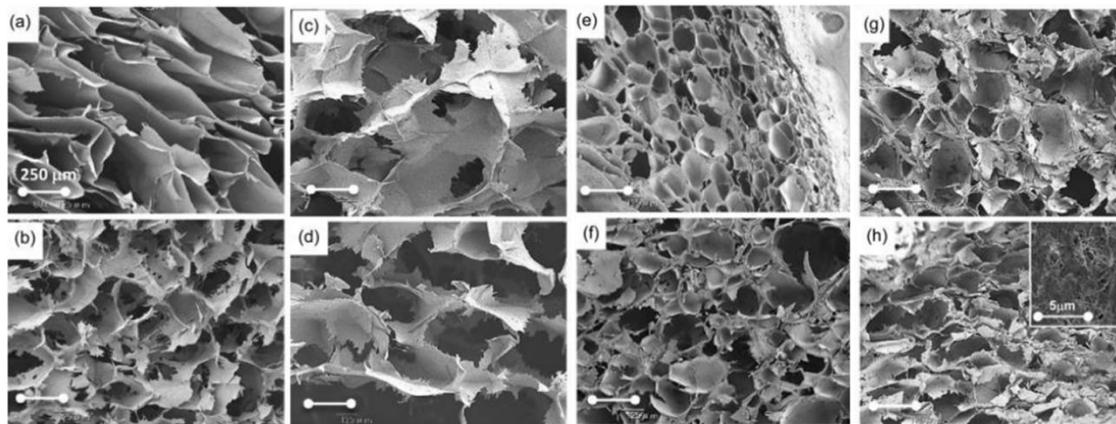
	Thick wall	Transfer	Thermoformed	Processed
<b>Thickness</b>	5 to 10 mm	3 to 5 mm	2 to 4mm	
<b>Material</b>	Recycled paper	Recycled newspaper	Virgin wood fibers	molded fiber products that require some type of secondary or special treatment like additional printing, coatings or additives
<b>Molded process</b>	One single mold	One forming mold and one transfer mold	Multiple heated molds, dried in the mold	
<b>Surface property</b>	One side smooth, Another side rough	Both sides smooth, Better dimensional accuracy	Good dimensional accuracy, smooth rigid surface, resemble thermoformed plastic material	
<b>Typical application</b>	Non-fragile and heavy items	Egg trays or electronic equipment	Healthy and beauty, medical and scientific products	

Molded pulp products have developed quickly in recent years due to favorable environmental, social, and economic incentives (Curling *et al.* 2017; Didone *et al.* 2017; Gouw *et al.* 2017). More active research and development activities are still ongoing, for instance, the use of additives for value-added products (Didone *et al.* 2017), as well as the use of agricultural wastes and/or recycled fibers replacing virgin wood fibers. Curling *et al.* (2017) examined the feasibility of replacing virgin wood fibers with waste cereal straw fibers to fabricate thin flat pulp molded materials. Three different ratios (100% straw pulp, 80/20 straw pulp/kraft, 60/40 straw pulp/kraft) were studied. The results showed that the 60/40 sample had higher mechanical properties than the EPS control sample (although at a higher water absorbance), and it was found to be feasible to produce thin flat pulp molded materials with cereal straw fibers which possessed the required mechanical properties and biodegradability. Gouw *et al.* (2017) studied to produce molded pulp board using fruit pomace (FP) to partially replace recycled newspaper (NP). With the addition of optimum amount of CNF, their results

showed that FP could partially substitute NP to produce molded pulp board with better or similar properties to 100% recycled NP board.

### Bio-based Foam as Packaging Materials

Starch-based foams have been used to replace EPS (expanded polystyrene) and EPE (expanded polyethylene) (Salgado *et al.* 2008; Bénézet *et al.* 2012; David and Rum 2015). EPS and EPE, due to their non-biodegradability, pose a negative environmental impact. Starch-based foams are promising alternatives to EPS and EPE due to their biodegradability, low material cost, and low density (Stevens *et al.* 2010). Starch can be processed in various ways to make products having some properties similar to petroleum-based plastics. By processing it in different ways, starch can be employed to make packaging materials that resemble petroleum-based polymers in key respects. Starch can be used to obtain foams by processes including swelling, gelatinization, and network building with the use of extrusion and compression/explosion technologies (Glenn and Orts 2001; Carr *et al.* 2006), as shown in Fig. 2 (Ago *et al.* 2016). The main technical challenges with starch bio-foam are low elasticity, high stiffness, high brittleness, and high water absorption (Shogren *et al.* 2002; Svagan *et al.* 2011; Phaodee *et al.* 2015). In order to improve the strength and water resistance, researchers have added mineral fillers, wood fibers, resin, or coating with wax and other materials (Andersen *et al.* 1999; Fang and Hanna 2001). Thanks to their excellent mechanical properties, cellulose fibers can be used as additives to the starch matrix to increase the strength properties, such as tensile strength, stress performance, and toughness (Salgado *et al.* 2008; Bénézet *et al.* 2012; Li *et al.* 2014). Besides that, fibers together with components such as chitosan or natural latex can create synergistic effects to further improve the functional properties of the bio-foam (Kaisangsri *et al.* 2012; Phaodee *et al.* 2015).



**Fig. 2.** Scanning electron microscopy (SEM) micrographs of cross sections of LCNF(Lignin-containing cellulosic nanofibrils)/AP(Amylopectin) biofoams: BP(lignin-free CNF)/AP50(50% starch) (a); N(2.3% lignin CNF)/AP50(50% starch) (b); M(6.2% lignin CNF)/AP50(50% starch) (c); F(9% lignin CNF)/AP50(50% starch) (d); BP(AP-free) (e); N(AP-free) (f); M(AP-free) (g); and F(AP-free) (h); Reprinted with permission from Ago *et al.* (2016). *ACS Sustainable Chemistry and Engineering* 4(10), 5546-5552, Copyright (2016) American Chemical Society.

Many factors can contribute to the mechanical properties of a fiber to be used as reinforcement in polymer composites. These include fiber-matrix adhesion, the volume fraction of the fibers, the fiber aspect ratio ( $l/d$ ), and the fiber orientation (Carr

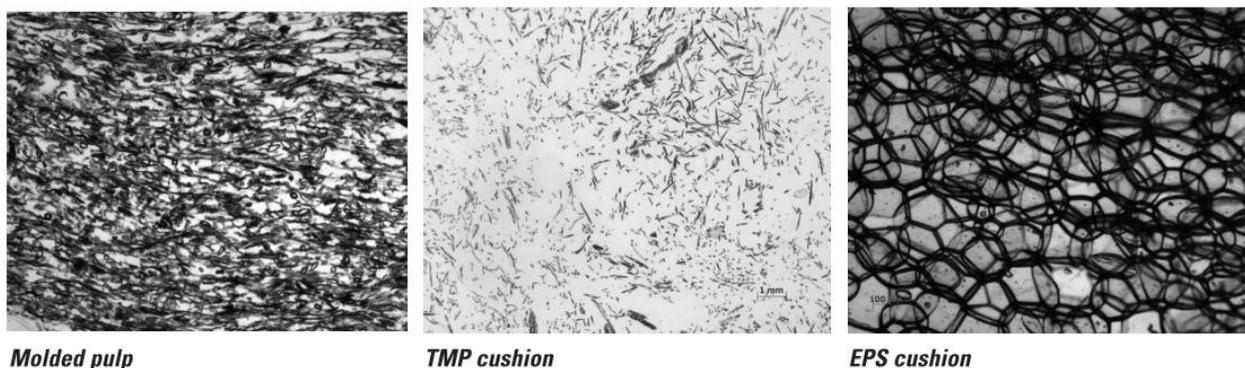
*et al.* 2006). Specifically, the excellent adhesion between fibers and starch plays an important role to improve the strength of bio-foams. Although fibers and most starch blends appear to be compatible because of the chemical similarity, the incorporation extent and distribution of fibers in the starch matrix, as well as the molecular interaction between cellulose and starch determine the adhesion. Wollerdorfer and Bader (1998) found that a fiber content of 25% in starch matrix improved the tensile strength of composite, with an increase up to 35%. Lawton *et al.* (2004) added aspen fibers to backed cornstarch foam to upgrade its mechanical properties. The fiber content of the starch-fiber composite foam ranged from 2.5% to 45%. With increasing cellulose fiber content, the strength of the foam trays increased up to a fiber content of about 15%. When trays contained more than 30% fibers, the tray strength became lower. The lower tray strength was attributed to non-uniform distribution of fibers when the fiber content was high. Phaodee *et al.* (2015) also thought that an uneven nanocellulose distribution within the biofoam structure could cause failure during elongation tests. This was despite the fact that an increase of nanocellulose content from 10% to 20% substantially enhanced flexural strength and modulus of bio-based foam from 1.4 and 74.4 MPa to 14.4 and 533.5 MPa, respectively. Svagan *et al.* (2011) prepared bio-foams using freezing/freeze-drying technology and improved the strength of bio-foams with cellulose nanofibers. There were two reasons leading to the improved strength. On the one hand, cellulose nanofibers could form a strong network structure in the starch matrix to soften the matrix. On the other hand, the molecular interaction between cellulose and starch was favorable to promote the cellulose nanofibers dispersion and increase the adhesion of nanofibers-matrix.

In order to further increase the strength of bio-foams with fibers, synergistic effects were created by adding other components. Kaisangsri *et al.* (2012) developed biodegradable foam trays from cassava starch blended with kraft fibers and chitosan. The foam produced with 30% kraft fibers in a matrix with cassava starch and 4% chitosan had properties resembling that of polystyrene foam. The density, tensile strength, and elongation of the starch-based foam were 0.14 g/cm<sup>3</sup>, 944.4 kPa and 2.43%, respectively. The tensile strength of the cassava starch-based foam increased significantly with increasing fiber content and chitosan concentrations, probably due to the interaction of kraft fibers with chitosan and cassava starch. Crosslinking was used to make bio-based polystyrene foam-like materials with glutaraldehyde and thermal compression molding technique; the effects of lignin, natural rubber latex (NRL), nanocellulose, and talc were studied relative to production of biobased foam using cassava starch as matrix (Phaodee *et al.* 2015). Increasing nanocellulose content from 10% to 20% led to enhanced flexural strength and modulus of bio-based foam. The improvement was attributed to strong interfacial interactions between nanocellulose, crosslinked starch, and NRL.

A greater enhancement due to fibers in a starch matrix can be achieved by adjusting the processing parameters. Li *et al.* (2014) studied the impact of varying the mass ratio of fibers to starch, contents of plasticizer, foaming agents, and active agents on the compressive strength of cushioning materials with orthogonal experiments. Their results showed that the compressive strength was most significantly affected by the mass ratio of fibers to starch. Guo *et al.* (2015) also studied how forming parameters affected the tensile strength and stretching toughness of plates, using an orthogonal experimental design (Guo *et al.* 2015). They found that top mold temperature had a greater influence than other factors such as bottom mold temperature, forming time,

and drying time. Their results showed that biomass products could be used for cushioning packaging as a substitution for current packaging materials. Equivalent performance was achieved in comparison with EPE and EPS. A compression/explosion method for making molded starch-based foam physical and mechanical properties similar to the foam was developed and used in commercial food packaging (Glenn and Orts 2001). One of the advantages of this method over traditional puffing or extrusion technology is to help increase the incorporation between fibers and starch, leading to improved mechanical properties.

Fibers themselves can be made into bio-foams as well as reinforcement of starch-based foam. Huang *et al.* (2014) produced foamed cushioning materials with 20% cationic starch, 5% polyvinyl alcohol, 74% beaten fiber slurry, and 1% of magnesium stearate. Luo *et al.* (2017) prepared a porous, wood-fiber-based cushioning materials for packaging using poplar fibers and wood powder raw materials by hot-press molding. Eco-friendly cushioning materials were prepared with thermo-mechanical pulps (TMPs) from waste woods using a suction-forming method (Lee *et al.* 2010). The cushioning properties of TMP cushion containing cationic starch were somewhat less effective than EPS and it exceeded the performance of molded pulp. Figure 3 shows the cross-sectional views of the three packing cushions.

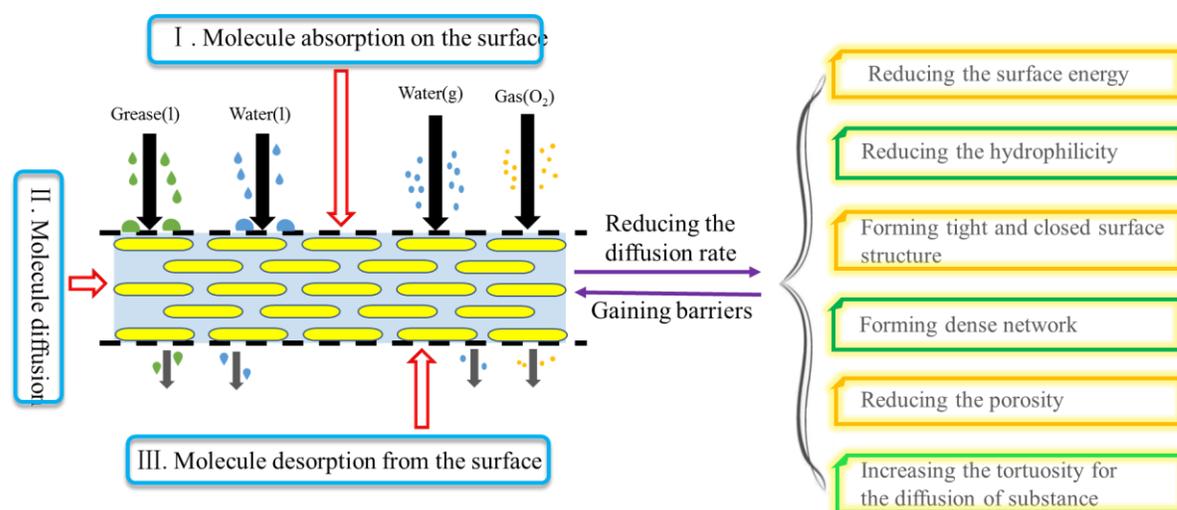


**Fig. 3.** Cross-sectional views of the three packing cushions; Reprinted with permission from Lee *et al.* (2010). *Tappi Journal* 9(7), 15-21.

## CELLULOSE FILM AND PAPER MATERIALS WITH BARRIER PERFORMANCE

Packaging materials serve an important function in preserving the quality and safety of food products. By providing sufficient barriers against water vapor, oxygen, grease, and microorganisms, food packaging can increase the shelf-life and reduce food waste (Barlow and Morgan 2013). Plastic films for use in barrier packaging are ubiquitous today, fulfilling a range of functions including moisture barrier, gas/aroma barrier, grease resistance, and chemical resistance (Bezigan 2016). Depending on what is most needed in a given application, different synthetic polymers can be utilized. These include polypropylene (PP) for mechanical properties and water vapor barrier, polyethylene (PE) for sealing and water vapor barrier, ethylene vinyl alcohol (EVOH) for barrier properties to gas and water vapor, polyethylene tetra phthalate (PET) and polyamide (nylon) for aroma/oxygen barrier with stiffness, and various others (Rosato

2013). However, those products can generate substantial waste disposal problems due to their lack of biodegradability. A lot of research and development activities have been carried out to make cellulose-based materials; these have achieved advantages such as biodegradability and barrier properties for packaging (Castro-Rosas *et al.* 2016; Shi *et al.* 2016; Urbina *et al.* 2016; Zhang *et al.* 2016; Chen *et al.* 2017; Ferrer *et al.* 2017; Hubbe *et al.* 2017). Four types of barrier performance often studied relative to packaging film requirements include grease and water resistance, as well as oxygen and water vapor barrier properties, as shown in Fig. 4. The different migration processes for the penetration of liquids (water and grease) and for the diffusion of gases molecules through a packaging film containing cellulose and its performances to gain barrier properties are illustrated in the figure.



**Fig. 4.** Migration process of molecules through a packaging film and its performances to gain barriers

### Regenerated Cellulose Films

Regenerated cellulose is prepared from dissolving pulp, which consists of high-purity cellulose (90 to 99%), by dissolution and then coagulation processes (Kumar and Christopher 2017). The process of cellulose regeneration, accompanying physical change and chemical reaction, not only provides the unsurpassed physical and chemical properties but maintains inherent renewability, biocompatibility, biodegradability, and environmental friendliness of cellulose. Regenerated cellulose can be obtained in different forms, such as powder, films, fibers, hydrogels, and spheres, *etc.*, depending on the change of the regeneration parameters (Wang *et al.* 2016). Regenerated cellulose films (RC films) have good thermal stability, optical transparency, good tensile strength and high oxygen barrier properties (as long as the conditions are dry), which endow RC films with great potential for use as a bio-based flexible packaging material (Yang *et al.* 2012; Cheng *et al.* 2017).

RC films are fabricated directly from cellulose solutions. The properties of regenerated cellulose films depend on cellulose feedstocks, solvents, and regeneration techniques. Solvents typically applied in cellulose dissolution include N-methylmorpholine-N-oxide (NMMO), ionic liquids and alkali/urea (Medronho and Lindman 2015). Shih *et al.* (2009) prepared transparent yellowish blend films from

cellulose and chitosan mixed in NMMO. The obtained films containing chitosan had high strength and non-diffusible antibacterial properties. Yang *et al.* (2011) developed transparent and bendable RC films from alkali/urea systems for bio-based packaging materials with high-oxygen barrier properties. Pang *et al.* (2014) prepared RC films using 1-allyl-3-methylimidazolium chloride (AmimCl), 1-butyl-3-methylimidazolium chloride (BmimCl), 1-ethyl-3-methylimidazolium chloride (EmimCl), and 1-ethyl-3-methylimidazolium acetate (EmimAc) as solvents, respectively. From their results, the ionic liquid EmimAc was shown to be a much better solvent for cellulose compared to other ionic liquids; the RC films from EmimCl exhibited the highest tensile strength.

Factors affecting regeneration techniques include coagulants, orientation, drying process, and so on. The coagulants can determine the optical transmittance, homogeneity of structure, porosity, and mechanical properties. The orientation is important to enhance the mechanical properties for cellulose films. The drying process affects the structure (such as porosity) and properties (such as mechanical properties, coefficient of thermal expansion and oxygen permeability) of the RC films (Wang *et al.* 2016).

Improvements in the barrier properties of RC films have been widely studied. For example, Yang *et al.* (2011) developed transparent and bendable RC films based on alkali/urea systems for bio-based packaging materials with high resistance to oxygen permeation. The effects of different cellulose sources, as well as different dissolution and regeneration conditions on oxygen permeabilities of the cellulose films were investigated. The lowest oxygen transmission rate of the cellulose films reached 0.003 mL· $\mu\text{m}^2$ /day/kPa when the film was prepared from a 6 wt% cellulose solution by regeneration with acetone at 0 °C. In another study, the transparent and flexible cellulose–clay nanocomposite films were prepared from cellulose /LiOH/urea solutions (Yang *et al.* 2014) . The nanocomposite films display high mechanical strength and Young's modulus, as well as low thermal expansion and oxygen permeability. The gas barrier properties of the films (minimum is less than 0.3 mL· $\mu\text{m}^2$ /day/kPa at 50% RH (relative humidity)) can be comparable to those of practical oxygen barrier films such as polyvinylidene chloride (0.4 to 5.1 mL· $\mu\text{m}^2$ /day/kPa at 50% RH). Isogai and coworkers (Yang *et al.* 2012) developed cellulose films with transparency, water repellency, and resistance to gas diffusion by surface modification of alkali/urea regenerated cellulose films through soaking in cationic alkylketene dimer (AKD) dispersion. The AKD-treated films prepared from an aqueous LiOH/urea system exhibited high water repellence and high gas barrier properties even under high humidity conditions. In addition, cellulose and other materials can be combined for the preparation of cellulose composite films. Wu *et al.* (2009) developed a series of novel biobased composite films derived from cellulose, starch and lignin by coagulating in a non-solvent condition from an ionic liquid, 1-allyl-3-methylimidazolium chloride. The composite films exhibited excellent mechanical and gas barrier properties. The favorable results were attributed to synergism between cellulose, starch, and lignin, as would be suggested by their mutual effectiveness in forming the structures of plant materials.

### **Cellulose Paper with Oil and Grease Barrier properties**

Grease barrier performance is becoming more and more important in applications involving direct contact with greasy foods such as fast foods, bakery

products, and pet foods. The use of fluorochemicals in such applications has been heavily questioned on account of their toxicity and resistance to biodegradation. Hence, developing new grease-resistant additives is an interesting research area. Cellulose fibers are a potential candidate for such applications due to their high cohesive energy density, molecule size, and the ability to form a homogeneous and continuous film (Kumar *et al.* 2014; Lavoine *et al.* 2014; Dai *et al.* 2017).

Aulin (2010) used micro-fibrillated cellulose (MFC) suspensions to coat paper base-stocks having different air-permeability. The goal was to prepare packaging materials with good resistance to oil. Using bench-scale rod coating equipment, the base papers were coated with an aqueous MFC dispersion having a concentration of 0.85 wt%. The MFC coating decreased the air permeability and increased the oil resistance. The coated paper having the lowest air permeability also exhibited superior oil resistance. Such results show that it is possible to form grease-resistant paper by application of MFC as an aqueous-based suspension. Satisfactory oil barrier properties require that the coating is continuous and homogeneous, such that it blocks the pores in the base-stock. Lavoine *et al.* (2014) reached a similar conclusion. They presented a comparison of bar coating and size press application of MFC. The grease resistance was measured according to the Kit test (T559 cm-02), in which paper is exposed to a dozen standard oils, which are numbered 1 to 12. Grease resistance increases with increasing Kit number. A Kit number of at least 8 qualifies a paper specimen as grease-resistant. In the study, the increase of the MFC coating weight improved the grease barrier (from a Kit number 0 to 5), but the values were not able to match the performance of a polyethylene-coated paper (Kit number 12).

Kisonen *et al.* (2015) prepared composite coatings with NFC and O-acetyl-galactoglucomannan (GGM) (from spruce wood), either with a novel succinic ester of GGM or with native GGM to enhance the grease and oxygen barrier. They synthesized succinic esters of GGM having two different degrees of substitutions (DS); this enabled control of hydrophobicity of the films. The NFC and NFC-GGM composite films were subsequently prepared by filtration on a fine membrane and dried using a Rapid Köthen Sheet Former. The coating formulation was prepared with a 15 wt% water dispersion of GGM or GGM-Su1 (low degree of succinic ester substitution), or 15 wt% ethanol dispersion of GGM-Su2 (high degree of succinic ester substitution) with 15 wt% of sorbitol (relative to GGM). The coating was applied with a bar coater. All such coatings achieved excellent grease resistance. To determine the grease resistance, a cylinder was placed on top of the film, and it was filled with colored rapeseed oil, while a sheet of absorbent paper was placed below. When NFC-GGM was tested at 60 °C and at 75% RH in 5-day-long tests, no oil penetration was observed. Both the NFC films alone and the NFC-GGM composite films coated with NFC-GGM-Su1 effectively excluded grease for a 6-day test at 50 °C and 50%RH. The NFC-GGM composite films coated with GGM-Su2 were impermeable for 11 days at 50 °C and 40% RH, and for 9 months at 23 °C and 50% RH in a follow-up exposure test.

Hassan *et al.* (2016) developed films from NFC and chitosan nanoparticles (CHNP), and these displayed excellent grease resistance. They used the mixture to coat paper sheets, and the grease proof properties of coated paper were improved. From their research results, excellent grease proof properties were achieved with films made from NFC or NFC/CHNP. Turpentine required more than 1800 s to seep through the film, which was sufficient to be classified as highly grease-proof. The superior performance was attributed to the very fine porous structure that resulted from the

nanocellulose/chitosan coating process. For comparison, the penetration time was about 70 s for coated paper. The result indicated that a continuous and homogeneous coverage to the base paper is essential to obtain good grease resistance.

### **Cellulose Paper with Oxygen and Water Vapor Barrier properties**

Nanocellulose is known to provide good oxygen barrier properties, which can be attributed to the dense network structure that is formed by the very small and flexible microfibrils. Compared to ordinary cellulose fibers, as used in papermaking, nanofibrillated cellulose has much higher surface area and aspect ratio. The decrease in permeability has been correlated to the high density within the film (Spence *et al.* 2010; Syverud and Stenius 2009). Kumar *et al.* (2014) evaluated the impact of raw materials and nanocellulose production process on nanocellulose film mechanical and optical properties. They found that cellulose nanofiber films had better barrier properties against oxygen. The cellulose nanofibers were prepared by refining and grinding bleached softwood kraft pulp. Cellulose nanofiber films were prepared by casting from an aqueous suspension followed by evaporative drying under controlled air conditions of 23 °C and 50% relative humidity (RH). The film thickness was approximately 25 µm. The oxygen transmission rate (OTR) of the cellulose nanofiber films was as low as 1.4 cm<sup>3</sup>/m<sup>2</sup>/day. This value is sufficient to meet the oxygen barrier requirements for packaging with modified atmosphere conditions.

However, the OTR of MFC materials will decrease drastically under the condition of high humidity because of the high hydrophilicity of MFC. To have a low OTR, MFC materials must have excellent barrier property to liquid water. Many researchers have been making efforts to impart high water resistance to NFC/MFC films.

Rodionova *et al.* (2011) developed cellulose nanofiber films with excellent oxygen barrier performance, and also good resistance to liquid water. To accomplish this, microfibrillated cellulose produced from kraft pulp was chemically modified by treatment with acetic anhydride. Acetic anhydride forms an ester bond with the OH groups on the cellulose molecules. This transforms the hydrophilic surface and makes it more hydrophobic. In their study, the OTRs of both pure and partially acetylated cellulose nanofiber films were between 4 and 6 cm<sup>3</sup>/m<sup>2</sup>/day, which fulfills the requirement of modified atmosphere packaging. Although the hydrophobicity improvement of the cellulose nanofiber surface was found as a result of acetylation, achieving the highest contact angle with water of 82.7°, the water vapor transfer rate (WVTR) was not improved by the acetylation. Both modified and unmodified samples exhibited similar WVTR (according to T448 OM-97), which exceeded 200 g/m<sup>2</sup>/day at 23°C and 50%RH. The WVTR for a packaging material such as PVDC, PE or PVC films is around 3.07 to 118.65 g/m<sup>2</sup>/day at 27°C and 100% RH for 12.7~18.3 µm thick film (Ferrer *et al.* 2017).

Chinga-Carrasco *et al.* (2012) introduced hexamethyldisilazane (HMDS) as an alternative for manufacturing hydrophobic cellulose nanofiber films. HMDS reacts in the gaseous phase with OH-groups, incorporating hydrophobic ethyl groups on the surface. OH-groups are abundant on cellulose nanofibrils. HMDS is thus a potential chemical for reducing the water wettability of cellulose nanofiber. Their results showed that the HMDS-modified films made of carboxymethylated MFC had a contact angle larger than unmodified ones (89° vs. 54°) for bleached cellulose nanofiber. At the same time, the oxygen permeability of the HMDS-modified films was less than 0.06

$\text{mL}\cdot\text{mm}\cdot\text{m}^{-2}\cdot\text{day}^{-1}\cdot\text{atm}^{-1}$ . This can be regarded as a satisfactory result for some packaging applications.

WVTR test results represent the ability to withstand high moisture environments. According to a study, merely changing the hydrophilicity of cellulose nanofiber cannot improve the WVTR to meet the requirements of the atmosphere packaging (Rodionova *et al.* 2011). The other factors such as the physical structure play an important role for the water barrier of MFC. Given the hydrophilic nature of cellulose, high humidity and moist conditions pose a great challenge. A promising way to meet the challenge is with layers of suitably water-resistant biopolymer films. The substances referred to in the literatures include polylactic acid (PLA), poly-(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV), and amylopectin solution, *etc.*

Meriçer *et al.* (2016) achieved robust, transparent multilayer composite films by coating cellulose nanofibers on different PLA substrates. This was made possible by activating the PLA film surface with an atmospheric plasma treatment. The oxygen barrier performance was improved by more than a factor of ten compared to neat PLA films at 35°C and 0% RH, and the permeability values were maintained at a humidity of up to 60% RH.

Sanchez-Garcia and Lagaron (2010) studied PLA nanocomposites, which were prepared using cellulose nanowhiskers (CNWs) or cellulose nanocrystals (CNCs) as a reinforcing element. The CNWs or CNCs made by acid hydrolysis of highly purified alpha cellulose were 60 to 160 nm long and 10 to 20 nm wide. The CNWs at 1, 2, 3 or 5 wt% were incorporated into a PLA matrix by means of solution casting from chloroform. The incorporation of the CNW decreased the water permeability of the PLA nanocomposites by as much as 82% and the oxygen permeability by as much as 90%. The highest barrier performance was found for composites with a CNW or CNC loading of 3 wt%. Rampazzo *et al.* (2017) reported that the permeability of CNC coating to oxygen and carbon dioxide was hundreds of times lower than common barrier synthetic polymers at similar thickness over a range of temperatures. Nanocomposites consisted with polypyrrole (PPy) and TEMPO-oxidize nanofibrillated cellulose (TOCN) had antioxidant activity and good barrier properties to the diffusion of oxygen and carbon dioxide gases and water vapor (Bideau *et al.* 2017). In general, nanocellulose has been found to be a promising reinforcement for the mechanical and barrier properties of biopolymer and synthetic polymer for food packaging (Bharimalla *et al.* 2017).

Hult *et al.* (2010) coated paper and paperboard, having various levels of air-permeability, with combinations of cellulose nanofibers and shellac. Shellac has the favorable characteristics of hydrophobicity, biodegradability, and renewability, such that it is a good candidate to be used commercially in combination with cellulose nanofibers. The OTR and WVRT of paper and paperboard decreased dramatically after being coated by cellulose nanofibers and shellac. For sample, the values of OTR and WVRT were 35275  $\text{ml}/\text{m}^2/\text{day}$  and 70.89  $\text{g}/\text{m}^2/24\text{h}$ , respectively, when a paper was coated with 2.21  $\mu\text{m}$  thickness of cellulose nanofiber. The values decreased to 5438  $\text{ml}/\text{m}^2/\text{day}$  and 8.14  $\text{g}/\text{m}^2/\text{day}$  respectively when the paper was coated with 2.21  $\mu\text{m}$  thickness of MFC and shellac. The improvement was due to the fact that the shellac top layer formed a homogeneous layer.

## SUCCESSFUL EXAMPLES OF COMMERCIAL PACKAGING PRODUCTS MADE OF NATURAL CELLULOSE FIBERS

In recent years, several fiber-based products have been launched by different companies to replace traditional plastic packaging (Fig. 5). These products not only have excellent functionality but also maintain the inherent advantages associated with natural fibers, including the biodegradability.

Trayforma boards from Stora Enso are used for food packaging (Storaenso 2017). They can be tailored to different designs, suitable for different applications. For example, the Trayforma boards used as bowls and plates are made up of top layer (kraft pulp), middle layer (kraft pulp + CTMP), bottom layer (kraft pulp); customer-designed coating may also be applied. The boards can be used as the original packaging materials to be heated in a microwave or conventional oven.

Södra has launched a bio-composite (Durapulp®), a mixture of cellulose pulp and the biopolymer polylactic acid (Södra 2016). The bio-composite is suitable for many industrial applications such as molding, air-laid, sheet or board, for packing food and consumer goods.

VTT is developing fiber foam technology (VTT, 2017). The main raw materials are wood fibers (virgin or recycled), while nanofibrillated cellulose can be used to enhance the specific properties. The involved processes include: preparing the feed stock, molding (into specific forms), dewatering and drying.

FiberForm packaging consists of 100% cellulose fibers (Billerudkorsnäs, 2017). Some unique properties, for example, a high stretching ability, could be particularly attractive for food packaging.

Carlsberg has developed the Green Fiber Bottle products that can be used for beer (Didone *et al.* 2017). The paper bottles are made by molding, free of inner liners, and completely biodegradable.

VTT has developed 100% bio-based stand-up pouches with oxygen, grease and mineral oil barrier properties by using different bio-based coatings on paper substrate, which is based on the enzymatic fibrillation of cellulose (HefCell) technology (Eagle 2017).

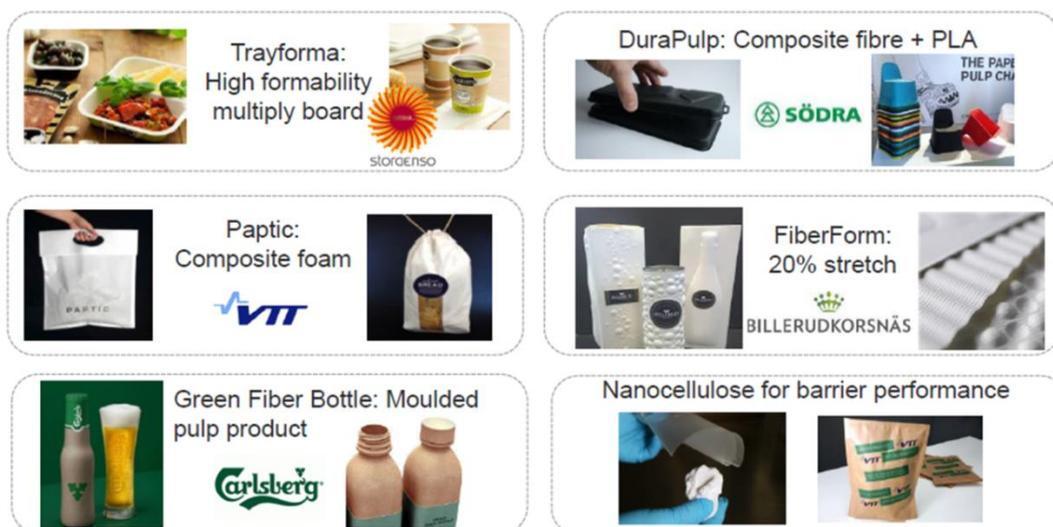


Fig. 5. Recent examples of packaging innovations using natural fibers

## FUTURE PROSPECTS, CHALLENGES, AND R&D NEEDS

As self-standing films, coatings, and bio-degradable fillers in composites, cellulose fibers provide novel and promising properties such as biodegradability, light weight, high mechanical performance, and high barrier properties for packaging materials. However, many challenges remain (Bharimalla *et al.* 2017).

Layers of cellulose nanofibers have been shown to achieve effective barrier properties, especially with respect to oxygen gas resistance. This generally can be attributed to a dense network structure. But under high humidity, the oxygen barrier property will decrease dramatically, largely due to the natural hydrophilicity of cellulose. Also, their hydrophilicity hinders the homogeneous dispersion in the composite matrix due to poor compatibility and interfacial adhesion when used as reinforcements for polymers of less hydrophilicity. Therefore, chemical modifications for fibers are desired in many instances. However, many of these methods are still at the lab scale. Furthermore, the drying of cellulose nanofibers is still a challenge (Hubbe *et al.* 2017). In comparison with plastics, the high cost of fiber-based packaging materials for many applications is still a huge obstacle to be overcome (Rujnić-Sokele and Pilipović 2017).

## CONCLUSIONS

Natural cellulose fibers are bio-based materials with good biodegradability and recyclability which are suitable for packaging applications, although they should be selected and processed accordingly to the specific needs of packaging products, taking into account the environmental, social and economic sustainability. Cellulose fibers can provide excellent mechanical properties when used in starch-based foams. They have excellent vibration and impact cushioning properties, which are not affected by extreme temperature or humidity when used as molded pulp products. Cellulose based materials can offer barrier properties to oxygen, carbon dioxide, and water vapor, as well as resistance to liquid water and grease, by choosing suitable processes. The application of cellulose fibers for some packaging products is technically feasible. The full commercial potential of cellulose fibers in packaging application will still have to be demonstrated in many cases.

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