

Factors Affecting the Dimensional Stability of Decorative Papers under Moistening

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A crucial problem for laminate producers is the dimensional instability of decorative papers during soaking in aqueous solutions, but the source of this dilemma is not completely understood yet. In this study, eight commercial decorative papers of similar fiber composition and sizing were analyzed for their structural, physical, and mechanical properties. These properties were examined for their correlations to the dimensional stability of papers when moistened, as assessed by the wet stretch dynamics. Structure-to-property relationships were evaluated by principal component analysis (PCA). Within the set of parameters examined, PCA revealed that fiber orientation and the content of fillers/pigments influenced the wet expansion of paper web and affected its margins and dimensions in longitudinal and transverse directions of the paper machine. These variables are discussed within the context of decorative paper engineering in order to produce high performance papers with regular wet expansion properties.

Keywords: Decorative paper; Dimensional stability; Swelling; Wet expansion; Principal component analysis

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INTRODUCTION

Laminate surfaces are materials of choice for a wide range of applications, as they provide functional and decorative solutions for different household and commercial requirements (Barash 2008; Dombo *et al.* 2010). Conventionally, decorative laminates contain three distinct layers of resin-impregnated papers (Fig. 1) joined together under heat and pressure until the resins are cured: (i) a core of kraft paper sheets impregnated with phenolic resin; (ii) a decorative paper with a printed design impregnated with melamine resin and; (iii) a translucent protective overlay sheet also saturated with melamine resin (Roberts and Evans 2005; Barash 2008; Nemli 2008; Kandelbauer and Teischinger 2010). Decorative laminates are widely used on kitchen counters, desk and table tops, and facing for walls, doors, and flooring, (Nemli 2008; Figueiredo *et al.* 2011) offering an elegant aesthetical effect with different textures, patterns, colors, and designs (Barash 2008). These features, combined with durability, easy maintenance, low cost, and high resistance to wear, stains, and abrasions, have led to a growing demand for laminates that have unique properties for indoor and outdoor design applications (Figueiredo *et al.* 2011). The increasing global trend dictates that laminates producers provide well-engineered products, where the high standards of surface appearance represent a major quality criterion.

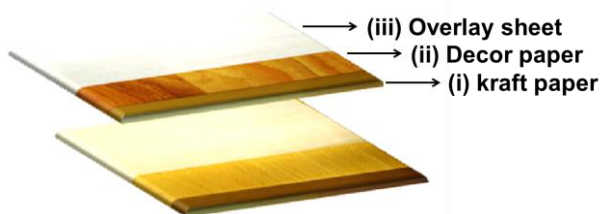


Fig. 1. Typical assembly of two decorative laminates

A crucial problem for laminates producers is the dimensional instability of resin-impregnated decorative papers. Dimensional instability strongly influences the end-use performance of paper, primarily in high-speed converting processes (Byrd 1972), causing paper curl, excessive shrinkage, and pattern misregister during the impregnated paper drying stage. During soaking in aqueous media, hygroscopic paper expands due to the swelling of fiber cell walls. Conversely, when the paper web is dried, it suffers linear deformation, contraction, or shrinkage (Uesaka *et al.* 1989; Nielsen and Priest 1997; Haslach 2000). Both wet expansion and shrinkage phenomena contribute irregularities and misfits on decorative paper patterns that strongly affect the final laminate surface quality, especially on wooden pattern floorings. For the latter applications, the docking of image patterns between two laminate pieces becomes difficult, if possible. Hence the wet expansion is a critical parameter for decorative papers with complex graphic images.

Due to the anisotropic orientation of the fiber network and to tensile stress, the wet expansion in the paper machine direction (MD) is generally less than in the cross direction (CD) (Nielsen and Priest 1997), leading to critical deviations from the specified paper margins that may lead to product inadequacy (Fig. 2) (Kandelbauer and Teischinger 2009). This behavior strongly influences the final design and appearance of a decorative laminate. The finished product performance depends not only on the impregnation process parameters but also on the specific qualities of raw papers and resins (Roberts and Evans 2005; Kandelbauer and Teischinger 2010; Bardak *et al.* 2011; Figueiredo *et al.* 2011). Thus, the factors influencing the dimensional stability of decorative papers require further research.

Conventionally, the dimensional stability of paper is assessed by measuring the hydroexpansion and/or hydrocontraction, which are related to atmospheric relative humidity and liquid (water), respectively. However, resin impregnation of decorative papers is a high-speed process under tension, and this information on the hydroexpansion and/or hydrocontraction is not completely adequate. Therefore, evaluating the dynamic wet expansion of paper under tension is a strategic procedure of utmost importance.

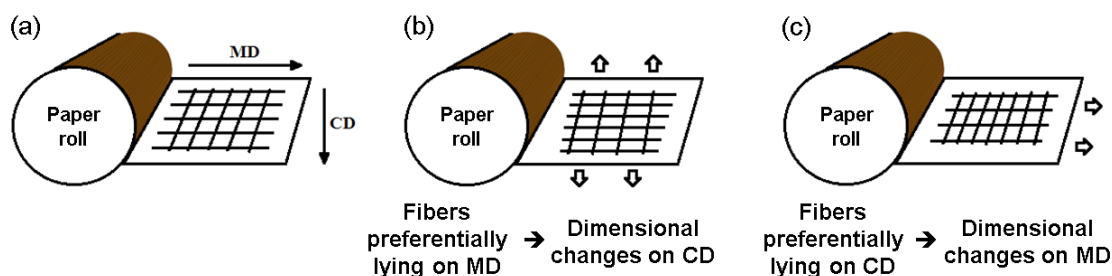


Fig. 2. Principal directions of paper web related to paper roll (a) and critical dimensional changes depending on predominant fiber orientation along the paper machine direction (b), or along the cross direction of paper machine (c)

In this study, multivariate analysis was used to assess the physical and structural properties that are responsible for decorative paper dimensional instability. Non-impregnated decorative paper sheets were characterized in terms of wet stretch dynamics and their structural, physical, and absorption properties. These properties were studied by principal component analysis (PCA) and correlated to paper web wet expansion.

EXPERIMENTAL

Materials

Samples of eight decorative papers (70 to 80 g/m²) with typical wooden patterns (W), used for the production of laminates by SONAE - Indústria de Revestimentos and supplied by different producers, were conditioned at 20 °C and 60% RH for 48 h. According to the producers' information, the papers were produced using similar proportions of hardwood/softwood bleached kraft pulps and contained low amounts of sizing agents (1%). The specific properties of the analyzed paper samples are presented in Table 1.

Table 1. Structural Properties of Wooden Patterns (W) Decorative Papers

| Properties | W ₁ | W ₂ | W ₃ | W ₄ | W ₅ | W ₆ | W ₇ | W ₈ |
|------------------------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|
| Grammage (g/m ²) | 70 | 70 | 70 | 70 | 70 | 70 | 80 | 80 |
| Bulk (cm ³ /g) | 1.14 | 1.15 | 1.14 | 1.12 | 1.13 | 1.15 | 1.07 | 1.10 |
| Ash Content (%) | 32.3 | 29.5 | 25.1 | 23.4 | 36.0 | 31.9 | 35.2 | 29.8 |
| Orientation (%) | 64.1 | 58.3 | 58.5 | 63.9 | 59.2 | 49.1 | 63.3 | 81.1 |

Physical and Mechanical Properties Analysis Methods

Grammage was determined in accordance with ISO 536. Thickness, density, and bulk were measured following ISO 534. Tensile strength and tear resistance were analyzed according to ISO 1924-2 and ISO 1974, respectively. Capillarity was determined using the Klemm method according to ISO 8787, and air permeation was measured by the Gurley method according to ISO 5636-5. Inter-fiber bonding was evaluated using the Scott test according to TAPPI T569 pm-00. All samples were analyzed in the longitudinal (MD) and cross directions (CD) of the paper machine (Fig. 2a). The fiber orientation was evaluated in percentage by ratio of the tensile stiffness index on CD and MD (Vahey *et al.* 2008). Higher fiber orientation is associated with higher paper anisotropy. The inorganic fillers/pigments content was determined by calcination at 525 °C, according to TAPPI T211 om-12.

Wet Expansion

Dimensional stability was evaluated measuring the dynamic wet expansion of decorative papers under tension using a wet stretch dynamics analyzer (model WSD 02, EMTEC, Leipzig, Germany). Paper strips of standard dimensions (210 mm x 60 mm) were tested for expansion in the CD and MD directions by one-sided contact with distilled water under adjustable tension (CD = 1 N; MD = 4 N). The paper surface exposed to the water was a 60 x 60 mm area, which was defined by a waterproof marker to keep the water inside the defined area. The paper strip was clamped to the device, and water penetration started after the paper contacted the water. During testing, the Emtec Measurement System software (EMTEC, Leipzig, Germany) recorded an expansion versus time graph, and the

wet expansion was expressed as a percentage. The average value was calculated from six measurements over paper web's half width.

Principal Component Analysis (PCA)

Principal component analysis is a multivariate data analysis method used to reduce the dimensionality of a large set of correlated variables into a new set of uncorrelated variables, the principal components (PCs). The PCs are ordered so that the first few retain most of the variation present in the original set of variables. Hence, the purpose is to explain the maximum amount of variance with the fewest number of PCs without considerable loss of information, *i.e.*, to simplify the original large data set by finding the dominant variables within it (Jolliffe 1986; Wold *et al.* 1987; Esbensen and Geladi 2010). Thus, PCA was used to detect the main physical properties affecting dimensional stability of decorative papers. Multivariate analysis (MVA) was computed using specific MVA software, Unscrambler® X 10.1 (CAMO, Oslo, Norway), that enables advanced regression, classification, and predictive modeling tools including PCA. Each analyzed sample corresponded to an object consisting of a data vector of 11 variables that represented physical properties. The dominant patterns were expressed in terms of a set of loadings and score plots.

RESULTS AND DISCUSSION

Dimensional Stability of Decorative Papers

During the impregnation of decorative papers with water-based resins and the consecutive drying stages, the margin dimensions are changed depending on dimensional stability. The irregularities in dimensional stability of different decorative papers are not fully understood, and they lead to the unplanned shutdowns of impregnation lines and production delays. Hence, the dimensional stability of decorative papers under tension is a crucial property of paper structure.

Eight selected decorative papers with wooden design patterns (W) were supplied by different producers; the properties affecting their dimensional stability were examined. Structural and physical properties were evaluated according to wet dynamic expansion (Fig. 3) and standard methods in the range mentioned in Table 2; PCA determined relationships between these properties and wet expansion parameters.

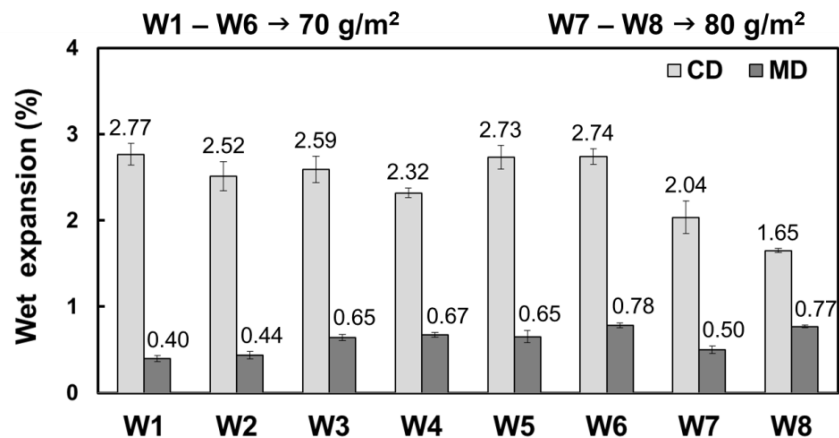


Fig. 3. Wet expansion of the 8 decorative papers measured on cross (CD) and machine (MD) directions. The grammage of various papers is indicated above the graph.

Due to the operational conditions on a paper machine, there is a tendency for fibers to become aligned along the machine direction (MD) (Brazier 1993; Ingalsbe 2001; Vahey *et al.* 2008). This anisotropic orientation of the fibers along MD leads to physical properties quite different from those measured in the cross machine direction (CD), and for this reason, all samples were tested along both MD and CD. Due to differences in the inter-fiber bonding in MD and CD, wet expansion in CD was 3 to 7 times higher than in MD. A notable difference in the wet expansion deviations along MD and CD (Fig. 4) can be explained by much higher anisotropy of fiber orientation in the CD than in the MD.

Table 2. Parameters of Decorative Papers Studied by PCA

| PC Number | Variable | Abbreviation | Range |
|-----------|---|--------------|-----------|
| PC1 | Ashes (%) | ASH | 23.0-36.0 |
| PC2 | Inter-fiber Bonding (J/m ²) | BOND | 43.0-73.0 |
| PC3 | Bulk (cm ³ /g) | BULK | 1.05-1.16 |
| PC4 | Capillarity (mm) | CAPIL | 18.0-27.0 |
| PC5 | Elongation (%) | ELONG | 1.0-4.0 |
| PC6 | Orientation (%) | ORIENT | 49.0-81.5 |
| PC7 | Air Permeability Gurley (s) | POROS | 15.0-24.0 |
| PC8 | Tensile Energy Absorption Index (J/g) | T.E.A. | 0.1-0.5 |
| PC9 | Tear Index (mN.m ² /g) | TEAR | 0.4-0.7 |
| PC10 | Tensile Index (N.m/g) | TENSILE | 14.0-35.0 |
| PC11 | Wet Expansion (%) | WET EXP | 0.40-2.78 |

Relationships between Variables

The properties affecting web dimensional stability were evaluated by PCA. The number of principal components (PC) was assessed by determining the percentage of at least 95% of the explained variation. PC1 through PC6 corresponded to *ca.* 97% of the total variability in both MD and CD, while PC7 through PC11 only accounted for about 3% (Table 2; Fig. 4). Nevertheless, plotting data sets with respect to the first two PCs provided a realistic representation of the data, revealing which parameters contributed the most to deviances. For instance, variables that lie close to each other in these plots exhibit a high degree of correlation. Because PC1 and PC2 represent the largest variations in the X-data (*ca.* 60%), the loadings and score plots distributed along the PC1-PC2 plane (Fig. 6) are the most useful for interpreting the existent correlations. Thus, only these planes were considered for further analysis. As all eight samples corresponded to decorative papers with similar patterns, the interpretation of the score plots was also omitted.

Relationships between the 11 analyzed variables can be identified on the loading plots for MD and CD (Fig. 5). A general analysis of the data for the MD and CD reveals some common patterns of distribution. On both plots, there was a constellation of T.E.A., ELONG, TENSILE, TEAR, and BOND variables lying close to the straight horizontal line intercepting the origin and characterized by high negative loadings on PC1 and residual contribution to PC2. These variables were strongly correlated to each other, indicating that papers with well-bonded fibers (BOND) exhibited higher tensile strength (TENSILE) and tear resistance (TEAR), and the elongation at break (ELONG) implied that more energy was required to break a sheet of paper (T.E.A.). These correlations conform to the general postulates of paper physics (D'A Clark 1985; Borch *et al.* 2002).

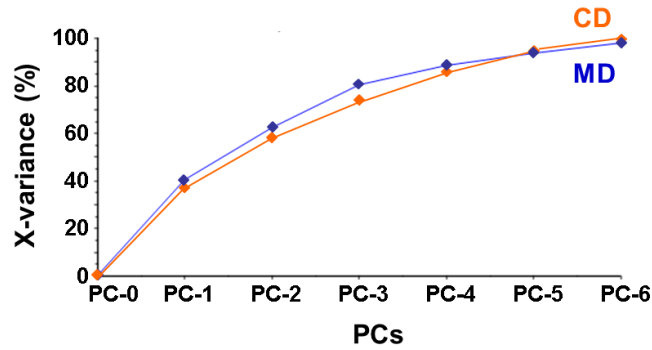


Fig. 4. Explained variance in the MD and CD for individual principal components

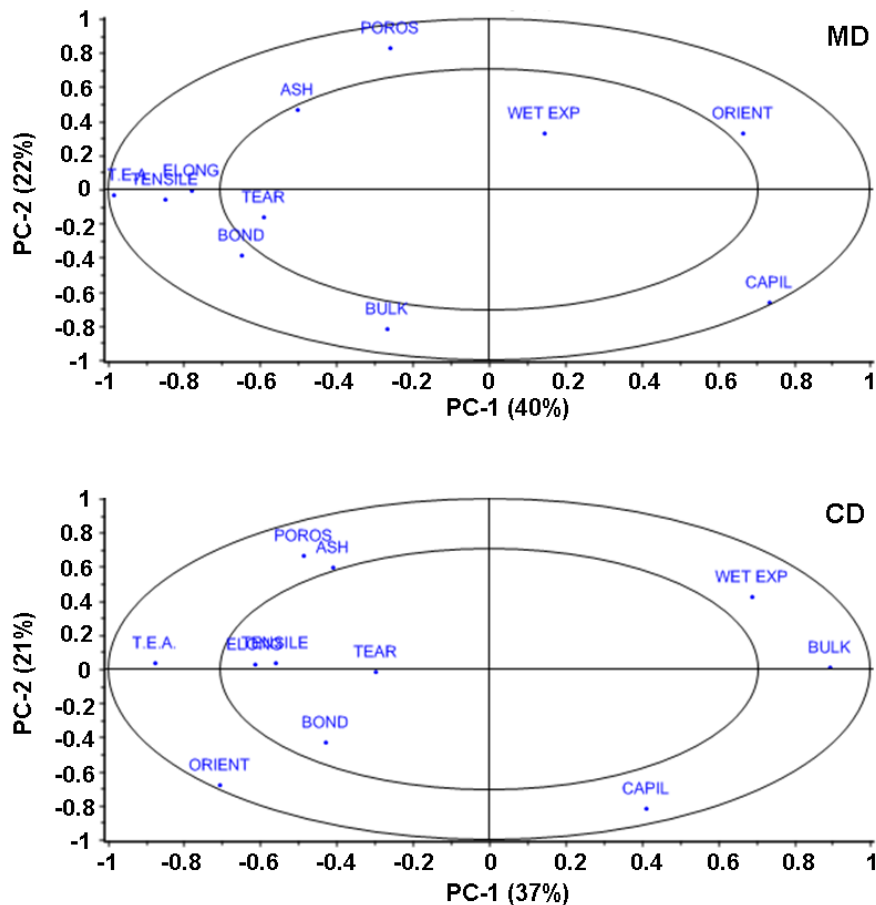


Fig. 5. Correlation loadings of wood-patterned paper samples in the PC1-PC2 plane for machine (MD) and cross (CD) directions

The distribution of this set of variables shows that the first principal component (PC1) was strongly related to the mechanical strength properties of decorative papers. Furthermore, the aforementioned variables were independent of capillarity (CAPIL), ash content (ASH), and porosity (POROS), which had high contributions to PC2 related to the water sorption properties of decorative papers. Some of the variables affecting the sorption ability (CAPIL and ASH/POROS) are located in diametrically opposed quadrants of the diagram, indicating a strong negative correlation between them, *i.e.* papers with the

excessive addition of inorganic fillers (ASH) exhibit higher Gurley test values (POROS), as an indication of higher air permeation of papers but lower capillarity (CAPIL).

The first relationship between ASH and POROS was expected because high ash content is associated with weakened internal hydrogen bonding (BOND) between fibers, which favors air permeation through the paper sheet. The second notable relationship between ASH and CAPIL is less evident and may be due to the strong interaction between fiber surfaces and inorganic particles. Thus, inorganic fillers adsorbed on the fiber surface not only weakened inter-fiber bonding, but also disturbed/deviated the capillary paths in the inter-fiber space. Rather than completely filling void spaces (pores, channels, *etc.*), water flows through paper as a bulk film that moves along channels formed by fiber overlaps (Roberts *et al.* 2003). This flow is interrupted by impermeable interfaces formed between inorganic particles and fibers (Portugal *et al.* 2010). Hence, inorganic fillers might decrease the water conductivity of paper, which is expressed as capillarity value (CAPIL) (Fig. 6). Apparently, the combined aforementioned factors led to a more pronounced wet expansion value (WET EXP) in the MD and CD directions, while the amount of inorganic filler increased (Fig. 6). Due to much lower expansion capacity of inorganic fillers than cellulosic fibers, the paper wet expansion grew slowly with the increase of filler content. Thus, papers with high inorganic filler contents (ASH) have weakened inter-fiber bonding (BOND) and higher air permeation (POROS). In turn, weak inter-fiber bonding indicates that during moistening, cell wall expansion changes the external dimensions of the sheet.

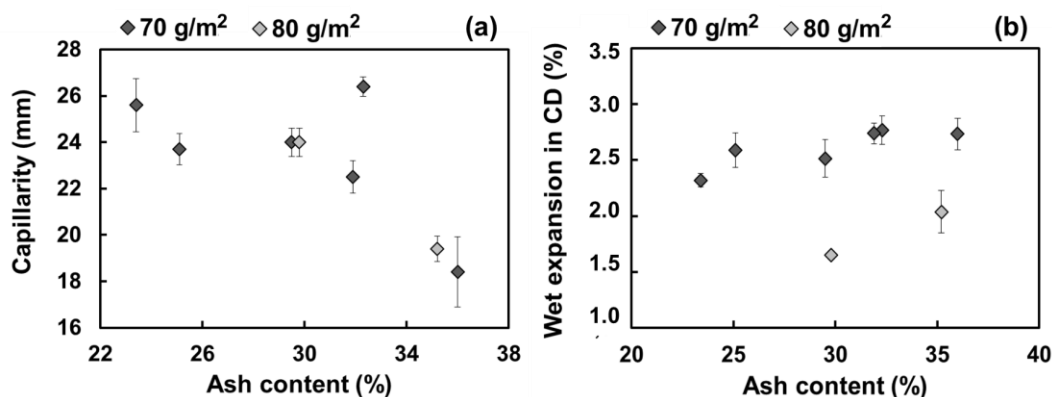


Fig. 6. Influence of paper fillers/pigments on (a) capillarity and (b) wet expansion in CD (including the deviation error bars)

Relationships between Paper Structure and Wet Expansion

The position of the ORIENT data point on both MD and CD loading plots suggests that the fiber orientation anisotropy is related either to PC1 or PC2, which exert strong influences on the mechanical properties and wet expansion of decorative papers, respectively. Samples with high ORIENT values exhibited an increased amount of fibers lying along the CD of the paper web and, consequently, presented enhanced mechanical properties on CD (Fig. 5). In regards to the WET EXP, papers with high ORIENT values and an increased proportion of fibers aligned in the CD had an increased tendency to swell along the MD and a decreased propensity to expand along the CD. This effect was confirmed by location of WET EXP and ORIENT in the same quadrant of the MD PCA plot and in the opposite quadrants in the CD plot (Fig. 5). Moreover, analysis of the MD loading plot shows that the variable WET EXP is poorly represented in the PC1-PC2 plane

because it is located in the inner circle of the ellipse. Thus, in this direction wet expansion does not contain enough structured variation to be discriminating for the decorative samples, as it is not influenced by either of the other depicted properties.

In general, the results confirm (Fig. 3) that wet expansion in the CD of decorative paper is crucial when compared to that in MD (Brazier 1993). Hence, the dimensional stability in the CD predetermines the wet expansion behavior of paper and may be improved while controlling the fiber orientation anisotropy by appropriately operating the paper machine. It is important to maintain this fiber orientation to avoid significant variations in the dimensional stability of produced papers. However, the improvement of strength properties in CD (increase in ORIENT parameter) should not critically weaken the mechanical properties of paper in MD that may result in the diminishing of tear resistance of the paper sheet during the impregnation process where the paper is always under tension. These factors must be balanced in order to provide resistant and dimensionally stable impregnated decorative sheets.

In addition to fiber orientation, the inorganic filler content affects the dimensional stability and should be considered during paper engineering. Increased inorganic filler retention in paper deteriorates its dimensional stability in the CD (Fig. 6). As the price of filler is typically around one third that of pulp fibers, however, diminishing the retained inorganic fillers/pigments in order to decrease the paper wet expansion increases the cost of paper production, and it may also negatively affect its consumer properties (opacity, printing quality, *etc.*). In any case, controlling the inorganic filler retention in decorative paper is important for maintaining its dimensional stability at the desired level.

CONCLUSIONS

1. Principle component analysis (PCA) of eight commercial decorative papers of similar fiber composition and sizing degree showed that their wet expansion under tension was strongly affected by the fiber orientation and filler/pigment content.
2. The wet expansion of decorative papers under moistening by water in the cross machine direction (CD) was up to 7 times higher than in the machine direction (MD), thus predetermining the wet expansion behavior.
3. Both the excessive addition of inorganic fillers and the predominant alignment of fibers along the machine direction (MD) were responsible for diminishing the paper web dimensional stability in the cross machine direction (CD) under moistening. These factors contributed to an increased change in the external dimensions of the paper sheet in this direction during the impregnation process, when the web was under tension.
4. A rough comparison of dimensional stability in decorative papers of the same grammage was performed by a simple capillarity test in the CD of paper. Lower capillarity was associated with higher wet expansion in CD. This quick test can alarm the laminate producers for eventual changes in wet expansion of newly supplied decorative paper, thus obligating them to take measures for correction of operation conditions during paper impregnation with aqueous resin solutions to control the wet expansion of impregnated paper.
5. The discussed effect of fiber orientation and the content of fillers/pigments on the wet expansion of decorative papers can help paper producers to define new engineering

strategies towards production of high performance decorative papers that maintain both dimensional stability and mechanical resistance.

ACKNOWLEDGMENTS

This work was developed at CICECO - Aveiro Institute of Materials and is funded by ERDF Funds through the Operational Competitiveness Programme - COMPETE for the project "Development of second generation high pressure laminates" (2GLAM – FCOMP-01-0202-FEDER-23006). The authors thank the Portuguese Research Institute on Forestry and Paper (RAIZ) for the provided technical assistance.

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Article submitted: October 1, 2015; Peer review completed: November 23, 2015; Revised version received: November 27, 2015; Accepted: December 21, 2015; Published: January 13, 2016.

DOI: 10.15376/biores.11.1.2020-2029