Mechanical Characterization of The Coating Layer of The Coated Paper

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ABSTRACT

Goal of this work was evaluation of mechanical properties of the coating layer on coated papers.
The estimated mechanical and physical parameters of the coating layer were the Young's modulus (E) and the strain (ε). These two parameters cannot be measured directly for a pigmented coating layer, and therefore were derived from a series of mechanical tests performed on both the coated paper and the base paper. Measurements in tensile strength, two-point's bending stiffness and dimensional stability have been performed. The methodology used involves the principles of a three-layered structure, in which all layers are assumed to be uniform and homogeneous.
The results showed a greater Young's modulus and a lower strain for the coating layer as compared to the base paper. It was shown that the coating layer acts as a mechanical reinforcement of the complex, performing higher strength, particularly for the tensile strength and the bending stiffness. The coating layer improves the stability of the material, when coated paper is submitted at high relative humidity.

Introduction

There are many kinds of coated paper grades depending on their final use. Paper with pigmented coatings designed for printing is one of the most common. The pigmented coatings were developed to provide a paper surface with better appearance and printability (1).

During the printing processes and converting operations, e.g., folding and book binding, mechanical properties are also important, since the paper is submitted to great stress and strain (2). For good paper runnability, without breaks production, tensile strength and bending stiffness are very important properties, in particular during printing and finishing operations (3). Dimensional stability is also important performance parameter in paper converting and utilization since paper is very hygroscopic (4). Consequently, when coated paper is submitted to different relative humidity environments, its mechanical behavior may decline; in consequence, breaks and printing quality problems may occur.

The Young's modulus is a mechanical parameter which provides a measure of the rigidity of a solid material. It is especially important given that it is associated with several other mechanical properties. Young's modulus of the coating layer significantly affects the tensile strength, stiffness and dimensional stability of the coated paper, since the coating layer can provide a higher powerful linkage during mechanical behavior than the base paper. However, as the density of the coating layer is two times larger than that of the base paper, the mechanical properties may decrease when the coat weight increases.

Plentiful research has been done in printability of coated paper and less emphasis has been put in mechanical properties (5). Nevertheless, many studies have been published reporting the study of mechanical properties of coated paper (6-8), but not regarding the mechanical properties of the coating layer.

The knowledge of mechanical properties of the coating layer will allow estimating its contribution on coated paper properties and afterward improving the mechanical performance of the coated paper in the printing and converting processes. Some researchers have investigated the mechanical parameters of the coating layer by using base films instead of paper (9, 10). The results showed overload mechanical values for the coating layer, because this approach does not take into account the permeability and porosity of the base paper since these films are impermeable.

In the present study it is assumed that coated paper is a laminate with parallel layers. Two side coated paper (C_2S) has a central layer corresponding to the base paper, this is composed by a fibrous material with preferred in-plane orientation which can contain fillers. The two outside surface layers correspond to the coating, a mix of pigments, adhesive and some others additives. The three layers are bonded by complex interfaces, where adhesion and mechanical phenomena may play an important role. These interfaces are formed due to the penetration of the coating into the base paper. However, in this study these interfaces were not taken into account.

In order to simplify this study, the complex C_2S is considered symmetric. It is also assumed that the neutral plane is located at the middle of the complex. Consequently, coated paper can be considered an orthotropic composite in which, the inner and outer layers have their own mechanical behavior, since they are formed by different materials.

Theory

The mechanical properties of a laminate material depend on the

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properties of its constituents and their distribution, as well **as** on physical and chemical interactions.

In this paper, we carried out a mechanical analyses of the C_2S considering that the coated paper is a symmetrical composite sandwich structure. A macro mechanical approach was followed and all layers were assumed uniforms and homogeneous, consisting of a paper sheet as a middle layer (the support or base paper) and two porous coating layers (top and bottom layers), as it is shown in **Fig. 1**. In this schematic, E_s is the Young's modulus of the base paper and E_c represents the Young's modulus of the coating layer which we intended to estimate. The thickness of the support and coating layers are expressed by t_s and t_c, respectively. The corresponding strain was denoted by ε_s and ε_c, likewise for the support and the coating. In that order, L_0 and L_f correspond to the initial and final length of the complex.

Tensile strength behavior of a complex material like a coated paper can be seen in terms of the Young's modulus of each of its constituents. The relative ratio of each constituent can lead us to the equivalent elastic module of the material. Thus, we admit that the force acting on the coated paper during the trial can be decomposed in two constituents: one component due to the base paper (F_s) and the other supported by the covering coating (F_c), which can be given by the following equations:

$$F_s = E_s \times t_s \times \varepsilon_s \qquad [1]$$

$$F_c = E_c \times t_c \times \varepsilon_c \qquad [2]$$

Admitting that both deformations are equal, given that are submitted to traction/elongation at the same time, the equivalent Young's modulus for the coated paper is given by the next equation:

$$E_{cp} = \frac{E_s \times t_s + 2 \times E_c \times t_c}{t_s + 2 \times t_c} \qquad [3]$$

Where the subscript "cp" refers to coated paper.

Dimensional stability is a measure of paper's tendency to stretch or shrink, especially when affected by changes in moisture content produced by the wetting in the printing process, or even with over time in high relative humidity storage.

Some mechanical properties have potential to foresee dimensional stability. DeRuvo et al. (11) had correlated Young's modulus with dimensional stability. Silvy (12) used a more practical method foreseeing the dimensional stability of papers using the mechanical properties.

Dimensional behavior in the plan of a coated paper was studied utilizing the same model previously described. The central layer is essentially constituted by cellulose, which has great affinity for water and thus increasing the possibility of strain. The two exterior layers, corresponding to the coating, are of hydrophobic material, and therefore they are more resistant to humidity changes. Subsequently we can assume that in the complex there are opposite forces. On one hand we have the free strain of the base paper ($L_0\ \varepsilon_s$) and on the other hand the free strain of the coating ($L_0.\varepsilon_c$) which slows down the strain of the base paper. The true final length is given by the strain occurred into the coated paper. The residual strain of each layer can be given by the following equations:

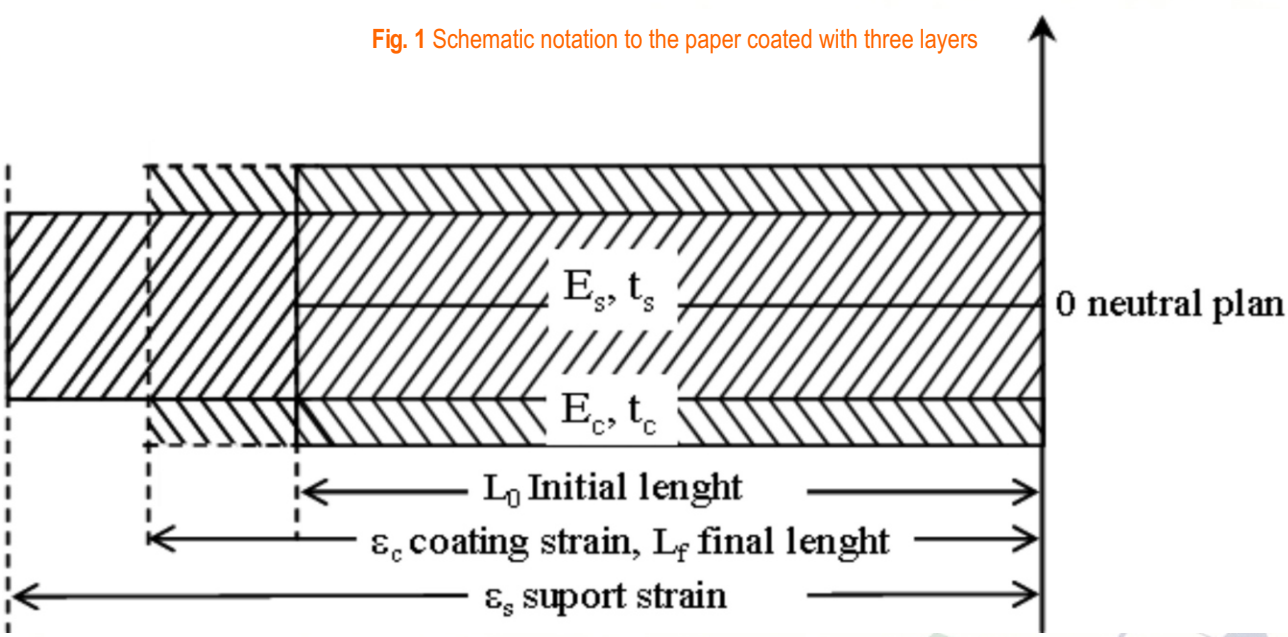
$$\varepsilon_{residual\ support} = \frac{L_0 \times (1+\varepsilon_s) - L_f}{L_0} \qquad [4]$$

$$\varepsilon_{residual\ coating} = \frac{L_f - L_0 \times (1+\varepsilon_c)}{L_0} \qquad [5]$$

As the three layers of the coated paper are submitted to the same force at the same time, the actuate forces in the base paper and coatings are equal and given by the following equations:

$$F_s = E_s \times t_s \times \left[\frac{L_0 \times (1+\varepsilon_s) - L_f}{L_0}\right] \qquad [6]$$

Fig. 1 Schematic notation to the paper coated with three layers



$$F_c = E_c \times t_c \times \left[\frac{L_f - L_0 \times (1 + \varepsilon_c)}{L_0} \right] \quad [7]$$

The final equation to calculate the coated paper strain (ε_{cp}) is given as following:

$$\varepsilon_{cp} = \frac{E_s \times t_s \times \varepsilon_s + 2 \times E_c \times t_c \times \varepsilon_c}{E_s \times t_s + 2 \times E_c \times t_c} \quad [8]$$

Bending stiffness behavior is a tension with small deformation perpendicularly to the plane of paper, where the outer surface part of the bending is stressed and the inner surface part is compressed. The zone called the neutral plane is found between these two parts, which is not subject to any type of force. In a factual homogeneous and symmetrical material this axis is situated in the geometric centre.

In the particular case, where elastic properties are assumed constant along the sample thickness, i.e., in the perpendicular direction to the plane, the equation for pure bending stiffness (S) can be simplified and written as (13).

$$S = \frac{E \times t^3}{12} \quad [9]$$

However, for many materials like coated papers, this equation cannot be applied, because the elastic properties change in the z direction (thickness). In these cases, the stiffness of the material may be given by addition of all layers. In symmetrical structures with three layers, bending stiffness can be determined through the following equation:

$$S_{cp} = \frac{E_c \times t_{cp}^3 - E_c \times t_s^3 + E_s \times t_s^3}{12} \quad [10]$$

Where "S_{cp}" refers to the stiffness of coated paper.

Kajanto (14) gave a similar equation to estimate bending stiffness of a three-layer structure.

RESULTS AND DISCUSSION

We have measured the properties described previously for the base paper of each of the coated papers in order to minimize errors, as well as the properties of the papers coated with different formulations.

Table 1 shows an example (**$P_{CW}B_{16}C_{A75}$-MD**) of the calculation performed in accordance with the developed algorithm. Estimated values of mechanical parameters for the coating layer and coated paper are represented in italic data. The sum of relative deviations between all of the calculated and experimental values are not presented but were always below 5% for every one of the iterative processes.

We observed a higher Young's modulus for the coating layer as compared to that of the base and coated papers. This result was expected since the experimental Young's modulus of the coated paper is greater than that of the base paper. The coating layer contributed to an improvement in the mechanical properties analyzed. Concerning the strain, the coating layer has approximately the same deformation as the coated papers and base paper. In this particular case **the strain values are small** and similar, so, for this particular example, no strong conclusion can be made.

The overall results obtained for Young's modulus and strain are shown in **Table 2.**

We can observe different values of the Young's modulus and strain for the coating layer in both directions of the plane. That is, the Young's modulus of the coating layer is greater for machine direction (MD) than for cross direction (CD) and the strain is the opposite. Although the Young's modulus and strain of the coating layer are quite different in both paper directions, similar values were obtained for the coating layer between formulations. These results show that the differences in the constitution of the formulations are not significant to affect the final value of the estimated parameters. Concerning the anisotropy of the coated paper, it seems that the arrangement of the coating material composition gives a rather homogeneous layer, leading lower Young's modulus ratio between both directions of the paper. The base paper is undoubtedly an anisotropic material due to the fibres orientation, giving a MD/CD ratio exceeding two units; however, the coating layer reduces slightly this anisotropy on coated papers. Coated papers show a Young's modulus ratio of approximately two and the coated layer

Table 1
Results obtained in the iterative calculus for $P_{CW1}B_{16}C_{A75}$-MD

Papers	Experimental paper properties		Estimated properties for:	
Base paper of $P_{CW1}B_{16}C_{A75}$-MD	Thickness(mm)	0.135	**coating layer**	
	Y. modulus (MPa)	5328	*Y. modulus (MPa)*	*6900*
	Strain (%)	0.090	*Strain (%)*	*0.090*
	Bending stiffness (mNm)	0.980	**paper coated**	
$P_{CW1}B_{16}C_{A75}$-MD	Thickness(mm)	0.137	*Coating thickness(?m)*	*1.65*
	Y. modulus (MPa)	5366	*Y. modulus(MPa)*	*5366*
	Strain (%)	0.085	*Strain (%)*	*0.090*
	Bending stiffness (mNm)	1.153	*Stiffness (mNm)*	*1.156*
Base paper of $P_{CW2}B_{16}C_{A75}$-MD	Thickness(mm)	0.131		
	Y. modulus (MPa)	5254		
	Strain (%)	0.09		
	Bending stiffness (mNm)	1.050		
$P_{CW2}B_{16}C_{A75}$-MD	Thickness(mm)	0.140	*Coating thickness(?m)*	*6.95*
	Y. modulus (MPa)	5415	*Y. modulus(MPa)*	*5411*
	Strain (%)	0.090	*Strain (%)*	*0.090*
	Bending stiffness (mNm)	1.271	*Stiffness(mNm)*	*1.269*

Table 2
Young'S modulus and Strain for the coating layer

Formulations	MD		CD	
	E (MPa)	ε (%)	E (MPa)	ε (%)
$P_{CW1}B_{12}C_{A75}$	6900	0.09	4330	0.15
$P_{CW2}B_{12}C_{A75}$				
$P_{CW1}B_{12}C_{A65}$				
$P_{CW2}B_{12}C_{A65}$				
$P_{CW1}B_{16}C_{A75}$				
$P_{CW2}B_{16}C_{A75}$				
$P_{CW1}B_{16}C_{A65}$				
$P_{CW2}B_{16}C_{A65}$				

around one and a half.

Regarding **Table 2**, we can observe that the CD strain values are higher than the MD strain values. However, the gap between these values is lower for the coating layer than for the base and coated papers, for which the gap between the values exceeds twice as much. This outcome was also noticed on the overall experimental results for the base and coated papers, as seen in **Table 3.**

Table - 3
Young'S modulus and Strain for the Base and Coated papers

Base paper and Coated paper	Y. modulus (MPa)		Strain (%)	
	MD	CD	MD	CD
Base paper	5396	2495	0.09	0.22
$P_{CW1}B_{12}C_{A75}$	5548	2598	0.09	0.21
Base paper	5411	2701	0.08	0.18
$P_{CW3}B_{12}C_{A75}$	5585	2857	0.08	0.18
Base paper	5328	2791	0.09	0.17
$P_{CW1}B_{16}C_{A75}$	5366	2833	0.09	0.17
Base paper	5254	2620	0.09	0.17
$P_{CW3}B_{16}C_{A75}$	5415	2806	0.09	0.17
Base paper	5274	2529	0.09	0.23
$P_{CW1}B_{12}C_{A65}$	5310	2567	0.09	0.22
Base paper	5228	2609	0.09	0.22
$P_{CW3}B_{12}C_{A65}$	5376	2726	0.09	0.21
Base paper	5256	2635	0.09	0.18
$P_{CW1}B_{16}C_{A65}$	5451	2712	0.09	0.17
Base paper	5462	2501	0.09	0.16
$P_{CW3}B_{16}C_{A65}$	5660	2688	0.09	0.16

Still in relation to overall results, MD strain values are approximately the same for base, coated paper as well as for the coating layer. This result is due to the predominant effect of the base paper. Although, CD strain values are slightly lower for the coating layer when compared to the base paper and coated paper. This is a consequence of the small coating deformation effect as compared to the bigger fibre deformation.

Regarding the variables chosen for the coating formulations, it seems that the ranges of values chosen don't have a significant impact on the results. Nevertheless, some researchers have documented that mechanical properties depend on the polymeric binder, pigment type, shape and size distribution (15, 16). Also the coating packaging arising from the pigment shape and adhesive level may give different porosity and the effect of porous structure of coating on mechanical strength is also documented (17). Nevertheless, as already stated, in this study these effects were not perceived.

In future work, it would be interesting to perform formulations with a range of variables broader than would be possible.

Conclusions

The present study is based on the idea that the coating layer of a complex material can be advantageously analyzed, using simultaneously the concepts of mechanics of solids and an analytical calculation tool. Three equations, 3, 8 and 10, were developed in order to calculate Young's modulus and strain of a coating layer through the measurement data of the following properties: thickness, bending stiffness, tensile strength, and dimensional stability. An iterative method was used for solving these equations and estimate the unknowns, respectively Young modulus and strain of the coating layer.

Based on the results can be drawn the following conclusions:

Coating layer has a Young's modulus significantly higher than base and coated papers. The largest difference was observed in the cross direction of the coating layer, whose value is about two times higher than the values of base and coated papers.

Coating layer strain provides different outcomes for both directions of the paper. MD strain is the same as the base and coated papers, while the CD strain is smaller on the coating layer than on the base and coated papers.

The coating layer shows a lower anisotropy as compared with the base and coated papers, since its Young's modulus ratio is one and a half while the base and coated papers exhibit an average MD/CD of two. Also strain results show similar conclusion, the difference between SM and ST strain is smaller for the coating layer than for the base and coated papers.

When exposed to moisture or to external forces, coating layer undergoes less deformation than the base or coated papers. This means that when the coating is applied on the base paper, its mechanical properties improve in terms of Young's modulus and deformation.

Experimental

Materials

In this study different coating formulations were prepared and applied to both sides of the base paper. For the production of coated papers, industrial uncoated paper produced with *Eucalyptus globules* was used and four coating colors at 65% solids by weight dispersed in water. For all formulations the mass proportions of optical brightening (0.5%), insolubilizer (0.3%), cobinder (0.3%) and dispersant (0.07%) were kept constant. The relative amount between precipitated calcium carbonate (PCC) and kaolin, as well as, the percentage of binder, styrene / butadiene (S / B) latex and acrylic latex were changed, giving us two variables. **Table 4** display the coating color formulations, the relative amount between PCC and Kaolin varied according to the values 75/25 and 65/35. The binder content is expressed relatively to the pigments.

Table - 4 Formulation of the Four Coating Produced.

Products	Color 1	Color 2	Color 3	Color 4
PCC	75	65	75	65
Kaolin	25	35	25	35
Latex S/B	6	6	8	8

Latex acrylic	6	6	8	8

Each formulation was characterized in terms of solid content, viscosity, temperature and pH, in order to keep all these parameters almost constant. Work values were as follows: solids content of 65%, viscosity of 1000 mPas (at 100 rpm), temperature 20ºC and pH of 8.5.

The coating colors were applied on the base paper at the CTP (Centre Technique du Papier) pilot coater. The four formulations were applied at two coat weights (third variable), giving us eight different samples of coated paper. The nomenclature of the paper samples was symbolized as follows P_{CWX} - B_Y - $C_{AB,}$ where: $\mathbf{P_{CWX}}$ is referred to the paper with X **c**oat **w**eight; **X** assumed the values 1 (12 g/m^2) and 2 (20 g/m^2). $\mathbf{B_Y}$ is the binder content, **Y** refers to the percentage of binder relative to the pigments and assumed the values 12 or 16%. $\mathbf{C_{AB}}$ was the proportion of pigment A (PCC) relative to the total pigment and took the value of 75 (75% PCC and 25% Kaolin) or 65 (65% PCC and 35% Kaolin). These coated papers and the base paper were tested in terms of structural and mechanical properties, after conditioning at 50% of relative humidity and 23 ºC of temperature. Making use of the mechanical tests, such as, tensile strength, bending stiffness and dimensional stability, we have analysed the coated and uncoated papers.

Using the values of Young's modulus and strain resultants from the mechanical tests we try to estimate mechanical parameters of the coating layer. For this propose, the uncalendered C_2S was considered as a laminar structure, formed by three homogeneous and uniform layers.

Methods

The uniaxial tensile strength was performed according to the ISO 1924/1 Standard. The measurements were carried out using a dynamometer Instron universal testing machine. Tensile tester measures the load applied as a function of elongation of the sample strip. Bending stiffness was measured using a L&W bending tester with the ISO 2493 Standard. This equipment utilizes the two point's principle in which one end of the sample is fastened in a clamp and the sample is loaded at a given distance from the clamp. The sample is forced to curve with a bending angle pre-established. The measurements were carried out under the following conditions: bending angle of 15º, length of 25 mm, width 38,1 mm and a bending rate of 5º/sec.

Dimensional stability measurements were carried out at the Varidim system, developed by Serra Tosio and Chaves (18), according to ISO 8226 Standard and follow TECHPAP technical information (19). This equipment gives the length variation in real time and can work in a moisture range between 15 and 90%. In the present study, a plan of humidity was used with the following points: 20, 50, 65, 85, 65, 50 and 20%, each humidity has been maintained for 75 minutes. The thickness of the samples was measured using a micrometer tester and following the Standard **ISO 534:1988.**

In order to determine the coating layer parameters, Equations 3, 8 and 10 were used. As we have two different coat weights for each of the formulations and assuming that the Young's modulus and strain are the same for each formulation and independent of the coat weight (9, 20), we have six equations (three per coat weight) with three unknown parameters, the Young's modulus (E_c), the strain (ε_c) and the thickness (t_c) of the coating layer. In order to solve this system of equations, an iterative approach in Excel solver was used.

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