Three-dimensional forming of plastic-coated fibre-based materials using a thermoforming process

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Abstract
Three-dimensional (3D) forming of fibre-based materials has been a topic of growing interest over recent years and 3D forming processes using hydroforming, press forming and deep drawing processes have been widely explored. Thermoforming as a potential alternative method for forming these materials remains, however, relatively understudied. This research attempts to provide a fundamental understanding of the thermoforming limitations of plastic-coated paperboards. In the work, a variety of commercial paperboards are subjected to experimental tests with different forming parameters and moulding depths. Shape accuracy, maximum acquired depth, thickness distribution behaviour and damage mechanisms are used to evaluate thermoformability, and the results linked to the material properties and forming conditions. The research findings indicate that the plastic-coated paperboards studied are thermoformable but only in simple geometric shapes and with low mould depths. Unlike plastic, thermoforming can result in thickness increase in plastic-coated paperboards, which is thought to be a result of out-of-plane auxetic behaviour of paperboards. Paperboard thermoforming was also found to be hindered by rupture, blistering and curling defects. Tensile strain at break is the key factor determining thermoformability. Additionally, the density of the paperboard can impact the heating step and the rate at which the moisture content of the material changes during the forming process. Furthermore, it was observed that changes in process parameters affected materials differently, with the direction and rate of change differing based on the material being used.

KEYWORDS
3D forming, fibre-based materials, paperboard, thermoformability, thermoforming

INTRODUCTION
Increasing awareness of the importance of environmental sustainability has led to widespread recognition of the need to reduce global use of plastic, which is a non-biodegradable, oil-based product, and invest in sustainable alternatives. Fibre-based composites are known as a possible alternative to plastic; while retaining the beneficial properties of plastic, they contain a high proportion of biodegradable fibrous material, resulting in a lower climate impact. As fibre-based products begin to replace plastic ones, improvements are needed in the area of conversion of these materials into versatile shapes. Therefore, 3D forming of fibre-based materials has been a topic of increasing research...
interest over the past few years. Despite extensive work, fundamental challenges still exist owing to the poor formability and stretchability of current fibre-based materials, which are poorly able to withstand plastic deformation without incurring damage.\(^1\) Research has thus focused on the development and refinement of materials and manufacturing processes to resolve limitations to the achievable depth and geometrical shape of the final products.

Material development has examined potential improvements in the formability of fibre-based materials via mechanical or chemical modification of the fibres,\(^2\) the addition of additives such as gelatin and agar to the fibres,\(^3\) modification of the coatings, and the use of lubricants.\(^5\) Strategies to optimize and enhance manufacturing methods for fibre-based formable materials have also been the subject of much research attention. Hydroforming, press forming, deep drawing, hot pressing and thermoforming are known methods for 3D forming of fibre-based materials. Nevertheless, the focus of research has been primarily on the first three methods.

In hydroforming, the material is formed in a female mould using a rubber membrane; however, the limited extensibility of paperboard material leads to low forming ratios (height to base diameter) in this process. Until recently, the method was generally only used at laboratory scale, and the focus has predominantly been on the production of defectless double-curved products through the use of extensible materials and optimization of the hydroforming process itself.\(^6\)–\(^8\) The second method, press forming, can produce deeper products than hydroforming. In press forming, the material is drawn into the female mould using a male mould. At the end of the movement of the male mould, compression force is applied, pressing the material in the clearance between the moulds. Wrinkling, dimensional inaccuracy and fracture are common defects in this process, and mitigation efforts have focused on selection of appropriate process parameters and improved design of the heating system and creasing pattern.\(^9\)–\(^13\) The third method, deep drawing, differs from press moulding only in relation to its immediate compression, which results in more uniform wrinkling distribution, and as such the patterning requirements are eliminated.\(^14\)–\(^15\) Furthermore, by using materials that exhibit enhanced compressive deformation, deeper products can be produced than by press forming.\(^16\)

Thermoforming, which is the focus of this research, is a forming method in which the material is heated to its softening point, after which it is stretched and induced to reproduce the mould shape by employing pressure difference or mechanical forces. The thermoforming process was primarily developed for plastics, and it is extensively used in the plastics industry, where its high production speed, low pressure and temperature requirements for forming customized products make it a cost-effective and popular manufacturing method.\(^17\) In the area of the production of fibre-based products, however, thermoforming has received little research attention. The lack of work can partly be attributed to the nature of the process, which largely matches the properties of plastic materials, and as noted by Östlund,\(^18\) the challenges posed by the porous structure of fibre-based materials. Thermoforming of fibre-based materials has thus far been researched for relatively flat moulded part products.\(^19\) Some work, also, has been done in the area of thermoforming of fibre-based composites in mixed structures, such as wood polymer composites (WPC), where the focus has been on thermoformability investigations using finite element methods.\(^20\)–\(^21\)

To date, thermoforming of plastic-coated fibre-based materials has been relatively understudied. Although the plastic coating allows these materials to be thermoformed, thermoforming of plastic-coated fibre-based materials faces certain limitations. Therefore, the primary objective of this research work is to gain a deeper insight into the thermoforming limitations of plastic-coated fibre-based materials by comparing the results for such materials with those for full plastic materials. Additionally, the effects of material properties and three key process parameters, that is, forming temperature, forming time and forming pressure, on forming results are explored. The knowledge gained can provide a foundation for future research investigating process and product development.

### 2 | EXPERIMENTAL

#### 2.1 | Materials

Two types of materials were used in this research: multilayer plastic and plastic-coated packaging paperboard. APET/EVOH/PE multilayer plastic was selected as a reference material due to its excellent thermoformability, which allows the authors to fully demonstrate the thermoformability limitations of coated fibre-based materials. The paperboards selected for study were commercial polyethylene (PE)- and polyethylene terephthalate (PET)-coated boards from various suppliers to enable analysis of their thermoformability differences. Plastic material, as named Ref in Table 1, is AMB Packaging (San Daniele del Friuli, Italy) AMBAR Rigid 38/6BA5 TS; Paperboard 1 is Flextrus AB (Lund, Sweden) Paperlite 300/65 HB; Paperboard 2 is Stora Enso (Imatra, Finland) Duplex 248+47EB41; Paperboard 3 is Stora Enso (Imatra, Finland) Prime BARR 248+43EB31; and Paperboard 4 is Stora Enso (Imatra, Finland) Trayforma Performance 350 + 40 WPET. Table 1 presents the measured properties of the materials used. In addition, Figure 1 presents scanning electron microscope (SEM) images acquired on cross sections of each material to highlight their layer structure and their differences.

All the materials were stored at constant humidity chamber of 80% relative humidity (RH) for the experiments and the measurements were made at 23 °C ± 1 °C and constant humidity of 50% ± 2% RH. Grammage measurement was performed according to ISO 536 standard using the analytical balance (Kern & Sohn, Balingen, Germany). The total grammage of the materials was measured, and the grammage of the paperboard layer was calculated based on the known plastic grammage. Thickness measurement was carried out according to ISO 534 standard using the digital micrometre (Messmer Büchel model 49-56, Veenendaal, Netherlands). Tensile tests were performed using the Shimadzu tensile tester (Shimadzu Corporation, Jiangsu, China) for plastic and L&W tensile tester...
The moisture content of the paperboard materials was measured using the moisture analyser (Adams Equipment PMB 53, New York, USA). Upon putting 3 g of the material into the moisture analyser, the material is gradually heated until the moisture is completely evaporated. The moisture content in a material is determined by the difference between its initial and final weight. Paperboard 1 consists of one layer of paperboard sandwiched between two plastic layers. In order to ensure that moisture content on that layer is equally accounted for in measurements, the material is delaminated from the sandwiched paperboard layer (Figure 2), allowing moisture to fully evaporate during measurements from both paperboard layers.

| Materials      | Plastic (Ref) | Paperboard 1 | Paperboard 2 | Paperboard 3 | Paperboard 4 |
|----------------|---------------|--------------|--------------|--------------|--------------|
| Composition    | PE/EVOH/APET  | PE/Paperboard| PE/Paperboard| PE/Paperboard| PET/Paperboard|
| Thickness (μm) | 285           | 437          | 429          | 369          | 474          |
| Total grammage of material (g/m²) | 363          | 412          | 310.5        | 301.5        | 395          |
| Grammage of paperboard (g/m²)     | N/A           | 357          | 280.5        | 271.5        | 355          |
| Grammage of plastic (g/m²)        | 363           | 65 (coating layer 60 + internal layer 5) | 30          | 30          | 40          |
| Moisture content (% M)            | N/A           | 5.9          | 7.5          | 7.9          | 6.3          |
| Tensile strength MD (kN/m)        | 15            | 24.2         | 23.1         | 23.8         | 30           |
| Tensile strength CD (kN/m)        | 15.5          | 14.7         | 15.4         | 11.1         | 14.2         |
| Strain at break MD (%)            | 4.5           | 16           | 2.1          | 2.6          | 2.3          |
| Strain at break CD (%)            | 4.8           | 10.9         | 6            | 7            | 5.5          |

FIGURE 1 Cross-sectional SEM images of the studied materials (material of the samples: Ref—plastic; P1–P4—paperboard) (The same scale is applied to all figures)

FIGURE 2 Delamination of Paperboard 1 for moisture content measurement

(Lorentzen & Wettre, Kista, Sweden) for paperboards in accordance with ISO 1924-3 standard.
2.2 | Thermoforming process

A VARIOVAC Primus (Zarrentin, Germany) thermoforming line (Figure 3) was used to perform the experiments. A 422-mm-wide roll of material was fed into the machine and clamped in the machine’s frame. It was then heated from the bottom until it softened. Next, the material was immediately transferred to the forming station within approximately 1 s. It was subsequently placed over the female mould (Figure 4) with top dimensions of 236 mm x 105 mm and adjustable depth up to 30 mm, where it was formed by applying pressure difference. The length of the mould (236 mm) aligns with the machine direction (MD) of materials and the width (105 mm) aligns with the cross-direction (CD) of materials. The formed part was then transferred to the cutting section of the machine and punched out of the system.

The thermoformability of the materials was examined using variation of forming parameters and two thermoforming processes: vacuum thermoforming and a combination of vacuum and air-pressure thermoforming (Figure 5). During forming, the pressure was controlled directly using an air-pressure valve. A pressure sensor was used to indicate the actual value of the forming pressure in the forming chamber. Various different mould depths were used to assess each material’s capabilities in terms of achieving the shape depth. The range of process parameters investigated in this study is presented in Table 2.

The forming parameters were chosen on the basis of the results of preliminary experiments and the limitations of the machine, and the materials being formed. It should be noted that since studying the effects of four parameters at three levels by examining the interaction between them is time-consuming and costly when five different materials are used, this study concentrates on testing the effects of each parameter individually.

2.3 | Thermoformability analysis

Several factors must be taken into account when evaluating the formability of different materials, and no consensus exists regarding the right criteria for assessing formability. In this research, shape accuracy, maximum depth acquired, wall thickness distribution and damage mechanisms are considered indicators of material formability. Prior to the measurements, all the samples were conditioned at 23°C ± 1°C and constant humidity of 50% ± 2% RH. To ensure the inclusion of postdeformations and springbacks into the analysis, measurements were performed a few days after the experiments. For each set of parameters, measurements were repeated for six identical samples and average values were used to derive the results.

The shape accuracy was first assessed by visual inspection, and the samples were then analysed with the 3D measurement system (Keyence VR-3200, Osaka, Japan). This analysis was done to gain a better understanding of shape accuracy differences between the various materials and to provide quantitative values for this quality factor. In the evaluation, the defined area shown in Figure 6 was chosen and a 3D model generated. The CD profile was used for further measurement. Next, the profile depth and the corner angle derived from the side lines of the profile were measured; these amounts were, respectively, 12 mm and 60° in the mould design. In order to perform this 3D measurement analysis, the formed plastic samples were painted with a zinc colour to make them clearly visible to the measurement system and to overcome issues caused by reflections. Moreover, due to limitations of the system in the depth of analysis, the plastic sample was analysed in two steps, and the results were merged together.

The forming depth of all the samples was measured using a handheld digital calliper. The depth of the samples was measured from the edge of the tray to the midpoint of the flat bottom of each tray. Due to the shape distortion and curling experienced by some of the samples following the forming process, a metallic fixture was used to affix the tray edges on a table and to ensure straight placement of all samples and accurate depth measurement. The thickness distribution of the samples was measured using the digital micrometre (Messmer...
Büchel model 49-56, Veenendaal, Netherlands). Thickness measurements were taken at seven points over the CD midplane cross section of the samples, as shown in Figure 7. According to the original design of trays, Points 1 and 7 are located at the edges of the tray, Points 2 and 6 are positioned in the middle of the sidewalls, Points 3 and 5 are located at the bottom radiuses and Point 4 is located in the centre of the base of the tray.

Furthermore, damage mechanisms that can further hinder the forming of materials or restricts their use were identified through visual inspection. In this context, the plastic coating layer of the material was also examined. Addition of plastic coatings to paperboard has the primary function of enhancing the barrier properties of the material. Hence, this study also examines whether thermoforming has an adverse effect on the coating layer of the studied paperboards and its barrier performance. Using the dye penetration test, it was determined if thermoforming causes the formation of pinholes in coating layer and deteriorates its barrier properties. The experiment was conducted in which a colouring solution was applied to the coating side of the thermoformed tray, and the other side of the tray was inspected for penetration. The tests were conducted according to EN standard 13676 (2001) using a colouring solution consists of 0.5 gram of dyestuff E131 blue dissolved in 100 ml of ethanol (96%). Moreover, the SEM imaging was also used to investigate the changes in the coating layer of the materials after thermoforming. Prior to forming, materials were imaged from their coating side; later, thermoforming trays were also imaged from their coating side.

### 2.4 Optical analysis

The optical analysis carried out throughout this study was performed using the SEM Hitachi SU3500 (Tokyo, Japan), equipped with a tungsten filament. The plastic coating side of the materials was imaged using secondary electron imaging (SEI) mode at an accelerating voltage of 10 kV and a working distance of approximately 5 mm. To acquire these images, the samples were sputter-coated (Au/Pd target) using the Edwards Scancoat Six Sputter Coater. For the cross-sectional imaging of the samples, backscatter electron imaging was used in compositional mode (BSE-comp). The microscope was set to variable pressure mode with 15 Pa for BSE-comp settings. The working distance and accelerating voltage for this set of measurements were at 5 mm and 15 kV, respectively.

### 3 RESULTS AND DISCUSSION

#### 3.1 Thermoformability analysis

#### 3.1.1 Shape accuracy

Experiments showed that the plastic material can be fully formed, using appropriate process parameters, to complete shapes with no limitations. Furthermore, it was possible to form the plastic material by vacuum pressure alone, that is, without applying additional pressure, in as little as 0.5 s of forming time. However, the best forming results were achieved with 2 s of forming/heating. Paperboard materials, on the other hand, required a combination of vacuum and pressure forming, and they demonstrated clear limitations in replicating the desired shape, as illustrated in Figure 8. The best shape was achieved by Paperboard 1, followed by Paperboards 2–4, respectively. Indeed, the paperboard trays have a balloon shape rather than the demanded geometry. This result can be seen better in the 3D analysis.
of the samples presented in Figure 9, where the extent to which the paperboards have limitations in forming complex geometries can be clearly seen.

Figure 9A presents the result for the plastic material, and Figure 9B describes Paperboard 1 as an example. Comparison with the reference values in the mould (12 mm depth, 60° angle) shows that the plastic material can reproduce the mould geometry almost perfectly. The paperboard materials, however, achieved maximum depths ranging from 2.7 mm for Paperboard 1, 2.1 mm for Paperboards 2 and 3 and Paperboard 4 attained the lowest depth with only 1.3 mm. As for the angle of forming, Paperboard 1 showed the best result with 136°, while Paperboards 2 to 4 had values of 151°, 156° and 166°, respectively. These values were obtained for the samples formed with 110°C forming temperature, 2 s forming time and vacuum forming for plastic and 110°C forming temperature, 3 s forming time and vacuum plus 1 bar forming pressure for the paperboards. Unless otherwise specified, these are the forming conditions selected for comparison in the results.

3.1.2 | Forming depth

The plastic material could be easily formed into the complex shape of the mould. Increasing the depth of the mould to the maximum depth (30 mm) did not negatively affect the formability of the material. Increasing the mould depth with paperboard, however, does present challenges as the material might rupture. The best performance in this regard was achieved by Paperboard 2, where despite the temperature and time used (110°C forming temperature, 3 s forming time), no rupture occurred when it was moulded at a depth of 20 mm. At this depth, Materials 3 and 4 could be used without defect only up to 100°C and 90°C, respectively. From this perspective, Paperboard 1 was the weakest as it could only tolerate up to 18 mm mould with restriction in temperature of 100°C. However, looking at the initial moisture content of the materials (Table 1), Paperboard 1 had a lower moisture content than the other paperboards, which may explain why this material could not withstand higher moulding depths.

The 15 mm mould was the only moulding depth in which all paperboard materials could be used without any occurrence of rupture by changing the forming parameters. Figure 10 presents a comparison of the forming depth for the materials. Error bars indicate the standard deviation of the results found on measurements of six identical samples. Changing the forming parameters for the plastic material compromises the shape accuracy (e.g., in deep grooves), but the maximum forming depth is almost always achievable within the range of investigated forming conditions; therefore, only one bar in the figure is dedicated to the plastic material which shows the maximum depth achieved with this material. In line with the results for shape accuracy, Paperboard 1 achieved higher depth, while Paperboard 4 showed the
Weakest performance. The maximum depth achieved by each material for the range of process parameters investigated is given in Figure 10. It should be noted that the geometry of the mould used can have a significant effect on the maximum depth that can be achieved by the material; the maximum depth may thus vary when using another geometry.

Figure 10 also shows how the achievable depth changed when changing the process parameters. There is no straightforward evidence that increasing or decreasing the forming parameters has the same effect for all the materials. Depending on the material used, the effect of the forming parameters on the depth could vary. Consequently, experiments must be done separately for each material to find the optimal parameters for forming.

To find the extent at which each of the forming parameters affected the attainable depth for each material, statistical analysis with Minitab software was carried out (Figure 11). Based on multiple regression coefficient analysis, it can be concluded that increasing forming pressure is more likely to enable Paperboards 1, 3 and 4 to achieve greater depth. For Paperboard 2, however, the forming temperature affects the material more significantly. The higher effect of forming temperature on Paperboard 2 is further explained in Section 3.2, which discusses density effect on thermoformability of paperboard materials.

3.1.3 | Thickness distribution

It is important to investigate thickness distribution in thermoformed products. Generally, the stretching in the process reduces the material's thickness compared to its raw state, and if the process is not controlled properly, the material can become thin, which can significantly degrade the quality and strength of the formed part. Similarly, uniform distribution of thickness is essential for good quality products. Accordingly, in this study, thickness was measured over the cross-sectional midplane profile of the selected samples indicated in Figure 7.

As discussed in previous sections, paperboards exhibit weak forming, resulting in it showing a different profile than that shown in Figure 7. In particular, mapping Points 2, 3, 5, and 6 selected on the original design required more scrutiny. Figure 12 shows an example of mapping these points on a Paperboard 1 test piece. This was done by first identifying the point where the curved shape begins; it is where the flat bottom connects to the radius, and it is visually discernible. Next, an edge of a measurement spot with a diameter of 15 mm was placed on the point to measure the thickness of Points 3 and 5. On Points 2 and 6, the midpoint of the distance between the other edge of the measurement spot and the edge of the tray was detected. Points 1, 4, and 7 were measured the same way as outlined in the original design (Figure 7). Figure 13 presents the results of respective thickness measurement for the materials. The bars represent the standard deviation of the results from the measurements of six identical samples. As the studied materials are in different thickness ranges, and to better illustrate the thickness distribution differences, all the distributions have been combined into one diagram by assuming the thickness of each material before forming as zero.

The thickness distribution of the plastic material has a roughly symmetrical reduction in thickness moving from the top edge to the bottom because the material experienced more stretching as it became deeper. However, it is the bottom radiuses (Points 3 and 5) where the greatest thickness reduction occurred. Here, the material reached the full depth of the mould and then began to form the final shape, and the corners are the last points of the material that touched the mould, and therefore, they are the thinnest. The present result is consistent with earlier studies concerning the thermoforming of multi-layer plastics, where similar cross-sectional mould geometries were found.25,26 The distribution seen in this example represents what would be typically observed in a thermoforming process.

Two separate distributions, however, were noticed for the plastic-coated paperboards. With Paperboard 1, the same trend for thickness reduction as the plastic material occurred from the top edge to the side walls. However, in this case, the lowest thickness was achieved at Point 4 (bottom). This was because the material...
attempted to reach the depth, but the bottom radiuses were not formed, and therefore, Point 4 was the last location in contact with the mould. On the other hand, with Paperboards 2–4, an unexpected increase in the thickness was observed after forming the material. As shown in the diagram, a thickness accumulation is visible moving from the top edge to the side wall. Subsequently, from Points 2 and 6 to Point 4 (bottom), the materials in general showed a tendency toward decreasing thickness, but the bottom is still thicker than the edges.

The most likely explanation for the increase in the thickness for the Paperboards 2 to 4 can be attributed to a phenomenon known as out-of-plane auxetic behaviour of paper. When paper is stretched under tensile deformation (e.g., during thermoforming), thickening can occur with the stretching of the fibres, and this phenomenon has been termed auxetic behaviour. Fibre networks are characterized generally by curled fibres, while some fibres are found on top of each other and some below. Stretching a material causes the curled fibres to straighten, and with this change, the fibres on top of them are pushed up, resulting in the thickness increasing. In the example of this study, thickening occurred in Points 2 to 6; the reason is because the material starts to be stretched where the depth is changed as it is the case from Points 2 to 6. In addition, increasing in the thickness was greater on side walls compared to the bottom. This further suggest that rate of stretching is greater on side walls compared to the bottom. This could be attributed to the curved shape of side walls compared to the flat bottom found on paperboard samples (Figure 12). Other studies have also found an increase in the thickness of paper-type materials under tensile deformation. Verma et al. further demonstrated that the rate of this auxetic behaviour can vary depending on the structure of fibre networks and the method of preparing paper materials. Similarly, the rate of thickness increase between Paperboards 2 and 4 is different in the current study, as shown by Figure 12. The differing performance of Paperboard 1 could not be explained clearly, but its layer structure may partially account for its behaviour. In addition to the coating layer, Paperboard 1 has another plastic layer incorporated between layers of paperboards, which may cause the material to behave more like plastic. However, a further investigation regarding the mechanism behind this difference remains to be conducted.

Regarding the effect of the process parameters on the thickness distribution, the experiments indicated that in contrast to the forming depth, changing the forming parameters within the specified range had little effect on the thickness distribution. Statistical analysis of the results also showed that changes in the forming parameters had no statistically significant influence on the thickness. Hence, at the studied range of parameters, the elongation properties of the materials tend to dominate in thickness distribution of the coated paperboards and optimizing the process parameters has little influence.

3.2 Effect of material properties on thermoformability of paperboards

3.2.1 Tensile properties

Tensile properties can be considered one of the most important properties of materials for thermoforming. The formability of fibre-based materials is dependent on various mechanical properties, for example, elongation, compressive strain, compressive strength and substrate-to-metal friction, and the degree to which these properties cause changes in the formability varies widely depending on the forming equipment and forming parameters. Unlike press forming and deep drawing, thermoforming displays tensile deformation rather than compressive deformation; and therefore, elongation is the most critical of the above-mentioned properties. Basically, elongation is a measurement of extensibility, which is the degree to which the linear length of a material can be extended by elastic, viscoelastic or plastic deformation when external forces are present, and extensibility in tensile deformation is related to strain at break tensile properties.

Tensile tests indicated that Paperboard 1 has a sixfold higher strain at break value in the MD and a two-fold higher value in the CD than the other paperboards, which may partly explain its better shape conformance. Considering the forming angle in the deep grooves and the maximum achieved depth of the trays, correlation analysis indicates a strong relation between the CD strain at break properties of
the paperboards and their ability to form the mould shapes (Figure 14). According to the study by Östlund et al., the strain at break at CD is a dominant factor in the 3D formability of paper materials. The same trend is seen in the MD direction with correlation values of -0.86 and 0.80 for the forming angle and forming depth, respectively. Moreover, looking at the thickness distribution presented in Figure 13, and excluding the edges where no stretching occurred, it can be seen that the stretching of the materials is correlated with their strain at break properties. Based on the change in thickness, greater strain at break results in greater stretching ability. In Figure 14, this correlation can be seen in the CD direction, and correspondingly, the correlation value is 0.90 in the MD direction. It is nevertheless important to note, as demonstrated by the \( R^2 \) values, that the strain at break properties and formability factors do not follow an exact linear relationship because of the difference between uniaxial and 3D strain behaviour. However, these results suggest the higher strain at break properties are beneficial to improve thermoforming performance while the rate of change in thermoforming performance cannot be completely predicted using the uniaxial strain at break properties of the materials.

3.2.2 | Density

Thermoforming using roll-fed machines involves heating the material prior to forming. Thermoforming is generally used to form plastic materials, and this preheating step has the benefit of drying out moisture that has been absorbed by the plastic and is detrimental to its forming process. In fibre-based materials, however, the existing moisture has a crucial impact on the forming capability. The presence of moisture in paper material can make it softer and improve its formability as the greater its moisture content, the weaker the fibre bonds, and so the paper has a lower elastic modulus and tensile strength, making it easier to stretch and form. Consequently, to better understand the performance of paperboard materials in thermoforming, the changes in moisture content during the forming were investigated for the specified forming conditions (110°C forming/heating temperature, 3 s forming/heating time and vacuum plus 1 bar forming pressure). The moisture content of the material was measured immediately before forming from the material roll and directly after forming from the formed sample. The rate of change in moisture content of the materials was calculated accordingly and presented in Table 3.

Using the Fourier number, which is the ratio of heat conducted through a material to heat stored during the heating process, it can be possible to explain the rate of changes in the moisture content of the materials:

\[
\text{Fourier number} = \frac{\text{Thermal diffusivity of the material} \times \text{Heating time}}{\left(\text{Sheet thickness}\right)^2}. \quad (1)
\]

In the experiments in this study, the heating time was constant for all the materials, and the material thicknesses are fairly close; thus, the thermal diffusivity of the materials can be considered the determining factor. The thermal diffusivity of a material is an indicator of how rapidly its temperature changes when heated, and according to heat transfer laws, it is inversely related to the density of the material. Nevertheless, there has been disagreement regarding the dependency of thermal diffusivity on the density of paper materials. In contrast to the research conducted by Niskanen and Simula, which showed that the thermal diffusivity of paper material is inherently independent of its density, Morikawa and Hashimoto found an inverse relationship between these two variables. The reason for this is explained by the structure of paper materials, which are made up of cellulose and air; the higher density corresponds with fewer pore spaces and lower air content in the material. Air has a much higher thermal diffusivity than fibres, so the higher the density, the less air is present in the material, which translates to a lower thermal diffusivity. Accordingly, in this study as well, the density of paperboard layers is calculated to determine whether the discussed inverse relationship is valid when considering the changes in moisture content in materials.
To calculate the density of paperboards in the studied materials, it is necessary to know the thickness of the paperboard layer. As such, the cross-sectional SEM images of materials are used to determine the thickness of paperboard layers. The ratio between the thickness in the paperboard layer and the total thickness is determined by using the available length measurement tools for images (in this study with the help of Microsoft Visio). Accordingly, the paperboard layer’s thickness is calculated based on the measured total thickness of material, and its density is calculated by dividing the grammage of paperboard (shown in Table 1) by its thickness. As an example, Figure 15 demonstrates how the thickness of the paperboard layer of Paperboard 2 was determined. This method may not be able to provide an exact thickness of the layer due to variations in thickness, but it can provide a fair indicative value.

To assess the effect of density on heating of the material and the potential relationship with the change in moisture content, Paperboard 1 has been excluded from the study. This is due to the differing layer structure of this material in comparison to the other three paperboards as seen in Figure 1. Unlike the other three paperboards, this material has an internal plastic layer between two paperboard layers, resulting in a different mechanism of distributing heat. Moreover, to compare this material with other materials, density of each paperboard layer is needed for the calculation, which requires information regarding the grammage of each layer and that information is not available in this study. Thus, to provide a valid comparison, the relationship between density and moisture content change is examined between the three other coated paperboards.

The results of this study show that there is a strong correlation between the moisture content changes in material during thermoforming and their density (Figure 16). Therefore, when comparing all the materials for the same heating time, Paperboard 2 with the lowest density had the highest thermal diffusivity and rate of heat conduction, as well as the highest temperature increase, leading to a higher change in its moisture content. These results suggest that using denser paperboard materials can aid in moisture retention during the thermoforming process.

Conversely, however, when it is claimed that increasing density leads to better moisture retention in the material due to lower temperature rise, it might be argued that at the same time higher temperatures are needed to enable the material to be formed better. In this study, there is no information pertaining to the difference between the set heating temperature and the actual material temperature. Nevertheless, it can be assumed that with same heating time, the denser paperboard had lower actual temperature or that there was an uneven distribution of temperature in the material, which would account for the surfaces of the material reaching the required temperature, while the cores of the material did not reach the same temperature. Interestingly, this assumption is in accordance with the effect of forming temperature on the depth of trays. From Figure 11, it is apparent that the less dense material is more affected by forming

| Materials | Paperboard 1 | Paperboard 2 | Paperboard 3 | Paperboard 4 |
|-----------|-------------|-------------|-------------|-------------|
| Moisture content changes (% M) | 1.69 | 3.50 | 2.65 | 2.78 |
| Paperboard layer density (g/cm³) | N/A | 0.703 | 0.805 | 0.773 |

**TABLE 3** Moisture content changes and density of paperboard layers

**FIGURE 15** The method used to find the thickness of the paperboard layer

**FIGURE 16** Correlation between moisture content changes and density of paperboard layers
temperature than the denser material, which supports the hypothesis that for same heating time, the denser material did not experience a sufficiently great temperature increase that it could have a more significant effect on the forming performance. Accordingly, for denser materials, precision control of the heating stage and temperature distribution is of more importance since non-uniform heating directly affects the quality of the thermoformed parts.

As stated earlier, this study was unable to determine the relationship between density and moisture content change in Paperboard 1. However, the moisture content measurements show a lower change in this material compared to the others, which can be attributed to the layer structure in this material. A paperboard layer in this material is sandwiched between two plastic layers, and this protection may account for the lower change in moisture content for this layer and the lower average value for the entire material. The relatively smaller change in moisture content experienced by this material may be a reason it could achieve better forming, in addition to its better elongation properties. However, it is unclear to what extent this better moisture retention boosts formability due to the simultaneous effects of the forming parameters.

3.3 | Damage mechanisms

The most common defect observed in the thermoforming of the paperboard materials was rupture. The mode of rupture in the thermoforming experiment is shown in Figure 17, where the fracture occurs along the longitudinal MD. Defects such as rupture are caused by stress levels exceeding the strength of the material. The bonds within the fibres fail first, followed by the fibres themselves failing. In the thermoforming process, tensile deformation can also result in tensile failure and fracture formation. Rupture occurrence depends on both the material properties and process parameters. In this present study, rupture was most frequently caused by an increase in the mould depth. The moisture content of the material was also observed to be a very significant factor as low moisture content resulted in earlier rupture as the depth increased. Yet, in some cases, a drastic increase in forming temperatures could also cause rupture.

Blistering was another observed defect. Blistering can be caused by excessive heating of the material, as well as excessive moisture content where the water vapour cannot leave the uncoated side of the paperboard, which results in internal delamination and blistering. Blistering can be prevented by selecting appropriate processing parameters and applying a barrier coating with high adhesion to the paper. All studied paperboards showed blistering when formed at high temperatures, although these temperatures ranged according to the material. An example of this defect can be seen in Figure 17, which depicts blistering on Paperboard 1 after the forming temperature was increased to 140°C.

Curling was a defect that occurred during storage. Curl, as defined by Salmen, is a specific type of dimensional instability which occurs when the paper material is deformed over its elastic limit. According to Niini et al., this phenomenon has been also seen in press-formed products, and it is primarily due to hygroexpansion, stress relaxation and creep of materials while absorbing or desorbing moisture in different humidity conditions. In this study, in spite of having stored all the samples in the same condition, samples made of Paperboard 1 exhibited curling during storage. The higher plastic content of this material and its considerably better elongation properties make the material less stiff and more flexible, which is very useful in terms of forming but at the same time can cause curling and instability of the formed product during storage. Further investigation is

![Figure 17](image1.png) Rupture, blistering and curling in the thermoformed plastic-coated paperboard samples

![Figure 18](image2.png) SEM images of coating layer for Paperboard 3 before and after the thermoforming process
required to ascertain the ideal storing conditions to avoid curling in such fibre-plastic materials.

The dye penetration test was conducted on samples obtained under different forming conditions to analyse the potential damage to the plastic coating layer of the studied paperboards. The results showed that the thermoformed trays studied did not leak regardless of their type of coating (PE or PET) and the conditions under which they were formed. This can generally verify that the thermoforming process does not cause pinholes to appear in the coating layer of the materials. In addition, SEM imaging of thermoformed trays showed no surface defects after forming. As an example of this, Figure 18 illustrates SEM images of the coating side of Paperboard 3 before and after forming. Images also depict a smoother coating layer after the forming, which itself could be beneficial in possible subsequent sealing processes of thermoformed trays. Overall, this study found no evidence of pinholes or surface defects in the plastic coating of the thermoformed trays studied. Nevertheless, a more detailed investigation of the barrier properties of thermoformed trays remains to be conducted.

4 | CONCLUSIONS

This study evaluated the thermoformability of plastic-coated paperboards based on their ability to replicate mould shapes, the maximum achievable depth, thickness distribution behaviour, and damage mechanism. Further, the effect of material properties and process parameters on formability was discussed. A plastic material with excellent thermoformability was used as a reference to better visualize the limitations of the coated paperboards.

The thermoforming of paperboard is greatly restricted by the nature of the material, which contributes to its low stretchability. Plastic coating provides a certain thermoforming window to produce 3D products from these materials without causing structural damage. However, in comparison to fully plastic material, the thermoformability of plastic-coated paperboards is limited to simple geometric forms and shallow depths. Indeed, thermoforming of paperboard yields low-depth balloon-shaped products rather than the more complex geometrical shapes such as patterned-shapes and deep grooves that are possible when using plastic. In addition, although thermoforming tackles the issue of possible thickness reduction and thinning that is found in plastic materials, coated paperboards can undergo a reverse trend where there is an increase in thickness. Thickening is attributed to the out-of-plane auxetic behaviour of paperboard materials. Furthermore, damage mechanisms such as rupture and blistering pose further limitations to the thermoforming of paperboards, while curling disrupts post-usage of products. On the other hand, the plastic coating of the thermoformed trays tested showed no signs of pinholes or surface imperfections.

Empirical study and subsequent statistical analysis revealed that careful selection of the plastic-coated paperboard material is required to achieve acceptable thermoforming performance, with attention to be given to the tensile elongation and density properties. The higher the strain at break of the material, the deeper its attainable forming depth, and the better its stretch and shape conformity. Additionally, the higher density of paperboard results in less thermal diffusivity and a lower rate of heat conduction. Consequently, paperboard of higher density can experience lower moisture content changes for the same heating time than less dense paperboard. Correspondingly, however, it requires longer heating time to reach the required forming temperature.

The results of this study show that no simple change, increase or decrease, in the forming parameters has the same effect on all materials, and the degree to which parameter changes affect the materials varies. Therefore, the process conditions and tooling require specific adjustment to the material used to provide good thermoforming performance and yield defect-free products of high quality.

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CONFLICT OF INTEREST

The authors declare that there is no conflict of interest.

DATA AVAILABILITY STATEMENT

Data are openly available in a public repository that does not issue DOIs.

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