Converting and its Effects on Barrier Properties of Coated Packaging Materials: A Review

Roman Lev,* Panu Tanninen, Johanna Lyytikäinen, and Ville Leminen

Considerable research is ongoing, examining opportunities for substituting plastic packaging with more sustainable alternatives, and some encouraging results have been achieved. Coated paper and paperboard demonstrate promising performance; however, several serious drawbacks still need to be overcome. Recent research in this area is reviewed in the current work, including mechanical and machinery aspects of paperboard converting, as well as barrier properties of coated materials before and after processing. The main objective of the study was to establish how coated paperboard behaves during converting operations and investigate what changes in its properties occur, considering not only the convertibility of the material as a whole but also effects on substrates and coatings. The results of the literature review show that creasing, folding, and the presence of forming stresses severely damage barrier and pigment coatings even if the paperboard-based product is reported as having good oxygen and water vapour barrier or oil resistance after production. Thus far, most materials cannot fully match the performance of plastic packaging materials due to a noticeable reduction in barrier properties after converting. The work presents factors linking the convertibility of coated materials and their subsequent barrier properties as valuable knowledge to support future development of sustainable materials.

DOI: 10.15376/biores.18.4.Lev

Keywords: Coated paperboard; Barrier coatings; Pigment coatings; Convertibility; Creasing; Folding; Formability

*Contact information: Packaging Technology, Lappeenranta-Lahti University of Technology LUT, 53850 Lappeenranta, Finland; *Corresponding author: Roman.Lev@lut.fi*

INTRODUCTION

Driven by sustainability trends, the packaging industry has devoted considerable effort towards the creation of more sustainable materials for a wide variety of different applications, and the innovation process is still ongoing. Among the materials tested, paper- and paperboard-based materials are considered to be the most promising due to their advantageous mechanical properties, light weight, recyclability, and renewability (Lavoine *et al.* 2014; Desmaisons *et al.* 2018; Poulose *et al.* 2022). The widespread utilization of paper and paperboard as packaging material can be explained by the production of boxes and containers for product transportation, but these fibre-based materials are less commonly used for packaging with direct food contact (Marsh and Bugusu 2007). Food packaging is a demanding area, since food products may contain oil and water, which highlights the main disadvantage of fibre-based materials – poor barrier properties. More commonly used plastic-based packaging material offers significant variability of shape, together with excellent resistance to penetration of chemicals through its surface, whereas the formability of paperboard is limited to simple-shaped products (Vishtal *et al.* 2014).

Still, an intermediate solution – plastic-coated paperboards – has proved commercially successful, especially in production of liquid packaging cartons and barrier-coated trays. However, their utilization is being increasingly questioned on grounds of the poor degradability and the eco-unfriendliness of the plastic coatings (Rastogi and Samyn 2015). This is apparently the main reason for a strong industrial interest in bio-coated paperboard: high volume of packages with plastic coatings is already being produced with utilisation of established tools and processes. It would appear that all that remains is to substitute plastics by bio-based solutions. For this reason, the study of fibre substrates and bio-based or bio-degradable coatings is an active research area with investigations focusing on advantages and drawbacks relative to conventional fossil-based forerunners.

The vast number of studies found in the literature testifies to the great interest in the field. Many different coated fibre-based substitutes for plastic-containing packaging materials have already been developed, some of which in their initial flat form have been shown to possess barrier properties comparable with those of plastics. However, the properties of packaging materials can differ drastically after converting – a process of transformation of raw materials into final packaging products such as trays, cups, boxes, *etc*. For example, coating failures after creasing operations, such as cracks, can reduce or nullify barrier properties; non-optimized forming parameters can cause ruptures that affect not only the coating but the substrate as well; and coating sticking caused by high temperatures can compromise product integrity. These examples of convertibility impairments can render novel materials unsuitable for packaging applications, and enhancement of convertibility has become a matter of concern. Development requires not only material property testing in the initial state, but also examination after various converting operations to estimate the integrity of the coated substrates and their resistance to different kinds of stresses.

Several reviews have been presented with partial coverage of the above-mentioned issues. The authors have examined topics including enhancement of the functional properties of packaging materials using barrier additives for coatings (Andersson 2008; Shen *et al.* 2014; Rastogi and Samyn 2015) and the use of essential oils in active food packaging (Sharma *et al.* 2021). Studies have discussed processes related to the material formed, such as damage mechanisms (Östlund 2018), forming factors (Vishtal and Retulainen 2017), and coating cracking (Panek and Hart 2022). The current work is an attempt to provide an overview of factors having the greatest effects on coated fibre-based materials at key stages of their lifecycle with consideration of material components separately and in interaction with each other. The work discusses three different topics: 1) the factors underpinning substrate integrity during converting and how they affect the coatings applied; 2) the performance of novel coatings before and after converting and how performance can be improved; and 3) the mechanical properties of substrates and coatings, and the result of their interaction with tooling during converting operations.

The terms "converting" and "convertibility" are not widely discussed in most studies, despite their marked effect on coated material performance and their potential to compromise the intended function of the final product. In many cases, studies examine coated materials as flat blanks and focus only on the barrier properties of the virgin coated material and the potential of the material for future use in more sustainable packaging. Additionally, if forming processes are discussed, it is usually done with reference to uncoated paperboard; yet, coatings can drastically change substrate performance in response to converting operations. The main unknown issues are: *How does material behaviour change during and after converting compared to the initial state?* and *What differences exist between forming of coated and uncoated substrates?*

For data collection and further discussion of the state-of-the-art of coated packaging material, the current study examined the following research questions:

– What shortcomings of sustainable coated packaging materials have been identified in comparison with conventional plastic-based materials? What characteristics of sustainable coated packaging material hinder their performance and their wider utilization?

– What properties are needed in various packaging applications and what have reported studies aimed to achieve?

– How do material properties affect converting and vice-versa, in particular, what impact does converting have on the coated surface of packaging material?

Several estimations have been made for better perception of the materials of interest. Coated paperboard is the most often characterised by its grammage, expressed in grams per square meter (gsm). The value is calculated for both substrates and coatings, allowing comparison of different materials. Thus, grammage of typically studied paperboard substrates is most often within 190 to 350 gsm, whereas grammage of bio-based coatings have varied even more – from 1 to 21 gsm (*e.g.*, Lavoine *et al.* 2014; Javed *et al.* 2018; Tanninen *et al.* 2021b). Plastic extrusion coatings, commonly used as references, were always heavier and started at least from 17 gsm (Lavoine *et al.* 2014; Leminen *et al.* 2018). The stretch at break of typical materials was measured in standard conditions and in average varied between 2.0 and 2.5% in the machine direction (MD) and 5.0 to 7.0% in the cross-machine direction (CD) (*e.g.*, Tanninen *et al.* 2015c). As regards the nature of the coatings, typical bio-based coatings are brittle, hydrophilic, sometimes possess poor film formability, low melting point, and may have insufficient barrier properties far before converting. After contact with converting tooling, the properties reduce even more. In contrast, plastic coatings are more flexible for converting in different environments, while still often being neither bio-degradable, nor recyclable.

The main aim of the research was to synthesize and summarize knowledge on the barrier, mechanical, and convertibility properties of coated substrates, identify their interactions, and review the current state of development in the area of sustainable coated packaging material. Such knowledge can provide valuable insights into the barrier properties of coated materials and offer a foundation for future work on their performance and convertibility development.

CONVERTIBILITY OF COATED FIBRE-BASED MATERIALS

The packaging material production stage is only the first step of paperboard material conversion to a final product. Subsequent processes include, among other operations, conditioning for optimal moisture content, blanks preparation by cutting and creasing, and material forming using a wide range of three-dimensional (3D) shaping techniques. These stages are not the final ones prior to a container serving as a package with a product inside it, but during the preparation and forming of the final package, coated materials experience the highest mechanical stresses, which can compromise the integrity of their components. As cutting, creasing, folding, and forming are converting methods that impose the greatest demands on coated fibre-based material, this work focuses on these processes. However, the results and conclusions of the current review are also applicable to packaging production stages applying lower stresses on the packaging material.

Tooling-material interaction is a specific area of concern in packaging industry operations. Converting coated materials to ready-made products is realized using a wide variety of machines and devices. An unsuitable process, machinery or tooling can compromise end-product quality even if a material has good barrier or convertibility properties. The mechanical properties of the material have the greatest effect on the initial formability and subsequent performance of a final package. Developments in tooling and greater knowledge of material properties are prompting packaging science towards the goal of improved sustainable substrates and coatings, but many trade-offs still need to be overcome, *e.g.*, when improvement of one property impairs another.

Converting by Cutting and Creasing

During converting processes, it is important to prevent the coated surface from being damaged, since the material will otherwise not be able to perform its intended function (Panek and Hart 2022). Evenly coated materials with a barrier or pigment layer quite often have good barrier properties or a smooth surface for printing. However, subsequent cutting and creasing operations apply high compression stresses to the surface and potentially impair the properties acquired during material production. Bio-based coatings, in particular, show poor resistance to creasing operations and often cannot withstand creasing-generated stresses (Tanninen *et al.* 2015c).

During creasing operations, lines are produced on the material surface (Fig. 1) by pushing the material into grooves using creasing rules. The blanks produced are then folded to a final package shape, which are expected to have undamaged edges (Domaneschi *et al.* 2017). Production of folding lines is also possible using other processes, for example, paperboard scoring (Leminen *et al.* 2021), and several works mentioned below have examined different paperboard treatment methods for package forming.

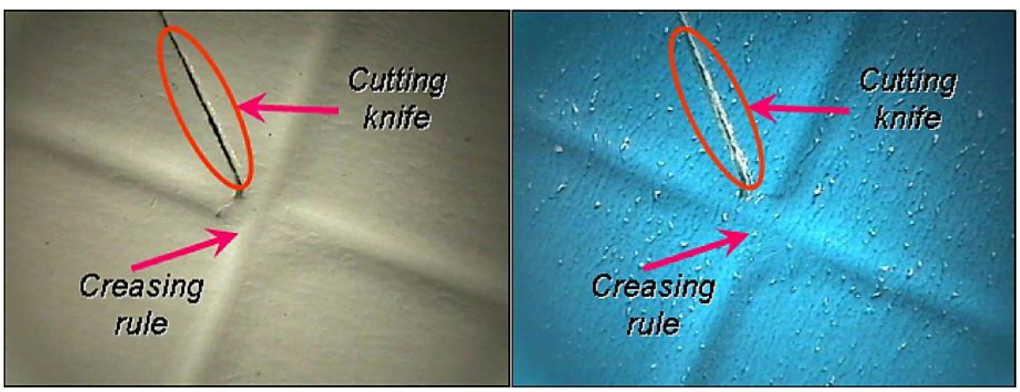

Fig. 1. Creasing and cutting lines on the printed and unprinted surface of coated paperboard. Modified from Kim *et al.* (2010); Reprinted from *Journal of Industrial and Engineering Chemistry*, 16(5), Copyright 2010, with permission from Elsevier.

Of the creasing processes available, two are the most frequently mentioned in the studies reviewed. One is flatbed die-cutting, which is the most suitable method for industrial production thanks to its speed; the other is wheel grooving on a digital table, which is useful for smaller production batches (Bota *et al.* 2022). Flatbed die-cutting and wheel-grooving methods aim not to damage the surface of materials. Instead, the processes act to generate inner substrate delamination that causes a reduction in fibre bond strength along creasing lines and reduces paperboard stiffness (Marin *et al.* 2022). In contrast, another creasing method – scoring – makes a partial cut through the material top layer, resulting in a folding line (Geis *et al.* 2014). Scoring can be done mechanically (*e.g.*, by knife) or by laser, resulting in continuous or dashed cuts, which respond to folding differently (Mentrasti *et al.* 2013). All four methods were studied by Leminen *et al.* (2021), and the results were reported to differ as regards the dimensions of the folding lines made: creased grooves were much wider than scored ones, but their depths were still comparable. According to the study, the flatbed die-cutting process performs best with wider grooves in the matrix (Leminen *et al.* 2019) and results in a lower force being required to fold the sample (see further sections).

Some literature provides information about the usage of creasing equipment and, particularly, the effects of equipment parameters on packaging performance. The main actions of creasing tools are executed by two parts: the male creasing tool, which can be a metal rule or a wheel of different widths, and the plastic matrix with a groove into which the material is pushed (Bota *et al.* 2022). Male tool width is measured in points (pt.), and the industrial recommendation for paperboard is most often 2 pt., which has been shown to produce the lowest percentage of unacceptable trays (only 3%) (Tanninen *et al.* 2015a). Narrower rules demonstrated similar results but required operation within a very precise force range to prevent cutting through the coatings (Tanninen *et al.* 2015a). A clear correlation between paperboard thickness and creasing tools is generally observed: a 2 pt. wheel showed the best forming performance for thinner material, whereas a 6 pt. wheel resulted in better trays for thicker paperboard grades (Leminen *et al.* 2018). In the study by Tanninen *et al.* (2015a), however, narrower creases were seen to have better performance for thicker material. This means that not only creasing tool dimensions should be taken into account in production planning, as other factors such as the creasing process also affect final performance (Leminen *et al.* 2018). The dependence of the coated surface integrity on different creasing toolsets used was studied by Tanninen *et al.* (2015c). The authors tested six sets of creasing rules and grooves and found that pinholes formed on the coated surface regardless of the toolset. It would appear that surface defects formation is prevalent as a consequence of converting rather than because of the specific toolset used, and coatings contents has no notable effect on convertibility (Tanninen *et al.* 2014b, 2015c).

Foldability After Creasing

Creasing is an operation during which the paperboard experiences intentional local damage to reduce its stiffness along the creasing line, and such weakening consequently leads to easier blank folding. The "foldability" of creased blanks is determined by the force needed to fold them, which is dependent on factors including the method used for creasing, the depth of the creases, paperboard-based material content, and thickness. In general, wider and deeper creases result in better foldability due to a higher degree of delamination in the material. Flatbed die-cut materials therefore require less force for folding than blanks produced with a creasing wheel or scored, although it should be noted that the difference may be negligible (Leminen *et al.* 2021). Interestingly, a detailed study of folding response in relation to bending angle has revealed somewhat conflicting results. Mentrasti *et al.* (2013) compiled folding moment-angle diagrams, which peaked during highest resistance of the paperboard to bending. Well-defined line dimensions resulted in better folding performance and higher quality packages (Nygårds *et al.* 2014), but it seems that the severity of damage induced by creasing or scoring correlated with foldability as well. Thus, a continuous cut line significantly damaged the surface and resulted in the lowest folding moment and stiffness (in contrast to Leminen *et al.* (2021). The force needed to fold the material grew linearly with bending angle and peaked at 180°. While ensuring undamaged paperboard along the folding line, a dashed cut line resulted in the highest folding force being required, with a peak at 40 to 50°, and the force needed remained the same until

180°. The least damaging method – normal creasing – had the most complex response and peaked twice at 40 to 50° and 90°. A noteworthy finding was that creasing depth differed even when measurements were taken from different parts of the same sheet, which demonstrates paperboard anisotropy. For all cases, samples unloading showed a similar monotonic trend, as it was independent of the folding line type and depth. Studying the dependence of foldability on creasing method, Mentrasti *et al.* (2013) and Leminen *et al.* (2021) have reported inconsistent results in regard to which creasing methods resulted in the lowest and highest force being required to fold the samples. Still, the differences appear to be negligible, meaning that the creasing method does not seem to be a major factor affecting foldability. During production planning it is more important to consider the end use of the package as, for example, scoring-based folding approaches damage the material surface and may be unsuitable for two-sided coated substrates. Additionally, Leminen *et al.* (2021) discovered that boxes made with laser scoring withstood compression tests better, as higher delamination results in lower compression force. A smaller box height also contributes to higher compression strength (Csavajda *et al.* 2017). There thus arises a trade-off between material foldability and strength, as easier packaging production and better stiffness of packages have to be balanced for optimal performance.

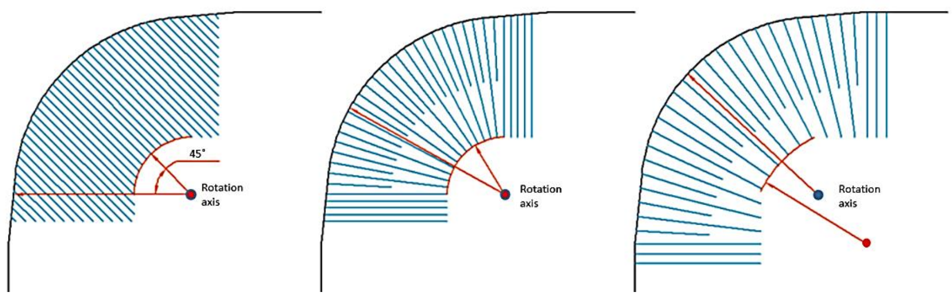

Fig. 2. Examples of creasing patterns available for production of paperboard tray blanks (Tanninen *et al.* 2015a).

In contrast to the creasing method, the creasing depth seems to have a much greater effect on foldability, as deeper creases reduce the bending stiffness of paperboard more effectively. If there is a rapid decrease in bending moment as a result of bending stiffness reduction, the creasing operation has performed its intended purpose (Nygårds *et al.* 2014). In comparison to uncreased materials, even creases with the lowest depth require less force to fold (Tanninen *et al.* 2021b), and peak of bending moment may occur before 20° (Huang *et al.* 2014). The presence of a matrix while creasing and the width of its creasing groove have a direct effect on foldability: without a matrix, the depth of creases is reported to be three times lower (Leminen *et al.* 2019). Adjustment of creasing depth based on the mechanical properties of the material helps to maintain surface integrity and prevent fold cracking (Bota *et al.* 2022). The creasing window is usually formed between two lines: the first one indicates the occurrence of effective delamination in the paperboard, and the second line defines the start of undesired in-plane crack formation. Such cracks are to be avoided, as they are formed by fibres torn out of the network during stress application, and the damaged fibres contribute to cracking of outer plies and compromise package integrity (Marin *et al.* 2022). Force values used to produce creases correlate with crease depths, and despite common assumptions, material density has greater effect on resulting creasing force than material thickness. Indeed, a stronger interface delaminates less and therefore requires higher forces to produce deeper creases (Marin *et al.* 2022; Huang *et al.* 2014). In this regard, it has been reported that in multi-layered materials weaker top and bottom layers help to form a good crease and prevent in-plane cracks (Nygårds *et al.* 2014), although the simplest layer structure still results in better formability (Ovaska and Geydt 2018). Coatings sometimes have no significant effect on creasing depth (Leminen *et al.* 2019) and creasing behaviour is mainly determined by substrate properties. It is notable that after a certain crease depth, deeper creasing does not provide any improvement in foldability and the application of higher creasing force is not necessary (Tanninen *et al.* 2021b). Additionally, the creasing pattern on a tray blank (Fig. 2) influences its foldability: the distance between the creases correlates with tray corner radius, but its lower limit should be set to at least 1 mm, as the coating integrity is otherwise compromised. A larger number of folds is beneficial for a smoother tray wall and radially positioned creases towards the rotation axis of a tray corner (Fig. 2, middle) are the best pattern for tray production (Tanninen *et al.* 2015a).

The ability of paperboard to fold is closely connected to its bending stiffness. When the stiffness lowers after creasing, a paperboard blank becomes easier to fold, but overall package strength is then compromised (Lavoine *et al.* 2014). Bending stiffness and maximum bending moment decrease with creasing depth, since deeper creases cause more delamination in the middle of the substrate (Huang *et al.* 2014; Leminen *et al.* 2019), which explains the better performance of deeper creases. It is often assumed that a package is folded only once around its creased lines during production, but in the case of further utilization, factors such as fatigue accumulation should be considered. Every single fold-unfold operation above the yield point generates structural damage (Na *et al.* 2020), as deformation dissipates energy while unloading, and part of the energy goes to irreversible degradation. Paperboard may have no visible cracks but it deforms constantly with increasing deflection (Wang and Sun 2018). Accumulated damage to the material can result in stiffness degradation (Yamada *et al.* 2017) and lead to ultimate bending fatigue failure. Stiffness degradation is dependent on loading cycles and obeys the following trend: at first, stiffness degrades gradually, but abrupt decrease in stiffness then occurs when close to the failure cycle (Wang and Sun 2018). Increasing the depth of the creases improves the foldability of the blank while the delamination cracks are well-defined along the folding lines, but at the same time, the failure stress value becomes lower, making the material weaker (Marin *et al.* 2022). While paperboard is known to be anisotropic and fibres are usually preferentially aligned in the machine direction (MD) during production (Nygårds 2005; Rhim 2010; Bota *et al.* 2022), bending stiffness, as most other mechanical properties, differs in the MD and cross-machine (CD) directions. It has been reported that bending stiffness is greater when the material is creased perpendicular to the MD, as more MD-oriented fibres bridge the crease zone and resist folding (Nygårds *et al.* 2014). This means that creases made parallel to the machine direction usually provide better foldability (Bota *et al.* 2022). At the same time, the bending moment in the MD decreases faster with higher creasing depth, because MD fibres help to transfer shear loading and contribute to bond failure (Huang *et al.* 2014). In analysis of folding and creasing, the force needed to fold paperboard material was found to level out in the middle of the depth range tested (Huang *et al.* 2014). This confirms the results of Tanninen *et al.* (2021b), where it was noted that producing deeper creases after a certain depth is not necessary as it does not improve foldability further.

Formability Affecting Factors

Folding of paperboard along a creased line can be considered a relatively simple operation (Panek and Hart 2022), and it is actively used in processes such as the production of folding boxes from coated or non-coated paperboard. In its turn, the production of trays or similar containers utilizes 3D forming machinery, and the manufacturing process seems to be more complex due to the larger number of parameters affecting product performance. While many parameters result in complex trade-offs and constraints, for example, between visible forming quality and durability of a product, or dimensional stability and production speed, some parameters act synergistically to achieve better converting quality. Examples of such synergistic parameters include many that are closely connected with the mechanical properties of the materials used, such as temperature, moisture, and blank holding force (BHF) (Hauptmann *et al.* 2011; Hauptmann *et al.* 2017). Paperboard forming practices came originally from metal forming, with changes to account for the differences between the materials, in particular, the anisotropy of paperboard and its sensitivity to moisture and temperature change (Groche and Huttel 2016). Therefore, the most popular methods for 3D forming of fibre-based materials are press forming, hydro-forming, and deep drawing (Afshariantorghabeh *et al.* 2022). They differ mainly by mode of material stretching for shape formation, but all three utilize heat and moisture to enable bond weakening in the fibre structure and adjustment of BHF to enhance formability.

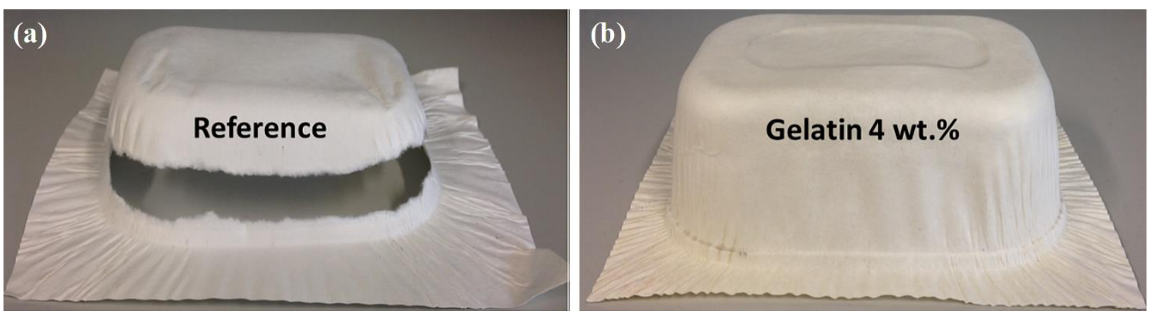

Fig. 3. Example of the additive effect on formability: (a) untreated and (b) treated with gelatine paper. Reprinted from Khakalo *et al., Reactive and Functional Polymers* 85, Copyright 2014, with permission from Elsevier.

Good forming quality, defined by shape stability, surface smoothness, and absence of ruptures and visible cracks, is achieved by bio-coated materials more often than with an intact barrier layer (Leminen and Tanninen 2015). The quality of formed products sometimes exceeds the performance of traditionally used plastic-coated substrates, for example, by providing a smoother tray surface (Leminen and Tanninen 2015). Additionally, boxes made from paperboard with microfibrillated cellulose (MFC) coating demonstrated compression strength comparable to polyethylene (PE) coated paperboard boxes, but with lighter coat weight and biodegradability properties (Lavoine *et al.* 2014). In addition to affecting barrier properties, it has been found that the incorporation of additives to coating films improves convertibility. For example, gelatine increases material flexibility (Fig. 3) (Vishtal 2015; Franke *et al.* 2021), latex improves material extensibility and enables self-healing of coatings under heat (Tanninen *et al.* 2014b; Leminen and Tanninen 2015), and talc reduces coating stickiness (Wang *et al.* 2018b), hydrophilicity, and pinhole formation (Leminen and Tanninen 2015). It is, however, important to optimize additive content in coating recipes as additives, together with excessive coat weight, can sometimes negatively affect barrier properties (Tanninen *et al.* 2014b). It is worth mentioning that if materials are used for cup forming, high oil and grease resistance (OGR) is not needed. This is because cups are usually used for non-greasy products and good sealability is of greater importance. It has been reported that addition of either talc, kaolin, or calcium carbonate to PLA matrix for coatings have no strong effect on hot bar or air sealing, and good results were achieved with all recipes used by Helanto *et al.* (2022). Thus, in some converting processes, materials with bio-based coatings demonstrate good performance, and more attention has to be paid to improvements to their barrier properties, particularly elimination of pinholes formation (Tanninen *et al.* 2015c; Leminen and Tanninen 2015).

Mechanical properties

Interaction between coated materials and the processes needed to convert them is dependent on the mechanical properties of the material, such as tensile strength, elongation, elastic modulus, *etc*. Numerous materials are being actively tested for comprehensive prediction of coated substrate behaviour during converting operations (Girlanda and Fellers 2007). As paperboard possess viscoelastic nature with time-dependent creep (Tanninen *et al.* 2021a) and a tendency to show spring back and walls deflection after forming (Hauptmann *et al.* 2011), materials with higher stiffness and lower recoverable strain component tend to have better formability (Vishtal *et al.* 2014). In this regard, to produce trays with higher stiffness, it is recommended to cut blanks along the MD, meaning that the ready tray's longer side will be parallel to the machine direction. In addition, it has been found that coated trays demonstrate significantly higher stiffness than non-coated ones due to greater thickness (Tanninen *et al.* 2021a).

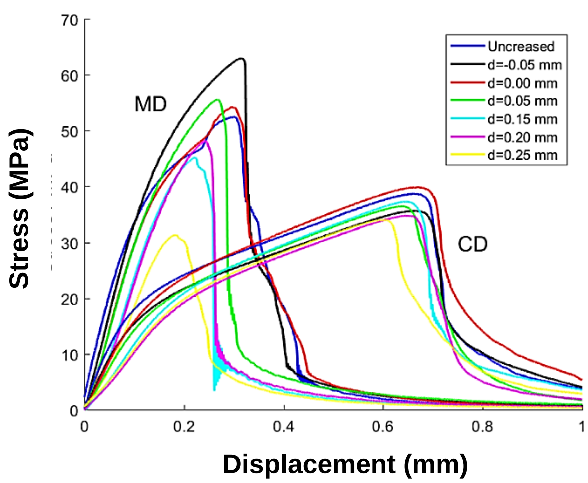

Fig. 4. Typical stress-strain graphs for paperboard-based materials: mechanical properties usually differ in the CD and MD directions. Reprinted from Marin *et al.* (2022), with permission from Wiley.

Initially, it may seem that the convertibility of a material is determined by its substrate, which is much thicker than the coating. However, experimental results show that the coating film applied often affects converting operations, causing changes in the mechanical properties (Javed *et al.* 2018). These properties may have conflicting effects on material performance as a whole. For example, increase in material tensile strength results in higher stiffness of a package, but at the same time, tensile strength affects paperboard extensibility, which is the characteristic responsible for a material's ability to stretch without rupture (Khakalo *et al.* 2014). It can be seen from the stress-strain graphs of several studies (Huang *et al.* 2014; Javed *et al.* 2018; Marin *et al.* 2022) that while one material fails at higher stress during tensile tests, but with lower strain, another material can behave conversely (Fig. 4).

In contrast, lower tensile strength and bending stiffness indicate better formability (Ovaska and Geydt 2018). However, lower coating elasticity does not always mean poorer resistance to converting operations (Javed *et al.* 2018). Although the inverse dependency of tensile strength and strain is a common trend, certain additives can reduce both parameters in some cases. Indeed, the forming result cannot only be described by product strength and bearable strain; other factors such as surface quality and visual appearance usually need to be considered as well (Franke *et al.* 2021). As the anisotropic nature of fibre-based substrates leads to an assumption of different mechanical characteristics in the MD and CD (Rhim 2010) (Fig. 4), these differences may affect coating integrity in response to higher substrate stiffness in one of the directions (Kim *et al.* 2010). In contrast, it has been found that strain or stress values of pure coating film separated from substrate may exhibit no difference between the CD or MD, and the coating itself can be considered as isotropic (Franke *et al.* 2021). At the same time, substrate and coating properties sometimes differ drastically. For example, lower coating elasticity may compromise forming quality, while substrate alone would perform better (Franke *et al.* 2021).

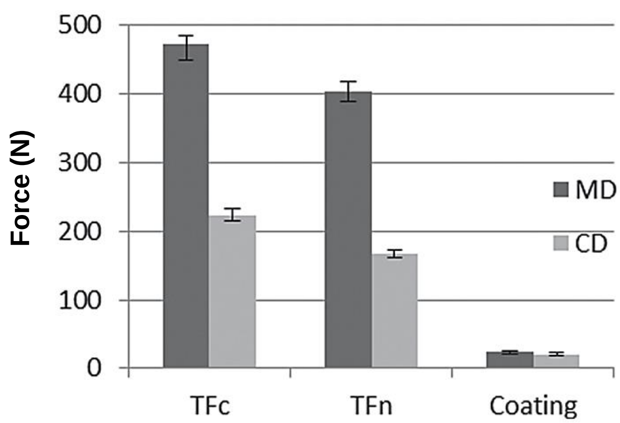

Fig. 5. Coating strengthening effect: the force, needed to break a coated sample (TFc) is higher than the sum of the forces needed to break the substrate (TFn) and coating separately. Reprinted from Franke *et al.* (2021) with permission from Wiley.

Quite interesting behaviour has been detected for a polyethylene terephthalate (PET) extrusion-coated paperboard when testing the substrate and coating separately and then comparing their performance individually and in combination as a coated paperboard material (Franke *et al.* 2021). Tensile tests with stretching in either the MD or CD direction alone showed that the sum of forces needed to break the substrate and coating film separately does not equal the force required to break the coated material (Fig. 5). The latter force was higher by 9.5% in the MD and 15.4% in the CD, indicating that the coating penetrates the substrate fibres and strengthens them. However, during a bulge test where the load was multiaxial, the sum of the forces, needed to break the substrate and the coating individually was almost equal to the force needed to break the coated material. In this case, the coating film thickness decreased as a result of multiaxial loading and the strengthening effect was eliminated (Franke *et al.* 2021). In this regard, surface treatment methods, for example, corona treatment, have been reported to promote adhesion between the substrate and coating and generate higher material stiffness (Morris 2008). It should be noted, however, that paperboard coating may also have an opposite effect: water-based coatings may penetrate the substrate and lead to a decrease in mechanical properties as a result of excessive fibre bond weakening. Due to lower infiltration of barrier to substrate, extrusion-coated materials have shown clearly higher tensile stiffness than dispersion-coated materials (Tanninen *et al.* 2015c). In addition, plasticizing additives, which usually reduce surface cracking, may migrate from the top of the coating to the bottom and closer to the substrate, and in this manner they can impair the performance of the material as a whole (Javed *et al.* 2018).

Blank holding force

Of the mechanical parameters that can be adjusted during 3D forming, the blank holding force (BHF) is one of the most discussed, as BHF seems to have the greatest effect on forming performance. BHF adjustability provides opportunities for achievement of better quality, but parameters can have inconsistent effects in different forming processes, and several authors trying to optimize BHF have found conflicting results despite using similar BHF trajectories.

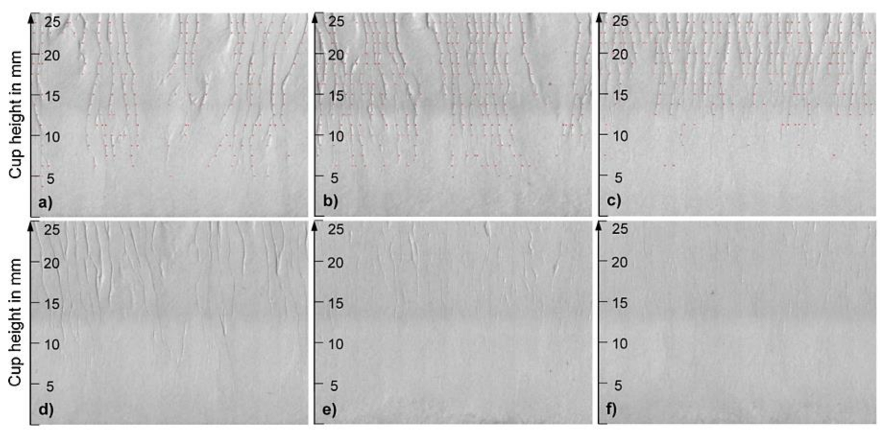

Fig. 6. Example of different wall quality after deep drawing using several BHF trajectories. Reprinted from Hauptmann *et al.* (2016), *Journal of Materials Processing Technology* 232, with permission from Elsevier.

In deep drawing, one forming quality parameter on which BHF has great effect is wrinkle distribution along the product height (Fig. 6). In the studies reviewed, the result of forming for a product like a tray is considered “ideal” if the surface is free of wrinkles and “good” if wrinkles appear closer to the top flange and their number is as high as possible. In press forming with pre-creased blanks, a smooth tray inner surface and flange serves as an indicator of a good result. Using high BHF was reported to be the best way to meet the quality goals as wrinkle distribution becomes more uniform with increased BHF until the tray wall appears free of wrinkles (Hauptmann *et al.* 2017; Leminen *et al.* 2018).

However, higher BHF facilitates forming only while material fibres are able to compensate internal deformation (Hauptmann *et al.* 2015) and forming occurs within an appropriate range of BHF values. When the BHF is too low, the paperboard blank folds insufficiently and the desired quality is not achieved; and too high BHF causes ruptures at the tray edge or corners (Tanninen *et al.* 2014b; Leminen *et al.* 2015). The material formed is not usually damaged by clamping itself, but a forming punch can tear the blank if it is held too tight. The amount of damage in the products formed increases as a function of BHF value if the force is set above a certain limit (Tanninen *et al.* 2020). Moreover, BHF is adjustable within the process duration, and it is possible to use several forces while forming. Variability of factors affecting the optimal BHF range for each process is the reason for the unpredictable behaviour of BHF compared to heating and moisture: while the best forming results are achieved with heated and moisturized blanks, the BHF trajectory affects forming differently in each case and is dependent on the particular process used.

The most basic force trajectory during forming is constant BHF, which does not require adjustment, although an optimal value or range has to be determined. Forming results for constant BHF are dependent on several factors, of which tray shape is one. It has been reported that during press forming BHF value has a clear correlation with the quality of the rim area to which the lid is attached (Leminen *et al.* 2015). Insufficient BHF was found to lead to unacceptable quality in rectangular trays, and air tightness after sealing them with a plastic film was compromised (Fig. 7).

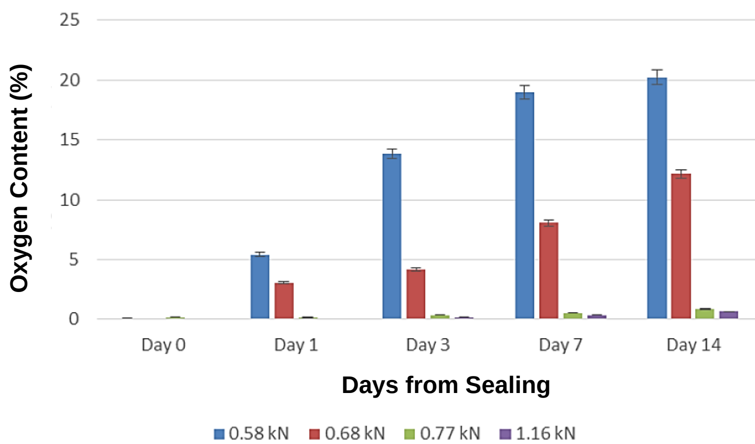

Fig. 7. Oxygen content increase inside rectangular trays as an indicator of sealability in response to the BHF value used during forming (Leminen *et al.* 2015).

When forming oval trays, it was found that they withstood higher BHFs, due to the less demanding geometry, and appropriate BHF had a wider operating window with good sealability between 520 and 1930 N compared to 580 to 1160 N for rectangular trays. Usage of the latter range produced unruptured trays, but sealability was still poor, which suggests that appropriate BHF range could be even narrower. For the processes of deep drawing, keeping BHF constant has been reported to result in the poorest forming quality compared to the further mentioned forming operations with variable BHF (Hauptmann *et al.* 2016).

A slightly more complex trajectory which does not keep BHF constant is a "linearly decreasing" trajectory, requiring force adjustment throughout the whole forming process. Typically, BHF is maximized at the beginning of forming and gradually decreases with a punch movement until reaching a minimum at the end of the process. Linearly decreasing force trajectory (*e.g.*, from 5 to 0.5 kN) has shown a slight improvement in surface quality compared to constant BHF (1.6 kN) in deep drawing (Hauptmann *et al.* 2016). During press forming, decreasing trajectory (175 to 75 N) was found to result in the best forming quality, *i.e.*, without ruptured trays (Tanninen *et al.* 2020). In another study by Tanninen *et al.*

(2018), however, the authors noted that decreasing BHF showed either too high force in the beginning, causing rupture, or too low force at the end resulting in deficient corner folding, and constant BHF was therefore recommended.

Several variations of trajectories aiming to keep the highest possible BHF have been introduced. Three options, "peak," "extended maximum," and "theoretical optimum" (Fig. 8), were tested by Hauptmann *et al.* (2016) for the deep drawing. The main differences between the three designs were either the starting point of BHF increase or the duration of its maximum value. It was found that tray surface improved visibly with advanced BHF control and became smoother. A modified "rupture optimized" trajectory was tested by Hauptmann *et al.* (2016) as well with higher BHF and smaller drawing clearance (0.4 mm instead of 0.45 mm), and it was found that the tray surface had almost no wrinkles at all (Fig. 6-f), possibly due to the increase in BHF. In contrast, a trajectory with peak BHF value in the middle of the press forming (Fig. 8, right, trajectory E) was found not to be the best option as it caused ruptured tray corners according to Tanninen *et al.* (2020).

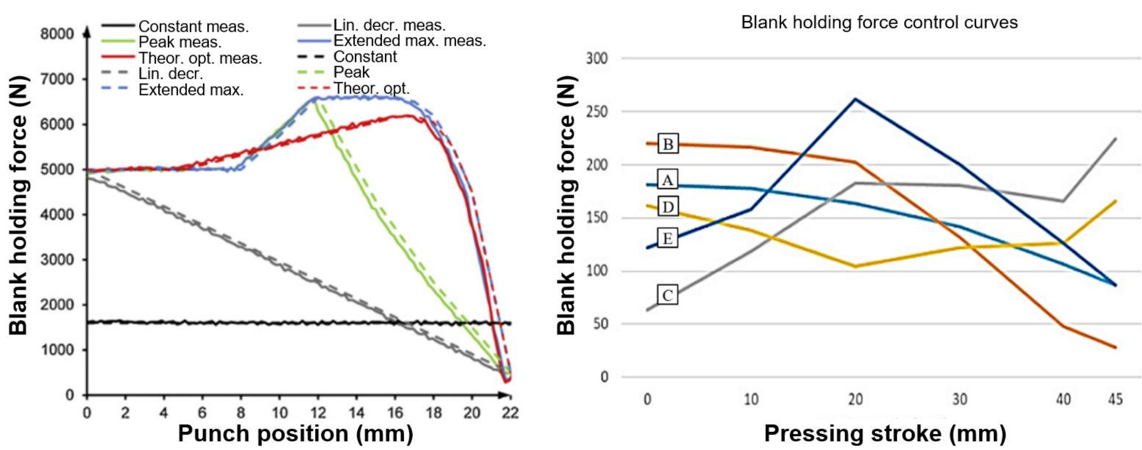

Fig. 8. Graphical representation of different BHF trajectories used in tests. Modified from Hauptmann *et al.* (2016), *Journal of Materials Processing Technology* 232 (left), with permission from Elsevier; and Tanninen *et al.* (2020) (right).

Together with BHF curve adjustment, mould speed has been reported to improve forming quality and raise average process BHF (Tanninen *et al.* 2018). In the study, stroke speed was kept at 400 mm/s until the mould contacted the surface of the blank and then decreased after contact. Thus, stroke speed was reduced during forming by 16% from 292 to 245.5 mm/s. This adjustment lowered the incidence of production of ruptured press-formed trays compared to the previous setup using constant male mould speed (300 mm/s) from 22% to 0% at 2 kN BHF and by half for 2.2 kN (from 70% to 35%).

The literature shows that using similar BHF trajectories can result in different forming quality, which can probably be explained by the specifics of the forming processes. In deep drawing, higher forces are typically used compared to press forming, due to the utilization of a fixed blank and the constant contact of the material with the mould. However, both processes are similar in that the BHF should not be increased close to the end of the punch movement, and trajectories with increasing force at the end have been reported to be the most unsuitable for forming, as they result in severe rupturing (Hauptmann *et al.* 2016; Tanninen *et al.* 2020). Particularly in Tanninen *et al.* (2020), the authors noted that the absolute value of BHF is less important than "its average magnitude throughout the pressing stroke movement," meaning that the smoother the changes in force, the better the product quality achieved.

Temperature and moisture effects on formability of substrates and coatings

The processibility of paperboard-based material during 3D forming is improved by heat input, which dramatically increases plasticity and "freezes" the material's shape after forming (Vishtal *et al.* 2014), thus decreasing deflection from the desired appearance (Hauptmann *et al.* 2011). Thermal softening weakens the interfibre bonds of the paper structure and enables easier fibre-to-fibre movement (Hauptmann *et al.* 2011; Tanninen *et al.* 2017). Besides its effect on shape formation, heating is reported to increase product stiffness after cooling (Tanninen *et al.* 2021a) and can activate self-healing properties of synthetic polymers (Leminen and Tanninen 2015). However, forming processes have their own temperature ranges, outside of which undesirable consequences can occur. Moreover, not only the absolute temperature value influences forming: other factors such as heating time, coating type, material stiffness, and thickness (Vishtal *et al.* 2014; Ovaska and Geydt 2018) may affect processing parameters significantly.

Taking particular examples, substrate heating was reported to decrease tray rupturing during press forming of a dual polymer coated paperboard (Tanninen *et al.* 2014b). Of twelve materials tested, only one showed adverse effects from substrate heating, while the convertibility of the others was improved to varying extents. The force needed to press a material into a cavity was reduced by 20% with four times temperature increase (from 23 to 80 °C) (Tanninen *et al.* 2017), meaning better energy efficiency and less demanding parameters. After a certain forming depth, intensity of heating and reduction in pressing force show clear correlation, resulting in 36 to 46% less force needed at 23 to 140 °C temperature change. Higher temperature affects the stiffness and dimensional quality of ready-made trays: under torsional load, rotational stiffness increases as a function of heat input (Tanninen *et al.* 2021a). However, excessive heating reduces the effect, highlighting the importance of temperature range optimization. With regard to visual acceptance, heating was reported to reduce the difference between the lower and upper flange diameters of a formed tray by 9.1 mm, which can be considered a remarkable increase in forming quality (Ovaska and Geydt 2018).

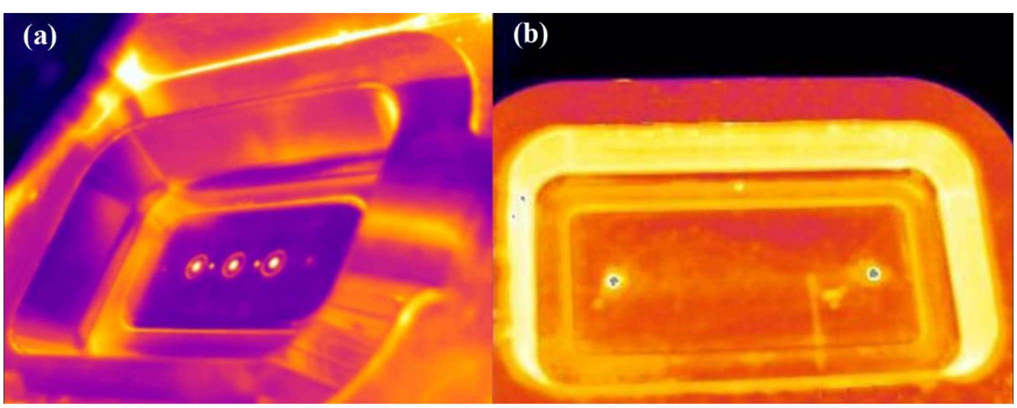

Fig. 9. Heat distribution of (a) traditional and (b) improved mould designs. Modified from Tanninen *et al.* (2014a) *Journal of Materials Processing Technology* 214(8), with permission from Elsevier.

From the machinery point of view, uneven heat distribution compromises the forming quality of trays, and traditional forming mould design assessment has encountered several difficulties, for example, uneven heating, inaccurate temperature control, quality issues affecting fixing, and complex fabrication (Tanninen *et al.* 2014a). New designs are therefore being developed to address these issues. For example, a novel design by Tanninen *et al.* (2014a) utilizes oil chambers, more evenly spread heating elements, several temperature sensors for heating control, and a reworked fixing system (Fig. 9). In usual designs, the temperature decreases gradually with every mould cycle and the quality of subsequent products is compromised. The new design resulted in 39% temperature drop improvement compared to a traditional design (-11 °C against -18 °C). Additionally, it substantially reduced variance in the outer dimensions of ready trays and the amount of scrap.

Moisture has a similar impact on fibre-based materials to temperature in regard to its effect on forming ability. Water affects the hydrogen bonds in the paperboard, which are responsible for the strength and stiffness of the material (Franke *et al.* 2021). When the material is sufficiently moisturized, the bonds are weakened, giving better flexibility and plasticity, but this process should be thoroughly controlled. Well-adjusted process conditions enable high plastic deformation (Groche and Huttel 2016) and enhance material elongation, which is highly desirable during forming. Additionally, moisture lowers the bending stiffness and increases strain at break for fibre-based materials (Östlund *et al.* 2011), as well as shifting maximum elongation values to lower temperatures (Khakalo *et al.* 2014). In particular, moisture has been reported to limit wrinkling during deep drawing (Östlund *et al.* 2011) and improve shape stability (Hauptmann *et al.* 2011). Optimal moisture range between 6.5 to 12% visibly improves wrinkles distribution, although the quality achieved does not improve further with higher moisture content (14%) (Hauptmann *et al.* 2017). Increase in moisture from 6 to 7% to 10 to 11% improves formability by up to 70% at the same temperature (Vishtal *et al.* 2014). However, the uncontrolled interaction between heating and material moisture content can result in detrimental effects on forming (Vishtal *et al.* 2014).

Poorly adjusted moisture can compromise the quality of a forming process. Too low humidity does not enable effective plastic transformation of the fibre network (Tanninen *et al.* 2014a), whereas excessive moisture sorption damages the paperboard hydrogen bonds, weakens the structure and causes swelling of fibres (Vishtal *et al.* 2014; Tanninen *et al.* 2015b). Despite water increasing the strain of the paperboard, giving better formability (Franke *et al.* 2021), it drastically reduces tensile strength (up to 30 times) (Desmaisons *et al.* 2018), which leads to lower dimensional stability, durability and poor-quality end products (Niini *et al.* 2021). In addition, excessive moisture decreases drawing depth for coated and uncoated paperboards. In multiaxial load tests, wet paperboard was found to burst at earlier stages of strain, although adding components such as gelatine or soap was reported to weaken this effect slightly (Franke *et al.* 2021).

The performance of moisturized materials depends not only on the precise water content, but also on the way in which the water is applied to the paperboard. One- and two-sided moisture coverage give different forming results and the application process appears to be more important than the overall moisture percentage in the material. Thus, blanks that had undergone one-sided water treatment were found to form better products than blanks treated on both sides, since the dry layer assisted in withstanding forming friction (Östlund *et al.* 2011). In another example, a roll moisture application method developed by Hauptmann *et al.* (2017) was shown to have advantages compared to the widely used approach of conditioning in a humidity chamber. In the roll application method, water was applied to blanks from two sides, either with two metal rolls or with a metal and rubber sponge roll. The first method demonstrated water intake of 11% after two repetitions, whereas the second showed 12% water intake already after one treatment. It was also found that blanks prepared using the roll method and having 8.3% moisture content during forming behaved similarly to those with 12% moisture content after climate chamber preparation, but the first approach required only some seconds compared to at least 24 h pre-conditioning at constant humidity. Such timesaving can be very beneficial in production processes.

Besides affecting the material substrate, heating and moisture also affect bio-based additives and coatings by changing their barrier and convertibility properties (Lyytikäinen *et al.* 2021; Ovaska *et al.* 2015). For example, wet sprayed agar or gelatine were found to soften under elevated temperatures, improving material formability by 15% if added individually, and by 18% when combined in comparison with the initial 12% (Vishtal 2015). The improved formability could be explained by an increase in the elongation of the materials treated and the rise in processing temperature until reaching a maximum; however, a drawback of this effect is reduced strength and durability (Khakalo *et al.* 2014). Gelatine was also reported to provide the highest drawing depth and less wrinkling by decreasing bending strength and enabling higher moisture content (Franke *et al.* 2021). Traditionally used plastic-coated materials benefit the most from higher plastic content at elevated temperatures due to the increased softening of polymers under heat. At the same time, formability could be dominated by the heat-softening properties of the fibre matrix rather than the coating content in plastic-free materials (Tanninen *et al.* 2017).

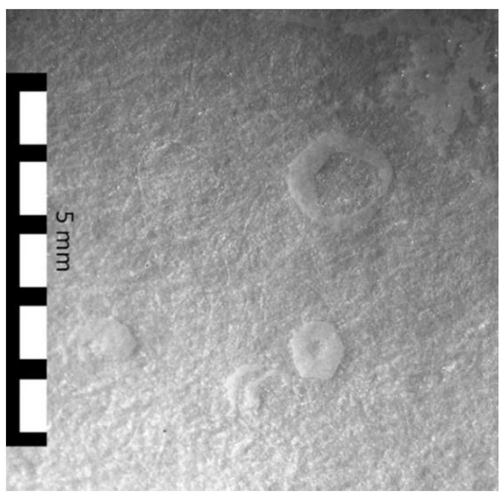

Fig. 10. Example of the surface damage due to sticking of the paperboard dispersion coating. Reprinted from Tanninen *et al.* (2014b), with permission from Wiley.

Despite it being possible to process fibre-based substrates at a wide range of temperatures, the permissible temperature range can be narrowed drastically by the coatings applied. The above-mentioned self-healing properties of polymer coatings while being heated only occur when the process temperature is thoroughly controlled. With excessive heating, coatings tend to melt and stick to the tooling (Fig. 10), thus compromising the surface quality (Ovaska and Geydt 2018). The melting temperature of coatings depends on parameters such as coating type, thickness, and composition. For example, bio-coatings developed in Tanninen *et al.* (2015c) started to stick to tooling at 150 °C, and Tanninen *et al.* (2014b) reported the existence of a severe sticking point for all the materials studied, starting from 110 °C on average. Polyethylene terephthalate (PET) coating has been found to withstand temperatures up to at least 180 °C, while some other coatings such as PE or coatings with pigments have greater heat sensitivity, which reduces the temperature processing range of the material as a whole (Ovaska and Geydt 2018). When considering the effect of temperature on coating structure, it has been seen that non-optimized converting temperatures can lead to delamination and can cause wavy hills on the coated surface, increasing its roughness by a factor of 5 (Ovaska and Geydt 2018). At lower temperatures (*ca*. 80 °C), nanoscale delamination occurred between the substrate and coating, which, however, affected a very small area and had negligible effect on material performance (Ovaska and Geydt 2018). If uncontrolled, excessive moisture content can create blistering of the plastic coating (Tanninen *et al.* 2014a), especially in pigmented barriers, which may have high moisture absorbency depending on the pigment type (Tanninen *et al.* 2014b).

The combined action of high temperature and moisture leads to moisture evaporation. Depending on the heating intensity, evaporation appears to have either a positive or negative effect. Forming quality is compromised, despite there being no ruptures in the samples formed, if heating conditions are insufficient (Tanninen *et al.* 2014b). In contrast, excessive heat evaporates too much water from pre-conditioned samples, causing their moisture level to approach that of the initial state (Vishtal *et al.* 2014). This lower moisture level negatively affects converting by reducing substrate wet softening and increasing paper-to-metal friction. The explanation for the latter is that sufficiently evaporated moisture acts as a lubricant between paper and metal during forming (Vishtal *et al.* 2014) and reduces the friction coefficient by 12 to 44% (Tanninen *et al.* 2017). For example, three converting processes were studied by Vishtal *et al.* (2014): 1) 2D spherical forming (2D SF), 2) 3D spherical forming (3D SF) and 3) 3D deep drawing (DD). The authors identified the optimal temperature ranges for the processes: below 100 °C for 2D SF, 110 to 165 °C for 3D SF, and 140 to 180 °C (cavity) and 60 to 100 °C (die) for DD. Thicker materials required higher forming temperature. The ranges given were greatly affected by the moisture evaporation processes: in more isolated from the atmosphere systems (DD and 3D SF tools) the evaporated moisture could not escape and provided good lubrication during forming, so the temperature could be increased for better material softening. On the other hand, open tools such as those used in 2D SF are operated at relatively low temperatures due to intensive moisture withdrawal from the system. It is worth mentioning that the heat input needed depends on the properties of the material as well, and moisture evaporation is more intensive in thinner papers and at higher temperatures (Östlund *et al.* 2011).

COATED SURFACE PERFORMANCE BEFORE AND AFTER CONVERTING

Even though paperboard is a prospective material that offers mechanical stiffness and flexibility for packaging production (Andersson 2008; Hauptmann *et al.* 2015; Tanninen *et al.* 2017), some packaged products contain water, grease, or oil, which require one of the main drawbacks of paperboard to be overcome – poor barrier properties (Tanninen *et al.* 2014a; Vishtal and Retulainen 2017; Bakker *et al.* 2022). In this regard, coating of fibre-based substrates has proven to be an effective approach to mitigate the penetration of substances through the base material (Fig. 11). Plastics coatings (such as PE or PET) have demonstrated good functionality in many applications (Jonhed *et al.* 2008;

Leminen and Tanninen 2015) and are now used for packaging at an industrial scale. In recent years, however, in line with efforts to move to sustainable production, coatings from petroleum-based derivatives have fallen out of favour, primarily due to environmental concerns like poor recyclability or lack of biodegradability (Rastogi and Samyn 2015).

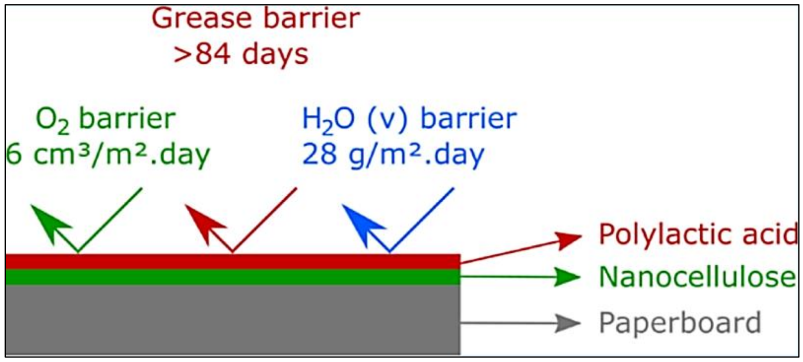

Fig. 11. Representation of barrier properties of a novel coated paperboard. Reprinted with permission from Koppolu *et al.* (2019).

To address the above-mentioned issues, attention has been directed to bio-based or bio-degradable agents that can theoretically replace plastic coatings in packaging. Biopolymers including lipids, polysaccharides, proteins, and biopolyesters are among the most promising potential materials to form new pathways for completely bio-based paperboard coatings (Rastogi and Samyn 2015). Several studies, *e.g.*, by Xu *et al.* (2022), Wang *et al.* (2018a), and Sun *et al.* (2019), have reported improvements in paperboard barrier properties from using coatings based on the aforementioned chemicals. However, while utilization of plastic coatings causes environmental issues (Rastogi and Samyn 2015), problems encountered using bio-based films are mostly associated with loss of initial properties after converting. This commonly happens due to surface damage after the application of mechanical forces during converting operations and the resulting large strains (Huang and Nygårds 2010; Barbier *et al.* 2012). Common issues include high brittleness (Tanninen *et al.* 2015c; Desmaisons *et al.* 2018), coating delamination (Leminen *et al.* 2021) and cracking (Javed *et al.* 2018), loss of barrier properties (Andersson 2008; Javed *et al.* 2018), hydrophilicity (Lavoine *et al.* 2014; Tanninen *et al.* 2015c; Rastogi and Samyn 2015), and poor material-tooling interaction (Tanninen *et al.* 2015c; Tanninen *et al.* 2014b; Ovaska and Geydt 2018). As development of sustainable materials and coatings is a relatively young branch of paper science, extensive research activities are being undertaken aiming to overcome the above concerns. Further investigation of questions such as coating resistance to stresses and barrier properties after creasing, folding and forming is required.

Barrier Properties of Coatings

Most studies investigating barrier properties focus mainly on three characteristics. The first is resistance of a packaging material to oxygen (O_2), which is an important feature since O_2 affects the properties of many packaged products, in the case of food packaging by reducing product freshness and shelf-life (Kerry *et al.* 2006). As fibre-based packaging materials are highly hydrophilic, the second property requiring attention is penetration of water vapour through the package wall, since moisture may impair the container’s stiffness and the storage conditions of the product (Song 2014). Finally, the third feature needed in many applications is oil and grease resistance, which prevents penetration of oils and allows package integrity to be maintained. Whereas plastic coatings provide an adequate barrier against all the substances mentioned, paperboard with its porous structure has no resistance to any of them, and work on development of sustainable coatings is actively ongoing to overcome this drawback.

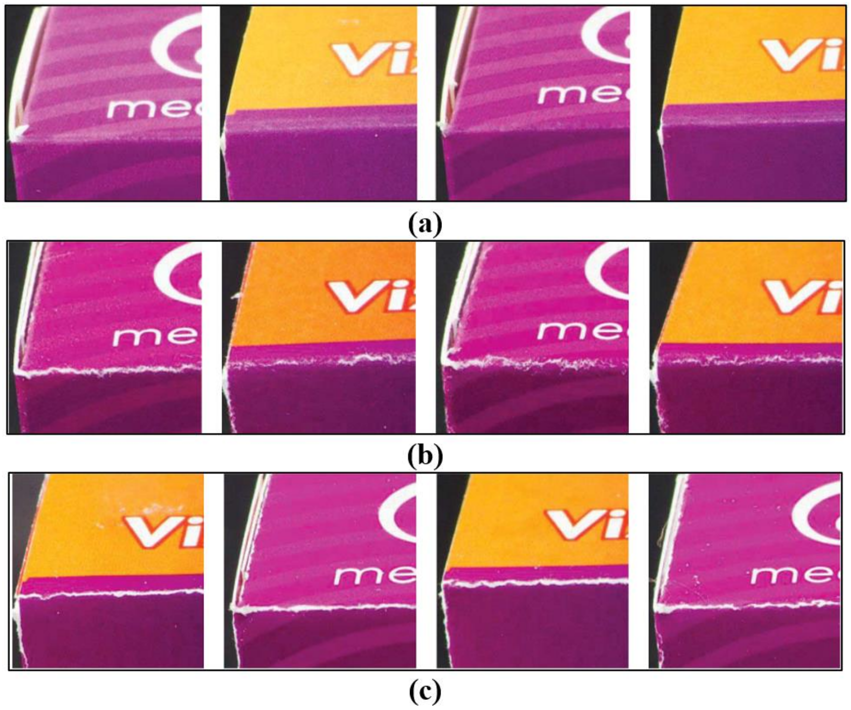

Fig. 12. Examples of paperboards after creasing and folding with: (a) good, (b) insufficient, and (c) poor cracking resistance. Reprinted with permission from Bota *et al.* (2022).

In addition to barrier layers, whose purpose is to protect the substrate from the penetration of oxygen, oils, *etc*., pigment coatings are also widely used, and converting cracks (Fig. 12) can develop on their surface as well (Marin *et al.* 2022). Pigment layers are applied on top of paperboard primarily to enhance printability and ensure attractive visual appearance. In this regard, cracking of the printed side clearly reduces paperboard product performance. Not all paperboards are prone to cracking and need a pigment layer for better printability, and some of them demonstrate acceptable surface quality and foldability without cracks when uncoated (Bota *et al.* 2022). However, printing may cause paperboard to be exposed to processes where the board is subject to high temperatures, compression and ultraviolet light (UV), which protects from scratching and rubbing but increases surface brittleness. Consequently, pigment coatings require improved parameter optimization as even small ruptures are visible on a pigmented surface (Tanninen *et al.* 2020). In addition to improving printability, pigment chemicals sometimes provide higher resistance of the surface to penetrating substances such as water vapour (Zhu *et al.* 2019).

Oxygen barrier

The barrier to oxygen refers to the ability of a package to inhibit O_2 transport through its wall, as the gas may be harmful for the contents of the package, for example, food (Auvinen and Lahtinen 2008). The severity of oxygen penetration is described by the value of the oxygen transmission rate (OTR) and oxygen permeability (OP). In study by

Christophliemk *et al.* (2017), a target value for OTR was set as less than 10 $cm^3/m^2 \cdot day \cdot bar$ to match requirements for various packaging applications. Several examples can be found where coatings drastically reduced the OTR of paperboard-based materials, *e.g.*, in works by Fukuzumi *et al.* (2009) or Aulin *et al.* (2010). The addition of nanofibrillated cellulose (NFC) to a polylactic acid (PLA) film was found to result in a reduction of oxygen penetration from 746 to 1 $mL/m^2 \cdot day \cdot Pa$ compared to pure PLA (Fukuzumi *et al.* 2009). By using MFC and lower relative humidity (RH) values, it is possible to achieve a very high oxygen barrier (Aulin *et al.* 2010), but a serious drawback of MFC is its high moisture absorbency, which is not desirable for packaging (Lavoine *et al.* 2014). A noteworthy finding from Lavoine *et al.* (2014) is that MFC had no effect on OTR when the pigment-coated substrate already possessed good oxygen resistance, meaning that sometimes coating not designed for barrier purposes can reduce the severity of oxygen penetration. Such an effect is most probably explained by the fact that uncoated paperboard usually has no oxygen resistance (Koppolu *et al.* 2019), and even the thinnest coating layer may improve the property considerably.

However, some coating chemicals do not have a significant effect on OTR in pure form without additives. For example, PLA films as an alternative to synthetic plastics are a subject of growing interest thanks to their excellent water resistance and biodegradability (Rhim and Kim 2009), but they have poor oxygen penetration inhibition properties (Fukuzumi *et al.* 2009). To compensate this drawback of a pure PLA coating, it is possible to incorporate additives such as talc, kaolin, and calcium carbonate to the coating recipe. The addition of talc has been reported to create a more tortuous path, preventing oxygen from penetrating through the coating layer (Sekelik *et al.* 1999) and resulting in a 32% OTR reduction at 4 wt% in PLA (Helanto *et al.* 2022) and an 8, 33 or 25% reduction at 1, 3 or 5 wt%, respectively, in PLA/PLC (poly(ε-caprolactone) (Jain *et al.* 2010). As can be seen, higher talc content does not mean lower OTR, and optimal concentration should be chosen. Kaolin and calcium carbonate have also been reported to improve the oxygen barrier, numerically by 56% at 3 wt% and 23% at 10 wt% in PLA, respectively (Helanto *et al.* 2022).

Another way to overcome the drawbacks of bio-coatings is by combining layers of different composition, resulting in synergetically improved effects. However, it should be remembered that reduction of OTR values does not guarantee successful subsequent use of a material in packaging production as depending on the final product application, good oil, grease or water vapour barrier properties may be required as well.

Oil, grease, and water vapour resistance

The main drawback preventing uncoated paper and paperboard from wider application (Yoo *et al.* 2012) is the lack of a barrier to resist penetration of oil and moisture (Jeong and Yoo 2020), which at high concentrations lower structural stability of the materials (Bota *et al.* 2022). Despite paper and paperboard already constituting more than one third of packaging worldwide (Huang 2017), plastics are still widely used, especially in the food sector, as grease resistance is one of the most important properties for such applications (Han and Krochta 2001). Therefore, one of the main research areas in development of paperboard-based packaging is improvement of the performance of novel coatings to enable them to match and even surpass plastic packaging in the nearest future.

Several standards for OGR measurements are used in the studies reviewed. ISO 16532-1 states that a material is greaseproof if it resists oil for at least 24 hours. According to the standard, time is measured with weight applied on an oil drop as without it OGR is much higher (Lyytikäinen *et al.* 2021). The Kit test method T 559-cm 02 (TAPPI 2012) utilizes a procedure involving an OGR check with 12 mixtures of castor oil, toluene, and n-heptane in different proportions, resulting in increasing penetration aggressiveness from 1 to 12. A sample is considered greaseproof if it attains a Kit number of 8 or above (Lavoine *et al.* 2014). Finally, ASTM F119 is used to compare penetration of different oils under different temperatures. This test does not provide an index of complete oil resistance but is useful for comparing the effects of coating compositions on OGR (Koppolu *et al.* 2019).

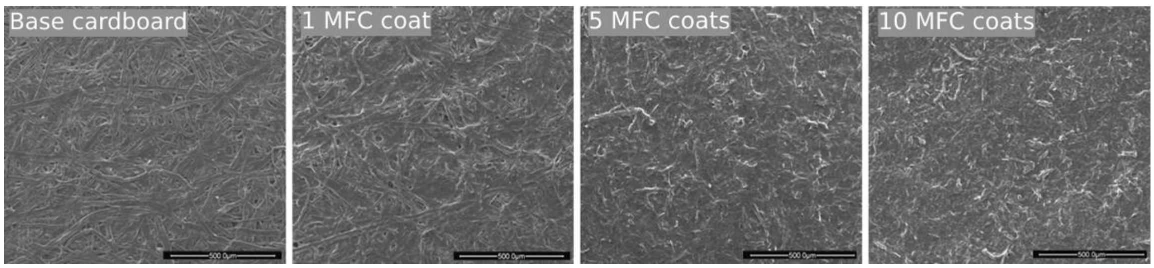

Fig. 13. Changes in paperboard surface with increase in the number of coating layers. Modified and reprinted from Lavoine *et al.* (2014), with permission from Wiley.

When application of a coating reduces the OTR value, it could be assumed that it will affect both OGR and water vapour transmission rate (WVTR), although not with the equal efficiency. As well as creating a good oxygen barrier, coatings can increase resistance to oil and water vapour penetration, but most often not to an extent suitable for packaging applications. For example, coating of a paperboard with MFC suspension (Fig. 13) increased its Kit value from 0 to 2.5, which, however, is insignificant compared with the value of 12 of the PE-coated reference (Lavoine *et al.* 2014). Coating solutions of MFC, EHEC (ethyl(hydroxyethyl)cellulose) and methyl nanocellulose (Me) were found by Lyytikäinen *et al.* (2021) to display an absence of a grease barrier for unevenly coated EHEC-MFC and MFC coated samples, and poor resistance to grease for Me-EHEC coated samples (no more than 2 h) while testing with ISO 16532-1. Five-layered Me and Me-MFC with even film showed 6 to 24 h resistance to palm kernel oil at 60 °C and 50% RH with pressure, but resistance dropped to 4 and 3 hours respectively when RH was raised to 70%. When palm kernel oil was replaced by rapeseed oil, the coatings resisted only 2 h at 23 °C and 50% RH with pressure, while still demonstrating greaseproof performance without pressure (Lyytikäinen *et al.* 2021).

The drawbacks of bio-based coating components could possibly be compensated by the combination of different barrier layers. For example, in the study by Koppolu *et al.* (2019) MFC or cellulose nanocrystals (CNC) coatings with potentially good oil and oxygen resistance were additionally covered with PLA or low-density polyethylene (LDPE) to compensate for their hydrophilicity. This combination drastically reduced WVTR and OTR through the materials and the paperboard demonstrated much higher OGR: CNC+PLA and MFC+LDPE showed no penetration after 90 days of the experiment (with ASTM F119). Whereas LDPE is a synthetic polymer with typically high barrier properties, the comparable performance of CNC+PLA is highly promising since both coatings are of bio-based origin. Similarly, work by Poulose *et al.* (2022) introduced potato fruit juice (PFJ) as a coating base for the first layer, and PLA / PLA+PBAT (Polybutylene adipate terephthalate) for the second layer (Fig. 14). In combination with the second layer, the hydrophilic and brittle surface of PFJ became smoother and resulted in improvements to barrier properties: 95% WVTR reduction, an increase in resistance to oxygen and doubled

OGR (31 days with the ASTM F119-82 procedure). The layers were applied separately as well, but equivalent results could not be achieved.

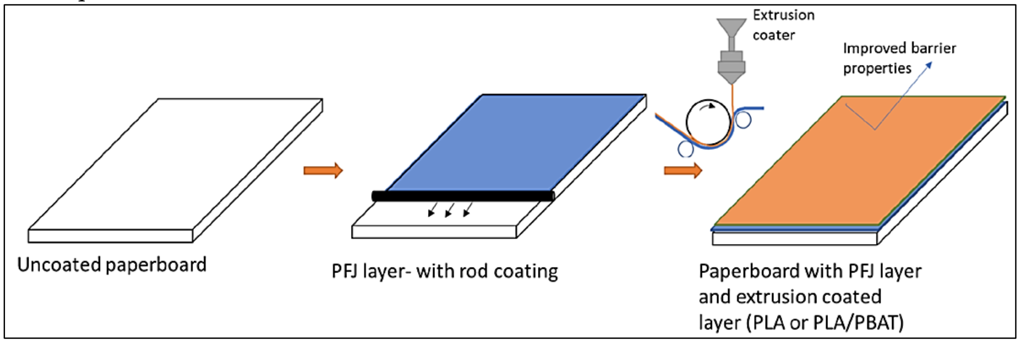

Fig. 14. Illustration of multi-layered coating process (Poulose *et al.* 2022). The study is operated under CC-BY license.

Additives can be used in bio-based coatings to increase oil and water vapour resistance. For example, along with good reduction of OTR, kaolin also affects OGR: non-converted material with a starch-kaolin coating demonstrated greaseproof performance with a measured Kit value of 8 (Tanninen *et al.* 2014b) or more than 24 h by time (Tanninen *et al.* 2015c). This result is comparable with latex-containing coating without kaolin (Tanninen *et al.* 2014b) and the PE-coated reference but with an even thinner layer. A noteworthy finding was that higher Kit values were achieved with lower coat weight of films with kaolin (Tanninen *et al.* 2014b). In comparison, hydrophobic starch- and polyolefin-talc coatings showed almost five times lower performance with around 5 h of OGR (Tanninen *et al.* 2015c). Nevertheless, talc or calcium carbonate can be used to lower WVTR with a 16 and 15% reduction at 1 to 5 and 5 to 10 wt% addition respectively, whereas the presence of kaolin in coatings had little effect on WVTR (Helanto *et al.* 2022).

Factors Affecting Coating Integrity

Maintenance of the properties of coated materials throughout their lifecycle is an issue of great concern. Factors compromising the barrier and surface properties may play a crucial role during production and converting processes, affecting both the substrates and coatings. Firstly, the integrity of the surface of a material is sometimes compromised by the coating process itself, and the process may result in a cracked film with ruptures or pinholes (Tanninen *et al.* 2015c; Leminen and Tanninen 2015; Javed *et al.* 2018; Lyytikäinen *et al.* 2021). Coating cracks are not always visible with the naked eye, and sometimes only microscopic imaging can reveal the smallest cracks, which still reduce barrier properties severely (Tanninen *et al.* 2015c, Javed *et al.* 2018). Further pressing, stretching, and the application of other forces during converting cause even greater damage. For example, several studies found that starch-based films did not provide any oxygen barrier already after coating due to cracks, although it was still possible to optimize their formulation, substrate, and drying conditions for better results (Javed *et al.* 2018; Christophliemk *et al.* 2017; Javed *et al.* 2016). Another possible defect is occurrence of pinholes from air bubbles in dispersions (Tanninen *et al.* 2015c), which are known to appear during drying of coating (Rissa *et al.* 2002). Several cellulose-based films have been reported to form uneven substrate coverage with subsequent absence of oil resistance (Lyytikäinen *et al.* 2021; Hult *et al.* 2010). Although the existence of pinholes makes OGR measurements impractical during tests (Tanninen *et al.* 2015c), evenly applied multi-layered coatings with visible pinholes may still resist oil penetration as the oil will not necessarily invade all layers, and OGR is therefore not compromised (Lyytikäinen *et al.* 2021).

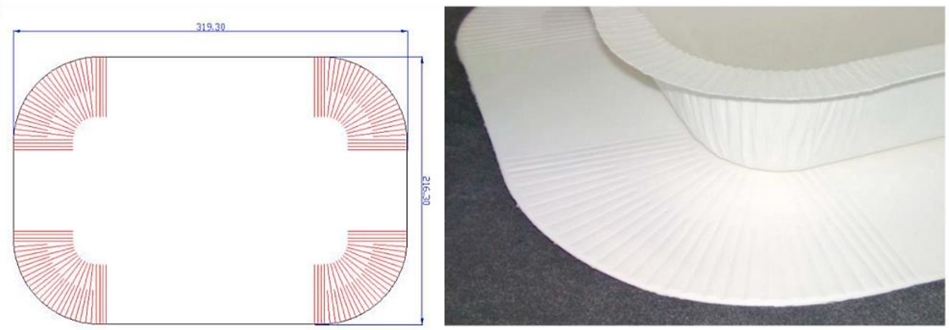

Fig. 15. Representation of a tray blank and ready tray. The tray corners are the parts deforming most (Leminen *et al.* 2013).

Measuring barrier properties after converting is a good quantitative indicator of the extent to which coatings have withstood converting stresses (Javed *et al.* 2018). Creasing, folding, and forming processes can result in surface pinholes and cracks (Tanninen *et al.* 2014b), which are caused by excessive tensile deformations of the material while being converted. If these deformations exceed the failure strain of the coating, it will crack (Zhu *et al.* 2019). In forming processes, the most severe stresses occur in the tray corners (Fig. 15) and in other areas where the converted material is stretched and/or compressed. These stresses can cause cracking of the coated surface, resulting in leaks in the package (Leminen *et al.* 2013). As an example, the already poor OGR of samples with HPC-based coatings was reduced by half, on average, after tray forming (Leminen and Tanninen 2015). Due to the brittleness of bio-based coatings, the cited authors had to reduce the force of die-cutting tools by 30% compared to a plastic-coated reference already at the blanks preparation stage as visible cracks appeared on the bio-coated surfaces otherwise. However, the starch-based coatings still had micro-cracks, and further force reduction was not possible because it could compromise subsequent forming (Leminen and Tanninen 2015). As discussed earlier, Me- and Me-MFC coatings demonstrated greaseproof behaviour without pressure application before creasing and folding but saw a reduction in grease resistance to 50 and 20 minutes respectively after converting due to mechanical rupture of the coating surface (Lyytikäinen *et al.* 2021). Coating with MFC was reported to have no stretching or cracking on the surface after converting (Lavoine *et al.* 2014), but this still does not compensate its initially low Kit value of 2.5.

In certain cases, coating cracks after creasing originate from failures of the paperboard substrate or pigment coatings under them. For example, studied in Javed *et al.* (2018) barrier coating cracked because the coating was applied either on a mineral layer, which was too brittle to withstand creasing, or directly on paperboard whose fibres were distorted by creasing and broke the coating barrier. Therefore, it is possible that despite the coating being able to withstand converting stresses, inner substrate failures may compromise the barrier quality nevertheless (Javed *et al.* 2018).

Some experiments have offered evidence that the cracking tendency in pigment and barrier coatings is dependent on the properties of the substrate, and it has been found that propagation of cracks in coated surfaces is affected by pulp type and its beating degree

(Youn *et al.* 2012). Pulp beating is a treatment which improves mechanical properties of pulp (Fig. 16), such as tensile strength, dimensional stability, and density (Zhao *et al.* 2017). Thus, in the study by Youn *et al.* (2012) a mixture of softwood (SW) and hardwood (HW) pulp at a ratio of 10:90 was found to have the best cracking resistance after folding. Despite a common assumption that stronger paperboard is more prone to damage under folding stresses, further SW addition reduced cracking resistance. As regards the effect of pulp beating, as the SW content in the pulp was only 10%, its treatment did not affect cracking resistance, while the increase in HW beating degree lowered it. Since beating improves the mechanical strength of the substrate, the folding stresses are transferred to the coating structure, compromising its quality (Youn *et al.* 2012). Additionally, printing and ink drying reduce surface flexibility, and the surface becomes even harder, resulting in more cracking (Van Gilder and Purfeerst 1994).

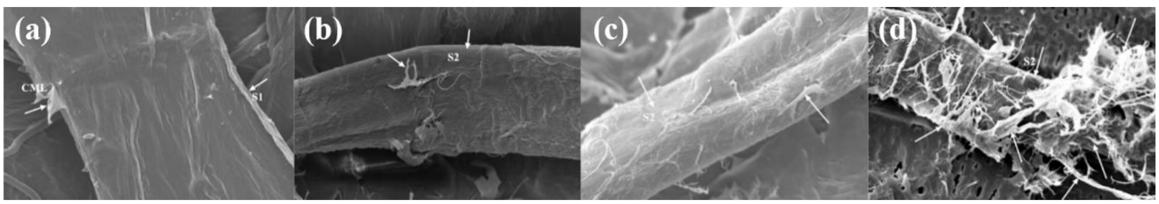

Fig. 16. Dependence of fibre surface on beating degree: (a) B0, (b) B2, (c) B4, (d) B7. Modified from Zhao *et al.* (2017), *Carbohydrate Polymers* 174, with permission from Elsevier.

To prevent coated surfaces from cracking, the same components as in barrier and pigment coatings could be used to improve flexibility for better converting and to maintain barrier properties. Thus, latex with smaller particles and a lower glass transition temperature (T_g) (the temperature at which a polymer changes its state from hard to soft (McKeen 2008) has been reported to provide better film formability, a smoother surface and a more flexible coating layer with higher cracking resistance. Latex with larger particle sizes and higher T_g resulted in the opposite performance (up to three times more cracking) (Kim *et al.* 2010). When latex was used as a binder (Rättö *et al.* 2012), the material demonstrated better crack resistance than with brittle starch (Tanninen *et al.* 2014b). The barrier properties of latex-based coatings can be improved by several pigments. Kaolins, which have good potential to resist oil penetration, and ground calcium carbonate (GCC), popular in pigment coatings (Kim *et al.* 2010; Zhu *et al.* 2019; Youn *et al.* 2012; Rättö *et al.* 2012), reduced WVTR through latex film according to Zhu *et al.* (2019). The results were better with lower pigment content and higher coat weight. However, while pure latex was more crack-resistant, the presence of pigments caused severe cracking of layers during folding and increased WVTR by at least twice. Kaolins still performed better that pure latex, whereas GCC did not.

Film formability and subsequent cracking tendency is dependent on the particle sizes of the pigments. For example, two types of GCC were tested by Rättö *et al.* (2012), GCC with broad particle size distribution (bGCC) and GCC with narrow (nGCC) particle size distribution. bGCC had more diverse sizes of particles, and smaller particles filled voids between larger particles, requiring less binder, which strengthened the coated surface (Lamminmäki *et al.* 2010). This effect improved cracking resistance. nGCC coatings were damaged more during creasing. In the same study, the authors tested clay pigment as well as GCCs. Despite clays having low strength for delamination and shearing and thus low resistance to cracking, the pigment still performed better than nGCC. Clay demonstrated performance closer to bGCC, which leads to the conclusion that sometimes factors other than pigment type alone may be involved in the results (Rättö *et al.* 2012).

CONCLUSIONS

This review discussed convertibility and barrier properties of coated fibre-based materials from several points of view. The goal was to provide a comprehensive overview of the most important issues arising during different stages of preparation and conversion of coated packaging materials. The work focused on examining factors that hinder or contribute to the convertibility of fibre-based substrates and coatings.

The most common converting operations for coated materials are creasing and folding. These actions are closely related, as effective creasing improves the foldability of material without excessive impairment of final package stiffness. Although there are several methods of creasing, they have negligible difference as regards their effect on foldability. Creasing depth plays a much more important role in convertibility improvement than creasing method. A window of creasing depth for efficient folding is formed between two thresholds: the lower bound is when sufficient delamination of the inner material structure occurs to reduce its bending stiffness, and the upper bound is when excessive damage leads to the onset of in-plane cracks formation, which compromises the integrity of the package and its surface. Experimental data has confirmed that after a certain depth, which depends on material properties, further creasing does not improve foldability but just damages the structure of the paperboard.

There is a similar dynamic when considering factors affecting the formability of coated materials. Namely, it is vital to adjust all forming parameters so that they fall within an acceptable range since parameters outside this range result in undesirable consequences. For example, the forming quality of coated substrates is reduced when forming temperature or material moisture content are too low for the process. On the other hand, overheated or over-moisturised materials become too weak to withstand converting, and failure of structural integrity occurs.

Forming of coated and uncoated paperboards differs considerably due to the effect of coatings on material properties. Coatings change the mechanical properties of substrates positively or negatively depending on the response to the nature of the films applied: coatings that result in excessive infiltration of water into the substrates weaken fibre bonds and impair material stiffness, whereas drier coatings such as those used in extrusion coating processes form strengthened structures. While high temperature is preferable for paperboard forming, it can damage the coated surface by melting it, causing delamination or sticking to machinery. Overall, there is always a trade-off between the formability of coated materials and their mechanical performance in a future package. However, the formability of sustainably coated paperboards is not a major problem in comparison to their barrier properties.

The resistance of novel bio-based coatings to oxygen, water vapour, and oil often appears to be sufficient prior to converting. However, creasing, and folding operations can compromise the integrity of the barrier or pigment coatings by loading them with stresses which the coated surfaces cannot withstand. Even when the size and type of converting equipment is chosen based on the material properties, it has been found that the most prevalent factor related to surface damage is the fact of conversion rather than the machinery used. Bio-based coatings require further development to reach the convertibility of more flexible plastic films. Several directions for improvement are being investigated, for example, mixing additives to dispersion coatings, designing more advanced tools, and optimizing converting parameters.

In conclusion, although a relatively extensive body of literature examines novel coated materials and describes their properties in unconverted form, far fewer sources discuss the convertibility of such materials and their subsequent surface behaviour. The surface of materials coated with bio-based coatings is damaged more easily during converting operations than materials coated with conventional plastic coatings, which reduces their potential for wider application. Since conversion processes often compromise the integrity of materials, it is extremely important to investigate the behaviour after creasing, folding, and forming of materials with novel coatings to ensure adequate performance for packaging applications.

ACKNOWLEDGEMENT

This research was funded partly by Business Finland, project "SUSBINCO", which is gratefully acknowledged.

REFERENCES CITED

Afshariantorghabeh, S., Kärki, T., and Leminen, V. (2022). "Three-dimensional forming of plastic-coated fibre-based materials using a thermoforming process," *Packaging Technology and Science* 35(7), 543-555. DOI: 10.1002/pts.2650

Andersson, C. (2008). "New ways to enhance the functionality of paperboard by surface treatment - A review," *Packaging Technology and Science* 21(6), 339-373. DOI: 10.1002/pts.823

Aulin, C., Gällstedt, M., and Lindström, T. (2010). "Oxygen and oil barrier properties of microfibrillated cellulose films and coatings," *Cellulose* 17(3), 559-574. DOI: 10.1007/s10570-009-9393-y

Auvinen, S., and Lahtinen, K. (2008). "Converted paper and paperboard as packaging materials," in: *Paper and Paperboard Converting* (2nd Ed.), J. Kuusipalo (ed.), Finnish Paper Engineers' Association/Paperi ja Puu Oy, Helsinki, Finland, pp. 286-332.

Bakker, S., Aarts, J., Esteves, A. C. C., Metselaar, G. A., and Schenning, A. P. H. J. (2022). "Water barrier properties of resin-stabilized waterborne coatings for paperboard," *Macromolecular Materials and Engineering* 307, article 2100829. DOI: 10.1002/mame.202100829

Barbier, C., Rättö, P., and Hornatowska, J. (2012). "Coating models for an analysis of cracking behaviour between folded paper and creased board," in: *12th TAPPI Advanced Coating Fundamentals Symposium Proceedings, Co-located with the 16th International Coating Science and Technology Symposium, ISCST 2012*, pp. 5-16.

Bota, J., Pibernik, J., Kovačević, D., and Brozović, M. (2022). "The effects of flatbed creasing tool on printed paperboard," in: *Proceedings - The Eleventh International Symposium GRID 2022*, pp. 657-663. DOI: 10.24867/GRID-2022-p71

Christophliemk, H., Johansson, C., Ullsten, H., and Järnström, L. (2017). "Oxygen and water vapor transmission rates of starch-poly(vinyl alcohol) barrier coatings for

flexible packaging paper," *Progress in Organic Coatings* 113, 218-224. DOI: 10.1016/j.porgcoat.2017.04.019
Csavajda, P., Böröcz, P., Mojzes, Á., and Molnár, B. (2017). "The effect of creasing lines on the compression strength of adjustable height corrugated boxes," 9(1), 15-22.
Desmaisons, J., Gustafsson, E., Dufresne, A., and Bras, J. (2018). "Hybrid nanopaper of cellulose nanofibrils and PET microfibers with high tear and crumpling resistance," *Cellulose* 25(12), 7127-7142. DOI: 10.1007/s10570-018-2044-4
Domaneschi, M., Perego, U., Borgqvist, E., and Borsari, R. (2017). "An industry-oriented strategy for the finite element simulation of paperboard creasing and folding: Finite element simulation of paperboard creasing and folding," *Packaging Technology and Science* 30(6), 269-294. DOI: 10.1002/pts.2298
Franke, W., Leminen, V., Groche, P., and Varis, J. (2021). "The effects of pretreatment and coating on the formability of extrusion-coated multilayer paperboard-plastic composites," *Packaging Technology and Science* 34(2), 105-116. DOI: 10.1002/pts.2542
Fukuzumi, H., Saito, T., Iwata, T., Kumamoto, Y., and Isogai, A. (2009). "Transparent and high gas barrier films of cellulose nanofibers prepared by TEMPO-mediated oxidation," *Biomacromolecules* 10(1), 162-165. DOI: 10.1021/bm801065u
Geis, T., Klingelhöller, C., and Hintze, W. (2014). "Constant depth scoring of fibre reinforced plastic structures to prevent delamination," *Procedia CIRP* 14, 205-210. DOI: 10.1016/j.procir.2014.03.103
Girlanda, O., and Fellers, C. (2007). "Evaluation of the tensile stress-strain properties in the thickness direction of paper materials," *Nordic Pulp & Paper Research Journal* 22(1), 49-56. DOI: 10.3183/npprj-2007-22-01-p049-056
Groche, P., and Huttel, D. (2016). "Paperboard forming – Specifics compared to sheet metal forming," *BioResources* 11(1), 1855-1867. DOI: 10.15376/biores.11.1.1855-1867
Han, J. H., and Krochta, J. M. (2001). "Physical properties and oil absorption of whey-protein-coated paper," *Journal of Food Science* 66(2), 294-299. DOI: 10.1111/j.1365-2621.2001.tb11335.x
Hauptmann, M., Kustermann, T., Schmalholz, M., Haug, H., and Majschak, J.-P. (2017). "Examination of the transferability of technological key features of paperboard deep drawing towards the application in fast-running packaging machines," *Packaging Technology and Science* 30(1-2), 21-31. DOI: 10.1002/pts.2270
Hauptmann, M., and Majschak, J.-P. (2011). "New quality level of packaging components from paperboard through technology improvement in 3D forming," *Packaging Technology and Science* 24(7), 419-432. DOI: 10.1002/pts.941
Hauptmann, M., Wallmeier, M., Erhard, K., Zelm, R., and Majschak, J.-P. (2015). "The role of material composition, fiber properties and deformation mechanisms in the deep drawing of paperboard," *Cellulose* 22(5), 3377-3395. DOI: 10.1007/s10570-015-0732-x
Hauptmann, M., Weyhe, J., and Majschak, J.-P. (2016). "Optimisation of deep drawn paperboard structures by adaptation of the blank holder force trajectory," *Journal of Materials Processing Technology* 232, 142-152. DOI: 10.1016/j.jmatprotec.2016.02.007
Helanto, K., Talja, R., and Rojas, O. J. (2022). "Mineral-filled biopolyester coatings for paperboard packaging materials: Barrier, sealability, convertability and biodegradability properties," *Nordic Pulp & Paper Research Journal* 37(1), 212-221.

DOI: 10.1515/npprj-2021-0076
Huang, H., Hagman, A., and Nygårds, M. (2014). "Quasi static analysis of creasing and folding for three paperboards," *Mechanics of Materials* 69(1), 11-34. DOI: 10.1016/j.mechmat.2013.09.016
Huang, H., and Nygårds, M. (2010). "A simplified material model for finite element analysis of paperboard creasing," *Nordic Pulp & Paper Research Journal* 25(04), 505-512. DOI: 10.3183/NPPRJ-2010-25-04-p505-512
Huang, J. (2017). "Sustainable development of green paper packaging," *Environment and Pollution* 6(2), 1. DOI: 10.5539/ep.v6n2p1
Hult, E.-L., Iotti, M., and Lenes, M. (2010). "Efficient approach to high barrier packaging using microfibrillar cellulose and shellac," *Cellulose* 17(3), 575-586. DOI: 10.1007/s10570-010-9408-8
Jain, S., Reddy, M. M., Mohanty, A. K., Misra, M., and Ghosh, A. K. (2010). "A new biodegradable flexible composite sheet from poly(lactic acid)/poly(ε -caprolactone) blends and micro-talc," *Macromolecular Materials and Engineering* 295(8), 750-762. DOI: 10.1002/mame.201000063
Javed, A., Rättö, P., Järnström, L., and Ullsten, H. (2018). "Crack analysis of barrier coatings based on starch and starch-PVOH with and without plasticizer," *Nordic Pulp & Paper Research Journal* 33(2), 336-347. DOI: 10.1515/npprj-2018-3039
Javed, A., Ullsten, H., Järnström, L., and Ernstsson, M. (2016). "Study of starch and starch-PVOH blends and effects of plasticizers on mechanical and barrier properties of coated paperboard," *Nordic Pulp & Paper Research Journal* 31(3), 499-510. DOI: 10.3183/npprj-2016-31-03-p499-510
Jeong, S., and Yoo, S. (2020). "Whey protein concentrate-beeswax-sucrose suspension-coated paperboard with enhanced water vapor and oil-barrier efficiency," *Food Packaging and Shelf Life* 25, article 100530. DOI: 10.1016/j.fpsl.2020.100530
Jonhed, A., Andersson, C., and Järnström, L. (2008). "Effects of film forming and hydrophobic properties of starches on surface sized packaging paper," *Packaging Technology and Science* 21(3), 123-135. DOI: 10.1002/pts.783
Kerry, J. P., O'Grady, M. N., and Hogan, S. A. (2006). "Past, current and potential utilisation of active and intelligent packaging systems for meat and muscle-based products: A review," *Meat Science* 74(1), 113-130. DOI: 10.1016/j.meatsci.2006.04.024
Khakalo, A., Filpponen, I., Johansson, L.-S., Vishtal, A., Lokanathan, A. R., Rojas, O. J., and Laine, J. (2014). "Using gelatin protein to facilitate paper thermoformability," *Reactive and Functional Polymers* 85, 175-184. DOI: 10.1016/j.reactfunctpolym.2014.09.024
Kim, C.-K., Lim, W.-S., and Lee, Y. K. (2010). "Studies on the fold-ability of coated paperboard (I)," *Journal of Industrial and Engineering Chemistry* 16(5), 842-847. DOI: 10.1016/j.jiec.2010.05.001
Koppolu, R., Lahti, J., Abitbol, T., Swerin, A., Kuusipalo, J., and Toivakka, M. (2019). "Continuous processing of nanocellulose and polylactic acid into multilayer barrier coatings," *ACS Applied Materials & Interfaces* 11(12), 11920-11927. DOI: 10.1021/acsami.9b00922
Lamminmäki, T., Kettle, J., Puukko, P., Ketoja, J., and Gane, P. A. C. (2010). "Printing: The role of binder type in determining inkjet print quality," *Nordic Pulp & Paper Research Journal* 25(3), 380-390. DOI: 10.3183/npprj-2010-25-03-p380-390
Lavoine, N., Bras, J., and Desloges, I. (2014). "Mechanical and barrier properties of

cardboard and 3D packaging coated with microfibrillated cellulose," *Journal of Applied Polymer Science* 131(8), article 40106. DOI: 10.1002/app.40106
Leminen, V., Matthews, S., Tanninen, P., and Varis, J. (2018). "Effect of creasing tool dimensions on the quality of press-formed paperboard trays," *Procedia Manufacturing* 25, 397-403. DOI: 10.1016/j.promfg.2018.06.109
Leminen, V., Niini, A., Tanninen, P., and Matthews, S. (2021). "Comparison of creasing and scoring in the manufacturing of folding cartons," *Procedia Manufacturing* 55, 221-225. DOI: 10.1016/j.promfg.2021.10.031
Leminen, V., and Tanninen, P. (2015). "Convertability and oil resistance of paperboard with hydroxypropyl-cellulose-based dispersion barrier coatings," *Journal of Applied Packaging Research* 7(3), 91-100.
Leminen, V., Tanninen, P., Lindell, H., and Varis, J. (2015). "Effect of blank holding force on the gas tightness of paperboard trays manufactured by the press forming process," *BioResources* 10(2), 2235-2243. DOI: 10.15376/biores.10.2.2235-2243
Leminen, V., Tanninen, P., Mäkelä, P., and Varis, J. (2013). "Combined effect of paperboard thickness and mould clearance in the press forming process," *BioResources* 8(4), 5701-5714. DOI: 10.15376/biores.8.4.5701-5714
Leminen, V., Tanninen, P., Matthews, S., Pesonen, A., and Varis, J. (2019). "The effect of creasing method and tooling on the geometry of formed creases in the creasing process of coated and uncoated paperboard," in: *AIP conference Proceedings*, Vitoria-Gasteiz, Spain, 020014. DOI: 10.1063/1.5112519
Lyytikäinen, J., Ovaska, S.-S., Heiskanen, I., and Backfolk, K. (2021). "The role of MFC and hydrophobically modified ethyl(hydroxyethyl)cellulose in film formation and the barrier properties of methyl nanocellulose film," *Nordic Pulp & Paper Research Journal* 36(2), 312-322. DOI: 10.1515/npprj-2020-0099
Marin, G., Nygårds, M., and Östlund, S. (2022). "Experimental quantification of differences in damage due to in-plane tensile test and bending of paperboard," *Packaging Technology and Science* 35(1), 69-80. DOI: 10.1002/pts.2608
Marsh, K., and Bugusu, B. (2007). "Food packaging – Roles, materials, and environmental issues," *Journal of Food Science* 72(3), R39-R55. DOI: 10.1111/j.1750-3841.2007.00301.x
McKeen, L. W. (2008). "Introduction to plastics and elastomers," in: *Effect of Temperature and Other Factors on Plastics and Elastomers* (2nd Ed.), William Andrew Inc., Norwich, NY, USA, pp. 1-39.
Mentrasti, L., Cannella, F., Pupilli, M., and Dai, J. S. (2013). "Large bending behavior of creased paperboard. I. Experimental investigations," *International Journal of Solids and Structures* 50(20–21), 3089-3096. DOI: 10.1016/j.ijsolstr.2013.05.018
Morris, B. A. (2008). "Understanding why adhesion in extrusion coating decreases with diminishing coating thickness," *Journal of Plastic Film & Sheeting* 24(1), 53-88. DOI: 10.1177/8756087908089486
Na, Y. J., Paluskiewicz, S. A., and Muhlstein, C. L. (2020). "High-cycle fatigue damage accumulation in paper," *Communications Materials* 1(1), 26. DOI: 10.1038/s43246-020-0025-2
Niini, A., Leminen, V., Tanninen, P., and Varis, J. (2021). "Humidity effect in heating and cooling of press-formed paperboard food packages: Comparison of storing and heating conditions," *Packaging Technology and Science* 34(8), 517-522. DOI: 10.1002/pts.2578
Nygårds, M., Bhattacharya, A., and Krishnan, S. (2014). "Optimizing shear strength

profiles in paperboard for better crease formation," *Nordic Pulp & Paper Research Journal* 29(3), 510-520. DOI: 10.3183/npprj-2014-29-03-p510-520

Nygårds, M., Hallbäck, N., Just, M., and Tryding, J. (2005). "A finite element model for simulations of creasing and folding of paperboard," in: *Proceedings of the ABAQUS Users' Conference 2005*.

Östlund, M., Borodulina, S., and Östlund, S. (2011). "Influence of paperboard structure and processing conditions on forming of complex paperboard structures," *Packaging Technology and Science* 24(6), 331-341. DOI: 10.1002/pts.942

Östlund, S. (2018). "Three-dimensional deformation and damage mechanisms in forming of advanced structures in paper," in: A*dvances in Pulp and Paper Research, Oxford 2017, Trans. of the XVIth Fund. Res. Symp. Oxford, 2017,* W. Batchelor and D. Söderberg (eds.), FRC, Manchester, UK, pp. 489-594. DOI: 10.15376/frc.2017.2.489

Ovaska, S.-S., and Geydt, P. (2018). "Three-dimensional forming of multi-layered materials: Material heat response and quality aspects," *Journal of Applied Packaging Research* 10(4), 28-44.

Ovaska, S.-S., Geydt, P., Österberg, M., Johansson, L.-S., and Backfolk, K. (2015). "Heat-Induced changes in oil and grease resistant hydroxypropylated-starch-based barrier coatings," *Nordic Pulp & Paper Research Journal* 30(3), 488-496. DOI: 10.3183/npprj-2015-30-03-p488-496

Panek, J., and Hart, P. W. (2022). "Review of coating cracking and barrier integrity on paperboard substrates," *TAPPI Journal* 21(11), 589-603. DOI: 10.32964/TJ21.11.589

Poulose, S., Toriseva, J., Lahti, J., Jönkkäri, I., Hedenqvist, M. S., and Kuusipalo, J. (2022). "A green high barrier solution for paperboard packaging based on potato fruit juice, poly(lactic acid), and poly(butylene adipate terephthalate)," *ACS Applied Polymer Materials* 4(6), 4179-4188. DOI: 10.1021/acsapm.2c00153

Rastogi, V., and Samyn, P. (2015). "Bio-based coatings for paper applications," *Coatings* 5(4), 887-930. DOI: 10.3390/coatings5040887

Rättö, P., Hornatowska, J., and Barbier, C. (2012). "Influence of the distribution of the shape and size distribution of pigment particles on cracking in coating layers during creasing," *Nordic Pulp & Paper Research Journal* 27(4), 714-720. DOI: 10.3183/NPPRJ-2012-27-04-p714-720

Rhim, J.-W. (2010). "Effect of moisture content on tensile properties of paper-based food packaging materials," *Food Science and Biotechnology* 19(1), 243-247. DOI: 10.1007/s10068-010-0034-x

Rhim, J.-W., and Kim, J.-H. (2009). "Properties of poly(lactide)-coated paperboard for the use of 1-way paper cup," *Journal of Food Science* 74(2), E105-E111. DOI: 10.1111/j.1750-3841.2009.01073.x

Rissa, K., Vähä-Nissi, M., Lepistö, T., Savolainen, A. (2002). "Talc-filled water-based barrier coatings," *Paperi ja Puu* 84, 467-472.

Sekelik, D. J., Stepanov, E. V., Nazarenko, S., Schiraldi, D., Hiltner, A., and Baer, E. (1999). "Oxygen barrier properties of crystallized and talc-filled poly(ethylene terephthalate)," *Journal of Polymer Science Part B: Polymer Physics* 37(8), 847-857. DOI: 10.1002/(SICI)1099-0488(19990415)37:8<847::AID-POLB10>3.0.CO;2-3

Sharma, S., Barkauskaite, S., Jaiswal, A. K., and Jaiswal, S. (2021). "Essential oils as additives in active food packaging," *Food Chemistry* 343, article 128403. DOI: 10.1016/j.foodchem.2020.128403

Shen, J., Fatehi, P., and Ni, Y. (2014). "Biopolymers for surface engineering of paper-based products," *Cellulose* 21(5), 3145-3160. DOI: 10.1007/s10570-014-0380-6

Song, Z., Xiao, H., and Zhao, Y. (2014). "Hydrophobic-modified nano-cellulose fiber/PLA biodegradable composites for lowering water vapor transmission rate (WVTR) of paper," *Carbohydrate Polymers* 111, 442-448. DOI: 10.1016/j.carbpol.2014.04.049

Sun, H., Shao, X., Zhang, M., Wang, Z., Dong, J., and Yu, D. (2019). "Mechanical, barrier and antimicrobial properties of corn distarch phosphate/nanocrystalline cellulose films incorporated with Nisin and ε-polylysine," *International Journal of Biological Macromolecules* 136, 839-846. DOI: 10.1016/j.ijbiomac.2019.06.134

Tanninen, P., Kasurinen, M., Eskelinen, H., Varis, J., Lindell, H., Leminen, V., Matthews, S., and Kainusalmi, M. (2014a). "The effect of tool heating arrangement on fibre material forming," *Journal of Materials Processing Technology* 214(8), 1576-1582. DOI: 10.1016/j.jmatprotec.2014.03.001

Tanninen, P., Lindell, H., Saukkonen, E., and Backfolk, K. (2014b). "Thermal and mechanical durability of starch-based dual polymer coatings in the press forming of paperboard," *Packaging Technology and Science* 27(5), 353-363. DOI: 10.1002/pts.2036

Tanninen, P., Leminen, V., Eskelinen, H., Lindell, H., and Varis, J. (2015a). "Controlling the folding of the blank in paperboard tray press forming," *BioResources* 10(3), 5191-5202. DOI: 10.15376/biores.10.3.5191-5202

Tanninen, P., Leminen, V., Kainusalmi, M., and Varis, J. (2015b). "Effect of process parameter variation on the dimensions of press-formed paperboard trays," *BioResources* 11(1), 140-158. DOI: 10.15376/biores.11.1.140-158

Tanninen, P., Saukkonen, E., Leminen, V., Lindell, H., and Backfolk, K. (2015c). "Adjusting the die cutting process and tools for biopolymer dispersion coated paperboards," *Nordic Pulp & Paper Research Journal* 30(2), 336-343.

Tanninen, P., Matthews, S., Ovaska, S.-S., Varis, J., and Backfolk, K. (2017). "A novel technique for the evaluation of paperboard performance in press-forming," *Journal of Materials Processing Technology* 240, 284-292. DOI: 10.1016/j.jmatprotec.2016.10.002

Tanninen, P., Leminen, V., Matthews, S., Kainusalmi, M., and Varis, J. (2018). "Process cycle optimization in press forming of paperboard," *Packaging Technology and Science* 31(5), 369-376. DOI: 10.1002/pts.2331

Tanninen, P., Leminen, V., Pesonen, A., Matthews, S., and Varis, J. (2020). "Surface fracture prevention in paperboard press forming with advanced force control," *Procedia Manufacturing* 47, 80-84. DOI: 10.1016/j.promfg.2020.04.140

Tanninen, P., Leminen, V., Matthews, S., Niini, A., and Varis, J. (2021a). "Measurement of changes in torsional stiffness of press-formed paperboard packages induced by heat load utilizing a developed measuring device," *Packaging Technology and Science* 34(10), 641-650. DOI: 10.1002/pts.2601

Tanninen, P., Matthews, S., Leminen, V., and Varis, J. (2021b). "Analysis of paperboard creasing properties with a novel device," *Procedia Manufacturing* 55, 232-237. DOI: 10.1016/j.promfg.2021.10.033

TAPPI (2012). "Grease resistance test for paper and paperboard," TAPPI Press, Atlanta, GA, USA.

Van Gilder, R. L., Purfeerst R. D. (1994). "Latex binder modification to reduce coating pick on six-color offset presses," *Tappi Journal* 77, 230-239.

Vishtal, A., Hauptmann, M., Zelm, R., Majschak, J.-P., and Retulainen, E. (2014). "3D forming of paperboard: The influence of paperboard properties on formability,"

Packaging Technology and Science 27(9), 677-691. DOI: 10.1002/pts.2056
Vishtal, A., and Retulainen, E. (2017). "Deep-drawing of paper and paperboard: The role of material properties," *BioResources* 7(3), 4424-4450. DOI: 10.15376/biores.7.3.4424-4450
Vishtal, A., Retulainen, E., Khakalo, A., and Rojas, O. J. (2015). "Improving the extensibility of paper: Sequential spray addition of gelatine and agar," *Nordic Pulp & Paper Research Journal* 30(3), 452-460. DOI: 10.3183/npprj-2015-30-03-p452-460
Wang, C., Shi, J., He, M., Ding, L., Li, S., Wang, Z., and Wei, J. (2018a). "High strength cellulose/ATT composite films with good oxygen barrier property for sustainable packaging applications," *Cellulose* 25(7), 4145-4154. DOI: 10.1007/s10570-018-1855-7
Wang, Y., Yang, M., Shen, R., Shao, S., Chen, L., Gong, W., Shan, L., and Gao, C. (2018b). "Development of metoprolol tartrate-loaded sustained-release pellets: effect of talc on the mechanism of drug release," *Pharmaceutical Development and Technology* 23(7), 664-673. DOI: 10.1080/10837450.2016.1212881
Wang, Z.-W., and Sun, Y.-C. (2018). "Experimental investigation on bending fatigue failure of corrugated paperboard," *Packaging Technology and Science* 31(9), 601-609. DOI: 10.1002/pts.2390
Xu, K., Li, Q., Xie, L., Shi, Z., Su, G., Harper, D., Tang, Z., Zhou, J., Du, G., and Wang, S. (2022). "Novel flexible, strong, thermal-stable, and high-barrier switchgrass-based lignin-containing cellulose nanofibrils/chitosan biocomposites for food packaging," *Industrial Crops and Products* 179, article 114661. DOI: 10.1016/j.indcrop.2022.114661
Yamada, Y., Iwata, K., Kadowaki, T., and Sumiya, T. (2017). "Method of reduced variables for stiffness degradation process of unidirectional CFRP composites subjected to alternating bending," *Composites Science and Technology* 138, 117-123. DOI: 10.1016/j.compscitech.2016.11.011
Yoo, S., Lau, S. H., and Krochta, J. M. (2012). "Grease penetration and browning resistance of pulpboard and paperboard coated with whey protein," *Packaging Technology and Science* 25(5), 259-270. DOI: 10.1002/pts.977
Youn, H. J., Sim, K., Oh, K. D., Lae Lee, H., Han, C. S., Yeu, S. U., and Lee, Y. M. (2012). "Fold cracking of coated paper: The effect of pulp fiber composition and beating," *Nordic Pulp & Paper Research Journal* 27(2), 445-450. DOI: 10.3183/npprj-2012-27-02-p445-450
Zhao, D., Yang, F., Dai, Y., Tao, F., Shen, Y., Duan, W., Zhou, X., Ma, H., Tang, L., and Li, J. (2017). "Exploring crystalline structural variations of cellulose during pulp beating of tobacco stems," *Carbohydrate Polymers* 174, 146-153. DOI: 10.1016/j.carbpol.2017.06.060
Zhu, Y., Bousfield, D., and Gramlich, W. M. (2019). "The influence of pigment type and loading on water vapor barrier properties of paper coatings before and after folding," *Progress in Organic Coatings* 132, 201-210. DOI: 10.1016/j.porgcoat.2019.03.031

Article submitted: July 18, 2023; Peer review completed: August 5, 2023; Revised version received: August 29, 2023; Accepted: September 11, 2023; Published: September 18, 2023.
DOI: 10.15376/biores.18.4.Lev