## INFLUENCE OF LAYER COMPOSITION ON PROPERTIES OF RECYCLED BOARDS

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In the present study, three types of recycled boards with different top layer composition have been analysed. The quality of coated boards with different fibrous composition has been evaluated by determining their structural, mechanical, optical and surface properties. The slight increase in virgin fibre content in the top layer, from 10 to 20%, has been found to improve the surface, the optical and mechanical properties of coated recycled boards. A greater influence on the mechanical properties is exerted by the addition of a higher percentage of chemical pulp in the top layer of the three-layered structure of recycled boards, rather than by the addition of a forth layer from recovered paper and board. Additionally, the investigation aimed at determining the viscoelastic properties of the boards and assessing the influence of a 90% relative humidity on their tensile properties. Besides lowering tensile strength, humidity has even a more significant influence on the viscoelastic properties. Of the coated recycled boards under study, the influence of humidity has been found as less pronounced in the boards containing higher amounts of chemical pulp in the top layer.

Keywords: board, recycled fibres, viscoelastic properties, mechanical properties, humidity

#### INTRODUCTION

Thanks to its versatility, durability and relatively low cost, paperboard is used to package everything, from carryout food to pharmaceuticals, from cereals to hardware.<sup>1</sup> Considering the recent improvements in the quality, variety and availability of recycled as well as its content paperboard, competitive price and significant environmental benefits, the major consumer product industries (e.g., dry foods, home and personal care products) have largely switched to recycled materials. In fact, more than half of the products on the supermarket shelves are now packaged in recycled paperboard.<sup>1</sup> The board with a postconsumer recycled content is a high quality, cost-competitive packaging option with significantly lower environmental impacts than virgin board.<sup>2</sup> Although some mills produce 100% recycled paper, most of them mix their used pulp with some virgin fibre. However, the strength properties of pulp blends are not simply the average of the strength properties of their components. Depending on the nature of the components and on the specific property of interest, there

may appear positive or negative deviations from the average.<sup>3</sup> The main goal of this study was to investigate how a higher content of virgin fibres in the top layer of 3layered boards, or the addition of another layer made from recovered paper and board influences the mechanical properties of recycled boards.

Tensile strength is a very useful property describing the general strength of paper or board.<sup>4</sup> However, besides tensile strength, the viscoelastic properties of boards are very important, too. All polymeric materials, including paper and board. exhibit viscoelastic behaviour to a higher or lower extent. This means that, when the material is deformed, it will not behave like an elastic spring, but will partly show a viscous flow. One of the possibilities to obtain some information on the viscoelastic behaviour of paper or board is the study of a stress *versus* strain curve. This method was also applied in the present investigation for determining the viscoelastic properties of recycled boards with different composition and grammage.

The relationship between loading and

deformation, describing the characteristic behaviour of a material, is referred to as a constitutive relation. The load-deformation relation depends upon the magnitude of both stress and strain, on the rate of stress and strain application, and also on environmental factors, such as temperature and moisture content.<sup>5</sup> As paper is a highly hygroscopic material, the relative humidity of the surrounding environment and the moisture content generally affect the viscoelastic properties of paper to a great extent.<sup>6</sup> The moisture uptake modifies the properties of paper and board, among them the mechanical properties. At low moisture contents, paper and board become relatively stiff and brittle, whereas, at high moisture content, they appear as very ductile materials.<sup>7</sup> The second aim of the study was to assess the influence of relative humidity on the viscoelastic properties of boards.

### EXPERIMENTAL

#### Materials

The board samples investigated in the present study were produced by the Slovene company Količevo Karton d.d. from recycled pulp, by a multilayer technology. Three types of boards, KROMOPAK, KOLIPRINT and GRAFOPAK, with different composition of the top layer and different amounts of coating applied, were analysed. Boards KROMOPAK and KOLIPRINT are composed of three layers, board GRAFOPAK - of four layers. The middle and the back layers are composed of recycled fibres obtained from mixed waste paper and waste board, the middle layer also containing mechanical pulp. The top layer consists of bleached chemical pulp and recycled fibres from high-quality, sorted, white recovered paper. A higher percentage of chemical pulp is present in the top layer of the KROMOPAK board, compared to the other two boards. Board GRAFOPAK differs from the KOLIPRINT one as to the number of layers, since it includes a fourth, so-called protective layer, composed of recycled fibres obtained from recovered paper and board. The percentage of coating is the same in boards KROMOPAK and GRAFOPAK, while, in board KOLIPRINT, it is 4  $g/m^2$  lower. Three different basic masses (grammages), most commonly used, were selected for the analysis of each board type.

#### Methods

The boards were tested under standard climatic conditions (ISO 187), and their basic properties: basic mass (ISO 536), thickness and bulk (ISO 534), were determined. The surface roughness of the boards was determined according to ISO 8791-2, with a Bendtsen

roughness tester. Of the optical properties, ISO brightness (ISO 2470) and gloss (ISO 8254) were measured. The determination of strength properties included measuring tensile strength (ISO 1924-2), bursting strength (ISO 2759), tearing strength, by the Elmendorf method (ISO 1974), and bending stiffness by L&W 5° (ISO 5628). The tensile properties of the boards were measured with an Instron 5567 tensile testing machine equipped with a climate chamber. The samples were stretched at the same rate, 20 mm/min, at a temperature of 23 °C and different relative humidity: 50 and 90%. During stretching, several load and elongation data were recorded per second, until breaking of the sample occurred. From the measured load and elongation data, an average curve, subsequently converted into a stress-strain curve, was obtained. For numerical analyses, a computer program named DINARA<sup>8</sup> calculated the characteristic viscoelastic values (yield point, moduli, energy of deformation), giving continuous information on the behaviour of the material over the whole deformation range, up to breaking. The program scheme involves: plotting of a stress-strain curve, interpolation, integration, numerical differentiation and calculation of the values that determine the viscoelastic properties of the sample. The results of the numerical analysis of the curve are given as minimal, maximal and zero values of the function  $\sigma = f(\varepsilon)$ , its derivatives and integral values. The evaluation program DINARA was described in previous papers.<sup>9,10</sup>

## **RESULTS AND DISCUSSION** Structural and optical properties of boards

The tested boards differ in grammage and thickness, ranging from 214 to 493  $g/m^2$  and from 300 to 600 µm, respectively, as seen in Figure 1. The relationship between the increasing grammage and thickness is linear, the correlation coefficient being of 0.99. The bulk of the boards ranges between 1.18 and 1.46  $\text{cm}^3/\text{g}$ . The boards with the same composition and different grammage differ by up to 5% in bulk, while the boards with various compositions and the same or similar grammage differ by over 10%. Α modification in the chemical pulp content in the top layer can influence the bulk of the boards to some extent. The boards with a lower percentage of recycled fibre content in the top layer (KROMOPAK) have a higher bulk compared to those with a higher content of recycled fibres (KOLIPRINT or GRAFOPAK).

Of the surface and optical properties, roughness, ISO brightness and gloss of the board top layer were determined. Roughness was determined as Bendtsen roughness, involving measuring of the rate at which air passes between a flat circular land and the board surface. The results of the measurements are given in Figure 2. The higher is the value of the measured air flow, the higher will be the roughness of the board. All boards show quite a smooth surface, the smoothest one occurring in the board with the lowest percentage of recycled fibre content in the top layer (KROMOPAK). The process of pigment coating and the composition of the coating mixture were the same for all boards, the only difference occurring in the coating layer of board KOLIPRINT, which had a lower grammage. The lower coating layer thickness in board KOLIPRINT resulted in lower brightness (below 78.5%). The higher brightness of the board with a lower percentage of recycled fibre content in the top laver (KROMOPAK), over 85%, compared to other boards with a higher recycled fibre content, is the consequence of a higher percentage of bleached chemical pulp added to the top layer. The boards also differ in gloss, the measured values ranging between 40.3 and 50.5% (Fig. 2). The boards of the

KROMOPAK type, with the highest brightness, also have the highest gloss. The addition of a fourth layer from recovered paper and board (GRAFOPAK), in spite of the higher coating applied, improved neither the smoothness, nor the brightness or the gloss of the boards.

#### Strength and stiffness of boards

The bending stiffness of the boards is much higher in longitudinal (machine) direction than in transverse (cross) direction, which is due to fibre orientation. predominantly in longitudinal direction. The composition and grammage of the boards influence their bending stiffness. The influence of the composition on bending stiffness is evident when comparing boards KROMOPAK and KOLIPRINT and boards KROMOPAK and GRAFOPAK. The boards of the KROMOPAK type, which contain more chemical pulp in the top layer, are - at the same or similar grammage – stiffer than the boards of the KOLIPRINT and GRAFOPAK type, even if the latter contains one more layer (Fig. 3).

Table 1 Description of board samples

Board sample	KROMOPAK	KOLIPRINT	GRAFOPAK
Number of layers	3	3	4
Composition:			
- top layer (%)	80/20 sorted RP/CP	90/10 sorted RP/CP	90/10 sorted RP/CP
- middle layer (%)	80/20 mixed RP&B/MP	80/20 mixed RP&B/MP	80/20 mixed RP&B/MP
- back layer (%)	mixed RP&B	mixed RP&B	mixed RP&B
- protective layer (%)	/	/	RP&B
Coating $(g/m^2)$	30	26	30
Grammage (g/m <sup>2</sup> )	215, 275, 400	250, 350, 450	250, 300, 500





Figure 1: Grammage, thickness and bulk of board samples of different composition and grammage: KROMOPAK – 10% virgin fibres, 3 layers, 215-400 g/m<sup>2</sup>; KOLIPRINT – 20% virgin fibres, 3 layers, 250-450 g/m<sup>2</sup>; GRAFOPAK – 10% virgin fibres, 4 layers, 250-500 g/m<sup>2</sup>



Figure 2: Roughness, ISO brightness and gloss of board samples of different composition and grammage: KROMOPAK – 10% virgin fibres, 3 layers, 215-400 g/m<sup>2</sup>; KOLIPRINT – 20% virgin fibres, 3 layers, 250-450 g/m<sup>2</sup>; GRAFOPAK – 10% virgin fibres, 4 layers, 250-500 g/m<sup>2</sup>



Figure 3: Bending stiffness in machine (MD) and cross direction (CD) of board samples of different composition and grammage: KROMOPAK – 10% virgin fibres, 3 layers, 215-400 g/m<sup>2</sup>; KOLIPRINT – 20% virgin fibres, 3 layers, 250-450 g/m<sup>2</sup>; GRAFOPAK – 10% virgin fibres, 4 layers, 250-500 g/m<sup>2</sup>

Figure 4: Tearing strength in machine direction of board samples of different composition and grammage: KROMOPAK – 10% virgin fibres, 3 layers, 215-400 g/m<sup>2</sup>; KOLIPRINT – 20% virgin fibres, 3 layers, 250-450 g/m<sup>2</sup>; GRAFOPAK – 10% virgin fibres, 4 layers, 250-500 g/m<sup>2</sup>



Figure 5: Bursting strength of board samples of different composition and grammage: KROMOPAK – 10% virgin fibres, 3 layers, 215-400 g/m<sup>2</sup>; KOLIPRINT – 20% virgin fibres, 3 layers, 250-450 g/m<sup>2</sup>; GRAFOPAK – 10% virgin fibres, 4 layers, 250-500 g/m<sup>2</sup>

The grammage and thickness of the boards have a great impact on bending stiffness, too. The relationship between the increasing grammage and bending stiffness is linear, the correlation coefficient being over 0.95 (Fig. 3).

The tearing resistance of the boards was determined by the Elmendorf method, as the mean force required for continuing tearing of the already cut board. As shown in Figure 4, a slightly higher force is needed for tearing the boards in transverse direction, since this requires breakage of a higher number of interfibrillar bonds. The boards of the GRAFOPAK type exhibit the highest tearing strength, as they have one more, so-called protective layer and, consequently, more interfibrillar bonds have to be torn. The composition of the boards influences tearing strength, as the latter depends on the binding ability and number of bonds between fibres. A higher tearing strength is obtained when a fourth layer from recovered paper and board is added, instead of applying a higher content

of chemical pulp in the top layer. Tearing strength thus increases with the grammage and thickness of the boards, the correlation exceeding 0.8 (Fig. 4).

The bursting strength of the boards also depends on their composition, thickness and grammage. A comparison between boards with different composition and the same or similar grammage showed no great difference in bursting strength, only slightly lower values for the boards containing a higher content of chemical pulp in the top layer (KROMOPAK). The bursting strength of the boards depends much more on thickness and grammage. Bursting strength increases linearly with grammage, the coefficient being of 0.99 (Fig. 5).

As a strong linear relationship was obtained between bending stiffness, strength (tearing and bursting) and grammage, it has been suggested that the stiffness and strength of boards could be very well predicted for arbitrarily selected board grammage.

# Tensile properties of boards at 50 and 90% relative humidity

Besides breaking extension and breaking load, several other tensile properties could be obtained from the tensile test, when a continuous record of load versus elongation performed. The first characteristic is obtained from the curve is the initial resistance to stretching, represented by the slope of the initial straight part of the curve. The slope of the curve gives the modulus. which is a measure of the material stiffness and of its resistance to extension. An elastic modulus is obtained from the first derivative of the function, in the initial steepest linear part of the curve. All tested boards offer the highest resistance to stretching at a 0.29% extension in machine direction, and between 0.35 and 0.51% in cross direction, where the elastic modulus has the highest value. In machine direction, the slight tendency of lowering the elastic modulus with increasing the basic mass of the boards has been observed. In cross direction, where moduli have up to three times lower values than those for machine direction, the values of the moduli decrease with basic mass in the boards composed of 3 layers (KROMOPAK and KOLIPRINT) whereas, in those composed of 4 layers (GRAFOPAK), the moduli increase with the basic mass of the boards. The differences in the composition of the top layer and in the thickness of the coating layer seem to have some influence on moduli, too. Higher moduli are obtained at a lower grammage, and lower, respectively, at a higher grammage, for the boards of the KOLIPRINT type, in machine direction, in comparison with the boards with a higher content of chemical pulp in the top layer (KROMOPAK), where the difference between the moduli of the boards with different grammage is not so pronounced. In the boards cross direction. of the KROMOPAK type have higher moduli than the other two types of boards, which contain more recycled fibres, in spite of the low difference.

At a higher relative humidity (90%), the moduli for both directions have much lower values, up to twice lower in machine direction and up to 4 times in cross direction. Humidity has more influence on the boards with a higher content of recycled fibres, which is even more pronounced in the boards with a fourth layer from recovered paper and board.

The yield point is an important feature obtained from the second derivative of the stress-extension curve. Before the vield point, the extension of the material is elastic while, above the yield point, part of the extension is non-recoverable. Yielding occurs at an extension of 0.34% in machine direction, and between 0.5 and 0.6% in cross direction. The stress at which vielding occurs represents around 10% of the stress at break for boards, in machine direction, and around 20% of the stress at break in cross direction. The difference in yield stress among samples is up to 20% in machine direction, and up to 40% in cross direction. It seems that grammage and the different structure of the boards have no significant influence on the magnitude of the yield stress. On the other hand, relative humidity has a significant influence on the yield stress. At 90% relative humidity, the yield stress decreases up to 300%, versus the values obtained at 50% relative humidity. In the board containing a fourth layer from recovered paper and pulp, the decrease in the yield stress was less pronounced than in the 3-layered boards containing a higher percentage of chemical pulp in the top layer.

The boards also differ in the values obtained at the point of rupture, *i.e.* in stretch at break, stress at break and tensile energy absorption. Tensile energy absorption is a measure of the toughness of a material, as it is the total energy required to break the material. The higher value of tensile energy absorption was obtained for the boards with a higher content of recycled fibres in the top layer (KOLIPRINT), in cross direction, which means that the boards are tougher in cross direction, whereas the boards with a higher content of chemical pulp (KROMOPAK) are tougher in machine direction. The grammage of the boards also influences tensile energy absorption. With the increase of grammage, tensile energy absorption increased, too. A strong linear relationship exists between these two properties. When increasing humidity, tensile energy absorption decreases, up to 2.5 times, in both directions. The decrease was higher in the boards with a higher content of chemical pulp in the top layer, than that occurring in the boards with a higher content of recycled fibres.

In cross direction, the boards are more extensible than in machine direction, whereas, in machine direction, they have

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higher tensile strength. It seems that grammage has no significant influence on extension at break. The composition of the top layer and layer thickness has some influence on the extension properties of the boards. The boards with a lower content of recycled fibres in the top layer (KROMOPAK) are less extensible and have lower values of extension at break, as

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Figure 6: Elastic modulus in machine (MD) and cross direction (CD) of board samples of different composition and grammage: KROMOPAK – 10% virgin fibres, 3 layers, 215-400 g/m<sup>2</sup>; KOLIPRINT – 20% virgin fibres, 3 layers, 250-450 g/m<sup>2</sup>; GRAFOPAK – 10% virgin fibres, 4 layers, 250-500 g/m<sup>2</sup> at 50% and 90% relative humidity

compared to the boards with a higher content of recycled fibres (KOLIPRINT and GRAFOPAK). With increasing relative humidity, the boards became more extensible, at 90% relative humidity, extension at break being higher – up to 20%, and in cross direction – up to 50%.



Figure 7: Yield stress in machine (MD) and cross direction (CD) of board samples of different composition and grammage: KROMOPAK – 10% virgin fibres, 3 layers, 215-400 g/m<sup>2</sup>; KOLIPRINT – 20% virgin fibres, 3 layers, 250-450 g/m<sup>2</sup>; GRAFOPAK – 10% virgin fibres, 4 layers, 250-500 g/m<sup>2</sup> at 50% and 90% relative humidity



Figure 8: Tensile energy absorption in machine (MD) and cross direction (CD) of board samples of different composition and grammage: KROMOPAK – 10% virgin fibres, 3 layers, 215-400 g/m<sup>2</sup>; KOLIPRINT – 20% virgin fibres, 3 layers, 250-450 g/m<sup>2</sup>; GRAFOPAK – 10% virgin fibres, 4 layers, 250-500 g/m<sup>2</sup> at 50% and 90% relative humidity



Figure 9: Tensile strength in machine (MD) and cross direction (CD) of board samples of different composition and grammage: KROMOPAK – 10% virgin fibres, 3 layers, 215-400 g/m<sup>2</sup>; KOLIPRINT – 20% virgin fibres, 3 layers, 250-450 g/m<sup>2</sup>; GRAFOPAK – 10% virgin fibres, 4 layers, 250-500 g/m<sup>2</sup> at 50% and 90% relative humidity

The tensile strength of the boards, similarly with tensile energy absorption, shows a strong linear relationship with grammage. They both increase with the basic mass, the correlation being higher than 0.9. Figure 9 shows that the higher content of chemical pulp in the top layer has a greater influence on tensile strength than the addition of a fourth layer. At 90% relative humidity, tensile strength is up to 2.5 times lower in both directions. Also, at 90% relative humidity, the boards with a higher content of chemical pulp in the top layer reach higher values of tensile strength than the boards with an additional fourth layer.

#### CONCLUSIONS

The analyses have shown that the coated boards produced mostly from post-consumer recycled fibres possess adequate surface and mechanical properties. It has been found out that a modification in the composition of the top layer has some influence on the properties of coated boards. Boards with a 10% higher amount of chemical pulp in the top layer have higher bending stiffness, tensile strength and elastic modulus in both directions, and lower extensibility and tensile energy absorption in machine direction. These boards also have the best surface properties, such as the lowest roughness, the highest brightness and gloss. The addition of a fourth layer, made from post-consumer recycled fibres, improves the tearing strength of the boards, although it has less influence on the mechanical properties than the addition of a higher percentage of chemical pulp in the top layer.

The mechanical properties of coated are strongly related to their boards grammage. A strong linear relationship was obtained between properties, such as bending stiffness. tearing resistance, bursting moduli, vield stress, strength. tensile strength, tensile energy absorption, and basic mass. The existence of a strong linear relationship suggests that the stiffness and strength of boards could be very well predicted for arbitrarily chosen board grammage.

The influence of a 90% relative humidity on the tensile properties of the boards is significant. Besides lowering the tensile breaking properties, with the decrease of elastic modulus and yield stress, the resistance to stretching and the limit between the elastic and non-recoverable extension is lowered, too. In the boards containing a higher percentage of chemical pulp in the top layer, the decrease in tensile strength and elastic modulus is less pronounced, compared to the boards containing a higher

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percentage of recycled fibres.

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