Effect of Density on In-plane Material Behavior: The Case of Laboratory Paper and Commercial Paperboard

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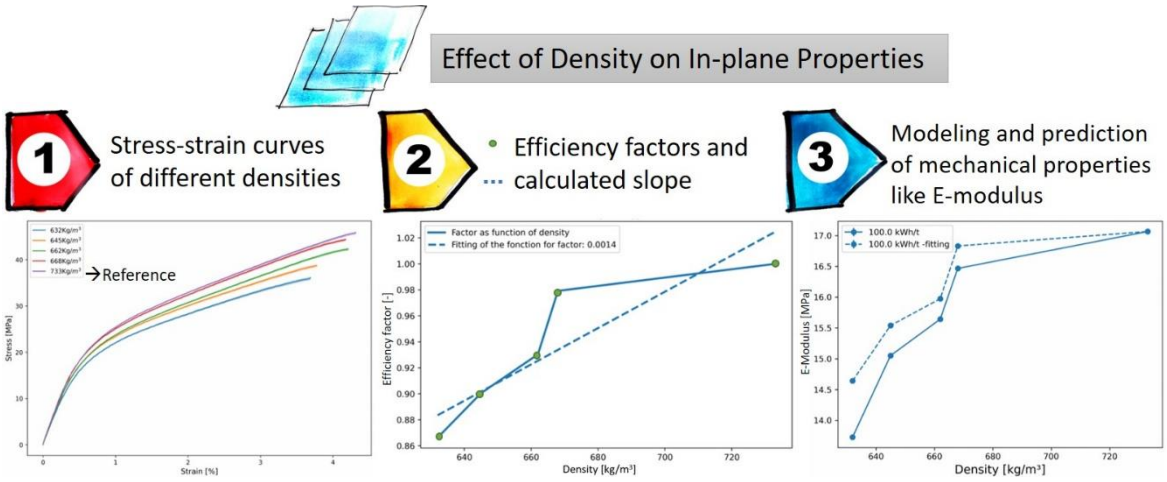
GRAPHICAL ABSTRACT



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Paper and paperboard are highly regarded for their recyclability and sustainability, but their inherent inhomogeneity presents challenges for material characterization and modeling. Despite being pressed during production, they remain compressible in the thickness direction, making density a key factor in determining mechanical properties. This study examines the effect of density and thickness compression on the in-plane mechanical behavior of paper and paperboard through uniaxial tensile tests on both laboratory paper with different refining energies and commercial paperboard with anisotropy. The results confirm that density significantly affects stress-strain response, elasticity, and plastic deformation. To capture this effect systematically, an efficiency factor is introduced that provides a quantitative measure of the density-dependent mechanical behavior to model the influence of density using a linear function. Incorporating efficiency factors refines the material modeling approach and improves predictions of stiffness and plastic stress. Higher refining energies result in a more homogeneous structure, reducing density-related variations, while commercial paperboard is less affected by fiber orientation and surface coatings. The proposed efficiency factor provides a new framework for optimizing and modelling the influence of the pressure and density on material parameters of fiber-based materials.

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Keywords: Density; Efficiency factor; Pressing; Compressibility; Refining; Paper; Paperboard

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INTRODUCTION

Paper and paperboard are widely used in packaging, logistics, and even construction due to their superior recyclability and sustainability compared to plastics. However, their inherent inhomogeneity – resulting from variations in fiber orientation, compositional distribution, and filler content – poses significant challenges for accurate material characterization, modeling, and process optimization.

The in-plane mechanical properties of the fiber-based materials paper and paperboard play a critical role in the performance of several 3D forming processes such as deep drawing, press forming, and hydroforming (Linvill and Östlund 2016; Östlund 2017). To more accurately characterize the in-plane mechanical properties of paper and paperboard, the effect of density and compressibility of paper must be considered. In 3D forming, such as deep drawing, the sample is subjected to pressure in the thickness

direction as it is compressed between the blank holder and die and as it flows into the die (Vishtal and Retulainen 2012). Especially when wrinkles occur, there is a large compressive force between the punch and die due to the accumulation of material. This is likely to affect both in-plane and out-of-plane material behavior and failure criteria, which is also important in simulating forming processes.

As can be seen from the definition of grammage, density is the value of grammage divided by thickness. The relationship between the grammage and thickness was investigated by experimental measurements of laboratory papers under different configurations. A correlation was found between the variance of local density and the variance of grammage (or "basis weight"), and it was sensitive to the mean grammage (Dodson *et al*. 2000). A model for the non-linear relationship between paper thickness and grammage was presented using routine grammage and thickness measurements combined with X-ray micro-chromatographic analysis (Bloch *et al*. 2019).

Before describing the mechanical properties of paper, it is necessary to introduce some important concepts (refining, wet pressing, rolling or calendering) in the papermaking process. Refining is a mechanical treatment of a pulp suspension to achieve properties suitable for papermaking. Refining makes the fibers flexible, which leads to more fiber-fiber bond development in the paper and improves tensile strength. Refining helps to straighten free fiber segments in the paper to some extent and improves papermaking (Annergren and Hagen 2009). However, refining also increases paper density, slows the drainage of water during sheet formation, and can increase residual stresses in the resulting paper. This is because refining makes the fibers more pliable and tighter together, reducing bending stiffness. Secondly, pulping increases fiber swelling and water retention, making web dewatering more difficult, reducing paper production efficiency, and increasing energy costs (Gimåker *et al.* 2011). Wet pressing of wet paper sheets and calendering or rolling of dry paper sheets are concepts that are easily confused in papermaking. During wet pressing, the paper passes through the press rolls, removing excess moisture and bringing the fibers closer together. A greater amount of inter-fiber hydrogen bonds are formed between the fibers in the course of subsequent drying of the sheet. As the moisture content decreases under pressure, the fibers come into closer contact and form more fiber bonds, improving the overall strength, flexibility, and durability of the paper (Paulapuro 2001). Calendering, or rolling, of dry paper sheets primarily affects the surface characteristics of the paper by passing the dry sheet through a series of rolls that apply high pressure to compress the fibers and reduce surface roughness (Litvinov and Farnood 2006). Since the paper is already dry, the pressure applied achieves densification primarily by reducing the air gaps between the fibers without creating new interfiber bonds.

Several studies have been conducted to investigate the effect of density on paper materials. The Z-direction compression characteristics of paper were measured using the micro-indentation technique, and the relationship between the apparent density and the average compression Young's modulus was investigated in (Pawlak and Keller 2004). The results showed that the correlation between local apparent density and local Young's modulus is not significant. Subsequently, a significant correlation was found between the surface roughness, local structure of the sheet, and the local Young's modulus. Girlanda and associates (Girlanda and Fellers 2007; Girlanda *et al.* 2012) evaluated the influence of density on the out-of-plane mechanical properties of paperboard. Their results indicate that paper properties are highly dependent on density. Specifically, they observed that higher density resulted in an exponential increase in Young's modulus, Z-direction strength, and a reduction in strain at break in the thickness direction.

Compressive stiffness was also studied at different temperatures and moisture contents. The effect of density on the in-plane properties has also been studied separately, since the in-plane and out-of-plane properties of paper materials are usually assumed to be decoupled. Henriksson *et al.* (2008) found that the Young's modulus of cellulose nano-paper increased with density, although the correlation was not particularly strong. Chapter 10 of (Niskanen 2012) also reviewed the influence of density on micromechanics. It was proposed that for a given paper grade at a given basis weight, density and Young's modulus depend on refining, wet pressing, and mixing different pulp components (Niskanen 2008). More specifically, the relationship between Young's modulus, yield strength, elongation at break, tensile strength, and density was investigated under various refining and wet pressing conditions. There are also studies on similar fiber materials, for example, the effect of density on the physical and mechanical properties of bamboo fiber sheet composites are evaluated. The results showed that as the density increases, the water absorption decreases, the thickness expansion first increases and then decreases, and the Young's modulus and shear strength under vertical and parallel loading increase (Zhu and Yu 2010).

In order to more easily quantify the effect of parameters such as density on the stress-strain curve, the concept of an "efficiency factor" has been proposed so that the stress-strain curves of different paper densities collapse to a single master curve when scaled by density. Seth and Page (1981) extended the efficiency factor parameter to the plastic mechanism to study the effect of various papermaking treatments on the stress-strain response of paper. They concluded that the nonlinear behavior of the paper stress-strain curve is mainly due to the properties of the fiber composition rather than the paper structure. Using the proposed efficiency factor, which is the ratio of the initial Young's modulus to the current Young's modulus for each curve, the corresponding experimental results for wet pressing and refining to change the number of bonds were compared (Borodulina *et al.* 2012). The experimental deviations indicate that fiber properties can be altered by refining in physical experiments. The concept of efficiency factor ground was also extended to characterize the changes observed in the tensile response of paper subjected to a previous strain in (Coffin 2012). By incorporating the efficiency factor ground into the constitutive equation and tracking the change in efficiency factor with strain, it is possible to account for the loss of compliance observed over the entire range of recoverable deformation.

The compressibility of paperboard in the thickness direction highlights the important effect of density on out-of-plane and in-plane properties. To predict and improve the performance of fiber-based materials, the effect of density should be understood and incorporated into the material modeling. However, traditional modeling approaches often fail to capture the subtle relationship between density variations and mechanical behavior, especially with respect to fracture resistance (Sanjon *et al.* 2024). In the authors' previous study (Leng *et al.* 2024), a numerical sensitivity study on the effects of three local structural distributions, *i.e.*, thickness, fiber orientation, and density, on the mechanical properties, including stress-strain curves, tensile strength, and strain at fracture, was conducted. The simulation results show that the density variable has the least effect on the mechanical properties, which differ very little from those of a homogeneous material. As mentioned in the paper, in the numerical simulation of uniaxial tensile tests, the density accounts for only a fraction of the gravity of the specimen, and thus the gravity bias has less effect on the mechanical properties compared to the homogeneous material model. However, this observation contrasts with experimental results where the strain distribution measured

using Digital Image Correlation (DIC) (Considine *et al.* 2005) of the specimens shows a correlation between regions of increased local strain and reduced basis weight. Hagman and Nygårds (2017) compared the formation and strain patterns of the paper and found that formation was the cause of the inhomogeneous deformations in the paper. More specifically, the deformation occurred in areas of lower density, which can be identified from the formation images, and then contributed to the development of the inhomogeneous deformation patterns. Krasnoshlyk *et al.* (2018) experimentally and numerically analyzed the fracture process of two fibrous web materials with different densities. The results show that high-density paper is able to localize sustained fracture to very small defects, whereas low-density paper requires rather large defects. It follows that the fracture process of paper and similar fibrous web materials is controlled by these rather large regions of low mass density. Stochastic simulations combining thickness, density, and fiber orientation variables of the fiber material show that the spatial variation of density has the largest effect on the local strain field, followed by thickness and fiber orientation (Alzweighi *et al.* 2021). However, the simulations in this work are in multiscale, *i.e.*, fiber, network, and sheet dimensions, so the modeling and computation of the model are more complex.

It is well documented that density is related to the in-plane mechanical properties of paper and paperboard, but a quantitative comparison and analysis, including pressing during processing and fiber orientation of commercial paperboard with high anisotropy, is still lacking. In addition, density variables have been shown to correlate well with fracture of the fiber-based material, and a representation of the effect of density is needed in numerical analyses. The purpose of this study is to investigate the influence of density and thickness compression on the in-plane mechanical behavior of paper and paperboard. To this end, the mechanical properties are investigated using uniaxial tensile tests on both laboratory-produced paper with varying degrees of refinement and densities, and commercial paperboard with controlled density and through-thickness compression. The results confirm that density has a significant effect on key material properties, including stress-strain response, elasticity, and plastic deformation. The differences in the force-displacement curves and stress-strain curves of the specimens before and after pressing are also evaluated. To systematically capture this dependence, an efficiency factor is introduced that provides a quantitative measure of how density variations affect mechanical behavior for the specific effects of refining, material, and fiber orientation

MATERIALS AND TEST METHODS

Laboratory Paper Production

Since the focus of this study is on the effect of density due to the inhomogeneity of the fiber-based material, Northern bleached softwood kraft (NBSK) pulp was used in the production of the laboratory paper, and the production parameters were kept consistent. In contrast to previous research, this study focused on a narrower range of densities under a specific refining energy level, with special emphasis on the inhomogeneity of the material. Therefore, laboratory paper produced in-house with four different refining energies was used to investigate the correlation between density and in-plane mechanical properties. The correlation between different densities and refining energies can facilitate a more comprehensive understanding of the material behavior.

The material under study is produced using highly versatile chemical pulp NBSK without the addition of any additives. This is a type of long fiber that is commonly used as a reinforcing pulp (Nanko *et al.* 2005). Refining increases the specific surface area of each fiber, which subsequently improves interfiber adhesion and chemical absorption. Four different refining energy inputs of a laboratory refiner (Voith LR 40) were used for the experiments: unrefined, a low refining input of 100 kWh/t, a medium refining input of 280 kWh/t, and a high refining input of 500 kWh/t. All materials are produced on the Rapid-Köthen automatic sheet former in the wet laboratory of the Chair of Paper Technology and Mechanical Process Engineering at the Technical University of Darmstadt, Germany, as shown in Fig. 1 (a). After papermaking, all laboratory papers were placed in a climate-controlled room for at least 24 hours. In order to achieve a greater range of density differences, the paper is rolled after production and conditioning, as shown in Fig. 1 (b), except for the smallest grammage at each refining capacity. The clearance between the top and bottom rolls was set at 0.15 mm, and samples were manually passed between two rolls five times.

Fig. 1. (a) Rapid-Köthen Automatic Sheet Former; (b) Rolling machine at PMV, TU Darmstadt

However, the thickness reduction is limited due to the spring-back after rolling of the paper in the thickness direction. The thickness of the material was determined according to ISO 534 with a defined contact pressure of 100 kPa, while the basis weight was measured according to ISO 536 by weighing a sufficiently large piece of paper. Five different grammages with four different degrees of refinement were produced for characterization (see Table 1). Five samples are produced for each test point and the standard deviation (SD) of thickness, grammage, and density are also listed.

Table 1. Basic Information on Laboratory Papers (average of 5 samples)

Refining Energy input (kWh/t)	Thickness (SD) (mm)	Grammage (SD) (g/m^2)	Density (SD) (kg/m^3)
0	0.188 (0.004)	107 (2.321)	572 (9.107)
	0.210 (0.004)	129 (1.958)	615 (18.464)
	0.230 (0.004)	147 (0.722)	637 (8.779)
	0.229 (0.002)	154 (0.471)	675 (4.900)
	0.239 (0.004)	164 (1.256)	686 (9.843)
100	0.153 (0.003)	96 (0.954)	632 (4.679)
	0.181 (0.004)	117 (1.167)	645 (17.189)
	0.202 (0.003)	135 (1.302)	668 (10.817)
	0.228 (0.002)	150 (1.738)	662 (3.607)
	0.240 (0.001)	176 (3.508)	733 (14.616)
280	0.158 (0.003)	112 (1.093)	710 (7.522)
	0.185 (0.001)	134 (1.320)	723 (7.136)
	0.203 (0.002)	156 (1.354)	769 (10.906)
	0.209 (0.002)	174 (3.055)	832 (13.829)
	0.240 (0.006)	212 (5.209)	882 (16.676)
500	0.137 (0.002)	107 (2.139)	787 (26.911)
	0.171 (0.002)	133 (0.913)	775 (5.968)
	0.199 (0.002)	152 (1.841)	766 (8.614)
	0.225 (0.005)	179 (2.625)	794 (19.743)
	0.235 (0.001)	204 (2.198)	870 (9.355)

Table 2. Basic Information on Commercial Paperboards (initial → after pressing)

Paperboard	Thickness (mm)	Grammage (g/m^2)	Density (kg/m^3)
Mat. A	0.2	174	870
	0.24	186	775
	0.27	199	736
	0.285	216	757
	0.32	234	732
	0.36	261	726
	0.4	301	752
	0.44	333	758
Mat. B	0.245	194	792
	0.30	229	764
	0.41 → 0.36	205	745 → 848
	0.46	350	762
	0.54	386	714
	0.6	412	687
Mat. C	0.19	196	1034
	0.21	213	1015
	0.24	228	952
	0.245	239	975
	0.28	258	922
	0.31	276	890
	0.35	310	886
	0.4	334	836
	0.43	360	837
	0.47	396	843
Mat. D	0.29 → 0.26	250	862 → 962
Mat. E	0.34 → 0.31	310	912 → 1000

Commercial Paperboard

Three types of commercial paperboard with different grammages were used to investigate the influence of density on mechanical properties. Materials A and B are bleached virgin fiber boards with a three-layer fiber structure and a chemi-thermomechanical pulp (CTMP) layer in the middle layer. Material C is a coated solid bleached sulfate (SBS) board with a three-ply chemical pulp fiber structure. In addition to a specific grammage of Material B, this work examined two other types of commercial paperboard after pressing. Material D is a premium uncoated recycled paper made from 100% recycled fibers and is commonly used for printing purposes. Material E is manufactured from bleached kraft pulp and exhibits good extensibility and nearly isotropic properties, rendering it suitable for printing and processing with exceptionally deep embossing. The technical data are presented in Table 2. Specimens corresponding to one grammage each of materials B and materials D and E were pressed in the thickness direction using a compression plate with a force of 50 kN and a loading time of 30 seconds to investigate how such dry-pressing affects mechanical properties. Therefore, the measured thickness and calculated density before and after pressing are also included in the table.

Material Characterization Method

In contrast to commercially produced paper, laboratory paper produced from the Rapid-Köthen Automatic Sheet Former is isotropic, obviating the necessity for specimens to be cut in multiple directions for uniaxial tensile testing. The laboratory paper, originally 205 mm in diameter, was cut into strips with a dimension of 120*25 mm^2 tensile test specimens using a cutting plotter. Commercial paperboard was cut in three directions: machine direction (MD), 45°-direction, and cross-machine direction (CD). The Zwick/Roell Z100 material testing machine, equipped with a high-resolution video extensometer was used for the tensile test. The clamping length is 90 mm, while the deformation evaluation length is 70 mm. Due to the geometric limitations of compression plates, the specimen geometry of pressed samples was different from before and is 60*30 mm^2, so the compression pressure was about 28 MPa. Then the clamping length for the following tensile test was 20 mm, the evaluation length was 5 mm, and the tensile speed was 20 mm/min. The tensile testing was conducted in a room with a temperature of approximately 23 °C and a relative humidity of 42%. Four replications have been performed for each series of experiments.

Introduction of Efficiency Factor and Modeling the Effect of Density

The efficiency factor $f(\rho)$ is a dimensionless quantity introduced to quantify the influence of density variations on material properties. It serves as a scaling parameter that adjusts mechanical properties relative to a reference state, allowing for the systematic analysis of density-dependent behavior. The efficiency factor has been widely used in materials science to model variations in elastic and plastic properties as a function of density changes.

In the previous work (R. S. Seth and D. H. Page 1981), the efficiency factor was originally defined as,

$$f_i = \frac{E_i}{E_{ref}} \quad (1)$$

where E_i represents the elastic modulus of a sheet with low bonding, and E_{ref} is the elastic modulus of the well-bonded reference sheet with the highest density. This formulation allows for the comparative assessment of mechanical properties across materials with different bonding characteristics.

To establish a quantitative relationship between the efficiency factor and density, the following formulation is introduced,

$$f(\rho) = slope \times \frac{\rho}{\rho_{ref}} \quad (2)$$

where *slope* denotes the linear dependence of the efficiency factor on density, and ρ_{ref} represents the reference density. The slope in this equation characterizes the rate at which the efficiency factor varies as a function of density, allowing modeling of local density effects on material behavior.

Building upon this framework, the efficiency factor can also be expressed in terms of the stress-strain relationship,

$$f(\rho) = \frac{\sigma}{\sigma_{ref}} \quad (3)$$

where σ represents the strain-stress response at a given density ρ, and σ_{ref} is the reference strain-stress curve.

Using this efficiency factor, the elastic modulus, which describes the stiffness of the material, is formulated as,

$$E_{modulus}(\rho) = E_{modulus_{ref}} \times f(\rho) \quad (4)$$

where $E_{modulus}(\rho)$ is the elastic modulus at density ρ, $E_{modulus_{ref}}$ is the reference elastic modulus at ρ_{ref}, and $f(\rho)$ is the efficiency factor at density ρ. This formulation enables the estimation of elastic modulus for materials with varying densities based on the reference modulus and the density-dependent efficiency factor.

Beyond the elastic modulus, the plastic stress σ_p is also analyzed, as it describes the material's resistance to plastic deformation. Similar to the elastic modulus, the plastic stress at any given density ρ is related to the reference plastic stress through the efficiency factor as,

$$\sigma_p(\rho) = \sigma_{p_{ref}} \times f(\rho) \quad (5)$$

where $\sigma_p(\rho)$ is the plastic stress at density ρ, $\sigma_{p_{ref}}$ is the reference plastic stress at ρ_{ref}, and $f(\rho)$ is the efficiency factor at density ρ. This equation facilitates the modeling of plastic deformation behavior across different densities by scaling the reference plastic stress using the efficiency factor.

By employing these formulations, the impact of density variations on material stiffness and plastic deformation behavior can be systematically quantified, thus providing a comprehensive framework for characterizing density-dependent mechanical properties.

RESULTS AND DISCUSSION

Influence of Density of Laboratory Paper on Mechanical Properties

The stress-strain curves of laboratory paper with different densities but the same refining level are illustrated in Fig. 2. It is clearly visible that the refining level had a great influence on the mechanical properties of paper.

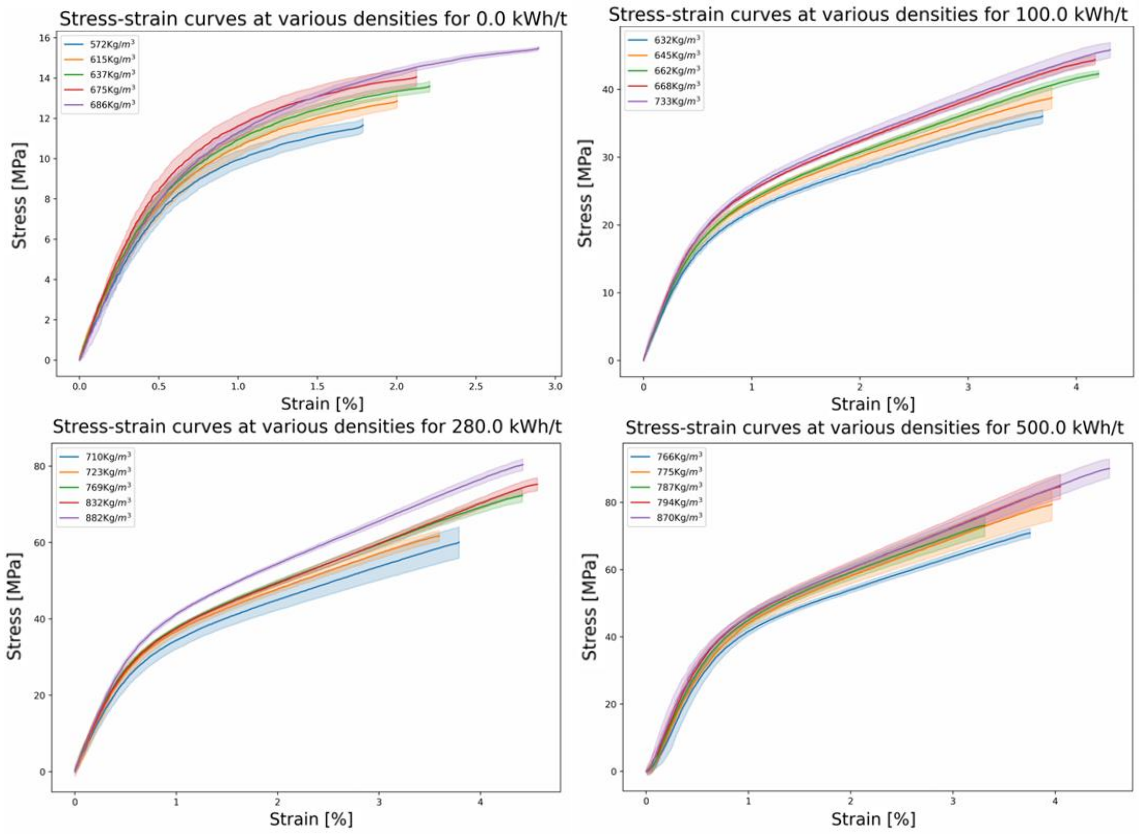


Fig. 2. Stress-strain curves at different densities without refining and with refining capacity 100, 280, 500 kWh/t

A comparison of elastic modulus is shown in Fig. 3 with solid lines, where the different densities were obtained using a rolling machine with specific rolling clearance. The effect of density on the elastic modulus shows a monotonically increasing trend at low refining energy; however, the trend was not obvious at high refining energy. Using the calculated efficiency factor, which will be discussed in detail in the later section, it is possible to characterize the effect of density variation on the elastic modulus in addition to the stress-strain curve. The dashed lines show the approximation of the elastic modulus using the efficiency factor to the highest density as a reference, for the elastic modulus at different densities. The overall description is satisfactory.

The results also show that as density increased, the stress-strain curve also increased, indicating greater tensile strength and greater maximal strain in most cases, as illustrated in Fig. 4. However, when the refining energy was higher, especially when it reached 500 kWh/t, the curve was no longer monotonous due to data deviations in the middle density. The reason for this could be that at a certain refining level, the fiber lengths are always the same and therefore there is no difference in the stress-strain curve.

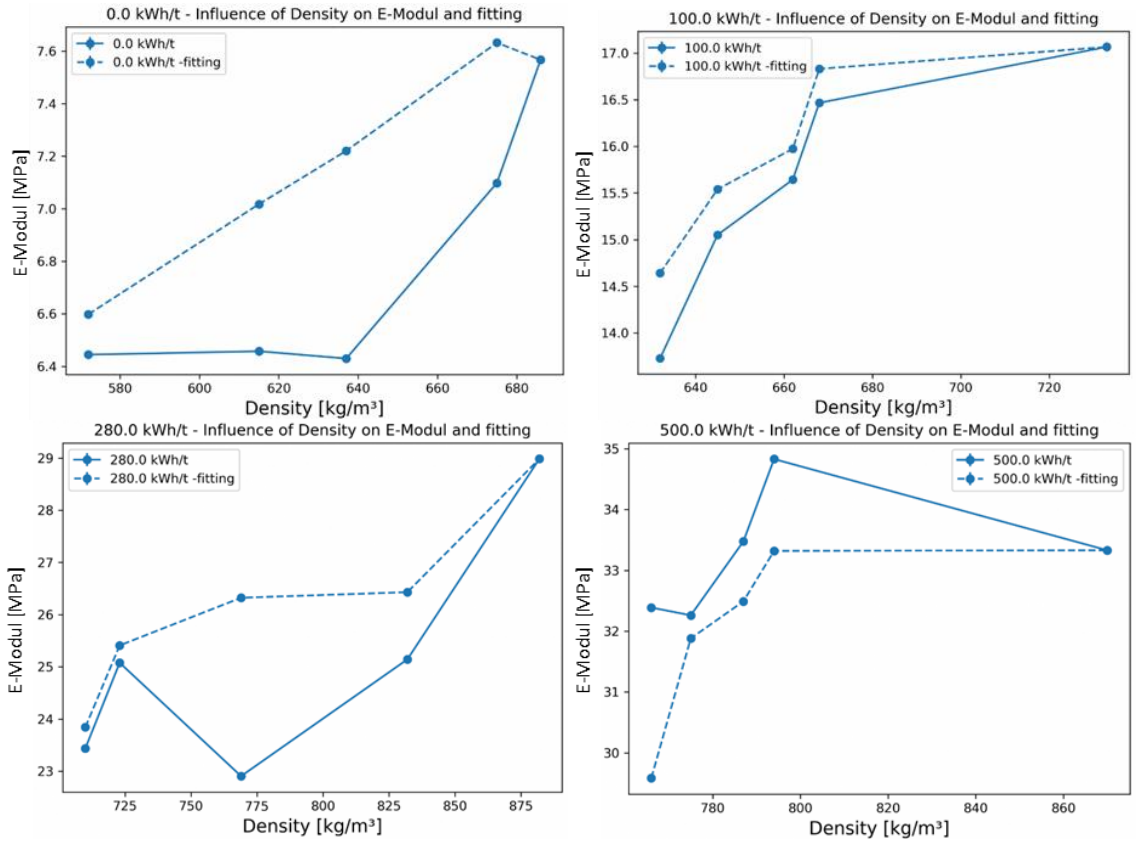


Fig. 3. Elastic modulus at different densities without refining and with refining capacity 100, 280, 500 kWh/t and the fitting using above efficiency coefficient

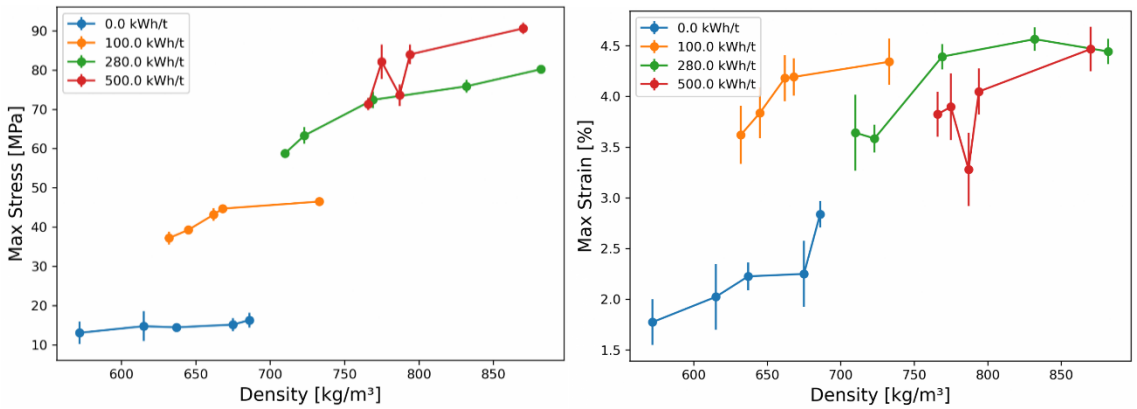


Fig. 4. Maximal stress and maximal strain values at different densities without refining and with refining capacity 100, 280, 500 kWh/t

These figures also show that refining had a significant effect on the mechanical properties, increasing the maximal strain and tensile strength of the material. The mechanical properties of unrefined laboratory paper were significantly lower. The refining capacity had some effect on the maximal strain, but the tensile strength increased significantly with increasing refining capacity. This was only reduced at the increase of the refining capacity from 280 to 500 kWh/t. Refining had a significant effect on the thickness and apparent density of the paper, with density increasing and thickness decreasing as

refining capacity increased (see Table 1), which is also consistent with the literature (Kibblewhite 1973).

Figure 5 shows the results of confocal light microscopy measurements of the lowest grammage of laboratory paper for different refining capacities. It is clear that the fibers became finer as the refining capacity increased. However, when comparing the 280 and 500 kWh/t refining energy levels, the difference in the degree to which the fibers were refined was not as great as the difference between the previous two images. Typical fiber lengths without refining can reach 1.2 to 1.5 mm with a width of 0.5 to 0.7 mm. At a refining capacity of 100 kWh/t, typical fiber lengths reach about 1 mm and widths of about 0.4 mm. At refining capacities of 280 and 500 kWh/t, optical surface measurements are already challenging for fiber identification, especially at 500 kWh/t, but the most obvious fibers do not exceed 0.8 mm in length and less than 0.3 mm in width.

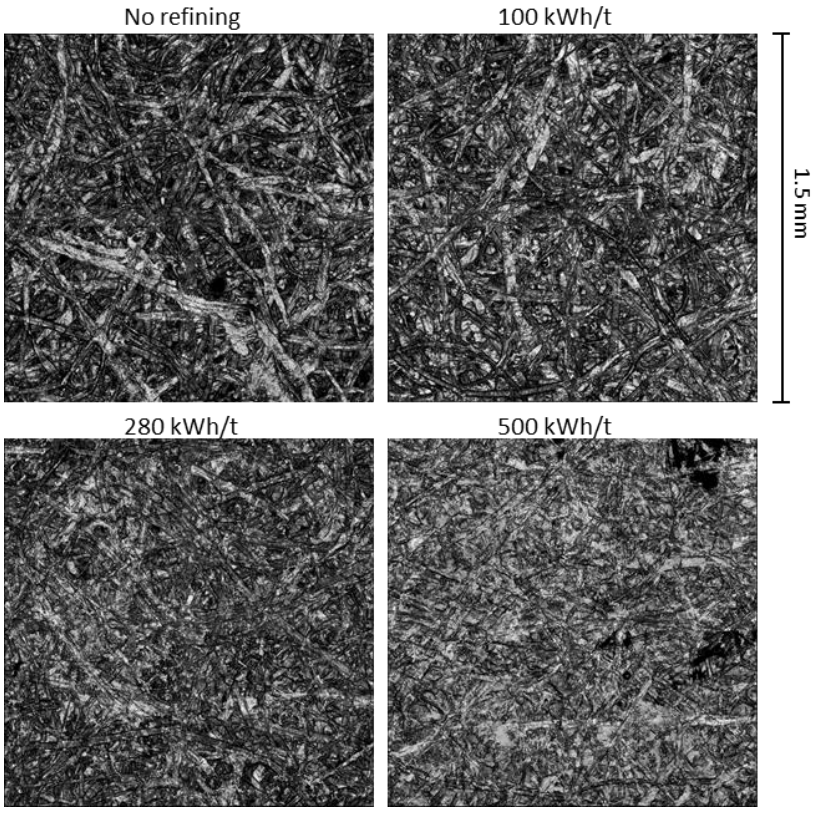


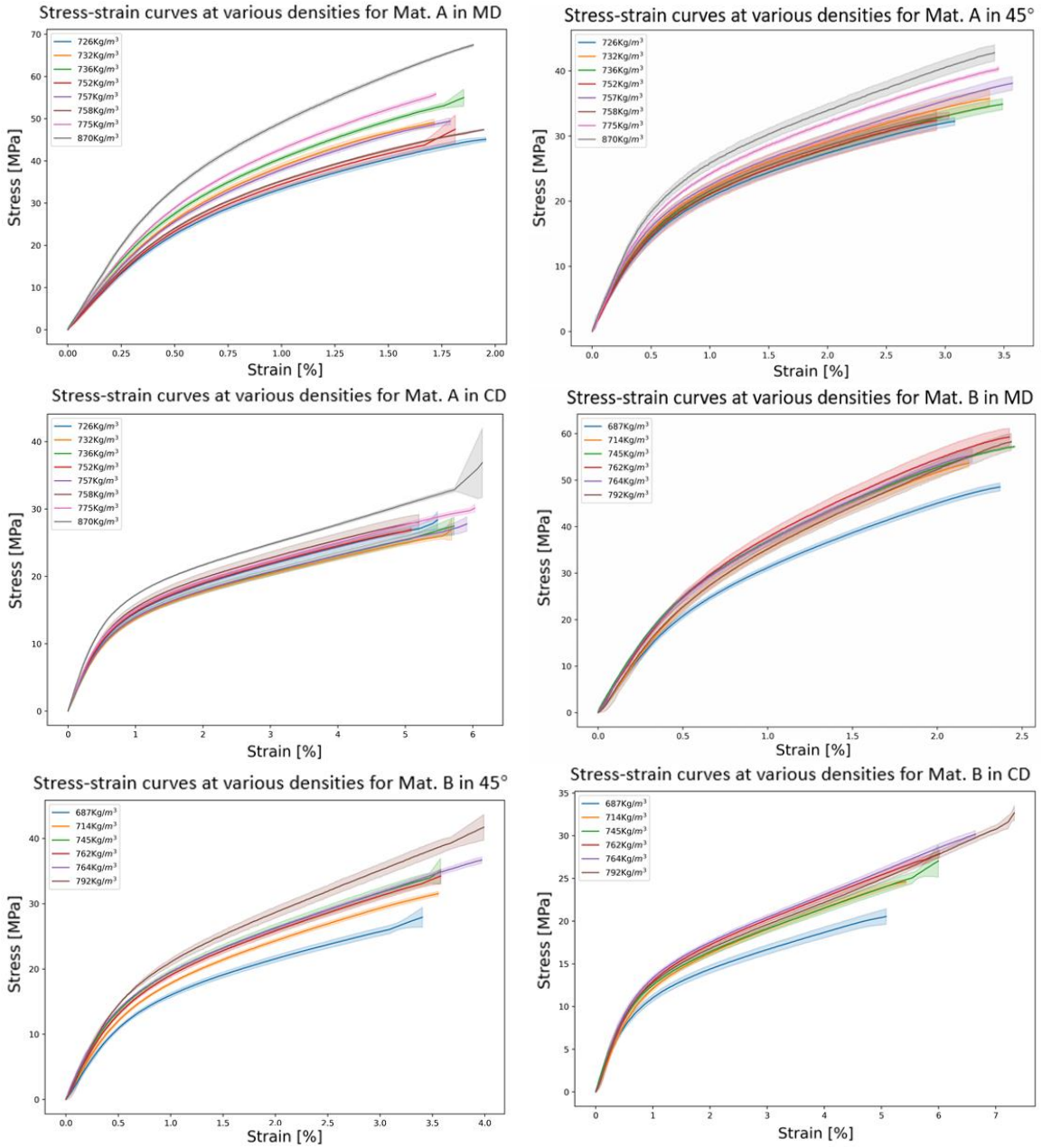
Fig. 5. Confocal microscope images of sample with different refining capacities

Refining increases the flexibility and surface area of the fibers, which increases the bonding between the fibers, resulting in an increase in the number of fiber bonds (Umair *et al.* 2020). As a result of this phenomenon, the paper density increases, leading to a reduction in paper thickness and an increase in tensile strength. Refining also caused differences in surface roughness with R_a ranging from 3.77, 2.83, 2.14 to 2.13 µm, *i.e.* increasing refining capacity results in smoother paper surfaces. Since the samples used for the measurements are directly from the papermaking process without rolling, the change in surface roughness is due to refining. As the refining capacity increased, the surface roughness of the samples decreased, which explains another reason for the decrease in thickness and resulting increases in density.

Influence of Density of Commercial Paperboard on Mechanical Properties

Unlike self-made laboratory paper, the exact production parameters of commercial paperboard are not known. What is certain, however, is that different commercial paperboard products undergo different calendering treatments, which increase density but do not create a large number of new hydrogen bonds and may even break some of them. This is the major difference from the refining of the pulp when producing paper in the laboratory as mentioned earlier.

The results for commercial paperboard were similar to those for laboratory paper, but the correlation is slightly weaker, as shown in Fig. 6.



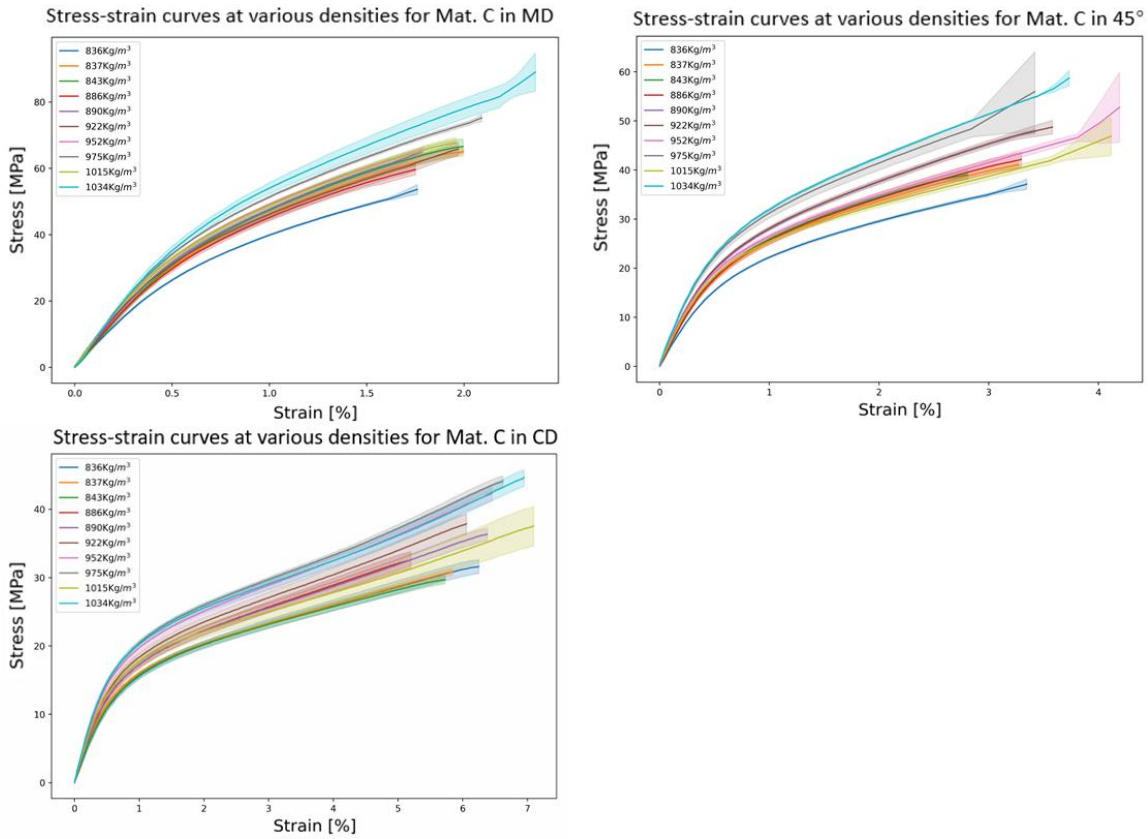


Fig. 6. Stress-strain curves at different densities for material A, B, and C in all three directions

The range of density variation for commercial paperboard was small, but it can still be seen that tensile strength increased with density, although there were a few exceptions. Three different test orientations were chosen to present results for only three materials, but the orientation of the tensile test had a negligible effect on density, so it is sufficiently representative. Similarly, the effect of density on elastic modulus is shown in Fig. 7 with solid lines. For commercial paperboard, the elastic modulus tended to increase with increasing density, but the deviation was more compared to that of laboratory paper. The dashed lines show the fitting of elastic modulus using calculated efficiency factor. For commercial paperboard, the fitting results of the efficiency factor were also satisfactory. The effect of density on tensile strength and maximum elongation is shown in Fig. 8. These plots resemble the results for laboratory paper, despite some data bias. When focusing on individual materials, it can be observed that the trends of the curves in the three directions were essentially similar, *i.e.* fiber orientation had little effect on the differences in fracture properties due to density differences.

Influence of Pressing on Mechanical Properties

The effect of additional pressure applications on dry commercial paperboard was also studied, since dry-pressing is a typical operation during certain converting processes during commercial paperboard, *i.e.* dry-forming applications. For example, the paperboard is usually pressurized in the thickness direction when it is in the gap between the blank holder and the die and between the punch and the die during deep drawing. In subsequent studies, there is potential to apply the results to localized pressure application to reduce the inhomogeneity of the paperboard and improve the mechanical properties.

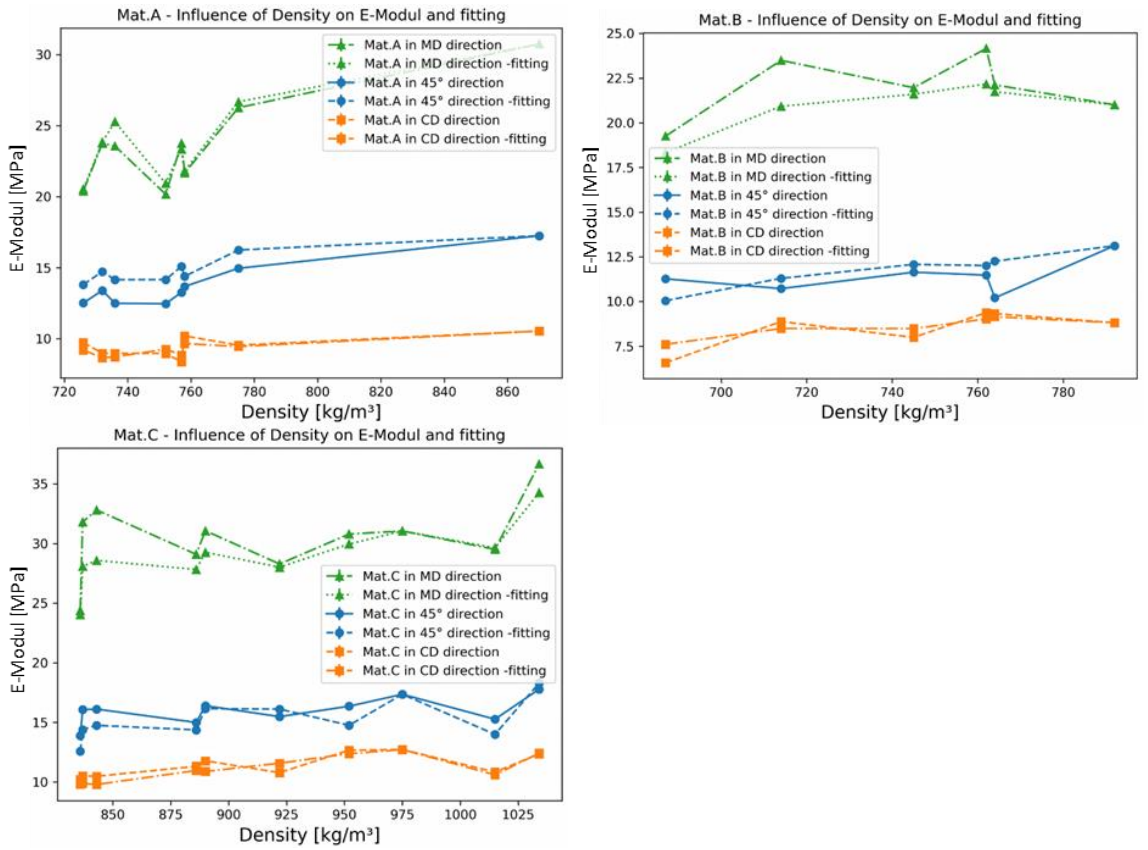
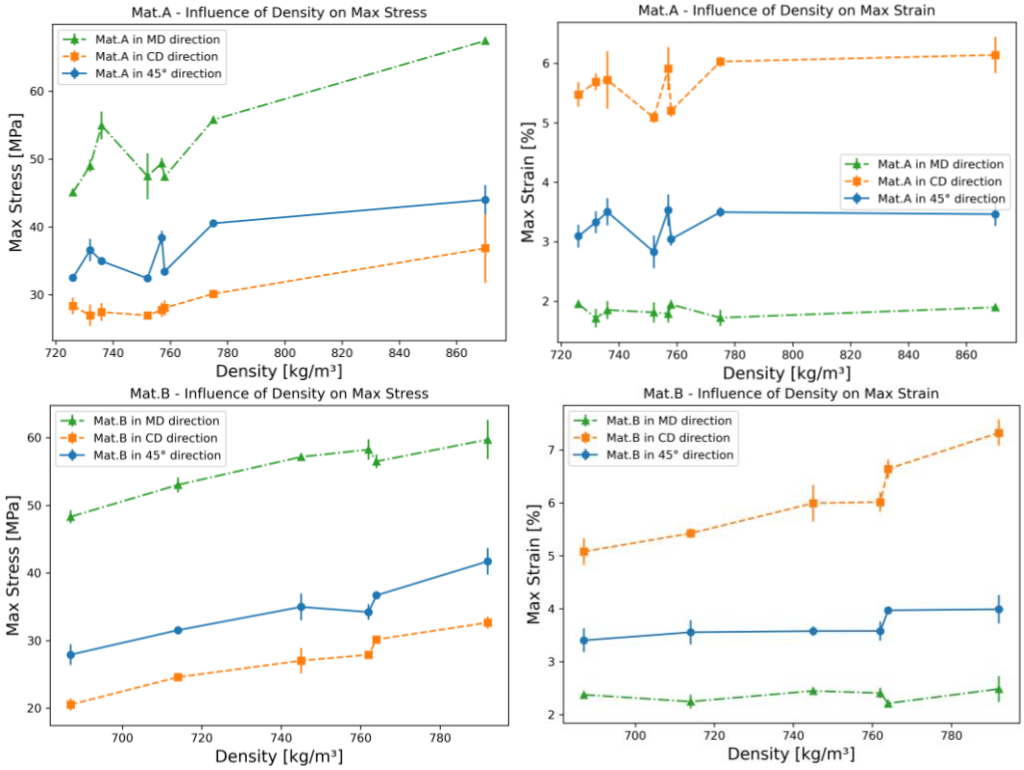


Fig. 7. Elastic modulus at different densities for Material A, B, and C in all three directions and the fitting using above efficiency coefficient



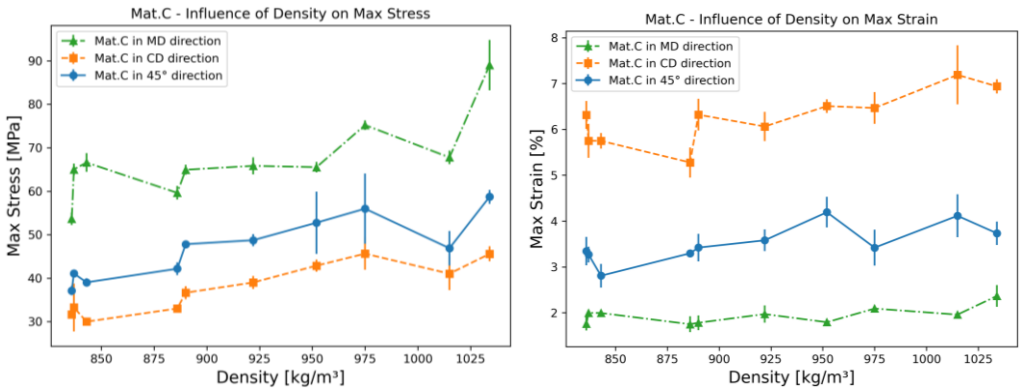


Fig. 8. Maximal stress and maximal strain values at different densities for material A, B, and C

Uniaxial tensile tests were also carried out on the unpressed and pressed samples. The resulting stress-strain curves of three materials are compared in Fig. 9. It can be seen that the stress-strain curves of the paperboard changed after being subjected to pressing. These changes are attributed to the densification of the samples.

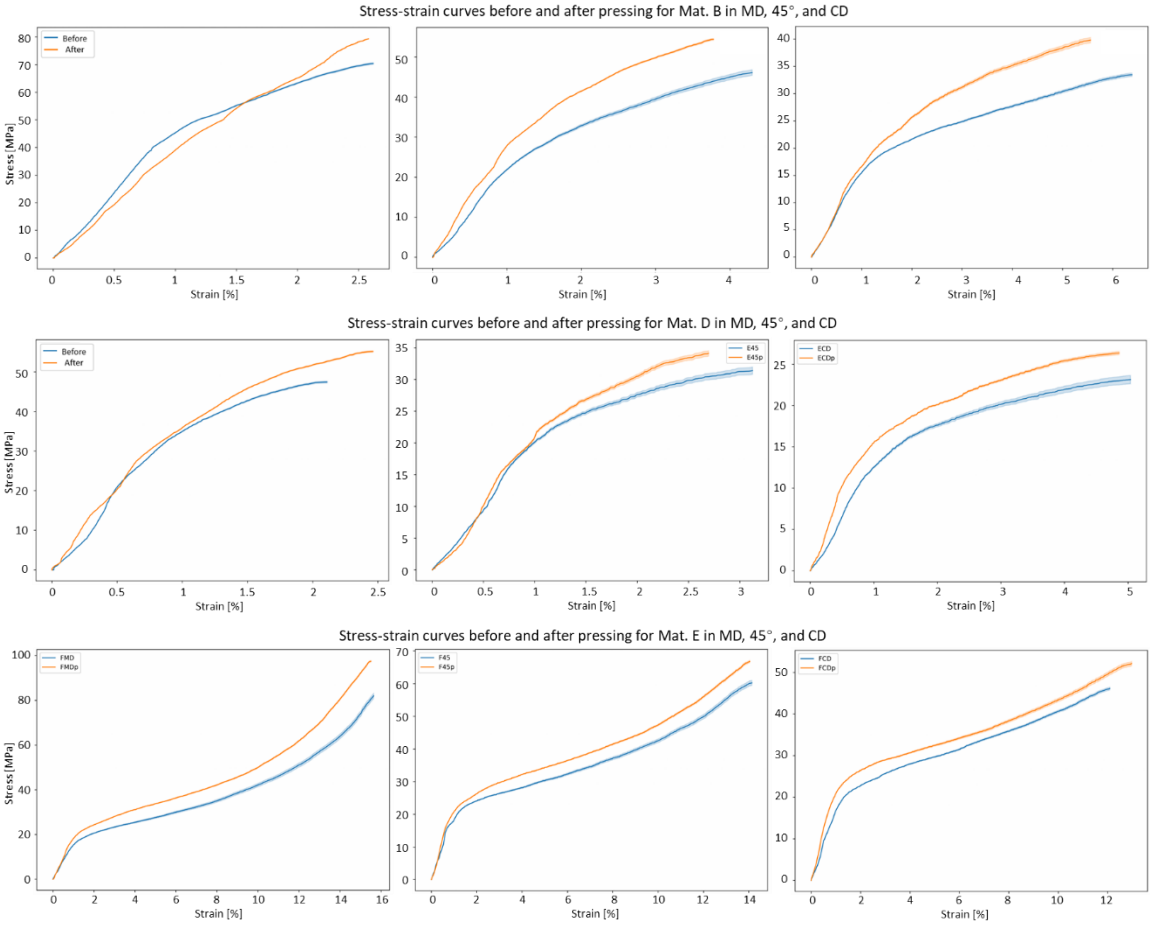


Fig. 9. Stress-strain curves of samples with and without pressing for material B, D, and E in 3 directions

Out-of-plane compression causes the material to have a higher in-plane modulus of elasticity, which is the slope of the linear phase of the stress-strain curve, and an increase in tensile strength but a usual decrease in maximal strain. The thickness reductions of materials B, D, and E after experiencing pressing in the thickness direction were 17%, 12%, and 13%, respectively. Of these, Material B underwent the greatest thickness reduction, with the corresponding greatest increase in ultimate strength, but also the greatest loss in ultimate elongation. However, the increase in fracture strength was significantly greater than the loss in fracture elongation. Materials D and E, on the other hand, showed a smaller change in properties due to a smaller reduction in thickness. This is another indication that density has a strong influence on in-plane mechanical properties and that it is possible to improve the properties and formability of the materials, for example by localized pressing.

In this case, the percentage increase in tensile strength was related to the percentage decrease in thickness, *i.e.*, the percentage increase in density, indicating the correlation between the in-plane and out-of-plane properties of the material. Compression in the thickness direction forces the fibers into the existing voids of the porous fiber structure, resulting in a significant reduction in the porosity of the board. In a previous study (Stein 2019), computed tomography (CT) scanning of material B was performed before and after pressing, as shown in Fig. 10. The black color in the image represents the penetration of X-rays into the air, and the white color is the penetration into the paper fibers, showing that the density of the samples increased significantly after pressing. The unpressed areas appear darker due to the higher penetration, while the pressed areas appear lighter due to the absorption of more radiation.

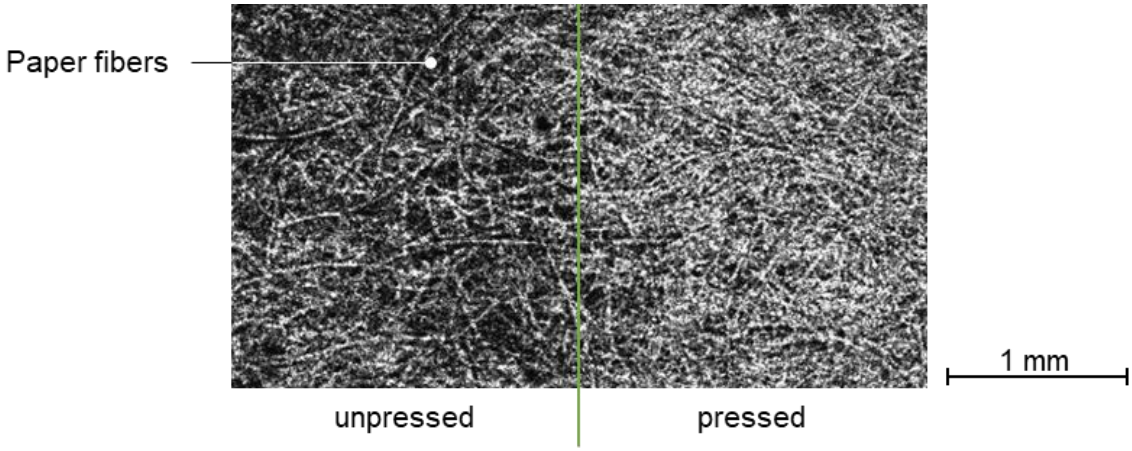


Fig. 10. Computed tomography images of samples before and after pressing (Stein 2019)

The tensile strength of the samples increased significantly after compression due to the increase in the contact area between the fiber surfaces, and the compression of the colleague fibers also created a new contact area at the fiber surface within the fiber lumen. The microscopic images showed that the fibers were flatter in cross section after compression, *i.e.* the fibers that collapsed during papermaking were further flattened by out-of-plane pressing. In addition, the maximal strain of the densified specimens decreased slightly in most cases. The possible reason for this is that a high degree of compression in the thickness direction breaks the bond between the fibers and also breaks the fiber walls, making the material more defective. During the tensile test, the eventual fracture of the specimen does not occur instantaneously, but rather as an accumulation of small defects, so that the compressed material reaches the maximal strain more quickly. In a tensile test,

the effective thickness of the material decreases, and it should be noted that this is different from an increase in apparent thickness due to delamination. Thus, the compressed material will reach the maximal strain more quickly for the same thickness as the material before compression. On the other hand, some specimens showed no reduction due to more closed internal voids and tighter bonding between the fibers, which has the potential to offset some of the loss in elongation.

Efficiency Factor Due to the Effect of Density

Stress-strain curves are a useful tool for describing the mechanical properties of materials in terms of their elasticity and plasticity, as well as for numerical simulation. To quantitatively compare the differences in mechanical properties of paper at different densities, the efficiency factor is utilized. Figure 11 is an example of the fitting of a stress-strain curve and its plastic part of a density of 710 kg/m^3 with a refining energy input of 280 kWh/t, using the maximum density of 882 kg/m^3 as a reference. Using the efficiency factor of 0.8227 and the reference curve, the stress-strain curve and its plastic part of other densities can be well described using only the reference data (in this work the highest density). The behavior of the plastic part of the material can also be accurately described by a reference curve and an efficiency factor calculated from the stress-strain curve.

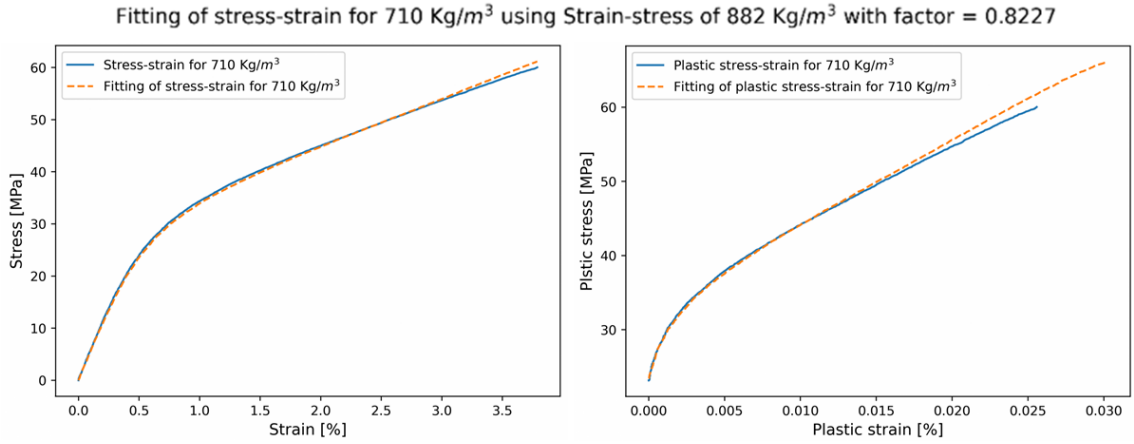


Fig. 11. Efficiency factor of the (plastic) stress-strain curve of 710 kg/m^3 using of 882 kg/m^3 as a reference

The fitting curves for laboratory paper are shown in Fig. 12. They are the connecting lines of the fitted factors for each density, and the dashed lines represent the fitted slopes of the fitted curves according to Formulation (2). It can be seen that as the refining capacity increased, the value of the efficiency factor became smaller, *i.e.* the influence of density became weaker. However, at a refining energy input of 500 kWh/t, the efficiency factor was a little overestimated at higher densities. Since the refining energy input of 500 kWh/t belongs to a high degree of homogenization of the fiber suspension, *i.e.*, the fibers were treated at a high level, the uniformity of the paper produced was also very high. As shown in Fig. 5, the typical length and width of fibers become smaller as refining capacity increases. When the material was highly homogenized, the effect of density on the mechanical properties becomes weaker.

For commercial paperboard, the results of efficiency factors and the slope of the efficiency factor as function of the density in three directions are shown in Fig. 13. The values of the efficiency factor were in the range of 0.0009 to 0.0013. The values were

always smaller than when the refining energy input was greater than 100 kWh/t. Materials A and B were uncoated and had a higher density factor compared to coated material C. It can be seen that the surface coating played a role in the extent to which density affects mechanical properties.

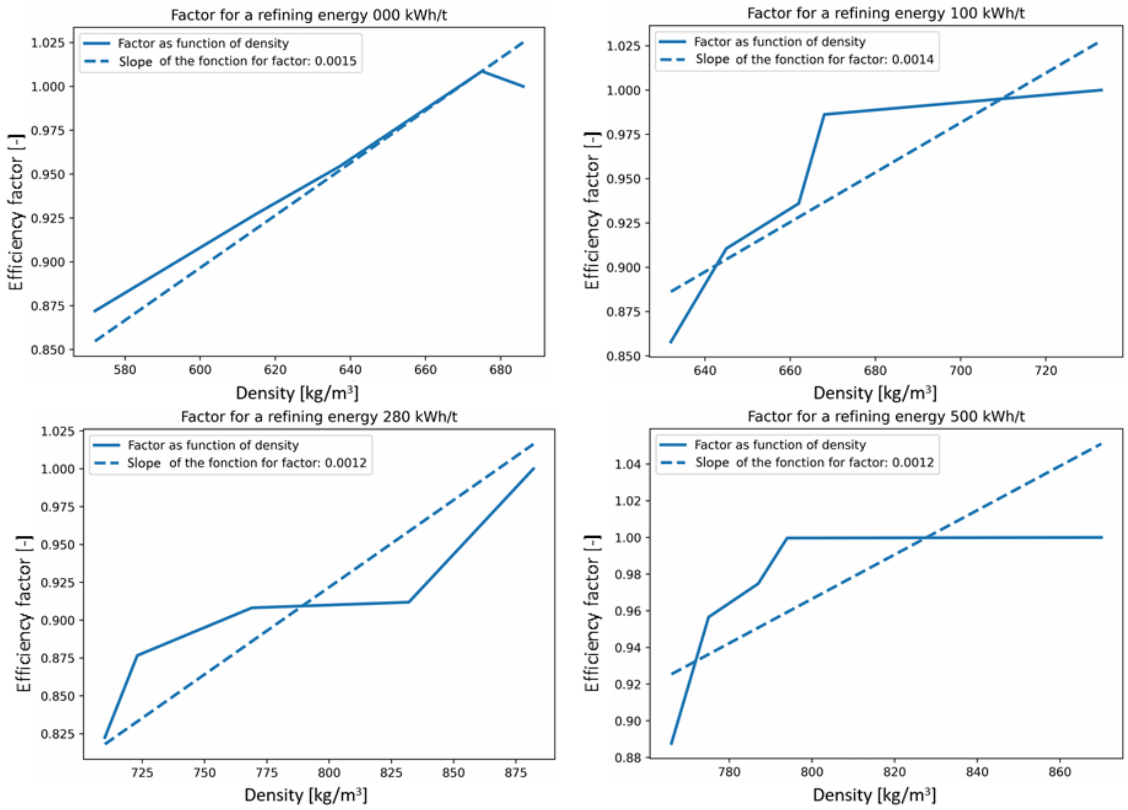
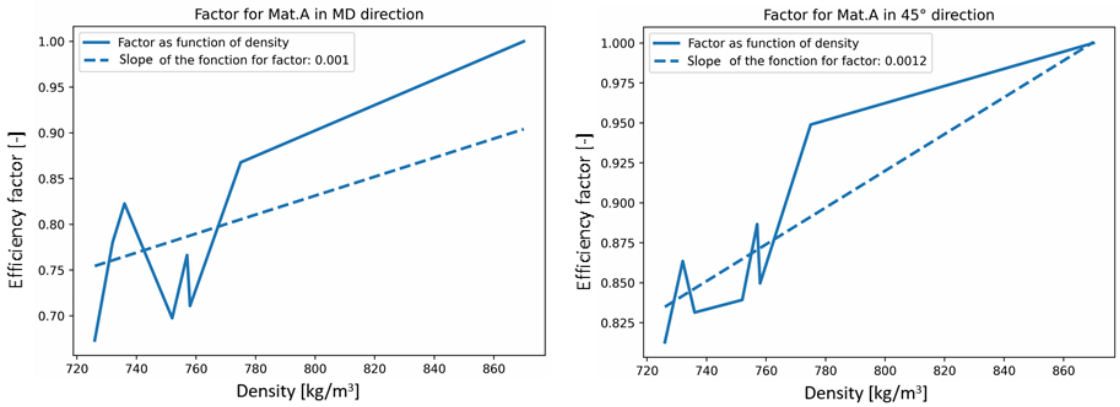


Fig. 12. Linear function to approximate the efficiency factor with the density without refining and with refining capacity 100, 280, 500 kWh/t



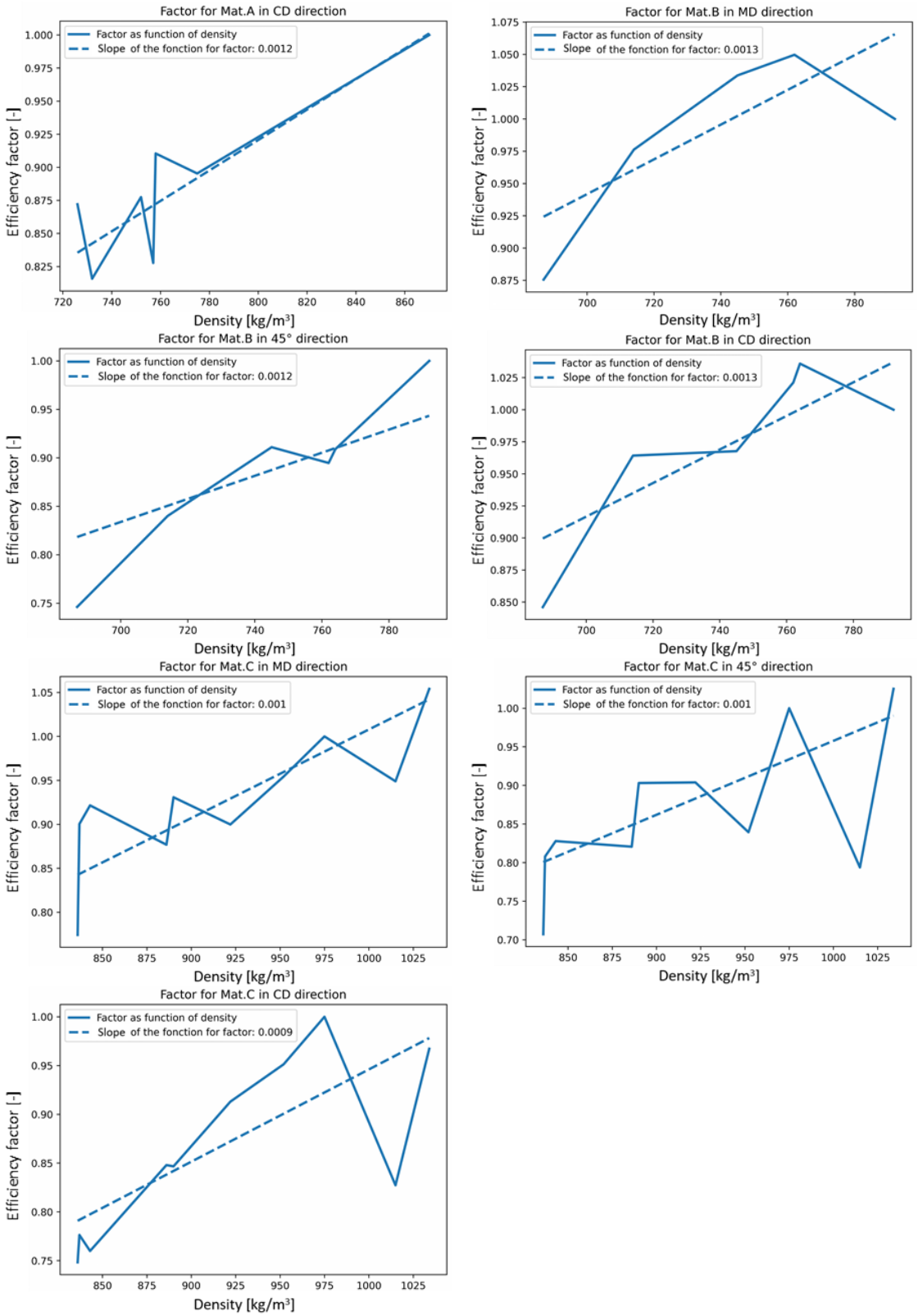


Fig. 13. Linear function to approximate the efficiency factor with the density for Material A, B, and C in all three directions

DISCUSSION

By analyzing the stress-strain curves, it can be observed that the higher the density, the higher the strength of the material usually is. In order to quantify the differences observed in the stress-strain curves, the authors have introduced an efficiency factor that takes into account the mechanical properties of the elastic and plastic components of the paper. By comparing the variation of the slope for laboratory paper at different refining levels, it was found that the accuracy of the fit decreased as the refining level was increased. As the refining level increased, the material became more homogeneous and the role of density variables decreased (see Fig. 14 left). In practice, however, a refining level of 500 kWh/t is very high and is not commonly used in large-scale production.

As for commercial paperboards, it was found that the proposed efficiency factors can also be used to describe the effect of density variations on the mechanical behavior by studying two uncoated paperboards and one coated paperboard. As shown in Fig. 14 (at right), the effect of fiber direction on the efficiency factor was not significant. In addition, the influence of the coating (Mat. C) on the density was considered to be influential, but further tests on coated and uncoated paperboards were used to demonstrate that this conclusion is necessary. The knowledge of the influence of density on the in-plane mechanical properties can be incorporated into stochastic material modeling, which would lead to a better understanding of the role of local density on material response and local failure.

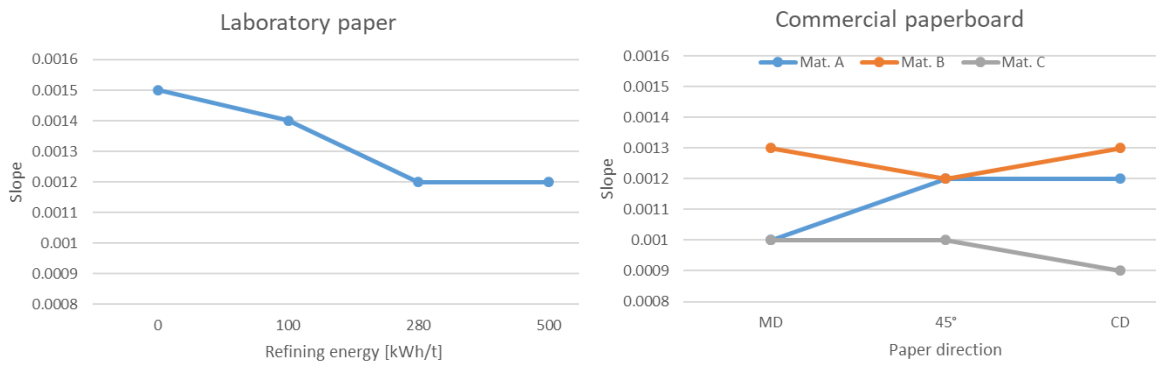


Fig. 14. Comparison of slope for laboratory paper with different refining energies and commercial paperboard with different materials and directions

The results show that using efficiency factors and reference data, it is possible to describe or predict the mechanical behavior of paper produced from the same pulp composition and refining energy at different densities. For example, multiplying the efficiency factor by the reference elastic modulus gives the estimated elastic modulus at density. The same is true for the stress-strain curve and its elastic part, which is of interest for material modeling and optimization. By utilizing this effective factor, the relationship between density and material properties can be described more precisely, enabling a refined modeling approach for inhomogeneous materials. This factor serves as a key parameter in predicting mechanical behavior and scaling properties such as stiffness and plastic stress across different densities.

The stretchability of commercial paperboard before and after out-of-plane pressing was also tested. The results show that densification also improves the mechanical properties of the material, *i.e.* it significantly increases the tensile strength of the material,

but it may also lose some of the elongation. Since paperboard is also subjected to out-of-plane compression in 3D forming processes, especially deep drawing, the results are useful for improving deep drawing process parameters and product quality. Furthermore, the combined effect of compression and humidification still needs to be studied because of the complex functions involved, such as the different compressibilities of water and air.

CONCLUSIONS

1. The degree of fiber refining has an important effect on the density and mechanical properties of the paper produced. Overall, an increase in refining capacity results in an increase in the density of the paper, and the mechanical properties, mainly tensile strength and maximal strain, usually increase as well. However, the response is not linear.
2. In this work, the density of a sample was increased without significantly changing the number of hydrogen bonds, *i.e.* only post-processing of the paper, such as calendering or rolling, is considered. The increased densities in this way of laboratory paper and commercial paperboard increase the stress-strain curve, *i.e.*, the maximum stress.
3. By applying high pressure in the thickness direction for a period of time of the three commercial paperboards, it was found that the application of compression to the dry paperboard increased the tensile strength while potentially reducing the maximal strain.
4. This study also described the elastic modulus, stress-strain, and plastic stress-strain curves using efficiency factors, which allows a quantitative description of the mechanical behavior of the elastic and plastic compositions based on density effects. Using this efficiency factor, it is possible to predict the mechanical properties of materials at different densities under the same manufacturing conditions. This can simplify material characterization and subsequent modeling.
5. The introduction of the effective factor provides a more advanced framework for understanding and modeling the mechanical behavior of paper-based materials. This approach enhances the accuracy of material characterization and supports the optimization of processing parameters in industrial applications.
6. The effect of material and fiber orientation on the efficiency factor in refining and commercial paperboard was also obtained. It was found that refining energy had a strong effect on the extent to which density affects the mechanical properties of the material, while the directionality of industrial paperboard had a weak effect.
7. This approach provides the basis for stochastic modeling and fracture prediction of materials, with an emphasis on accurately reflecting the effects of density inhomogeneities in the simulations.

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