Molded Pulp Products for Sustainable Packaging: Production Rate Challenges and Product Opportunities

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Molded cellulosic pulp products provide eco-friendly alternatives to various petroleum-based packaging systems. They have a long history of reliable usage for such applications as egg trays and the shipping of fruits. They have recently become increasingly used for the packaging of electronics, wine bottles, and specialty items. Molded pulp products are especially used in applications requiring cushioning ability, as well as when it is important to match the shapes of the packed items. Their main component, cellulosic fibers from virgin or recycled wood fibers, as well as various nonwood fibers, can reduce society's dependence on plastics, including expanded polystyrene. However, the dewatering of molded pulp tends to be slow, and the subsequent evaporation of water is energy-intensive. The article reviews strategies to increase production rates and to lower energy consumption. In addition, by applying chemical treatments and processing approaches, there are opportunities to achieve desired end-use properties, such as grease resistance. New manufacturing strategies, including rapid prototyping and advances in tooling, provide opportunities for more efficient form factors and more effective packaging in the future.

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INTRODUCTION

Molded pulp products, which often are made with recovered cellulosic fibers, are being increasingly used for the packaging and shipping of three-dimensional objects (Didone *et al.* 2017; Su *et al.* 2018). Such products, which are formed by the dewatering of cellulosic pulp suspensions, can have a wide range of three-dimensional shapes (Ma *et al.* 2004). Since about 1890, molded pulp systems have been used for egg cartons (Keyes 1890). More recently, there has been growing usage of molded pulp systems for packaging of electronics and specialty items (Marcondes 1997, 1998). Not only can molded pulp products be made from photosynthetically renewable and/or recycled materials, but they also can be easily recycled. Molded pulp products also face some important challenges. The focus of the present review article is on such challenges as relatively slow rates of production. Much of the time required to produce each molded pulp item involves its dewatering on a screen surface, followed by the energy-intensive process of drying. Publications describing ways to speed up molded pulp processing, including relevant strategies used in papermaking operations, are reviewed in this work. Further challenges are related to meeting a range of specifications for such properties as strength and water resistance. Accordingly, published approaches including chemical treatments of fiber suspensions are reviewed with respect to product goals.

Packaging is an important aspect of the economy. Along with providing basic protection of the goods inside, packaging helps in promoting its value. It can offer convenience in handling and display (Gutta *et al.* 2013). The global packaging market is set to reach over $1 trillion by 2021 (Smithers 2018). However, packaging has also been the subject of concerns about environmental sustainability. Every year, large amounts of packaging materials are being used with the expectation of single usage and disposal. Large portions of them are made of non-biodegradable and non-renewable materials such as plastics, glass, and metals in packaging applications. Even their incineration methods affect the environment.

Due to their low cost, light weight, high performance, and ease of processing, consumers and businesses have become used to disposable plastics. Usage of these materials has resulted in the accumulation of plastic products in the environment during the last several decades (MacArthur 2017; Schneiderman and Hillmyer 2017; Horejs 2020; MacLeod *et al.* 2021). Of the 8.3 billion metric tons plastics that has been produced since 1950, about 60% has ended up in either a landfill or the natural environment (Ashraf *et al.* 2021).

The small plastic fragments with less than ≤ 5 mm dimensions are defined as microplastics (Duis and Coors 2016). The accumulation of these microplastics, often due to mechanical breakdown, poses a great threat to marine organisms and also indirectly affects the ecosystem by absorbing hydrophobic pollutants due to its large area to volume ratio (Chatterjee and Sharma 2019). In light of such challenges, the focus is to find renewable, recyclable, biodegradable alternatives that can replace non-biodegradable and non-renewable packaging materials, such as expanded polystyrene (Ting and Chang 2014). Many businesses currently are moving from linear to circular economy by developing a new processing chain of materials and manufacturing; by such means the waste, emissions, and energy usage can be minimized by narrowing and closing energy and material loops (Garcia and Robertson 2017; Zhang *et al.* 2021). Considering all these factors, paper and cellulose-based materials are becoming more attractive, sustainable options for packaging materials (Didone 2017). Among these materials, molded fiber and its products have attracted increasing amount of attention, specifically to replace expanded polystyrene (EPS), which is also known as Styrofoam™.

A typical molded pulp packaging item generally can be described as having a relative low density, moderate resistance to compression, and good insulating properties, especially when dry (Anon. 2011; Abhijith *et al.* 2018). It generally has good shock absorbing ability (Wever and Twede 2007). Unlike expanded polystyrene (EPS), the cushioning or shock absorption property is often achieved by the inclusion of a number of dimples on the pulp sheet; more specifically, it was related to the perimeter of the dimples rather than their area (Eagleton and Marcondes 1994). Recently, the emphasis on cost-effective and eco-friendly packaging has been accelerated by increasing levels of electronic commerce, which has increased the amounts of packages individually sent to people's homes (Abukhader and Jönson 2003). In such applications, packaging can play a pivotal role in brand's reputation and the consumer's expectation. As more and more consumers embrace online shopping, the amount of related waste is also set to increase.

BACKGROUND

Before considering research related to the challenges and opportunities in molded pulp products, this section will provide some essential background. Published review articles and chapters have considered some aspects of molded pulp production and attributes (Emery and Emery 1966; Paine 1991; Anon 2011; Didone *et al.* 2017; Abhijith *et al.* 2018; Su *et al.* 2018; Zhang *et al.* 2021). For instance, Didone *et al.* (2017) place emphasis on the potential of hot-press processes, which can be important when the goal is to achieve increased mechanical strength. Abhijith *et al.* (2018) focused on potential use of fungal mycelia for molded pulp products. Su *et al.* (2018), in their brief review, covered a broader area that included not only molded pulp products but also bio-based foams. Zhang *et al.* (2021), in another brief review, focused on emerging applications and environmental sustainability. Relative to some other technologies, such as papermaking in general, the amount of scientific publication related to molded pulp products has been small. Though the present article has a limited focus on production rates and product attributes, the authors hope to encourage others to help fill the gap of information related to this important area of technology.

Molded Pulp Product Classes

During the many years of industrial production of molded pulp products, there have been some advances with respect to both the processing methods and the types of products that can be achieved (Keyes 1890; Twede and Wever 2007; Didone *et al.* 2017). For a long time the technology was limited to low-cost retail products, *e.g.* egg trays and boxes, and disposable food service wares (Chiang 1993; Cullen Packaging 2012). With advancement of technologies and considering its environmental benefits, its demand and market are presently increasing (Xiao and Shao 2015). Molded pulp products are being used in the packaging of industrial products such as electrical and electronic appliances, *e.g.* printers and computers (Noguchi *et al.* 1997; Ma *et al.* 2004; Wever and Twede 2007). Companies including Seventh Generation™, Pangea Organics®, and Paper Water Bottles have launched packages such as laundry-detergent bottles, packages of soap, and prototypes of water bottles to replace plastics (Didone *et al.* 2017). Molded pulp can also be used as a cushioning material for protecting the product during transportation, *e.g.* molded honeycomb paperboard (Ma *et al.* 2010).

According to the International Molded Fiber Association (IMFA), molded pulp products can be categorized in various groups based on the manufacturing process and quality of materials. The first one, which is made out of kraft and recycled paper and has a typical wall thickness ranges from 5 to 10 mm, is known as "thick wall." The procedure for making such products is illustrated in Fig. 1. As shown, the process starts with an aqueous suspension of fibers in a vat. A porous mold is inserted into the vat, and vacuum is applied to draw fibers (and any other additives) toward a screen surface. The mold is then withdrawn from the vat, still with the damp molded product attached. The molded pulp product is then released from the mold and subjected to oven drying. Thick-wall products mainly serve as support packaging for non-fragile, heavy items (*e.g.* furniture and vehicle parts) during shipping.

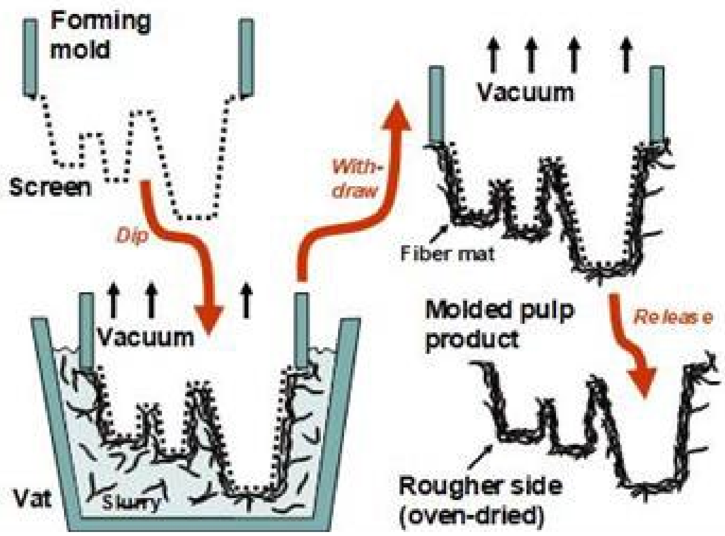

Fig. 1. Schematic illustration of thick-wall process for molded pulp production, involving vacuum forming, with separate drying in an oven

The second type is known as "transfer molded." These have thinner walls ranging from 3 to 5 mm with relatively smooth surfaces on both sides and better dimensional accuracy. These are typically used as egg trays and packaging for electronic equipment. The third type is the most recent approach known as "thermoformed" ("thin-wall"). Such products having thickness ranging from 2 to 4 mm with good dimensional accuracy, and smooth, rigid surfaces. This type of product utilizes heated molds, within which the initially formed product is pressed, densified, and dried. Such products can be used as a substitute for thermoformed plastic items. This process is illustrated schematically in Fig. 2. As shown, the process using for such items begins in essentially the same way as was shown in Fig. 1. However, for transfer-molded products the mold – with the damp molded product attached – is manipulated so that it gets pressed against an opposing surface that matches the intended shape of the product. As shown, the transfer mold surface is often heated, as implied by the term thermoformed. A key difference, in comparison to thick-walled molded pulp products, is that transfer-molded products tend to be relatively smooth on both the inside and the outside.

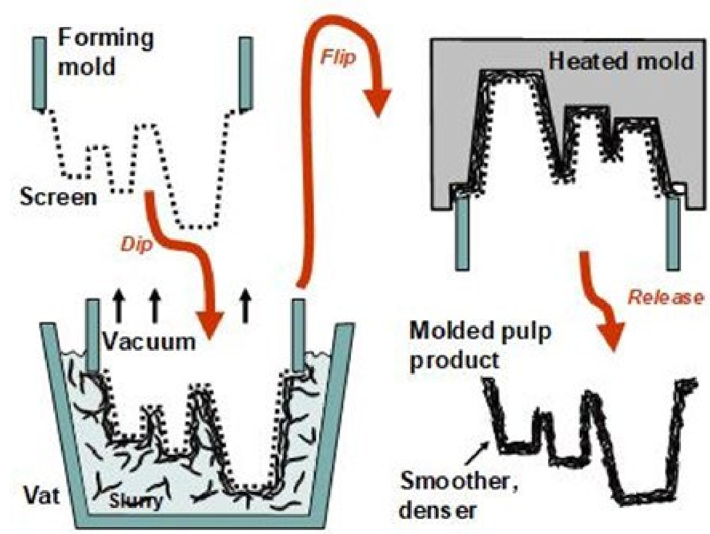

Fig. 2. Schematic illustration of transfer molded process, incorporating thermoforming conditions for initial drying

The final type is the "Processed," which refers to the previous items that require some further or special treatment such as additional printing, coatings, or additives ("International Molded Fiber Association," n.d.).

Transfer molded and thermoformed products

By transferring the initially formed item to a mating surface, the structure then can be pressed. Not only does the side initially facing away from the screen become much smoother, but also there is an opportunity to squeeze more water from the material. The improvements in accuracy and dimensional stability can expand application range of the molded pulp products (Didone *et al.* 2017). When using a thermoforming procedure, the goal is to at least partially dry the molded pulp product while it is still being pressed (Didone *et al.* 2017). In principle, such a process can achieve higher strength and higher rates of production.

Paine (1991) describes an alternative forming procedure in which pressurized air is used to dewater fiber suspensions against a screen. High temperatures can be used, such that solids contents of over 50% can be achieved during the forming step. In a related technology, a flexible diaphragm can be forced into the interior of a specialized mold (Kucherer 1995; Buxoo and Jeetah 2020; Saxena *et al.* 2020). The water is thus forced outwards through a set of detachable mold segments. Hollow molded pulp products, such as bottles, can be made in this way.

Property Ranges of Molded Pulp Products

Differences relative to flat papermaking

In light of characteristic differences in their forming processes, one might expect that the properties of molded pulp products will differ from those of flat paper products prepared from similar fibers. As a result of a higher typical solids content (consistency) of the fiber slurry, in comparison to typical flat papermaking conditions, one can expect that fibers will be oriented in all directions, rather than being mainly oriented in a plane. Figure 3 illustrates the likely mechanism whereby formation of paper from a dilute suspension tends to give a paper sheet in which fibers are mainly oriented in the plain of the sheet.

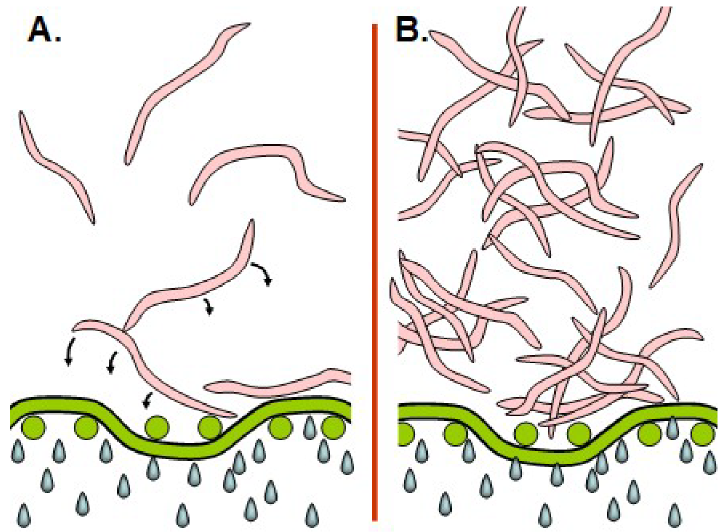

Fig. 3. Concept of why cellulosic fibers tend to become oriented in the plane of the sheet when formed from a dilute (< 1%) consistency suspension (A), whereas they tend to have more out-of-plain orientation when formed at higher consistencies (B)

Other differences between typical molded pulp products and paper grades, such as printing papers, relate to the types of fibers that are utilized. For instance, as will be discussed later, the fibers employed in molded pulp production are likely to be stiffer (less refined), compared to when making such products as printing papers or linerboard. Recycled kraft fibers, which can be considered for molded pulp products, will have experienced hornification, which results in an increase in stiffness of the wet fibers, leading to a bulkier mat structure (Zhang *et al.* 2002; Hubbe *et al.* 2007b). Recent work has shown promise for decreasing reliance on traditional refining of recovered kraft pulp fibers, thus making it possible to maintain higher drainability (Debnath *et al.* 2021). High-yield fibers, which are available from a wide range of woody materials (Salem *et al.* 2020), likewise are widely used for molded pulp products. Such fibers have a lower conformability in the wet state compared to refined kraft fibers due to their high content of lignin. The lignin is inherently rigid and it helps to resist internal delamination of the cell walls of high-yield fibers during refining.

Another important comparison one can make is with glass fibers. Westman *et al.* (2010) found that structures prepared with cellulosic fibers were much weaker than what could be achieved with glass fibers, which are individually very strong.

Differences relative to expanded polystyrene and related issues

The most critical properties of molded pulp products are mechanical strength and cushioning ability. From this perspective, molded pulp products can have similar properties to expanded polystyrene. One of the main jobs of a typical molded pulp packaging structure is to cushion the contained items against breakage. To accomplish this, the packaging needs not only to hold the packaged item, but it also needs to provide somewhat reversible deformation. Ideally, the function of a cushioning property is to minimize the damage of a product from any physical impact or vibration by absorbing the shocks arising during transportation. The cushion curves obtained by Pacific Pulp showed that molded pulp was actually a better protective cushion than the expanded polystyrene.

The performance shown on a cushion curve is dependent upon the manufacturer of the material and the thickness, size, and shape of the cushion. The drop test showed that if a package is exposed to shock, then the molded pulp package will be capable of absorbing that shock and the product would be protected (Eagleton and Marcondes 1994). It also provides excellent blocking and bracing functionality (Mabie and Camuel 2010).

Molded products often are designed with dimples, which are able to crush upon impact, thus absorbing some of the shock (Eagleton and Marcondes 1994). The cited article assigned molded pulp products only an intermediate rating in terms of shock-absorbing ability, noting that expanded polystyrene has a much greater ability to recover after crushing. Manufacturers can adjust the structure of molded pulp products, such as the addition of ribs, in an effort to improve the cushioning performance (Marcondes 1997). However, further testing showed that the transmission of shock though molded pulp material can be relatively high (Marcondes 1998). According to Paine (1991), molded pulp packaging products can be effective because they are able to fail sacrificially by distortion and crushing, ideally such that the contents of the package remain undamaged.

The apparent density within molded pulp structures falls within a wide range, which is understandable based on the wide range of process conditions. For example, a bulky, low-density structure is appropriate for thick-walled biocontainers intended for planting of seedlings (Aguerre and Gavazzo 2016). The cited study found apparent densities in the range of 0.28 to 0.69 g/cm^3. Values of about 0.6 g/cm^3 were judged as optimum. Howe (2010) noted that the densities of typical molded pulp products are higher than that of expanded polystyrene, which is unfavorable from the perspective of the mass than needs to be transported. Paine (1991) reported a range of apparent density of 0.2 to 1.0 g/cm^3 for various molded pulp products.

Evaluation of elastic modulus and stretch

The mechanical strength characteristics of molded pulp products can be revealed by stress-strain testing. Jacobson (2017) compared the strength characteristics of molding pulp structures formed with three kinds of pulp, namely kraft fibers, thermomechanical pulp (TMP), and recycled fibers. The unrefined kraft fibers, when formed into molded pulp sheets, achieved a tensile breaking stress of only about 5 MPa, whereas after refining (called “activation” in the article), the strength reached 37 to 47 MPa. These differences are shown in Fig. 4. Meanwhile, the TMP achieved breaking stresses in the range 25 to 27 MPa, and the recycled pulp reached 15 to 18 MPa (apparently with no additional refining having been applied). Hua *et al.* (2020) evaluated the Poisson’s ratios and determined the failure mechanisms of molded paper materials by means of digital images correlations and uniaxial tensile testing. They found that the drying of molded pulp under pressure within a mold led to higher Poisson’s ratio. In other words, stretching of the material in one direction led to a greater contraction in the other direction when the extent of bonding had been increased. The most important modes of tensile failure were found to be cracking and buckling. The mechanical properties also can be expected to depend on the presence of any defects, such as post-forming instabilities, cracks, wrinkles, or water-pockets (Jacobsen 2017).

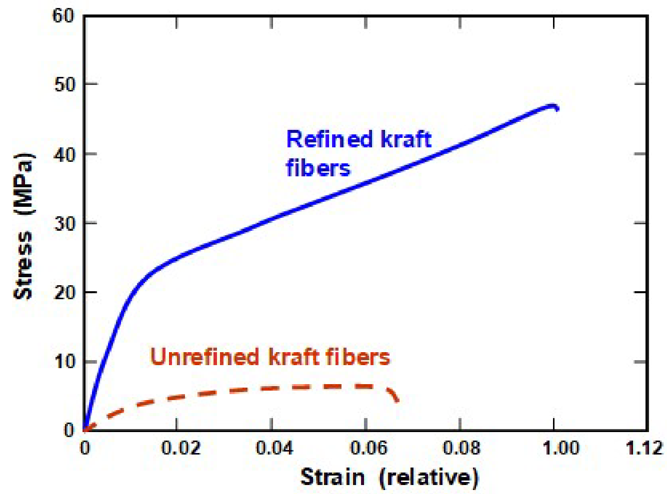

Fig. 4. Effect of mechanical refining on the stress-strain curves of molded pulp structures prepared from refined and unrefined kraft fibers. Figure redrawn based on more detailed plots reported by Jacobson (2017)

Jacobsen (2017) also compared the stress-strain curves for kraft sheets prepared under papermaking conditions, in comparison to when the same material was prepared as a molded pulp item. As shown in Fig. 5, the paper sheets exhibited a much higher initial modulus of activity within the plane of the sheet.

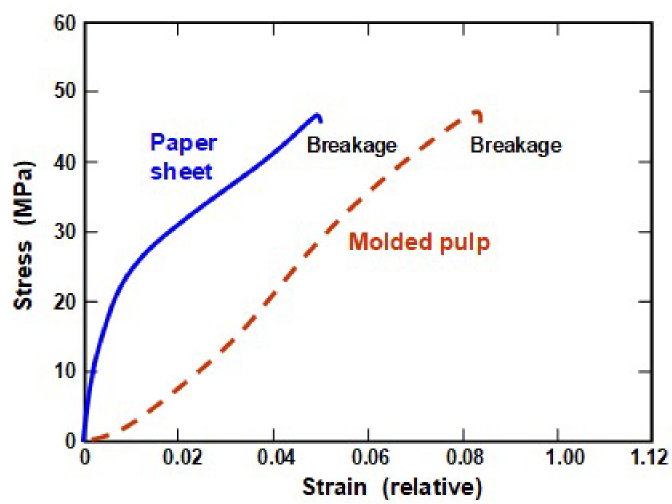

Fig. 5. Effect of slurry consistency (*i.e.* whether the sheets are formed as paper or as a molded pulp product) on the stress-strain curves of molded pulp structures prepared from refined and unrefined kraft fibers, respectively. Figure redrawn from more detailed plots reported by Jacobson (2017)

Such a result is consistent with the predominantly planar orientation of the majority of fibers in a typical paper structure. By contrast, the higher consistency that is used when forming a molded pulp structure is expected to lead to a more random distribution of fiber orientations with respect to the three orthogonal directions. In addition, restraint of sheet contraction during drying of a conventional paper sheet (as in the standard drying of paper handsheets) is expected to have a straightening effect on the fibers, which makes a further contribution towards enhancing the within-plane elastic modulus.

Compression strength is expected to be important for the performance of molded pulp products (Anon. 2011). Ji and Wang (2011) conducted short-span compression tests of molded pulp material (apparent density *ca.* 0.54 g/cm^3), studying the effects of apparent density and loading rate. Peak compression stresses in the range of 2 to 2.4 MPa were observed. The peak stress increased from about 2.4 to about 3.0 MPa with the increase in rate of compression from 0.008 to 0.08 reciprocal seconds. This observation is an indication of a somewhat visco-elastic response of the molded pulp material. Xiao and Shao (2015) found increasing peak compression stress of molding pulp with increasing temperature. Since equilibrium moisture content tends to decrease with increasing temperature (even at constant 50% relative humidity), the higher compression strength results at higher temperature can probably be attributed to lower moisture contents. These results are consistent with those of Sørensen and Hoffmann (2003), who observed strong decreases in static compression strength of molded pulp sheets with increasing relative humidity at controlled temperatures.

Viscoelastic effects of molded pulp material have been considered further. Ji *et al.* (2008) used the term emplastic (or adhesive) to characterize the material they studied. In other words, the material appeared to have a component of glue-like behavior (agglutinizing) when subjected to tensile stress.

Materials

In order to make a molded pulp product, one starts with a fiber slurry that may contain mostly water and about 3 to 5% fibers. Though the pulp molding industry uses similar fibers as the paper industry, the forming processes, fiber forming characteristics, densities, and structural functionality are different (Hunt 1998). Both virgin and primary fiber derived from wood or non-wood plants and recycled or secondary fiber, derived from waste paper and paperboard are used. The most used sources of natural fiber from plants include sugarcane bagasse, jute, flax, pineapple leaf, kenaf, bamboo, sisal, abaca, oil palm, rice husk, coir, coconut, and hemp (Westman *et al.* 2010). Both the physical and chemical properties of some of these fibers, such as high specific strength and stiffness, impact resistance, flexibility, and modulus, make them an attractive alternative over the traditional materials. For example, sugarcane bagasse fiber exhibits high specific strengths and modulus, cost-effectiveness, low density, and low weight, which make it a promising choice of raw materials by the industry (Sabdin 2014). In general, relatively long fibers provide strength, toughness, and structure, whereas shorter fibers often can provide high bulk (low density), closeness of texture, and smoothness of surface. Fiber length distribution influences a number of properties as well. For example, a short fiber length distribution will form a finer structure compared to a distribution with long fibers. Furnishes with a fiber distribution having more long fibers can increase the thickness of product. In the case of recycled fiber, furnishes with more old corrugated container (OCC) pulp content tend to be thinner than those prepared from old newspaper (ONP) pulp, even if their fiber length distributions are similar. This is because OCC fibers have a significant portion of kraft fibers that are more flexible and conformable when wet, and these tend to be compressed more, thus forming a thinner product (Kirwan 2013). Considering the strength properties, with each subsequent reprocessing cycle of kraft fiber, there is a decrease in mechanical properties attributed to the resistance of fiber swelling when rewet after being dried (Hubbe *et al.* 2007b). Wet end addition of lignin-containing micro and nano fibrillated cellulose in containerboard was found to offset the decreased strength associated with this by increasing the relative bonded area (RBA) of the paper (Tarrés *et al.* 2017; Starkey *et al.* 2021). Chemical compositions of some wood and nonwood fibers are given in Table 1 (Salem *et al.* 2021).

Table 1. Chemical Composition of Feedstock

Raw material	Cellulose (%)	Klason lignin (%)	Extractives (%)	Ash (%)	Hemicelluloses (%)
Eucalyptus	46.7	27.9	4.3	1.3	19.8
Hemp hurds	43.0	24.4	2.2	1.4	29.0
Bamboo	43.2	26.7	2.2	0.8	27.1
Hardwood	44.6	26.9	3.7	1.1	23.7
Softwood	46.0	28	3.0	1.0	22.0

Cellulose, the major component of fiber, has a high tendency to form intra-and intermolecular hydrogen bonds (Fengel and Stoll 1989). As a hydrophilic polymer, it has a high capacity for water absorption as well as limits to its barrier properties (Alavi *et al.* 2015). Hemicelluloses contribute to many intrinsic fiber properties, such as the swelling, fibrillation, bonding ability, and resistance to a hornification tendency (Pere *et al.* 2019). Resistance to hornification means that the material is more capable of swelling again after it has been dried, then placed back into water. Lignin is a rigid hydrophobic polymer, which is mostly removed during the kraft pulping process. During hot-pressing processes, the retention of a certain amount of lignin may contribute to the stiffness and water resistance (van de Wouwer *et al.* 2016). Lignin can also improve the mechanical properties, waterproofness, thermal stability, and adhesive properties of fiber materials in the polysaccharide-based materials represented by wood. Hence, light delignification has been developed to produce higher bonding. When the content of lignin was decreased from 24.9% to 11.45%, the density increased by 6.0%, the tensile strength increased by 22.0%, the bending strength increased by 23.9%, and the water contact angle increased from 64.3°–72.7° to 80.8°–84.3° (Wang *et al.* 2018).

Since the range of targeted properties is very broad, a wide range of fibrous materials have been considered for molded pulp production in various studies. Researchers have prepared molded pulp items from kraft pulps (Aguerre and Gavazzo 2016; Jacobsen 2017; Dislaire *et al.* 2021), high-yield wood or bamboo pulps (Jacobsen 2017; Wang *et al.* 2021), and chemi-mechanical pulps (Kirwan 2013; Zhao *et al.* 2020; Dislaire *et al.* 2021). As noted earlier, recycled pulps are very widely used by the molded pulp industry. Types of recycled fibers that have been studied for production of molded pulp items include old newspaper (Corwin 1972; Ahn 1994; Gavazzo *et al.* 2003, 2005; Dislaire *et al.* 2021), recycled cardboard (Dislaire *et al.* 2021), and old corrugated containers (Kirwan 2013; Aguerre and Gavazzo 2016).

Dislaire *et al.* (2021) compared six different pulp types in parallel tests, but for unknown reasons did not apply any refining action to three types of kraft pulp that were considered. Thus, they found the highest elastic modulus (*ca.* 1.2 GPa) for tensile tests of bleached chemithermomechanical pulp, which was followed by recycled cardboard pulp, recycled newspapers, and then the three unrefined kraft pulps.

A particular challenge when using recycled paper for production of molded products is that there may be a mismatch between the pH conditions of different waste paper types. Modern printing papers often are prepared under alkaline (pH 7.5 to 9) papermaking conditions with the presence of 20% or more of calcium carbonate filler (Hubbe and Gill 2016). At the same time, the mixture of recovered fibers also is likely to contain paper that has been prepared under acidic papermaking conditions, with aluminum sulfate as the primary buffering agent (Ehrhardt and Leckey 2020). Acidification of the calcium carbonate will result in its partial or complete dissolution, with the release of Ca^{2+} ions, and this can contribute to water hardness and various deposit problems in the mill. When there is sufficient calcium carbonate present to dominate the pH, the alkaline pH conditions may interfere with the action of rosin and aluminum sulfate additives, which have traditionally been used in molded pulp products (Kucherer 1995). When the amount of calcium carbonate is substantial, then the best approach often is to control the pH in the alkaline papermaking range and employ hydrophobic agents other than rosin (see later discussion).

Issues related to the choice between acidic and neutral-alkaline pH conditions of processing are illustrated in Fig. 6.

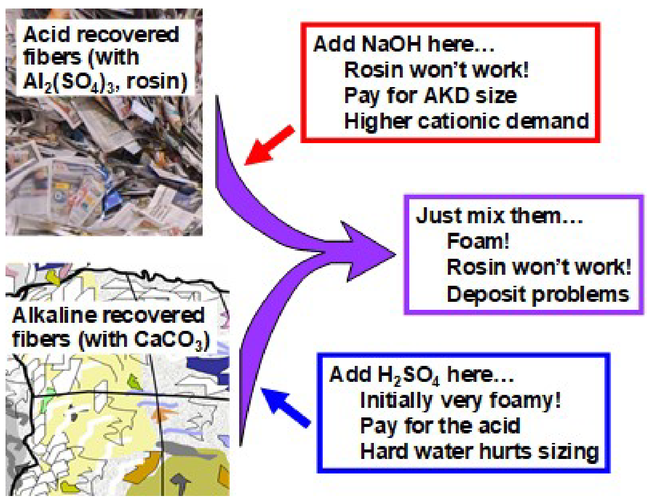

Fig. 6. Snapshot view of some key issues of concern when utilizing fiber sources that may contain acidic fiber materials (extract pH 4 to 5.5) and other materials that contain substantial amounts of calcium carbonate (extract pH 7.5 to 9)

As shown, the manufacturer is faced with some difficult compromises. Merely using the acidic and alkaline recovered fiber materials without pH adjustment can be expected to result in serious foaming issues. This is because the calcium carbonate mineral filler in the alkaline paper will be dissolving, especially if the amount of acidic material has a dominant effect on pH. Dissolution of $CaCO_3$ releases carbon dioxide gas (contributing to foam). It also dissolves Ca^{2+} ions, contributing to water hardness. In cases where the amount of acidic material is minor, a reasonable approach would be to add base (usually NaOH) to the water used to dilute the pulp. The rosin size already present in the fibers will no longer contribute to hydrophobization. Rather, an alkaline sizing such as alkylketene dimer (AKD) will need to be used. On the other hand, if the amount of alkaline paper in the mixture is minor, then one might opt to add some sulfuric acid to the dilution water used in pulping the recovered fibers. Large amounts of foam can be expected, at least initially, as the calcium carbonate dissolves. The very high levels of dissolved calcium carbonate can be expected to make it more difficult to hydrophobically size the paper with rosin and alum. Lab work and production-scale trials may be needed in order to decide which option makes more sense.

Non-wood fibers

Considerable research attention has been focused on the potential usage of non-wood fibers. These have included sugarcane bagasse (Jeefferie *et al.* 2011; Waranyou 2014), various types of straw (Curling *et al.* 2017; Hart 2020; Prasertpong *et al.* 2021), invasive grass (Chen *et al.* 2012), hemp (Buxoo and Jeetah 2020), and mycelium (fungal biomass) (Abhijith *et al.* 2018). As the price of global wood pulp is rising and the advancement of electronic media are decreasing the recycling of fibers from paper waste, the demand of alternative sources of raw material for creating the molded pulp packages has been continually increasing (Gouw *et al.* 2017; Johnston 2016). Based on the data of 80/20 straw pulp/kraft pulp mix based molded material, significantly better tensile properties were obtained compared to expanded polystyrene (modulus of 0.47 MPa for an 80% straw mix compared to 0.16 MPa for EPS). Along with strength properties, molded pulp has shown great biodegradability (20% mass loss after only 4 weeks covered in unsterile soil) (Curling *et al.* 2017). Be Green Packaging has introduced material made of six different resources, including bulrush, wheat straw, sugar cane, and bamboo (OAS Cataloging-in-Publication Data, 2016). Non-wood plants such as *Spartina alternifolia*, an invasive species, have been utilized recently. Thermo-mechanical pulping was utilized for this wild grass, and it shows good mechanical and cushioning property after mixing with other chemical pulps such as bamboo (Chen *et al.* 2012). Another study has shown the potential of fruit pomace (FP) as a source of fiber for partially replacing recycled newspaper (NP) to create molded pulp products. Along with certain amount of CNF, the FP based molded pulp board showed better or similar properties to 100% NP-based board (Gouw *et al.* 2017). A molded cup made out of composite of hemp–pineapple peels in 40:60 combination showed good strength property. A coating of beeswax having thickness of 0.70 mm on that cup was adequate to retain cold water for 30 min (minimum) without any leakage. The cup can degrade in both active soil and damp sand environments within 5 and 6 weeks, respectively. This means that fiber isolation from fruit peel wastes and hemp leaves to produce eco-friendly, biodegradable disposable paper cups is a viable approach (Buxoo and Jeetah 2020). Another vegetative part, the mycelium, can be grown in a mold to form different shapes for different items. It can grow quickly into a desired density. In addition, it can be dehydrated to stop further growth. After its useful life as a packaging material, it can be left out in the backyard for decomposition within a few weeks. However, there are challenges of maintaining a consistent density with a raw material that is a living organism and acceptancy by consumers (Abhijith *et al.* 2018).

Residues from forestry, agriculture, or discarded materials

The term residues has been used for lignocellulosic materials such as solid waste, forestry and agroindustrial waste, newspaper (ONP), office paper (OWP), corrugated cardboard (OCC), pine sawdust, eucalyptus sawdust, and sugar cane bagasse (Sengupta *et al.* 2020). These have been utilized to prepare molded products, and the properties that were evaluated were density, tensile, bursting, tearing, compression, stiffness, wet tensile, permeability, and water retention. It was shown that the OWP pulps increased strength properties, OCC pulps increased tear and wet tensile, ONP pulps increased stiffness, and reinforcement materials increased permeability. By adjusting the proportions of different pulp types, it becomes possible to reach the objectives of different product grades. For instance, a mixture of pulp OWP/OCC in a 50/50 proportion was been found to be optimum for some products (Aguerre and Gavazzo 2016). Mushroom root or mycelium-based materials have the potential as well to become the material of choice for a wide variety of applications, with the advantage of low cost of raw materials and disposal of polystyrene posing an environmental issue. The mycelium can be grown in a mold to form different shapes for different items. It can grow quickly into a desired density. In addition, it can be dehydrated to stop further growth. After its useful life as a packaging material, it can be left out in the backyard for decomposition within a few weeks. However, there are challenges of maintaining a consistent density with a raw material that is a living organism and acceptancy in consumers (Abhijith *et al.* 2018).

Fiber development

Processing technology, including pulping and refining operations, can affect the chemical composition of fiber. Chemical pulp has reduced lignin and can be almost free of lignin (Su *et al.* 2018). The contrast between the two main forms of pulping is illustrated in Fig. 7. As emphasized in the figure, although mechanical pulping retains almost all the raw material in the prepared pulp (*i.e.* "high yield"), the costs of electricity are generally high. That is because it takes electricity to run the mechanical refiners. By contrast, the fibers come apart easily after chips of wood are cooked in a digester with a mixture of NaOH and Na_2S (*i.e.* kraft pulping). The pulping can dissolve much or almost all of the lignin, as well as a substantial fraction of the hemicellulose, sometimes reducing the yield to values as low as 40%. Kraft pulps have excellent bonding ability, especially after an initial cycle of mechanical refining. However, some of the strength advantage of the kraft pulp fibers is lost with each cycle of paper production and recovery (Hubbe *et al.* 2007b). As mentioned earlier, the greater stiffness of both recycled kraft pulp and high-yield fibers can contribute to bulkier, less dense fiber structures, which can contribute better to the cushioning of packaged goods.

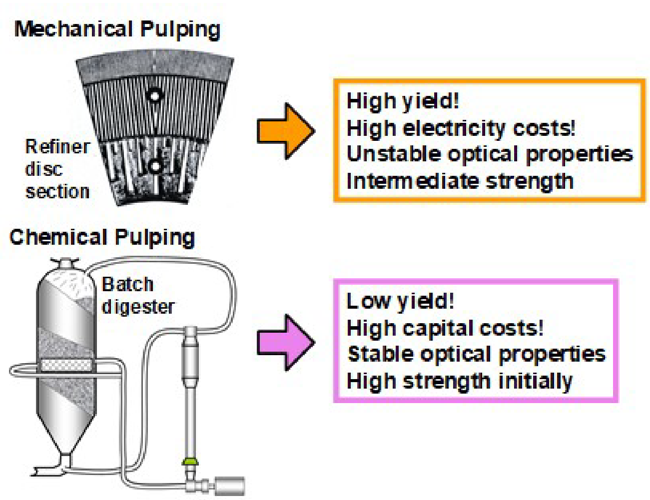

Fig. 7. Contrasts between the two major classes of cellulosic fibers that can be used in molded pulp production

The mechanical refining of pulps is well known to play a dominant role with respect to achieving various specified properties of paper products (Gharehkhani *et al.* 2015). Though the same general trends are likely to hold true with respect to molded pulp products, there are some important distinctions. First, typical apparent densities of molded pulp items (especially when not hot-pressed) are lower than those of common paper and paperboard products, such as printing grades and linerboard. As refining energy is applied, typically the resulting apparent density increases. Second, the combination of relatively high mass per unit area and the fact that the molded pulp items are individually formed places a high premium on the ease with which water can be removed by vacuum.

Nevertheless, refining can be a straightforward way to achieve the strength specifications, including the elastic modulus values needed for different molded pulp products. For example, Prasertpong *et al.* (2021) judged that a freeness value in the range 348 to 423 mL would give suitable tensile properties in typical cases when refining delignified rice straw. Such numbers represent a relatively low level of refining treatment.

The mechanism by which refining of a kraft fiber affects structural properties, and thereby affects dewatering as well as inter-fiber bonding ability, is represented in Fig. 8. The left-hand image represents the cross-section of an unrefined kraft fiber, which keeps the shape of a native fiber in wood, except that the fibers have been separated and most of the lignin has been removed. The repeated shearing and compression of wet bunches of fibers cause internal delamination of the fiber cell wall, especially the S2 layer, which accounts for most of the thickness of a typical fiber from wood. The delamination process is accompanied by swelling of the cell wall, as well as an increase in the conformability of the refined wet fibers. At the same time, the outer parts of the fiber, *i.e.* the primary (P) layer, the S1 sublayer, and outer parts of the S2 layer, become fibrillated. As shown, some of those fibrils break off and become part of the fines content of the suspension. In addition, the increased conformability of the cell wall leads to a tendency towards a ribbon-like cross-section of refined kraft fibers (Molin and Daniel 2004; Debnath *et al.* 2021).

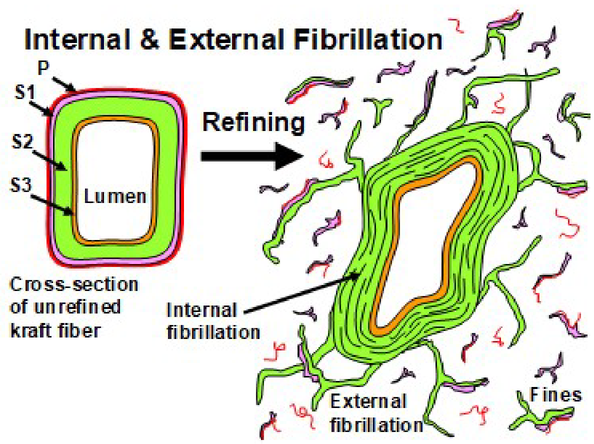

Fig. 8. Depiction of changes in the cross-sectional view of a kraft fiber subjected to mechanical refining action, leading to delamination within the cell wall (especially the S2 layer), external fibrillation, peeling off of cellulosic fines, and development of a ribbon-like shape

When cellulosic fibers are subjected to a very high level of mechanical refining, the extensive shearing and compression eventually will produce micro- or nanofibrillated cellulose (NFC) (Lavoine *et al.* 2012; Abdul Khalil *et al.* 2016). Klayya *et al.* (2021) esterified NFC with lactic acid. The treatment gave more favorable dewatering rates and strength. Because nanofibrillated cellulose is known to greatly increase the time required to remove water from pulp mats, such findings are worth noting.

Additives to the suspension

In addition to the fiber materials, the suspension that is used to prepare molded pulp properties often will contain chemical additives. Three main categories of such additives can be called binders (or strength aids), hydrophobic substances, and flocculants.

Starch varieties are widely used as binders to increase the strength properties of molded pulp products (Noguchi *et al.* 1997; Jeefferie *et al.* 2011; Waranyou 2014). Though relatively little has been reported pertaining to optimal usage of starch in molded pulp products, a great deal is known based on general paper industry applications of starches. In particular, cationic starch products are effective in increasing product strength when added to the pulp slurry at levels of about 0.5 to 1.5%, based on solids relative to fiber solids (Howard and Jowsey 1989; Chemelli *et al.* 2020). In cases where it is important to retain a substantial portion of the initial strength even when the material becomes wet, it can be advantageous to add wet-strength agents. For example, polyamidoamine-epichlorohydrin resins, which are ideal for neutral to moderately alkaline pH conditions, can be used (Su *et al.* 2012). Such resins cure and provide some covalent cross-linking within the structure when dried under hot conditions. Crosslinked glyoxylated polyacrylamide (GPAM) polymers have the combined advantages of crosslinked polymers and glyoxal groups. The glyoxal content of the polymer produces a hemiacetal structure between the aldehyde group and the hydroxyl groups in cellulose in the papermaking process, which reduces the expansion and deformation of rewetted paper. Addition of GPAM not only considerably improved the dry strength of paper sheets, but it also significantly enhanced their wet strength (Yuan and Hu 2012). In the case of coated papers, the coating layer can include either starch products or synthetic latex (*e.g.* styrene-butadiene resins) to serve as a binder for mineral pigment particles, including clay or calcium carbonate. Such materials often become incorporated into molded pulp products due to recycling of the coated paper or paperboard. Some of the most prominent strengthening and binding agents are represented in Fig. 9.

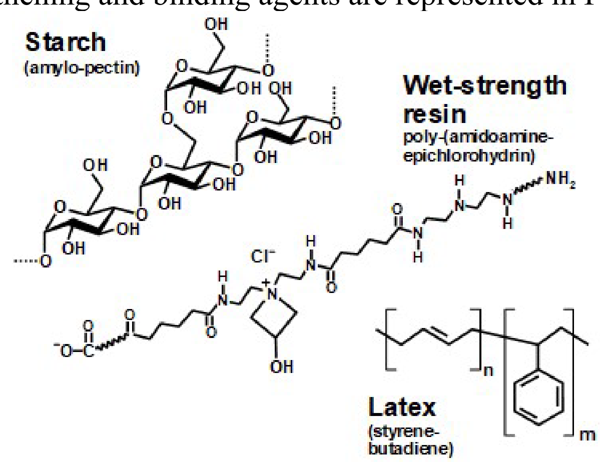

Fig. 9. Representation of various binding agents likely to be present in recovered paper, including dry-strength agents, wet-strength agents, and coating binders

Hydrophobic substances

Many molded pulp items, especially those used for food shipping or service, can benefit from hydrophobic treatment. Some of the major hydrophobic agents used in molded pulp applications can be classed as sizing agents, lignin, and waxes. Rosin products have traditionally been used in molded pulp products, in combination with aluminum sulfate (papermaker's alum) (Kucherer 1995). However, as noted earlier, complications can arise when the main fiber supply is recovered paper. The calcium carbonate filler that is present in a large proportion of recovered paper renders the pH too high for effective usage of rosin and alum. For this reason, alkylketene dimer (AKD) is likely to be the most practical sizing agent to be added to the pulp slurry before forming and drying the product (Ehrhardt and Leckey 2020). Waxes, when needed, are typically applied to the surface of a molded pulp product (Waranyou 2014; Buxoo and Jeetah 2020).

Lignin, one of the three main components of wood, can be described as a natural phenolic resin. Due to its low price, generally hydrophobic nature, and its potential contribution to bonding when sufficiently heated, it has been considered as an additive for thermoformed or heat-cured molded pulp products. Liu *et al.* (2021) achieved favorable results when adding enzymatic hydrolysis lignin to the fiber mixture when forming molded pulp products. The system was treated with ferric hydroxide and hydrogen peroxide to activate the lignin. Strength increases were observed. Because of the relatively high basis weights of many molded pulp products, it can be expected that the lengths of time and temperature conditions will be enough to cause the lignin to flow (Back and Salmén 1982). Such flow, especially when combined with pressure, can be expected to contribute to bonding within the structure.

Cationic emulsions of maleic anhydride derivatives of fatty acids such as oleic acid, as well as abietic acid, can be used as a potential hydrophobic sizing agent as they are low cost, ecofriendly, abundant and most importantly the presence and self-assembly between the hydrophobic tails of fatty acids, that are responsible for the sizing effect may be considered. Addition of aluminum sulfate to the fatty acids will help to attach to the fiber surfaces and contribute to making the paper hydrophobic (Bildik Dal *et al.* 2020).

Flocculants

As noted by Gavazzo *et al.* (2003), flocculating agents can have a large influence on the success of a pulp molding process. Two classes of flocculant have been employed routinely in such production. When using virgin fibers, such as mechanical pulps or chemi-mechanical pulps, it can be cost-effective to use aluminum sulfate as the main flocculant. In addition to its role in the fixing of rosin size (see earlier), the aluminum sulfate can contribute to a bulky, porous mat structure, which will tend to drain well and to retain the fine particles during the vacuum application. Within a pH region of about 4.5 to 6, the ratio of aluminum ion to OH^- ions can be optimized to form oligomeric species of alum, in addition to $Al(OH)_3$ precipitate; these conditions can be effective to flocculate the mixture (Strazdins 1986, 1989). Part A of Fig. 10 represents the three aluminum species that are likely to contribute to the flocculation of cellulosic suspensions under weakly acidic conditions.

The other main class of flocculating agent includes very-high-mass copolymers of acrylamide. When such additives are prepared with about 3 to 10% of cationically charged monomeric groups, they can be used directly as flocculants for cellulosic materials, which typically have a negative surface charge (Hubbe *et al.* 2009). Such additives are believed to function by bridging of the macromolecular chains between the adjacent surfaces in the suspension. The polymeric flocculating agents are a favored choice when the fiber suspension has a pH in the range of about 7 to 8.5, *i.e.* the neutral to alkaline pH range. Later sections will focus on the role of flocculants in promoting dewatering and in decreasing the rate at which the wetted surfaces of production equipment become contaminated with deposits.

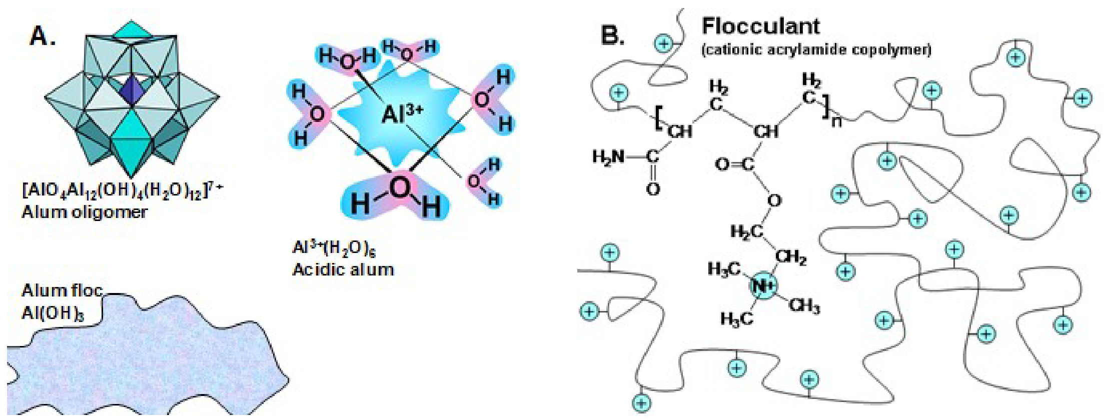

Fig. 10. A. Three species of aluminum that are likely to contribute to flocculation of cellulosic fiber suspensions; **B.** Representation of a very-high-mass cationic copolymer of acrylamide (flocculant or retention aid). Note that the cationic groups are randomly distributed in such polymers.

Manufacturing Processes

Reviews of manufacturing processes

Having discussed product classes, typical properties, and key components and additives for molded pulp products, the remaining background topic to be covered here is the manufacturing processes. As described in earlier review articles (Emery and Emery 1966; Porteous 1977; Ebmeyer 1994; Anon. 2011; Didone 2020), although not all molded pulp items are made in the same way, there are some common features. For instance, molded pulp operations start with the mixing a fiber suspension, often with a consistency of about 4% (Hunt 1998). The fiber suspension is supplemented with selected additives from among those mentioned in the previous subsection. The suspension is then dewatered in a mold, usually by suction, but sometime by use of pressurized air (Paine 1991). Next, there is an option of mating the mold with an oppositely matching mold structure, so that the molded pulp structure becomes squeezed between the two, expelling more water (Didone *et al.* 2017). The final step is drying, which can either be within the mold (press drying or thermoforming) or after the product is released (unconstrained drying) (Anon. 2011). The steps just mentioned will be considered below in more detail.

Vacuum forming in three dimensions

Vacuum is the most common means by which fiber solids are drawn toward a screen surface during production of molded pulp items (Eagleton and Marcondes 1994; Anon. 2011). Figure 1, shown earlier in this article, provides a schematic illustration of such a process. Porteous (1977) reports that as much as 85% moisture content may remain in the molded pulp item at the end of the vacuum application. Though the consistency (filterable solids content) of the suspension can fall in a wide range, its value is determined by the desired mass per unit of area, as well as the volume provided within the mold equipment. Thus, whereas paper machines often operate with a headbox consistency of about 0.5%, the consistency during molded pulp manufacturing is often about four to ten times higher. Another key difference is that the wet web of paper will pass over multiple dewatering units, such as hydrofoils or forming blades, as well as suction boxes having increasing levels of vacuum (Hubbe *et al.* 2020). By contrast, a typical molded pulp production operation entails a single application of vacuum at a single level. The implications of this difference will be considered in a later section, when considering factors affecting the production rate.

Pressurized air molding

Different designs of equipment can make it possible to achieve nearly equivalent results with pressuring air in place of vacuum (Porteous 1977; Paine 1991). Kucherer (1995) describe a Swiss company that used such processing in the period between 1925 and 1969 to make such molded pulp items as cans, tubes, and bottles, which were called "blow-molded". By usage of hot air, such a method can achieve at least partial drying of the formed object (Paine 1991; Anon. 2011). Paine (1991) reports that solids contents in the range of 50 to 55% could be achieved in such operations, with the remainder of the water being removed in a secondary drying step. Kumamoto and Otani (2001) and Kumamoto *et al.* (2002) patented a related technology in which air pressure is used to inject a balloon into the interior space of a multi-part, detachable mold system, thus forcing a fiber suspension outwards to the screen surfaces. Because the flexible membrane presses against the interior, that surface becomes much smoother than would have been achieved by the other methods described so far in this section.

Precision forming with pressure

The terms transfer molded and precision forming are used in cases where, after the application of vacuum, part of the mold rotates so as to mesh with an oppositely-shaped structure that presses against the layer of molded pulp (Didone *et al.* 2017; Hurter 2017). Such a process was shown schematically earlier in this article in Fig. 2. As implied by the word "precision", such a procedure provides an opportunity to achieve more exacting replication of structural details, including smoothness of the side facing away from the initial screen surface (Saxena *et al.* 2020).

Drying and curing

Challenges associated with the drying of molded pulp items include the three-dimensional shapes, non-uniform thicknesses, and non-uniform densities of the formed objects. Conventional steam-heated cylinders, as used in production of most ordinary paper, are unsuited to the job of evaporating water from molded pulp products. That is unfortunate, since such technology has become highly optimized, with efficient recovery of latent heat from the evaporated vapors (Hubbe 2021). Unconstrained drying means that heat is applied to the item after it has been released from the mold (Anon. 2011). This can be done in an oven, typically with circulation of air to promote even drying.

Terms including press-drying and thermoforming can be used when heat is applied during the transfer operation from the initial vacuum forming to the mated surface (Sutton 1978; Anon. 2011; Sridach 2014; Waranyou 2014; Wang *et al.* 2017; Didone *et al.* 2020; Klayya *et al.* 2021). As the molded pulp item is pressed between the two surfaces, it becomes drier and denser simultaneously as the water continues to be expelled through microchannels in the mold surfaces. Didone *et al.* (2020) estimated that the drying operation requires about 8 to 20 times as much energy in comparison to the initial forming process with vacuum. A particular challenge that faces manufacturers when employed press-drying procedures is the likelihood of undesirable delamination of the product (Didone *et al.* 2020). The mechanism of such delamination has been studied in detail in connection with earlier efforts to speed up the drying or ordinary paper (Lucisano and Martinez 2001). In principle, such delamination can be minimized by slowing down the process, giving the vapors more time to escape through the porous openings of the mold before the mold surfaces are separated.

Processing challenges

Though the manufacturing steps outlined above are being effectively used to produce a wide range of molded pulp items, there are some important challenges that seem to stand in the way of rapid expansion in manufacturing. These include issues related to production efficiency, the relatively high amounts of energy needed to evaporate water, and the relatively slow processes involved in individually forming, dewatering, and drying each molded pulp item. The next section addresses factors affecting the production rate.

FACTORS AFFECTING THE PRODUCTION RATE

Cost issues

Factors related to the drainage rates of water, the evaporation of water, as well as operational efficiency issues can be expected to have a major impact on the production rate of a molded pulp operation. Such issues have been considered in various articles on the topic (Porteous 1977; Luan *et al.* 2019; Klayya *et al.* 2021), but there has been a need to discuss such issues in an integrated manner.

As illustrated in Fig. 11, a factory based on continuous processing needs to cover different kinds of costs in order to remain economically viable. Fixed costs can include insurance, lighting, salaried labor, and debt payments on capital items related to the production. Semi-variable costs can include some contract labor. The most important variable costs will be the materials used in manufacturing (Hubbe and King 2009).

An important topic to consider, while discussing the various factors affecting water removal rates, is whether some of what has been learned in studies of the papermaking process can be borrowed and applied in the field of molded pulp production. A key finding from such studies is that more rapid dewatering, along with less stratification of the density of the wet fiber mat, often can be achieved by applying repeated pulses having gradually increasing intensity (Hubbe *et al.* 2020). Gentle suction, by means of hydrofoils, can be quite effective while the wet web of paper consists of mostly water and lacks strength. Increasing vacuum can be applied as the wet web reaches higher solids contents, along with greater ability to resist applied forces.

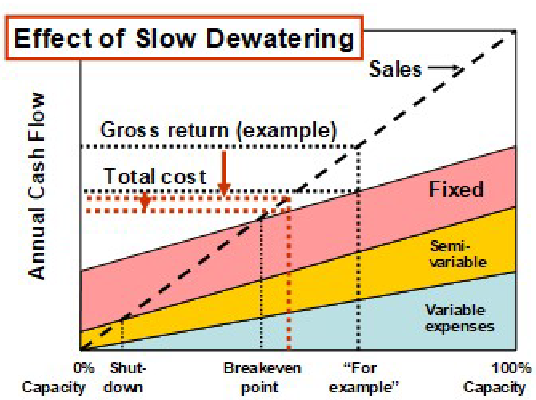

Fig. 11. Illustration of how production rate is necessary to cover different kinds of costs and maintain profitability in a manufacturing business. The bold dotted lines indicate an assumed "normal" operating condition. The bold red lines indicate what would happen if slow dewatering made it necessary to decrease the speed of production. The downward arrows represent before-tax income for the "normal" operations and the slower operations.

Various versions and implications of this hypothesis will be considered in the discussion that follows, while keeping in mind that testing or implementation of the hypothesis may be limited by the available equipment in specific cases. These issues are highlighted in Fig. 12. As shown, a typical flat paper machine system will include several hydrofoils, each providing a separate pulse of very low vacuum, followed by low-vacuum flat boxes (often with increasing vacuum), then high-vacuum boxes the progressive vacuum levels, and finally two to four felted wet-press nips, which again typically have increasing intensities.

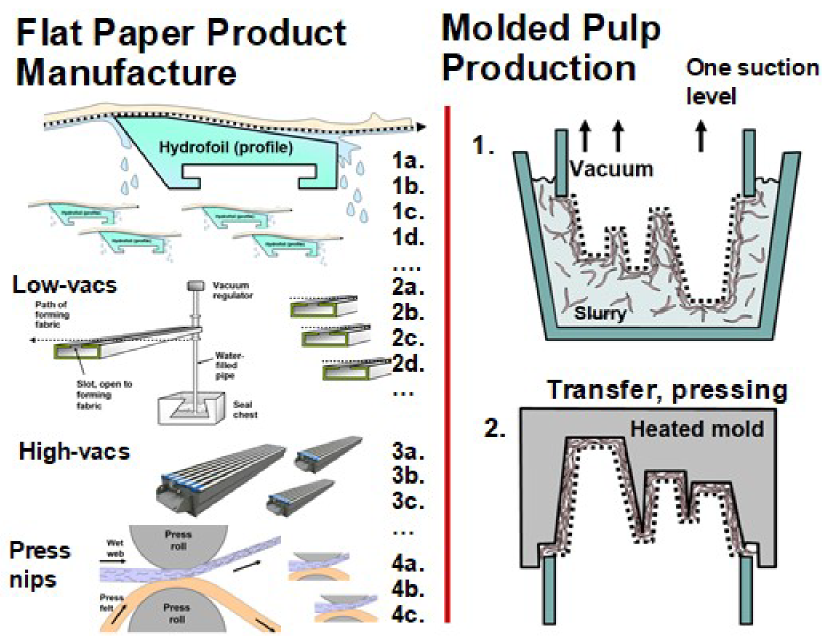

Fig. 12. Contrasting the number of vacuum or pressure instances in typical papermaking *vs.* typical molded pulp production

Kozeny-Carman Analysis

Imagine a molded pulp operation in which the screen surface of a mold has just come into contact with the fiber suspension but no vacuum has yet been applied. At that instant, the mixture adjacent to the screen surface will be relatively uniform and most of it will consist of water. At that point, the water can rush quickly toward the screen as soon as the vacuum is applied. Almost immediately, a mat begins to form, and the resistance to flow increases. According to Darcy's law, the rate of flow is expected to be proportional to the applied pressure, times a permeability coefficient, and divided by the fluid viscosity, *i.e.* the viscosity of water (Darcy 1856). Kozeny and Carman (Kozeny 1927; Carman 1937, 1938; Carrier 2002) are credited with the first rudimentary attempts to account for the permeability coefficient. The theory assumes a uniform bed comprised of incompressible, sphere-like particles. The model is illustrated conceptually in Fig. 13. Notably, when predicting the flow rate, the adjustable parameters include the applied pressure, the fluid viscosity, and the radius of the particles, and the porosity (fractional void space) of the bed. Based on such a model, it is easy to propose that the resistance to flow would be expected to increase in proportion to the thickness of the mat than has accumulated on the screen surface of the mold.

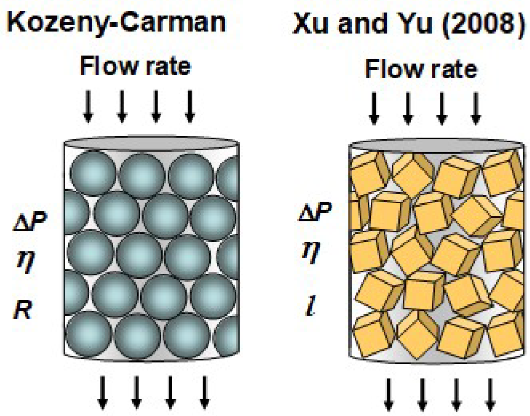

Fig. 13. Conceptual illustration of assumptions made in the Kozeny-Carman and Xu-Yu models to predict the flow rate through a packed bed, based on the applied pressure, the fluid viscosity, a measure of particle size, and the porosity of the bed

When attempting to explain the resistance to the flow of water through a suspension of particles, several important points can be learned from the Kozeny-Carman equation (Pal *et al.* 2006). The first is that the resistance to flow tends to increase with increasing specific surface area (area per unit mass) of the particles. If one assumes that the particles are identical in shape and non-porous, then it follows that smaller particles will contribute to greater resistance to flow. High levels of fine particles are known to slow down the dewatering of cellulosic fiber suspensions (Kullander *et al.* 2012). Luan *et al.* (2019) reported that fines contributed to longer dewatering times during production of molded tableware. The Kozeny-Carman theory also explains why heating up of process water will tend to increase the dewatering rate. That is because water's viscosity decreases markedly with increasing temperature. The Kozeny-Carman theory also provides a rationalization of why the resistance to flow would increase with increasing mat consistency.

In an attempt to make the Kozeny-Carman analysis more realistic and to apply it in more situations, some aspects of fractal theory have been used (Costa 2006; Xu and Yu 2008). Costa (2006) derived a model in which the two fitting parameters were the Kozeny-Carman coefficient and a fractal exponent. The model gave an improved fit to data from the permeability through beds of pumice particles having differing porosity. Likewise, Xu and Yu (2008) found that a fractal model corresponding to a square geometric model gave the best fit to practical data. This approach is illustrated, in an approximate way, in Fig. 13. Note that the cubes shown in the illustration are meant to represent cubic in a fractal sense, rather than representing actual cubes. The original Kozeny-Carman model assumed spherical particles of equal size, and such a model did not fit the data as well.

Mat Structure and Permeability

The Kozeny-Carman analyses, as described above, tend to give misleading predictions for wet cellulosic fiber mats. The particles in the mat or bed are assumed to be incompressible and are assumed to remain in fixed, uniformly distributed positions. Real mats of cellulosic fibers exhibit compression. Such compression implies that the fibers can become deformed and pressed together, possibly sealing off some of the flow channels. Ingmanson (1952, 1953), when applying the Kozeny-Carman approach to the dewatering of paper, introduced a way to account for compressibility. Further progress, related to incorporating issues of compressibility and different shaped particles into the prediction of flow resistance, are covered in an earlier review article (Hubbe and Heitmann 2007).

The structure within a fiber mat can be expected to have a large effect on the flow resistance. In principle, relatively stiff fibers can be expected to contribute to a bulky mat structure having relatively open channels available for flow. Support for this concept includes the fact that mechanical refining of fibers tends to render them less stiff in the wet state (Paavilainen 1993), and it also tends to slow rates of dewatering (Gharehkhani *et al.* 2015). Based on the theoretical work of Philipse and Wierenga (1998), there is a huge range of packing that can be achieved by a random assembly of rod-like particles, depending on the assumptions that one makes in how they interact.

Lindström (1989) proposed that a bulking effect is associated with frictional effects among fibers during the formation of a fiber mass during the dewatering process. It is known that such frictional effects tend to produce a greater volume of sediment when particles or fibers settle from a suspension and form a cake (Kline 1967; Alince and Robertson 1974; Gruber *et al.* 1997). That is because the fibers each tend to stick to each other upon contact rather than sliding into a densely packed form. In practice, such effects can be achieved in molded pulp production by the use of flocculants, *i.e.* retention aids or alum treatment (Gavazzo *et al.* 2003). Such effects will be discussed when considering factors affecting the rate of release of water from pulp suspensions and mats.

Flow Resistance through the Screen

In addition to the main resistance to flow provided by the fiber mat itself, it is also worth considering the flow resistance provided by the screen and other structural aspects of the mold. Though one can expect a wide diversity is the details of mold construction, a metal wire mesh screen often faces the fiber suspension (Cullen Packaging Ltd. 2012). Such a screen is illustrated in Part A of Fig. 14. The cited article describes a metal screen having 50 μm wires with 50 μm spacing between the wires. These details are in a similar range to a 200-mesh screen, as has been used in the preparation of standard handsheets (TAPPI Method T205). The pressure loss during flow of incompressible fluid (such as water) through such a screen can be estimated using equations obtained by Brundrett (1993) over a range of Reynolds numbers.

The mold structure itself, which supports the screen, generally has much larger but far less numerous perforations, allowing for release of water, in response to the applied vacuum or pressure. Although a simple cylindrical drilled hole could be considered, other hole profiles may be preferred. For example, conical-profile holes are used in the type of screen that is used for the Dynamic Drainage and Retention Jar, which has been widely used for many years for evaluation of retention and drainage aid for papermaking (Britt 1981; TAPPI Method T 261 cm-94). Part B of Fig. 14 illustrates both the Britt jar apparatus and a profile of the screen detail. By presenting the smallest part of the openings towards the fiber suspension, such a format minimizes the chances that material will get stuck in a passage. In addition, it seems likely that flow resistance would be less compared to when the liquid has to pass through cylindrical holes matching that minimum diameter.

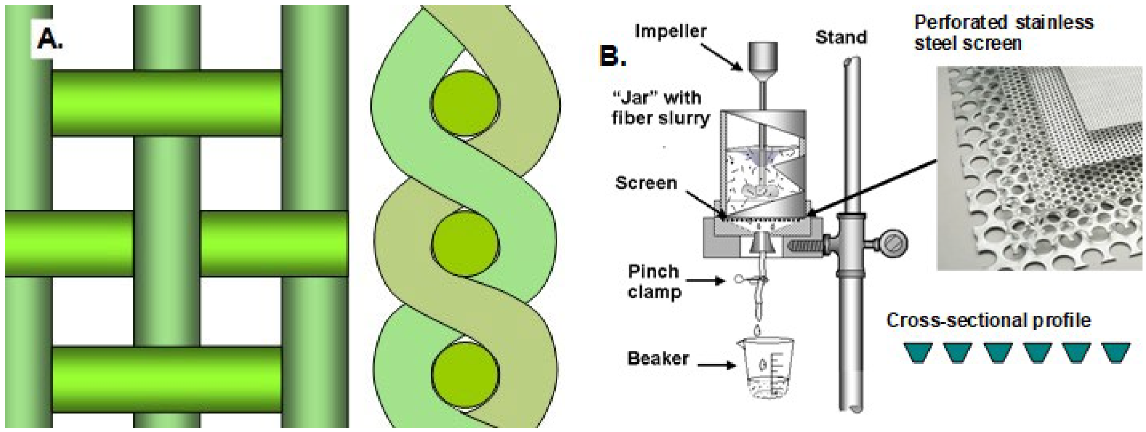

Fig. 14. A: Plain-woven screen for which calculations of flow resistance are available;
B: Illustration of the Britt Dynamic Drainage/Retention Jar, showing also a cross-sectional view of the conical holes of the screen

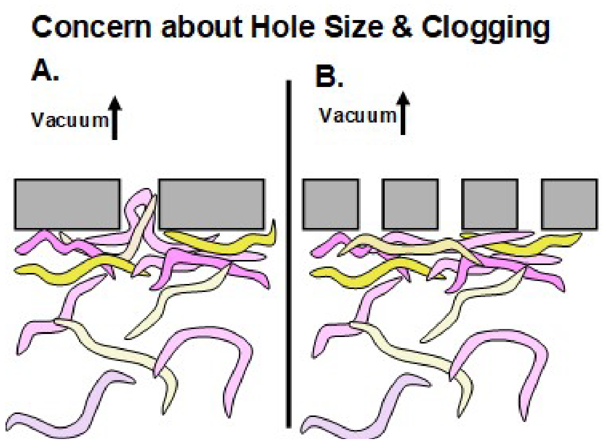

Fig. 15. Concept of how the size of opening at a screen surface may influence the degree to which fibers landing on the screen tend to droop into the throats of the openings

In addition to the flow resistance of the screen itself, many papermakers have experienced a phenomenon that has been called sealing. This happens when individual cellulosic fibers are able to droop into the openings of a forming fabric, thus slowing down the flow. A pictorial theory to explain this phenomenon was advanced by Kufferath (1983). He envisioned openings within a forming fabric as each being like a funnel. Fibers able to get into the throat of the funnel were anticipated to have a large adverse impact on dewatering, whereas those lying across the rims of the funnel did not contribute greatly to the sealing of flow through the fabric. Such a concept leads logically to an expectation that a relatively small size of opening might be ideal – small enough to discourage entrance by drooping parts of fibers. This concept is illustrated in Fig. 15.

Deposits of pitch, stickies, or scale, *etc.*, within a screen or multiply perforated surface of a mold also can be expected to decrease flows. Such issues will be considered later, when discussing deposition and deposit-control strategies.

Effects of Fines

When evaluating the freeness of papermaking pulps, the rate of dewatering is profoundly sensitive to the presence and amounts of cellulosic fines (Hubbe 2002; Cole *et al.* 2008; Chen *et al.* 2009). As noted by Gess (1991), the adverse effect of fines on dewatering tends to be greatest when both the content of fines and the basis weight are high. These are expected to be issues of great concern when preparing molded pulp products from recovered paper. Not only are the basis weights generally at the higher end of what papermakers often deal with, but also the recycled pulp may include fines-rich components, such as mechanically pulped fibers. Not only is the fines content often relatively high in recycled pulp mixtures, but there may be large hour-to-hour or day-to-day variations in fines content.

The types of fines having the greatest adverse effect on dewatering are the fibrillar fines produced by delamination of the outer layers of fibers during their refining (Hubbe 2002; Cole *et al.* 2008; Hubbe *et al.* 2008). Not only are these the most slender fines, contributing to a high specific surface area, but they are also long enough to easily become stuck as they are drawn through a wet mat of fibers.

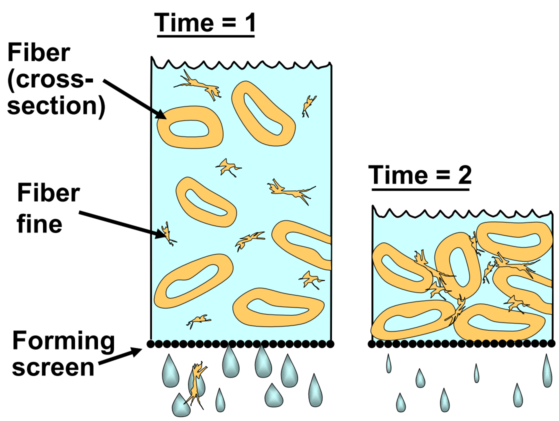

Fig. 16. Illustration of the choke-point mechanism to explain the adverse effect of cellulosic fines on dewatering rates, especially during low-shear dewatering of relatively heavy basis weight fiber mats when the content of cellulosic fines is relatively high

A mechanism to explain the adverse effect of fines on dewatering is illustrated in Fig. 16. To be able to show both the fibers (with lengths 1 to 3 mm) in the same view as the fines (defined to be less than 76 µm according to a screen separation assay (TAPPI Method T 261), the fibers are shown in a cross-sectional view. The idea implied by Fig. 16 is that unattached fines will tend to be drawn through the fiber mat by the flow. Some of the fines may get stuck in the choke-points of the mat, especially in parts of the mat that are denser. In addition to cellulosic fines, suspensions of fibers in a mill environment may contain substantial amounts of entrained air. A study by Lorz (1987) compared the drainage times when paper handsheets were formed with different amounts of air present in the mixture just before starting the drainage. Just 0.4% of air, on a volume basis, was enough to double the dewatering time. The effect can be attributed to bubbles blocking the drainage channels within the fiber mat. Problems related to entrained air can be addressed, in general, by using a multi-part strategy. One begins by minimizing unnecessary addition of anything likely to contribute to the stabilization of foam bubbles, *e.g.* surface-active agents and dissolved polyelectrolytes not attached to surfaces. The next step is likely the judicious addition of a low and steady dosage of a defoamer (Pelton 1989; Rekonen *et al.* 1990; Wilson and Wittich 2022). These products are typically emulsions of water-insoluble surfactants, sometimes formulated with hydrophobic particles, such as hydrophobized silica. Upon addition to the pulp suspension, the active ingredients have an affinity for the surface-active materials already associated with the walls of bubbles. Upon reaching those interfaces, the defoamer molecules spread rapidly, leading to the coalescence of adjacent bubbles. Although defoamers can be highly effective, their overuse can contribute to deposit problems and difficulties with the effectiveness of hydrophobic sizing agents. Further progress against the adverse effects of entrained area can be achieved by mechanically removing air from the fiber suspension (Matula and Kukkamäki 1998). This can be accomplished by devices in which vacuum is applied. Dissolved air thereby comes out of solution, and the entrained bubbles become greatly enlarged, so that they float to the water's surface and break.

Densification Dynamics and Rewetting

Densification of layer adjacent to screen

Whenever a wet mat of fiber is dewatered under pressure or vacuum, the structure becomes densified adjacent to the screen or felt into which water is flowing (Hubbe *et al.* 2020). The situation is represented, pictorially, in Fig. 17.

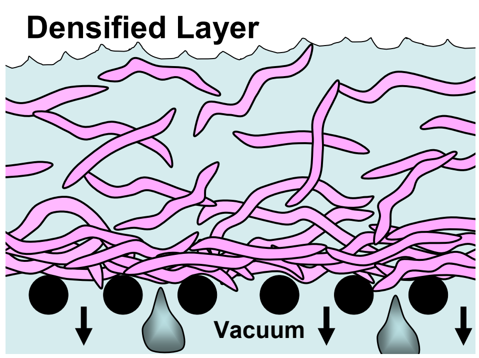

Fig. 17. Representation of how a densified layer can be expected to form adjacent to a screen from which water is allow to leave the material

The phenomenon depicted in Fig. 17 is particularly noticeable when the wet-web of paper passes through a single-felted wet-press nip (Paulapuro 2000, 2001; McDonald 2020). Chang (1978) noted that the effect tends to be strongest when there is strong resistance to flow, *e.g.* in the case of well-refined kraft fibers. Campbell (1947) was apparently the first to propose that such densified layers would be a key point of resistance to dewatering.

To explain the stratification phenomenon, is has been proposed that pressure within a wet web of paper during pressing is supported by two main components, namely mechanical pressure and hydraulic pressure (Wahlström 1969; MacGregor 1989). On the side of the paper opposite to the felt, any possibility of net flow of water is completely blocked by the presence of a solid press roll, and the hydraulic component of pressure can be expected to be dominant in that location. By contrast, on the side facing the highly permeable felt, there is continuous leakage of water from the fiber mat, and hence the solid material is forced to take up more and more of the applied load.

Though there are some key differences, it can be argued that a very similar situation results when a fiber mat is dewatered against a screen by vacuum or applied pressure, as in the case of molded pulp production. The flow of water, in response to the applied vacuum, dynamically pulls upon the fibers in the mat, squeezing them in the direction of the screen surface (Didone *et al.* 2017). So even though such densification may be difficult to measure, it is proposed here that it is likely to explain some key aspects of both molded paper production and high-vacuum dewatering of paper associated with suction boxes and couch rolls (Hubbe *et al.* 2020). Because molded pulp operations typically employ a single vacuum step, drawing the fiber suspension toward a mat under low hydrodynamic shear conditions, such systems are likely to be especially susceptible to the adverse effects of the densification and sealing aspects of fiber mats during dewatering.

Diminishing effects of vacuum application

In the beginning of this section it was hypothesized that the dewatering performance of molded pulp processes might be improved by application of pulses of gradually increasing intensity. Indeed, when using vacuum to remove water on a conventional Fourdrinier-type paper machine, it is common practice to employ progressively more vigorous conditions (Hubbe *et al.* 2020). Initially, hydrofoils, placed just below the forming fabric, provide pulses of relatively gentle vacuum (Meyer 1971). Next, after the solids of the wet web have risen sufficiently, low-vacuum boxes are used, in which the suction is provided by the siphoning action of water removed from the paper web, as it passes down a tube to a seal pit (Pitt 1987). Next, when the wet web is yet a bit stronger, high-vacuum suction is applied, using vacuum pumps (Johnson 1991; Räisänen 2000). Finally, the highest level of vacuum may be applied as the wet web passes over a vacuum couch roll (Johnson 1991), just before being transferred into the wet-press section of the paper machine.

When a steady vacuum is applied to dewater a fiber suspension, the flow has been shown to decrease to a low value after an initial period (Baldwin 1997). The cited article showed that an effective way to overcome such a problem is to increase the applied vacuum as the wet web passes over successive high-vacuum flatboxes. A possible explanation for this is that the higher vacuum is able to overcome resistance that was out of range of the lower vacuum at the previous suction box. A problem with that explanation is that one might expect the higher vacuum to simply tighten the sealing action of an already-densified layer pressed against the screen. It was proposed in an earlier article (Hubbe *et al.* 2020) that the key to such dewatering as a wet web passes over a series of vacuum flatboxes involves the partial re-expansion of the fiber mat in each interval between vacuum applications. The natural elasticity of the wet web can be expected to cause a spring-back effect, especially in the highly compressed layers. The expansion can be expected to draw air and maybe some water back into the sheet, rendering the material more permeable before it reaches the next vacuum application.

Having described a set of strategies that appears to work effectively for the manufacture of flat paper products, a question can be raised about whether parts of such a strategy can be effective for molded pulp processes. Most molded pulp production systems employ just one vacuum application and often an additional pressing step. Are there ways to incorporate progressive increases in vacuum or pressure during such production? Are there ways to incorporate multiple pulses, thus allowing for some elastic re-expansion and restoration of permeability between the pulses? These may be questions that can be considered by machine designers and researchers.

Rewetting issues

Another action that is implied by the elastic re-expansion of the mat, after the end of a vacuum application, is that some of the just-removed water may be drawn back into the material. Such rewetting has been considered for many years in relation to the wet-pressing of paper (Chang 1978; Jaavidaan *et al.* 1988; McDonald and Kerekes 1995). It has been studied in the case of vacuum dewatering at suction boxes (McDonald 1999; Granevald *et al.* 2004; Sjöstrand *et al.* 2015). Based on the cited studies, Fig. 18 shows a possible approach to estimate the maximum rewetting pressure that will be acting to draw water back into a fiber mat as it elastically expands after it has been compressed (*e.g.* by applied pressure or suction). In principle, suction can happen when capillary forces are preventing air from rushing into the mat and when there are continuous columns of water spanning the thickness of the mat. The mechanism starts to fail if and when the suction exceeds the maximum capillary pressure (Miller and Tyomkin 1986).

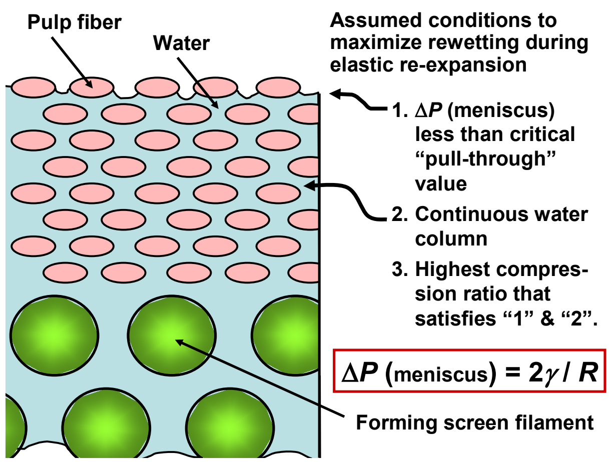

Fig. 18. Approach to estimating the condition of maximum rewetting in response to elastic re-expansion of a fiber mat at the moment when the compression force (pressure of vacuum) is discontinued

A different model can be applied if one assumes that effects due to elastic recovery of the mat has been completed. Because the sizes of the pores within the cellulosic fiber mat can be expected to be much smaller than those of a screen upon which the mat is being dewatered, the dominant capillary pressure will be acting to pull water back into the mat. The rate at which water flows into the pores can be estimated by using the Lucas-Washburn equation (Lucas 1918; Washburn 1921). This equation is often used to estimate rates of penetration of wetting liquids into porous materials. As illustrated in Fig. 19, the model assumes equal, cylindrical pores of radius R, an advancing contact angle of θ, and a dynamic viscosity of η. Key predictions from the Lucas-Washburn equation are that the maximum suction is associated with low pore size, but maximum flow increases with increasing pore size. The amount of liquid taken up by the porous solid is predicted to increase as the square-root of time.

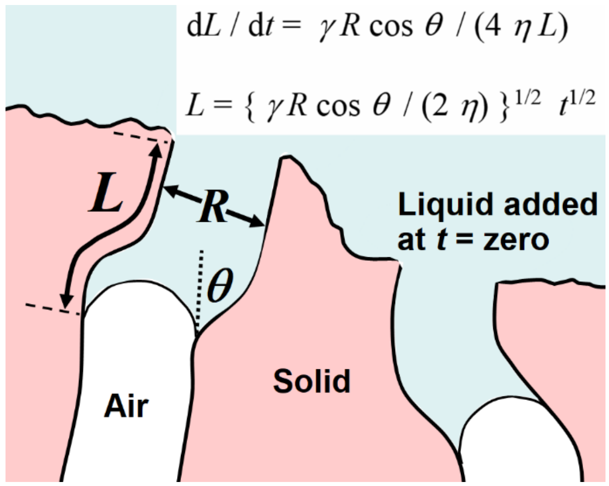

Fig. 19. Lucas-Washburn model and equations to predict the rate and extent (at a specified time) of permeation of a wetting liquid into a porous solid that is assumed to have simple, cylindrical pores

Although the re-expansion of a wet mat after vacuum compression provides a plausible mechanism to draw water back into the mat, there are some reasons to expect that such rewetting might be inefficient, especially in the case of typical molded pulp processing. Chief among these is the fact that the layer facing the screen surface will have been densified. Thus, I'Anson and Ashworth (2000) predicted that relatively little water would flow back into the mat during the relatively brief period available for that to occur during a papermaking operation. It is worth considering that longer periods of time might be available in typical molded pulp processes. Another limitation is that the drawing of liquid back into an expanding porous material will depend on capillary action, and such a mechanism will partly fail as air becomes drawn into the mat. Because typical molded pulp items are less dense than paper items such as printing paper and linerboard, there is a greater expectation that air will quickly enter into the expanding material if that is possible starting from the moment that the vacuum is released. Many molded pulp systems are set up such that air passes through the wet material before the vacuum is released. After the vacuum dewatering of a molded pulp item, it makes logical sense to continue the vacuum suction until the vat that held the fiber suspension has been moved away. A further consideration is that, due to the relatively thick nature of typical molded pulp items, the relative mass of water that is available to be picked up from a damp screen might be too small to be important. It seems that an enterprising graduate student could consider some of these issues and produce mechanistic evidence pertaining the range of pulp solids, basis weights, and fiber pad structures that are typical for molded pulp items.

Chemical-based Dewatering Strategies

The number of studies that have been carried out specifically related to molded pulp products is quite small, in comparison to those dealing with conventional papermaking, and among them, only a few have touched upon the use of chemical agents to promote faster drainage (Gavazzo *et al.* 2003; Klayya *et al.* 2021). Therefore, it will be assumed here that chemical strategies that have been found to speed up the dewatering during production of flat paper products are likely to have the same directional effects when applied to similar pulp mixtures under the conditions of molding. Since most of these issues have been reviewed elsewhere (Hubbe and Heitmann 2007; Hubbe *et al.* 2020), only what might be regarded as the most important aspects will be covered in this section.

Retention aids

According to Pelzer (2008), very-high-mass acrylamide polymers, *i.e.* retention aids or flocculants, are used on a majority of paper machines worldwide, including almost every grade of paper or paperboard. Such a flocculant, a cationic copolymer of acrylamide, was described earlier in this article (see Fig. 10). Though one of the most obvious reasons to employ a retention aid is to minimize discharge of valuable fine material into the wastewater, the optimized use of retention aids can help in terms dewatering rates (Britt and Unbehend 1980). In addition, optimized use of retention aids and related chemicals will minimize the amounts of fine and colloidal-size particulates present in the suspension, and the expected benefit may be a reduced rate of accumulation of deposited pitch or stickies (Bobu *et al.* 2002).

Britt and Unbehend (1980) showed that an adverse effect on dewatering can occur if insufficient hydrodynamic shear is applied after treatment of a stock suspension with retention aid, due to a high level of fiber flocculation. Fiber flocs created by the bridging action of an effective retention aid system may initially seem quite promising due to rapid initial dewatering. However, it was found that a high level of fiber flocs in the wet sheet was highly unfavorable with respect to vacuum dewatering. Air was sucked through the thin areas of the damp paper, and the floc areas remained wet. Therefore, when employing flocculants during production of molded pulp items, it can be important to follow up the chemical treatment with an optimized level of agitation of the fiber suspension. Whereas on a paper machine, this can easily be achieved by adding retention aid before a pressure screen (Hubbe and Wang 2002), such a device is not likely to be present (or not in the right position) during molded pulp processing. Thus, an additional shearing device, such as an in-line motor-driven agitator, is likely to be helpful before a molded pulp operation, which typically involves much lower levels of hydrodynamic shear.

The mechanism by which retention aids promote the dewatering of fiber suspensions appears to be related to the earlier discussion of the adverse effect of cellulosic fines on rates of dewatering. Unattached cellulosic fines, especially the fibrillar fines created during refining of pulps, have the potential to clog drainage channels within a pulp mat in the course of the dewatering process (Hubbe 2002; Cole *et al.* 2008; Hubbe *et al.* 2008). Of particular concern is the choking off the drainage channels in the relatively dense part of a fiber mat adjacent to a screen surface. At such locations, the mat will tend to be densest due to the somewhat longer time that has passed since its formation and due to the effects of vacuum or pressure on the mat. By anchoring fines onto the surfaces of full-length fibers, which are too long to migrate through the structure of the wet mat and get stuck, there is potential to defeat the choke-point mechanism. In addition, even if the suspension has been agitated strongly to break apart the fibers from each other and avoid excessive flocculation, the amount of hydrodynamic shear required to detach a fine particle from a fiber surface is very much greater than for a larger particle or fiber (Hubbe 1985).

High-charge cationic agents

When papermakers have a reason to promote rates of dewatering beyond what can be achieved just with a retention aid, as described above, the next option may involve a high-charged cationic polymer. The reason for considering a high-charge cationic polymer at this point, rather than aluminum sulfate (papermaker's alum), is that a majority of molded pulp products are being made with pulp from recovered paper, and many such pulps have substantial amounts of calcium carbonate. As discussed earlier, the ensuing alkaline pH conditions are not well suited to the usage of alum. Candidate cationic polymers that could be considered would include polyethylene imine (Bobu *et al.* 2002; Chen *et al.* 2011), poly(diallyldimethylammonium chloride) (Tripattharanan *et al.* 2004), and polyvinylamine (Hokka *et al.* 2014), among others. Optimum results are likely to be achieved with the addition rate is adjusted to obtain a zeta potential or charge demand of the filtrate not far away from zero (Horn and Melzer 1975; Bhardwaj *et al.* 2005; Hubbe *et al.* 2007a). This relationship is shown in Fig. 20.

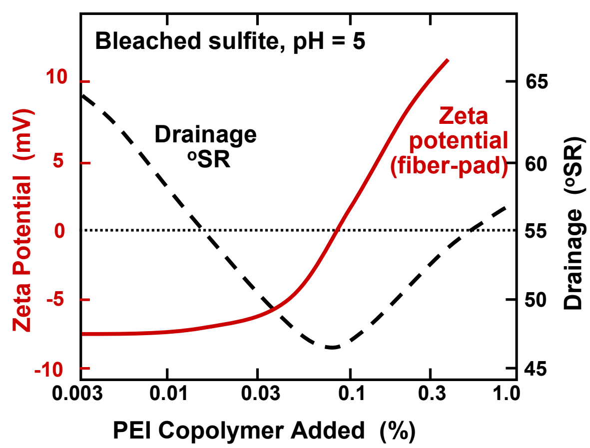

Fig. 20. Effects of high-charge cationic polymer addition on the dewatering rate (degrees Schopper-Riegler) and the zeta potential of a cellulosic pulp slurry (redrawn from original by Horn and Melzer 1975)

Micro-/ nanoparticles systems

Papermakers often are seeking rather large gains in dewatering rates, going beyond what can be achieved by combinations of retention aids and high-charged cationic polymers as just discussed. In such cases, a further step can be very promising. During the 1980s and 1990s, researchers showed great effectiveness when cationic flocculants are used in sequence with the addition of very small negatively charged particles (Andersson *et al.* 1986; Langley and Litchfield 1986; Andersson and Lindgren 1996; Hubbe 2005). Although there is some variation in how the systems are implemented, it is important that there be an addition of a high-mass cationic polymer to the furnish and a separate addition of a micro- or nanoparticle that has a highly negative surface charge and a very high surface area. Colloidal silica nanoparticles and sodium montmorillonite microparticles are added separately to the furnish, most often as the second additive. To work well, the system needs to be charge-balanced, and the amount of cationic polymer needs to be in a favorable ratio with the amount of the micro- or nanoparticle. The mechanism of action clearly involves an interaction between the cationic polymer, after its adsorption onto the fibers, and the negative surfaces of the minerals. As illustrated in Fig. 21, the cationic flocculant forms polymer bridges among adjacent solids surfaces. Then the remaining loops and tails of polyelectrolyte wrap themselves around the tiny negatively charged particles. The polymer bridges become progressively shorter during their progressive interaction with the tiny particles, which results in a general contraction effect. The system appears to function like a self-wringing sponge.

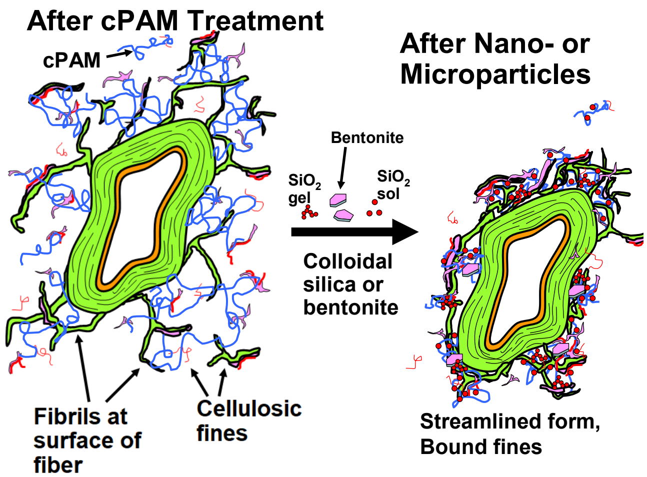

Fig. 21. Illustration of the mechanism by which combinations of high-mass cationic polymer and sequential addition of negatively charged nano- or microparticles can bring about a self-wringing sponge effect and contribute to more rapid dewatering

When considering using such an approach in the case of a molding pulp operation, a key question is whether there might be excessive flocculation of the fibers under the relatively gentile flow conditions that are typical in such operations. To minimize such a tendency, a modest first step would involve the selection of a colloidal silica product consisting of individually dispersed primary particles, *i.e.* a sol rather than a gel-type silica product (Hubbe 2005). Such products tend to favor drainage promotion effects in preference to flocculation effects.

Enzymes to promote dewatering

In cases where the main resistance to dewatering involves very slender cellulosic fine matter, *i.e.* essentially fragments resembling nanofibrillated cellulose, then cellulase enzyme treatment may be a promising strategy to achieve gains in drainage (Oksanen *et al.* 2011; Singh and Bhardwaj 2011; Piyush *et al.* 2013). The idea is to employ a sufficiently effective treatment such that it essentially dissolves the targeted very fine cellulosic material, while its action on the much larger and thicker cellulosic materials is not enough to cause serious damage. The plus side of such a strategy is that, unlike the systems just discussed, enzymatic treatments are unlikely to cause flocculation. The downside is that one needs to pay very close attention to the pH, enzyme dosage, and time of treatment. An excessive treatment would be expensive, and there would be potential serious damage to the fibers. In addition, enzymes more quickly become denatured and inactive when exposed under unfavorable temperature, pH, or other conditions.

Production Efficiency

Cost considerations

Molded pulp products generally can be regarded as a commodity business. The unit price of each molded pulp item is constrained by the marketplace, and many such items face stiff competition from competitive products made from plastic or from non-biodegradable combinations of materials. Each operation will need to cover a set of fixed costs, such as sunk costs (capital items purchased in the past and being paid off), insurance, and salaried labor, *etc.* By running such a system faster, there may be opportunities to increase revenue, since only the variable and semi-variable components of cost are expected to increase (Hubbe and King 2009). Figure 22 illustrates how interruptions in production can be expected to decrease the amount of production achieved within a period of time, such as a day. With less revenue, there would be less sales and less ability to cover the fixed, variable, and semivariable costs.

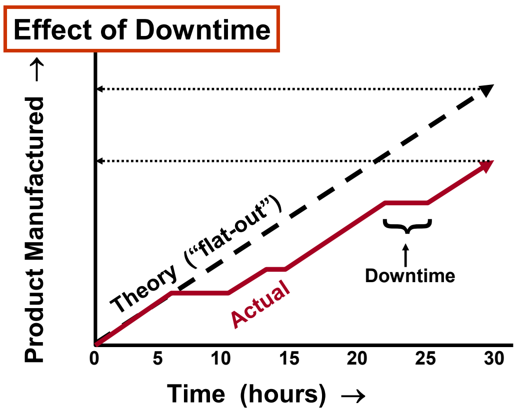

Fig. 22. Expected adverse effect on productivity when there are interruptions of operation of the main equipment, *e.g.* a molded pulp production line

The previous sections explored some ideas having potential to speed up dewatering, which often is a limiting factor in the overall production rate. However, the fact that a molded pulp system can run fast may be irrelevant during periods when it is out of service due to contamination or other problems. Therefore, the present section will focus on some effects of deposits and strategies to overcome such problems. Deposit problems on production equipment often can become more difficult with the passage of years, since there is often a progressive decrease in the quality and cleanliness of recovered fibers that can be obtained at low cost (Corwin 1972).

Deposits

Because molded paper products are often produced from recovered paper, and because a wide variety of virgin fiber sources are being considered, both pitch and stickies may be of interest to molded pulp producers. In addition, regardless of the fiber type, it is important to address issues related to biological slime and foam.

Luan *et al.* (2019) described serious pitch deposits during production of molded pulp from bleached sugarcane bagasse. A brown deposit formed during the hot pressing stage of the molded tableware production. These undesirable deposits caused serious damage to the production efficiency. Such problems are expected to increase the frequency of needed stoppages of production to clean the equipment, to reduce the quality of the products, and to reduce operational efficiency (Hubbe *et al.* 2006; Sohaili *et al.* 2016; Luan *et al.* 2019). Luan *et al.* (2019) studied such issues in a molded pulp operation. The study found that high contents of fines containing waterproofing agents and oil proofing agents were the main causes of the problems.

Control of deposits

As already mentioned in the context of increasing the dewatering rate of pulp mats, the optimized usage of a retention aid (Hubbe *et al.* 2009) is often the starting point for programs aimed at improving the productivity of a molded pulp or papermaking operation. From the perspective of deposit control, it is important to find out what dosage of retention aid is sufficient to reduce the rate of accumulation of deposited substances on forming equipment. This is likely to be a lesser treatment compared to the dosage required to achieve a high level of retention of fines in the product. When conducting trials to determine the needed chemical dosages, it is important to replicate the test conditions and to ensure that each tested condition is maintained long enough for the system to reach an equilibrium of chemical contents. Addition of a very-high-mass cationic copolymer of acrylamide (a cationic retention aid) at the level of about 0.01 to 0.05% on a solids to solids basis is often enough to decrease the turbidity and solids content of the water drained away during the forming process. In general, a moderate increase in the proportion of fines and colloidal material that is retained in the product, and thereby purged from the process water, can be regarded as a favorable indication. Figure 23 indicates the usage of cationic acrylamide copolymer, as well as some other additives, in an effort to reduce deposition of pitch and stickies onto processing equipment.

To counter-act the tacky or sticky nature of wood pitch, synthetic elastomers, and anything else that behaves similarly, various detackifying agents can be used (Hubbe *et al.* 2006; Sutman and Nelson 2022). Traditionally, talc has been used as a main treatment agent; its relatively hydrophobic surfaces allow the platy talc mineral to collect onto the surfaces of tacky and sticky particles. As noted in the cited articles, similar effects can be achieved in many systems by use of copolymers that are formulated to adsorb onto the surfaces of the tacky and sticky particles. The latter treatments may take advantage of a phenomenon called steric stabilization, whereby the surfaces of hydrophobic materials are coated by substances with tails and loops of hydrophilic polymer segments, which extend into the solution phase and prevent hydrophilic materials from agglomerating or coalescing (Hubbe *et al.* 2006).

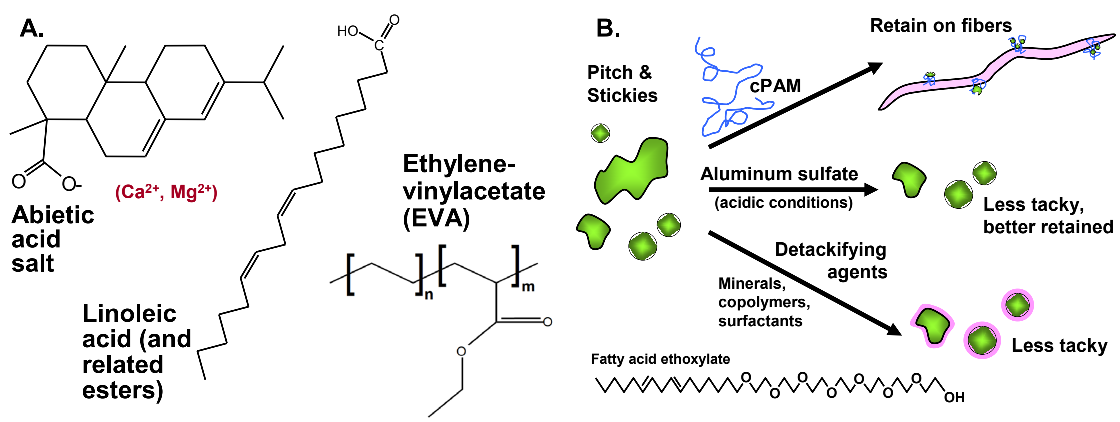

Fig. 23. Some commonly observe components of tacky and sticky deposits, as well as some commonly used types of additives for the control of tacky and sticky deposits when processing cellulosic pulp

Sometimes mill operators may notice that deposit problems are mainly localized on a specific piece of wetted equipment. In such cases, the problems often can be solved by direct treatment of those surfaces (Sutman and Nelson 2022). As depicted in Fig. 24, the materials that can be used as release agents include poly(dimethylsiloxane) and magnesium or calcium stearate. These can be prepared as emulsions or dispersions. The idea is to spray the screen with a highly dilute suspension of the release agent in the interval between vacuum applications.

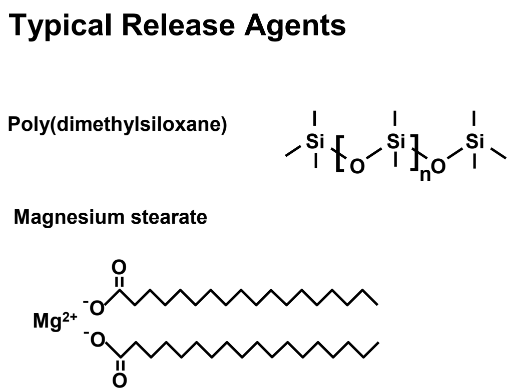

Fig. 24. Examples of commonly used release agents, which might be sprayed on the screen surface as a of dilute emulsion between suction steps

Release

When the molded pulp item is transferred from the initial suction element onto the facing part of the device, usually after a period of pressing, it is important that the release take place easily, without leaving material behind or causing delamination. Stickies or pitchy materials on the mold surface, as just discussed, may contribute to the problem. Whatever the cause, a release agent, such as those shown in Fig. 24, can be used as part of the solution. Such agents are routinely used, for instance, to optimize the creping of tissue paper (Valencia *et al.* 2020). Trials are recommended not only to select an important additive but also to be able to judge a suitable low level of addition so that the treatment does not lead to other issues, such as deposits or slipperiness. As illustrated in Fig. 25, it can make sense to gently spray the screen surfaces with highly dilute release agent in the interval between the suction cycles.

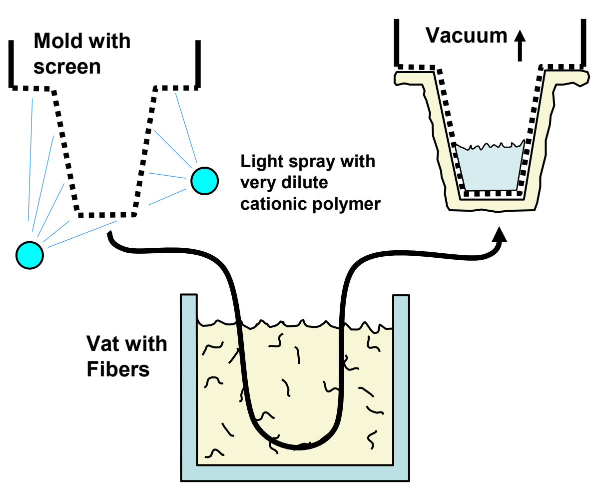

Fig. 25. Examples of commonly used release agents, which might be sprayed on the screen surface as a of dilute emulsion between suction steps

PRODUCT OPPORTUNITIES

In light of advantages such as the ability to use recycled, green materials, as well as full recyclability, it is reasonable to expect that the molded pulp industry can expand into increased volume and additional product areas. However, in each case, there needs to be a match between product requirements and physical or mechanical properties. This section considers such issues.

A Need for Standards

According to Marcondes (1997, 1998), a lack of standardization of molded pulp items may pose a barrier for companies considering alternative packaging options, especially for packaging of expensive items. At the time of that writing, the cited author wrote that there were insufficient specifications regarding such properties as shock resistance. The article introduced test methodology and recommended development of performance criteria. On the other hand, some researchers have been able to utilize available standards not limited to molded pulp items. For instance, Buxoo and Jeetah (2020) describe using common standards of papermaking for evaluation of molded cup products, which were prepared with fruit peels, hemp leaves, and a beeswax coating. An inherent challenge, in addition to some opportunities, lies in the fact that molded pulp products can be highly specialized. It can be difficult to set standards for a product that can have a seemingly infinite variety of shapes, thicknesses, and sizes. In some cases the underlying material strength properties, including modulus values and maximum stresses before failure will be capable of evaluation by use of sections cut from molded pulp products. In other cases, it might not be feasible to run standard tests, due to a mismatch between the item and the required shape needed for a test.

Optimum Structures

Some advances in computational approaches can be employed in the design of molded pulp products. For example, certain developers may be aiming to achieve the specified cushioning and strength with less material. Extensive research has been carried out on the parameters that affect such properties. Geometry plays an important role with packaging a product and the resulting static and dynamic strength. A structural factor approach has been developed that allows a modular design method to be used for molded pulp packaging design (Ma *et al.* 2004). For a long time it has been understood that performance of a molded pulp product can be improved by optimal usage of ribs and recesses, deployed in a manner to absorb shocks (Sutton 1978; Baker *et al.* 1994). Rather than developing such features by trial and error, it would be preferable to calculate the best candidate structures and then carry out simulations. Indeed, simulations have been carried out to predict the cushioning performance of molded pulp items (Ma, X. *et al.* 2004; Ma, S. C. *et al.* 2010; Geng and Xu 2011). Notably such approaches are not limited to product attributes. For example, Didone *et al.* (2020) used finite element analysis for study of dewatering issues in molded pulp production.

Opportunities Related to Curing

Compared to traditional papermaking, molded pulp products generally have a higher mass per unit area, which implies a relatively long period required for evaporative drying. Especially when such drying takes place within a press, there are opportunities for curing of various resins, both natural and synthetic, thus contributing to property development. In principle, the enhanced material properties then can be translated into product performance.

As was noted earlier, the lignin and hemicellulose components within woody materials can be made to flow under pressure in the presence of moisture, even at temperatures below the ambient boiling point of water (Back and Salmén 1982). Practical effects of such flow were demonstrated by Kunnas *et al.* (1993) in the course of studying a specialized drying process for linerboard. In the described Condebelt process, rather than drying the paperboard in contact with steam-heated cylinders, it was placed between a pair of continuous stainless steel belts, one heated and one cooled. The side with the cooled belt was provided with plastic fabrics, which gave sufficient space for condensate to collect. In addition to achieving much higher densities, the tensile index was increased by as much as 34% relative to conventional linerboard. Some of the concepts just described might well explain the relatively high levels of strength that have been achieved in some molded pulp systems involving heat and compression (Sabdin 2014). The cited author prepared structures from mixtures of pineapple leaf fiber and sugarcane bagasse waste using thermal compression.

Figure 26 shows the three categories of components within woody material that have the potential to flow and possibly contribute to bonding under the influence of heat (up to the boiling point of water), along with pressure and moisture. Though the extractives content of wood can be as low as about 1% based on mass (especially in temperate climates), it is well known that pitch-like materials can flow when heated. The ability of the hemicellulose and lignin to contribute to bonding of cellulosic materials, when subjected to heat and pressure, is well known from experiences in the manufacture of fiberboard, hardboard, and related products (Back 1987; Bouajila *et al.* 2005; Hubbe *et al.* 2018).

A question that might be considered by researchers is whether related gains in mechanical properties might be achieved in molded pulp structures that might not involve higher levels of density. Further opportunities for property development may lie in the enhanced curing of synthetic resins, such as the wet-strength resins to be considered below.

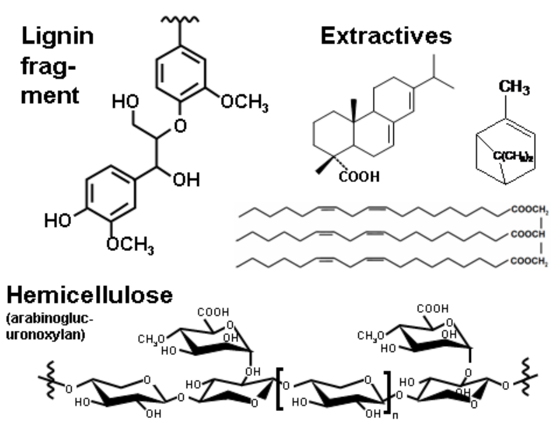

Fig. 26. Representation of the three component categories of woody materials that can be regarded as matrix materials, able to flow when plasticized by moisture. The lignin fragment includes a β-O4 linkage. The listed extractives are abietic acid, α-pinene, and a triglyceride fat comprising linoleic acid groups. The hemicellulose shown is common in hardwoods.

Strength Additives

Because molded pulp products often are intended to fill up space and provide cushioning, it can be advantageous to achieve increased strength by some means other than by densification. When papermakers rely on just mechanical refining and pressing to achieve product strength, a highly linear relationship has been found between the apparent density and the tensile strength (Guest and Weston 1990). In the cited study, the same linear relationship was found regardless of whether or not the kraft fibers had been subjected to drying and rewetting.

By usage of chemical bonding agents, there are opportunities to increase strength without as much reliance upon densification. For example, Howard and Jowsey (1989) showed that the dry-strength additive cationic starch mainly functions by strengthening the existing areas of contact between cellulosic fibers rather than by increasing the relative bonded area between adjacent fibers. It follows that by use of a bonding agent, one may be able to achieve a specified strength level with less refining of the fibers or with less wet-pressing. Some of the most promising additives to increase the bonding strength of cellulose fiber structures are starch and various wet-strength agents. The structures of these additives were shown earlier in Fig. 9. In addition, glyoxylated polyacrylamide (gPAM) products have shown promise for increasing the strength of various paperboard structures (Lu *et al.* 2020). Because the curing of gPAM involves the formation of covalent bonds (hemi-acetal), the strength is likely to exceed what can be achieved when using starch products, for instance, which rely on just hydrogen bonding. The hemiacetal bonds gradually come apart upon immersion in water, which means that the treatment material is easy to repulp. In addition, the permanent wet-strength agent poly-amidoamine epichlorohydrin (PAE) (Su *et al.* 2012) has good strength potential for such applications as molded pulp products.

Nanofibrillated cellulose has shown promise as an effective bonding agent, except that it is known to have a very strong negative effect on dewatering rates. Work by Rice *et al.* (2018) showed that the drainage effect could be overcome, while still achieving a large increase in dry strength, by first pretreating the nanofibrillated cellulose with cationic starch, and then using colloidal silica. The combination treatment was used for the strengthening of a kraft fiber structure. The bonding effect was sufficiently strong that it was possible to maintain a specified bending strength with little or no refining, while preserving a relatively low apparent density (*ca.* 0.4 g/cm^3) compared to a default value of *ca.* 0.51 g/cm^3 in the absence of the bonding system. The described system seems well suited for the production of cushioning products, for which a lower density may translate into saving in shipping costs.

Depending on the requirements, either the lignocellulosic fiber pulp by itself (as mentioned earlier) or composites made out of this natural fiber with different additives or a matrix polymer are being used to make these molded products. The natural fiber can play the role as a reinforcement for the composite (Mohammed *et al.* 2015). For example, pineapple leaf and sugarcane bagasse waste can provide strength and light weight for paper-based disposable composition applications (Sabdin 2014). Bamboo fabric-reinforced poly(lactic) acid (BF-PLA) composites have shown adequate mechanical properties (tensile, flexural and impact strength) and thermal stability (Fazita *et al.* 2015). Sometimes cellulose is isolated from the natural fiber to make a composite with another biopolymer. For example, a composite of sugarcane fiber cellulose (SCFC) and tapioca starch (TS) was synthesized along with glycerol as plasticizer for manufacturing disposable packaging food container. It was shown that addition of SCFC increased impact and flexural properties but decreased tensile strength properties at a combination of 41 wt% TS, 12 wt% SCFC and 47 wt% glycerol. Such composites can be formed as disposable bowls and plates that are microwave and freezer safe, as well as multipurpose containers (Jeefferie *et al.* 2011).

For frozen food, the paper pulp mold trays can be made with sugarcane bagasse, *Phragmites communis*, and recycled paper (Yeh and Huang 2006). During manufacturing the product, these are mixed in hydropower pulper equipment and through the vacuum-forming method to coat on a metal molding tool. After the molding operation, liquid acrylic, latex, resin, or ethylene vinyl acetate (EVA) are used with additives such as polypropylene, polyethylene, *etc*., for surface treatment. During exposure to the high temperatures, the surface of the resins and general compound coatings will dissolve easily. Then the toxic chemicals mix into the packed contents, where they can be seriously harmful for human health. Yeh and Huang (2006) added thermal-resistant additives (mixture of inorganic aluminum silicate; 50% to 80% content) and natural wax emulsion (1.5% to 9% parts relative to the fibrous material). The cross-linking agent was made with 0.75% to 5.4% parts of fluorochemical, high polymer compound (0.9% to 7.2%), 0.15% to 0.54% parts aliphatic polyamine, and 0.75% to 9% parts alkylacrylamide copolymers, relative to the amount of the fibrous material with amount equal to natural fibers. The thermal resistance additives contained anionic compounds. The surface of the paper pulp mold packing was adhered to a PET membrane by use of thermosetting adhesives. This provided not only for waterproof and greaseproof character, but also the ability to endure high temperature baking and subsequent rapid cooling. Foodstuffs packing produced by the above mentioned method can be heated with food at 220 °C in the oven for about 20 to 30 minutes and be stored at -35 °C (Yeh and Huang 2006).

Since molded pulp articles are widely used for variety of purposes such as packaging material, egg cartons, food service trays, beverage carriers, clamshell containers, plates, and bowls, additives are used to provide dimensional stability and physical strength from the molded pulp articles even when they are wetted. There exists an immediate need for improving the wet dimensional stability of molded pulp articles, especially in strongly alkaline environments. Hemmes *et al.* (2018) claimed the invention of adding a strength composition to the fiber slurry, forming a molded pulp article from the fiber slurry, and drying the molded pulp article, wherein the strength composition comprises at least one permanent wet strength resin. Wet strength resins such as polyamidoamine-epihalohydrin resins have a significant amount of reactive azetidinium groups, which provide the resin with a high cationic charge, which improves the retention of the resin to the fibers and provides the resin with a self-crosslinking ability. Preferably, the polyamidoamine-epihalohydrin resin has a charge density of 2.1 to 3.0 meq/g, determined at pH 7 by titration with potassium salt of polyvinylsulfate. When retained in the molded pulp article, the polyamidoamine-epihalohydrin resin self-crosslinks and forms a strong protection around fiber-to-fiber bonds, thus preventing the bonds from hydrolyzing, even in an alkaline environment (Hemmes *et al.* 2018).

Many biopolymers are being considered as additives or binders to improve the molded products properties. Molded pulp containers with a mixed adhesive of PVOH and starch (1:2) showed superior mechanical performance. The mixtures of alkaline pulp of sugar baggage and binders, which were conditioned to moisture level about 30%, had been formed by hot compression molding at 165 °C for 3 minutes. The water resistance of molded pulp containers was improved by the use of paraffin wax and PLA coating (Sridach 2014).

Liquid Holdout

Especially when molded pulp products are intended for use as tableware or for containment of liquids, it can be important to develop resistance to the permeation of water and aqueous mixtures. This can be a challenge in the case of molded pulp products, especially those that have a relatively low density and high porosity, as just discussed. As shown by Sørensen and Hoffmann (2003), the sorption of moisture into molded fiber tray can have a major negative effect on strength properties such as compression. According to the cited authors, the moisture content of molded pulp packaging can depend on the temperature during usage. By lowering the temperature of molded pulp package containing food, moisture condenses on the outer surface, which induces a large initial creep deformation.

The good news is that effective treatments are available and well known. When molded paper products are being prepared from virgin fiber materials, including either mechanical pulps or kraft pulps, it makes sense to rely on rosin sizing agents, such as cationic emulsions of fortified rosin (Ehrhardt and Leckey 2020). The use of a related system for the blow-molding of pulp-based containers has been reported (Kucherer 1995). For systems involving recovered pulp, which is likely to contain the alkaline mineral calcium carbonate, it may be necessary to switch to the use of a so-called alkaline sizing agent such as alkylketene dimer (Dumas 1981; Hubbe 2007; Ehrhardt and Leckey 2020). The preferred way to prepare each of these sizing agents, including the fortified rosin size, involves emulsification in a solution of cationic polymer, such as cationic starch or a synthetic cationic polymer. The positive ionic charge of emulsion particles stabilized in that manner generally can be retained at high efficiency on the generally negative surfaces of cellulosic materials.

Various amphiphilic copolymers can be used to enhance the water-resistant nature of paper-like products, often in combination with the treatments just described. The first generation of such additives were styrene-maleic-anhydride copolymers, and their most frequent usage has been in the surface sizing of paper. Amphoteric polymer sizing agents of this type contain both cationic and anionic groups, where the charge characteristics are pH dependent. This behavior shows high stability over wide pH range (Bung 2004). The sizing agents can be expected to migrate and orient and confer hydrophobic character to the surfaces during the evaporative drying process.

Addition of a water-repellent polymeric sizing agent was successfully used to provide water resistance without affecting other variables while making molded tray for food packaging from lignocellulosic material waste cereal straw fiber (Curling *et al.* 2017). The data showed that the pulp molded material containing up to 80% straw performed significantly better compared to expanded polystyrene in tensile properties (modulus of 0.47 MPa for an 80% straw mix compared to 0.16 MPa for EPS).

Barrier Performance

Grease resistance by chemical treatment

Since the present focus is on food-based packaging, molded products for food packaging also can benefit from barrier properties against water vapor, grease, or oxygen/ air, along with cushioning and mechanical strength. For food trays, the synthetic polymer polystyrene has been used for a long time. However, considering the environmental and health aspects, biobased or natural fibers are taking the place of polystyrene. In addition to fiber, which provides around 88% of the raw material for the product, generally there are a number of additives.

Because of the relatively porous nature of many molded paper products, efforts to increase the barrier properties will often involve some kind of coating application or a top ply. Ideally, the surface layer needs to fully cover a presumed porous structure of the main molded pulp material. The idea is to achieve major reduction in the characteristic pore size. The Lucas-Washburn equation (Lucas 1918; Washburn 1921), which is widely employed to understand hydrophobic sizing systems, predicts that resistance to liquid penetration can be increasingly effective as the characteristic size of pores becomes smaller. In principle, this can be achieved either by increased refining of cellulosic fibers, by use of fine and possibly platy mineral particles in a coating, or in the application of various copolymers in a size-press solution. The copolymers can include styrene maleic anhydride (SMA) and related additives. Such copolymers are intended to contribute to hydrophobicity after the surface layer, along with the whole structure, has been dried again (Bildik Dal *et al.* 2020; Bildik Dal and Hubbe 2021). Another option includes the use of AKD size, as discussed earlier, as a treatment of a surface layer.

When there is a need to resist the penetration by oils, as in the case of paper plates and some other food service items, the challenge becomes greater. Sizing agents such as AKD develop hydrophobicity, but not oleophobicity. It is well known that resistance to oils can be achieved by treatment with certain fluorocarbon compounds (Hubbe and Pruszynski 2020). Such compounds, when suitably anchored to the cellulosic surface, present such a low surface free energy that not even oils will be able to achieve a low enough contact angle to permeate into the pores. Jiang *et al.* (2017) fabricated a superamphiphobic paper by combining the control of fiber size and structure using plasma etching and fluoropolymer deposition. Although coating with a lower surface energy substance is a rapid, simple way of engineering oleophobic surface, long-chain fluorocarbon materials decompose into perfluorooctanoic acid (PFOA) that is bioaccumulative and toxic to humans (Wang *et al.* 2016). Due to health concerns and environmental concerns, the industry is very actively looking for other means of achieving oil resistance.

Grease resistance by refining and hydrophilic polymers

Before the emergence of fluorochemical treatments, the industry relied upon high levels of refining of the fibers, in addition to coatings with hydrophilic polymers, to achieve the high levels of density within paper needed to completely block oils from penetrating (Wright 1922; Kjellgren and Engström 2008; Hubbe and Pruszynski 2020). For example, Sheng *et al.* (2019) showed that effective resistance to grease could be achieved with a coating that included sodium alginate, carboxymethyl cellulose, and propylene glycol alginate, all of which are good film-formers and capable of forming strong hydrogen bonding associations when dried.

Goswami *et al.* (2008) showed good oil resistance of the handsheets, made of banana pulp and bamboo pulp fiber, where the air resistance was enhanced by increasing the beating degree to 80. Lu *et al.* (2016) enhanced oil resistance over a 24 h test period by combining cellulase pretreatment and refining, which can be explained by the film-forming behavior of microfibrillated cellulose. Kjellgren *et al.* (2006) prepared greaseproof paper having excellent air and grease resistance by coating with chitosan. It was shown that the improved grease resistance corresponded to the decreased air permeability. Sheng *et al.* (2019) prepared non-toxic fluorochemical-free greaseproof papers coated with sodium alginate (SA)/sodium carboxymethyl cellulose and SA/propylene glycol alginate. The resulting structure showed lower air permeability, which had a positive effect on the oil resistance of biopolymer coatings on cellulosic fibers substrate (Sheng *et al.* 2019). The larger polar component of surface energy gave a greater contribution to oil repellency for coatings with high surface energy. On the other hand, reduced surface energy of the final product was beneficial to improve oil and water anti-wettability. The coatings completely covered the base paper surface and penetrated into the inter-fiber networks to some extent, which altered the tensile strength and elongation at break of the coated paper (Sheng *et al.* 2019). Kisonen *et al.* (2015) prepared composite coatings with nanofibrillated cellulose (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 performance. 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. Hassan *et al.* (2016) developed films from NFC and chitosan nanoparticles (CHNP), and these displayed excellent grease resistance (Su *et al.* 2018).

Since industry is trying to promote sustainable packaging products, a number of bio-based polymers have been developed to improve barrier properties. Methylcellulose (MC) is a non-thermoplastic, water-soluble cellulose ether (Khan *et al.* 2010) with high oxygen barrier ability. As its hydrophilic nature would suggest, the water vapor barrier performance of MC is modest (Paunonen 2013). Liu *et al.* (2018) improved the water vapor barrier by grafting a coating of polyethylene-reinforced graphene oxide on a MC substrate (Helanto *et al.* 2019).

Surface plies or coatings

In principle, various different strategies might be selected as a means to apply a surface ply or coating layer during a molded pulp operation. For example, the material might be applied as a spray after the initial molded pulp layer has been formed and at least partly dewatered. Figure 27 depicts a process in which a molded pulp structure is formed by vacuum application in a first step, transferred to another section of the device (where it likely would undergo a pressing step), and then transferred to another vacuum step, which would likely involve use of a more highly refined cellulosic material. Although highly refined fibers would be expected to greatly slow down the dewatering process, the main structure will already have been formed. A mechanism called "healing" (Hubbe *et al.* 2020) will tend to draw cellulosic material toward any remaining flow channels, thus effectively plugging up the structure, even if the latest ply layer remains thin. Figure 28 depicts another strategy by which a spray process is used to deposit a layer onto an existing molded pulp item.

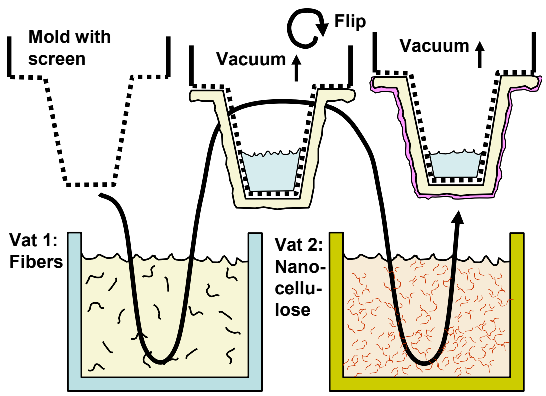

Fig. 27. Concept of a two-step vacuum suction system for the formation of a two-ply molded pulp structure, whereby the top ply might be relatively thin and dense

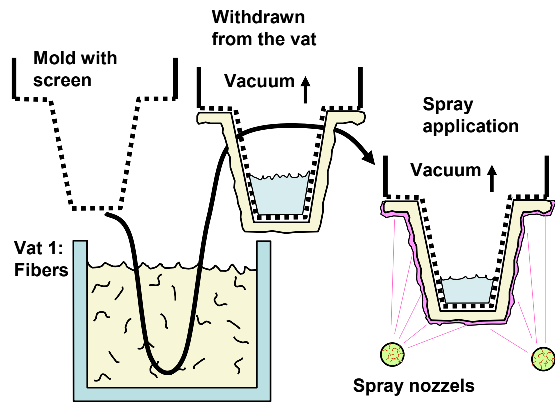

Fig. 28. Concept in which a spray system is used to apply a second layer onto a molded pulp structure

Nanocellulose application

The use of nanocellulose in the formulation of a coating or top ply for molded-pulp items can be viewed as a logical extension of the concepts just mentioned. In fact, some of the examples already discussed involved such materials. Nanofibrillated cellulose can be roughly defined as the product when mechanical shearing is applied at hugely higher levels than conventional refining. Tests have shown that films comprised of nanocellulose, when they are dry and undamaged, can achieve very high reductions in the transmission of oxygen gas, as well as oils (Aulin *et al.* 2010, 2012). An effective overall performance, including water resistance, can be achieved if another layer is applied that is formulated to protect the initial layer of nanofibrillated cellulose from exposure to water (Hubbe *et al.* 2017a; Tyagi *et al.* 2019; 2021).

For the improvement of barrier properties, nanocellulose particles can rely on their various properties. These including a high degree of crystallinity, a favorable length-to-width ratio of fibrils, the polar nature of the surface –OH groups on cellulose, and the ability of the fibrillar material to form a dense structure with high cohesive energy density upon drying (Lagarón *et al.* 2004; Dufresne 2012; Hubbe *et al.* 2017a). However, the uptake of moisture from surroundings is a significant drawback of nanocellulose, while facilitating other superior barrier properties (Lindström and Aulin 2014). As is typical with hydroxyl group-abundant biopolymers, dried films of untreated nanocellulose exhibit low water-resistance and high water vapor permeability when the relative humidity is high (Hubbe *et al.* 2017a). Crystallinity can contribute to barrier properties because it is impossible for molecules to penetrate the crystalline areas. Tyagi *et al.* (2019, 2021) took advantage of the exceptionally high crystallinity of cellulose nanocrystals in the formulation of a hydrophobic nanocellulose barrier layers for packaging applications.

Heat treatment also can improve wet strength, causing the film be become denser. This is possibly due to the aggregation or coalescence of adjacent cellulose chains (Pönni *et al.* 2012). Such coalescence makes the film less porous, which is beneficial in preventing leakage (Österberg *et al.* 2013; Hubbe *et al.* 2017a). Sharma *et al.* (2014) showed that heating films of nanofibrillated cellulose (NFC) for 3 h at 175 °C reduced the water vapor permeability by 50%. Xia *et al.* (2018) reported a ten-fold decrease in water vapor transmission rate (WVTR) when comparing 3 h post-treated TEMPO-oxidized nanofibrillated cellulose (TONFC) films to untreated TONFC films.

Nano-lignocellulose (NLC) is a form of nanocellulose produced from mechanical pulp. According to Spence *et al.* (2010), the presence of lignin increased water vapor permeability, due to increased porosity that compensates its hydrophobic nature. However, hot-pressing increases material density and thereafter imparts an improved oxygen barrier and surface hydrophobicity. Rojo *et al.* (2015) showed a reduction in oxygen permeability and surface wettability due to increase in material density as a consequence of hot-pressing of NLC films. Likewise, while adding lignin, the reduced wettability was found to correlate with a decrease in the London dispersion component of NFC surface energy (Helanto *et al*. 2019). To make it a low-cost sustainable option, lignin-containing cellulose nanofibrils obtained from hemp hurd waste were considered (Tyagi *et al.* 2021). The cited authors showed the effects of residual hemp (*Cannabis sativa*) hurd fibers based lignocellulosic nanofibers (LCNF) on barrier properties, obtained from hydrothermal, carbonate, and kraft treated pulp. The LCNF films and coatings showed much higher water contact angle (WCA) (80° to 102°) compared to the films produced from the bleached CNF. The water vapor transmission (WVP) rate also was lower. It was proposed that the presence of lignin led to better fiber defibrillation into nanofibers during the grinding process due to the radical scavenging ability of lignin.

Surface application of a nanocellulose-based formulation to molded pulp structures offers a promising way to develop barrier properties. Coating is the most common method used in the packaging industry for treatment of paper surfaces. The coating materials on the solid surface form a film with good adhesion to the surface to improve moisture and grease resistance (Kjellgren and Engström 2008; Hubbe *et al.* 2017a). Various coating techniques can be applied on to the surface, such as extrusion coating, solvent-based coating, and aqueous dispersion. In the case of molded pulp structures, spray and vacuum suction coatings are the two most user-friendly methods where nanocellulose or its composites can be applied either by a spray coater or by a two-step vacuum suction methods (Figs. 27 and 28). A spray coater in molded packaging can be applied to coat the outer surface to form a thin layer ranging from tenths of nanometers to a few micrometers. A study conducted by Hult *et al.* (2010) showed that a microfibrillated cellulose (MFC) coating on paper and paperboard significantly decreased air, oxygen, and water vapor transmission rates. Addition of a small portion of CNC in the composite helps to form a rigid network because of the crystalline nature of CNC. Such structures provide physical barriers against transport of water within the matrix. However, the key challenge of the spray coater for molded packaging would be the formation of a uniform layer due to the three-dimensional shape of the package. On the other hand, vacuum suction coatings can be applied by a two-step process whereby the molded package can be dipped in to the nanocellulose solutions to form a thin layer inside the package. The thickness of the nanocellulose layer will determine the overall barrier performance of the packaged materials. The key challenge here will be the dewatering and drying of the packaging materials.

Application of a nanocellulose suspension to a surface faces several challenges. The viscosity of a nanocellulose suspension can become unmanageably high with increasing solids content. The situation can be addressed to some degree by taking advantage of the shear-thinning character of such suspensions (Hubbe *et al.* 2017b; Li *et al.* 2020). Immediately after sufficient agitation has been applied to the suspension, it can be much more easily distributed by such means as spraying, extrusion, or passage through a coating system. Once spread, the material can be expected to undergo a process of immobilization, which likely involves the inter-diffusion among the nanocellulose particles.

Bioplastic composites and blends

Blending or forming a composite of biopolymers can help to overcome their individual limitations. For example, nanocellulose with other biopolymer blends that may include poly(lactic acid) (PLA) and composites are a promising approach to improve barrier properties. Such components can help to improve water resistance of nanocellulose. Blends of PLA and starch have been a topic of study (Yu *et al.* 2006; Johansson *et al.* 2012; Tang *et al.* 2012). Motivations for using starch in PLA formulations include a lower overall price and enhanced biodegradability (Johansson *et al.* 2012; Tang *et al.* 2012). In addition, blending poly(L-lactide acid) (PLLA) together with poly(D-lactide acid) (PDLA) improved thermal stability compared to PLLA or PDLA alone. The cited authors achieved a 50 °C higher melting temperature by making the blend of PLLA/PDLA (Yu *et al.* 2006). The L and D isomers also have an effect on the crystallinity and mechanical properties of the polymer. High crystallinity can be achieved with the L-form, whereas an amorphous character can be achieved with copolymers of D and L isomers (Andersson 2008; Helanto *et al.* 2019).

Another class of potential bioplastic is the polyhydroxyalkanoates (PHAs), which are a family of linear polymers that are biosynthesized by micro-organisms from various substrates. They are biodegradable and bio-compatible, resistant to water or moisture and to ultraviolet radiation, and they are good barrier to some gases (Panaitescu *et al.* 2018; Albuquerque and Malafaia 2018). However, there are major limitations on the functionalities such as low barrier properties towards many gases and water compare to plastics, thermal instability, brittleness (due to high glass transition and melting temperatures), stiffness, and poor impact resistance as food packaging. Various PHA blends have been developed to improve the performance and to offset the high price and offer more scope to expand their range of applications (Albuquerque and Malafaia 2018).

The compatibility between nanocellulose and other components of films can be enhanced through surface modifications. It has been shown that cellulose nanocrystals (CNC) dispersed in PLA and PHB blends significantly improved mechanical properties, crystallinity, and thermal stability of composites (Arrieta *et al.* 2014, 2015). The performance was found to depend on the efficient dispersion of the CNC within the matrix. Besides the increasing of mechanical strength, nanocellulose composites also lowered the water vapor permeability. This favorable performance was observed when the nanocellulose in the composites was less than 5.0 %. This low amount of nanocellulose can be dispersed in the relatively hydrophobic matrix material, thus hindering the passage of water vapor through composites. However, the water vapor permeability may increase with the increasing amount of loaded nanocellulose due to increased agglomeration of the nanocellulose (Abdulkhani *et al.* 2014).

Issues Requiring Attention

Though the preceding discussions make it clear that a great deal of progress has been made in understanding pulp molding systems, as well as the structures that can be prepared in such systems, there is a lot more work that needs to be done. This section will deal with some particular aspects of pulp molding where practical problems are deserving of attention.

Product quality is a key concern. Due to the viscoelastic plastic nature of paper and improper drying, the molded product may suffer from post-forming deformations. This problem can be solved to some degree by adjusting premolding dryness and post molding conditions; in addition, one can select fibers with a more elastic nature (Hauptmann and Majschak 2011). However, there is a need for further research, given the need to compete against plastic structures, which generally do not suffer from post-forming changes.

Wrinkling and cracks are visual defects that are caused by improper stress states and compressive forces acting transversely on the product. Sometimes, due to the effect of density of certain fiber types, high pressure, and high temperature, some sort of pockets may appear in the sample when water and vapor get trapped, which will result in a distinctly different surface finish (Jacobsen 2017).

Delamination has been observed in the pressing step, more specifically, impulse pressing step during manufacturing (Didone *et al.* 2020). A study proposed that such delamination involved damage to the cell walls of kraft fibers (Lucisano and Martinez 2001). Delamination was most prevalent at a distance of about one-third of the mat thickness away from the hot surface. Non-uniformity of the fiber mat, *i.e.* flocculation, appeared to contribute to the delamination. However, given the relatively high solids of fiber suspensions often used in pulp molding operations, fiber flocculation cannot be completely avoided.

As was noted in the Introduction of this article, the number of scientific publications related to molded pulp production is small in comparison to some other fields of manufacturing, such as papermaking. The present article has attempted to start filling the gap, with a focus on production rates and the achievement of a variety of product-related goals. The authors hope that other researchers will be likewise motivated to more fully fill in the picture in describing and improving this important field of technology.

CLOSING COMMMENTS

Molded pulp production is a relatively old technology, going back to the invention disclosure by Keyes (1890). However, it is a new technology as well, using computerized design to fashion efficient and eco-friendly packaging systems for electronic components (Marcondes 1997; Ma *et al.* 2004; Geng and Xu 2011). In addition to the technical issues that were the main focus on this article, the future of the molded pulp industry, as well as its expansion into new product areas are likely to be strongly affected by the availability and quality of cellulosic fibers. With the increasing dominance of electronic media, the molded pulp industry can be expected to have less and less access to the recovered newspaper furnish that has been a major raw material source in the past. Old corrugated container (OCC) pulp is widely available now, but there are other potential uses for OCC, such as the further processing of it to make absorbent paper grades (Zambrano *et al.* 2021). Thus, the price and availability of OCC is likely to change. Another challenge faced by the molded pulp industry is its relatively high usage of resources, especially water and energy. Because molded pulp products are prepared essentially one at a time, there is a large capital expenditure for equipment, relative to the tonnage of product that can be made per unit time with such equipment. The rate of production may not be enough to be able to afford the latest advances with respect to energy-saving strategies. By contrast, modern pulp and paper mills have been able to implement many strategies, such as heat exchangers networks, to reduce the total amounts of energy expended for a unit of production (Hubbe 2021).

Fortunately, the field of molded pulp seems to have an ace up its sleeve. The process, when it is used to make intermediate grades of packaging and cushioning materials, seems to be highly tolerant of using different kinds of fibrous material. This can conceivably include crop residues, many of which would otherwise be burnt. The feasibility of using varying types or qualities of fiber, and not needing to use extensive mechanical refining on those fibers, is a nice situation to be in when faced with shifting qualities and prices of fibers that can be recovered from consumers, businesses, or county recycling centers. Given the importance of molded pulp products with respect to packaging and other uses, as well as its environmental advantages over many competing technologies, one can expect the field to become increasingly important in future years.

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