THE EFFECT OF HARDWOOD FIBER MORPHOLOGY ON THE HYGROEXPANSIVITY OF PAPER

Iiro Pulkkinen,a* Juha Fiskari,b and Ville Alopaeus a

The purpose of this investigation was to study the effect of fiber characteristics, especially fiber wall thickness, on the hygroexpansion behavior of paper due to humidity changes. Five different eucalyptus Kraft pulp samples were studied with an OPTIDIM hygroexpansivity tester. As a reference, birch and acacia were included. In all, seven pulp samples were refined at low consistency (LC), using a Voith Sulzer refiner. Both anisotropic and isotropic sheets were investigated. The sheets were dried under restraint. The results showed that fiber wall thickness is an important factor in controlling the hygroexpansivity of paper through fiber network activation.

Keywords: Fiber wall thickness; Morphology; Hygroexpansivity; Fiber network activation.

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INTRODUCTION

Hygroexpansivity depends on a variety of pulp and papermaking parameters such as pulp type, fiber shape, beating conditions, fiber orientation, pressing, and drying tensions. The relationship between in-plane hygroexpansion and moisture content has been studied intensively in the literature (Brandon 1967; Brecht 1958; Gallay 1973; Uesaka 1990). It has been concluded that by understanding the influence of different papermaking parameters, hygroexpansion can be reduced to an acceptable level.

The hygroexpansivity of paper is dependent not only on the moisture sorption capacity of hemicelluloses, but also to a large extent on the structural factors reflecting the earlier stages of the papermaking process, all the way to pulping and bleaching. The structural features of fibers are responsible for sheet consolidation. Paper expands when the dimensional changes of fibers transfer to the dimensions of the macroscopic network (Gallay 1973; Salmén et al. 1987; Uesaka and Qi 1994). In general, increasing the degree of fiber-fiber bonding is known to affect mechanical properties to a great extent, but hygroexpansivity responds differently (Salmén et al. 1987; Salmén et al. 1985; Uesaka and Qi 1994). At higher humidity, where irreversible shrinkage is involved, the hygroexpansion coefficient sometimes shows a decrease with increasing density.

Earlier, Uesaka and Moss (1997) concluded that the major fiber geometry factors affecting paper hygroexpansivity are fiber width, wall thickness, and fiber length. Studies (Uesaka and Moss 1997; Courchene et al. 2007) have indicated that low microfibril angle (MFA) of wood fibers is beneficial to dimensional stability, but it could be that this becomes attenuated by the papermaking process (pulping, bleaching, beating and drying). Based on our previous findings, MFA and fiber wall thickness go relatively well
hand in hand (Pulkkinen et al. 2008). The fiber wall thickness and the fiber length are highly inter-correlated (Preston 1934; Dadswell 1958; Uesaka and Moss 1997; Wimmer et al. 2002), so it is logical that both high fiber wall thickness (McMillin 1973; Megraw 1997) and high fiber length (Uesaka and Moss 1997) result in lower hygroexpansion coefficient. According to Vainio et al. (2007), high MFA (low fiber wall thickness) would suggest that there would be more drying shrinkage, i.e. hence resulting in higher drying stress. However, the effects of MFA quite easily can be obscured by changes of other fiber properties (Watson and Dadswell 1964). Thin-walled fibers are generally believed to shrink more during drying (McIntosh 1961). The bonding ability of the fibers will determine how this translates into the hygroexpansivity of the fiber network.

When sheets are dried under restraint, the effect of the original fiber curliness disappears (Salmén et al. 1987). This is probably due to a straightening of the free fiber segments between the fiber crossings when dried under restraint, as stated by Giertz (1964). Giertz and Roedland (1979) have stated that the increase in tensile strength during drying is caused by increased fiber segment activation via increased swelling of fibers.

To evaluate the hygroexpansivity of different hardwoods, an OPTIDIM (Kajanto and Niskanen 1996) optical analyzer was used.

The main purpose of this investigation was to study the fundamental role of fiber wall thickness distribution on hygroexpansion in cases where drying in restraint is applied. The effect of fiber wall thickness on the activation of the handsheet was considered for the case where fiber swelling was approximately constant, the only aspect contributing to fiber network activation being the geometrical factors. As the sheets were dried under restraint, the whole activation potential of pulp fibers can be explored. Theoretical considerations on the role of bonding, drying shrinkage, and fiber segment activation are presented here.

EXPERIMENTAL

Materials

The study included five commercial eucalyptus pulp samples from different parts of the world, accompanied by birch and acacia as reference samples. The samples were E. grandis/E. dunnii (Uruguay), E. globulus (Portugal), E. urograndis (Brasil), E. urograndis/E. grandis (South Africa), E. grandis/E. saligna (Brasil), Birch (Betula pendula, Finland), and Acacia magnium (Indonesia). Some of the eucalypt pulp samples were mixtures of two species, 50%/50% for E. grandis/E. dunnii and 90%/10% for E. grandis/ E. saligna. The ratio of fibers in E. urograndis/E. grandis was unknown. The pulp samples were obtained in mill dried form.

Methods

Fibers were refined with a Voith-Sulzer disk refiner at a consistency of 4%. Fibers were refined up to 150 kWh/tonne with a specific edge load of 0.4 J/m. The handsheets were tested according to the following standards and methods:
• Apparent bulk density, ISO 534
• Tensile properties, EN ISO 1924

Isotropic handsheets of grammage 60 g/m² were made in a standard laboratory handsheet former according to the ISO 5269-1 standard. Anisotropic sheets were made with a dynamic sheet former (Uesaka et al. 2002) at KCL. The wire speed was set to 1000 m/min. The jet speed was set to 920 m/min (1.6 bar). The feed concentration was 4 g/l. The grammage of anisotropic sheets was 60 g/m².

**Water Retention Value (WRV)**

The WRV was determined by centrifuging a 1700 g/m² pulp pad at 3,000 g for 15 minutes in a commercially available centrifuge according to standard SCAN-C62-2.

**Fiber Saturation Point (FSP)**

The FSP was measured by solute exclusion (Stone and Scallan 1968) using a 2x10⁶ Dalton dextran polymer (Amersham Biosciences).

**Sheet Drying**

The isotropic sheets and anisotropic sheets were dried in contact with a plate to which the sheets were attached. This affected shrinkage during drying, which was done in a climate-controlled room (23 °C and a relative humidity of 50%).

**Measurements of Fiber Dimensions**

The FiberLab® measurement equipment consists of an analyzer and a sample unit. A CCD camera captures fiber images. The direct results are the length, width, and the thickness of the cell wall of the fiber. The calculated values are the curl index, coarseness, cross-sectional area, and volume index (FiberLab® 2001). Procedures of fiber and image processing to obtain fiber properties have been described in detail elsewhere (FiberLab® 2001).

10,000 fibers per sample were measured. A Fortran program was created to calculate the characteristics of the distributions, i.e. mean, standard deviation and skewness, of fiber length, width, fiber wall thickness, and fiber curl. It should be noted that the measurements are made for swollen fibers, and that the measured wall thickness is not the true wall thickness of the fibers. It is calculated as the mean value of both walls in the measurement interval.

The fines content was measured as the amount of fines detected by the analyzer (fibers shorter than 0.2 mm).

**Measurement of Dimensional Stability**

OPTIDIM is an optical method for measuring the dimensional stability of paper under varying humidity conditions. The sample, which was placed in a humidity control cabinet, after each humidity change and after equilibrium had been reached with the surrounding air moisture content and the moisture content of the sample, was put between weighted glass plates to prevent any out-of-plane dimensional changes and transillum-
inated with a bright light from the bottom, and a picture was taken from above with a 
CCD camera (Fig. 1). By placing the weight on top of the sample only before a picture is 
taken, the friction between the glass and the sample is minimized. The glass weight was 
chosen according to ISO 8226-2:1990 based on the grammage of the handsheets. The 
variations in brightness level caused by the formation of the paper were measured. The 
dimensions of the samples at low relative humidity were compared with the dimensions 
at higher relative humidity. The dimensional changes as a function of relative humidity 
were then calculated. The size of the samples was 70mm*70mm. Relative humidity was 
changed linearly from standard (50% RH) to low (10% RH), then to a high humidity 
(90% RH) and back to standard (50% RH) humidity for 4 h in each humidity condition at 
23 °C. The samples were also weighed at the moisture contents used. The moisture 
content of the paper sample was calculated based on reference samples (oven dried in 
105°C for two hours).

Six parallel sheets were prepared for each sample in each of the refining points. 
Hygroexpansivity of isotropic handsheets was measured as a geometric mean of cross 
directional (CD) and machine directional (MD) hygroexpansivity. The results are shown 
in Appendix A.

Figure 2 shows the hygroexpansion coefficient as a function of the moisture 
content of the anisotropic sheet in CD. As can be seen, the relationship between the 
hygroexpansion strain and sheet moisture content was approximately linear and 
reversible in the low-moisture-content range (10% RH → 50% RH), but in the high-
moisture-content range the relationship was non-linear and showed an irreversible 
shrinkage after the initial exposure to high humidity. In the literature, this is sometimes 
referred to as the release of internal stresses (Salmén 1987; Uesaka 1992).

The hygroexpansion coefficient (β) can be determined from the gradient of the 
linear part of the curves in the low-moisture range and is expressed as hygroexpansional 
strain divided by moisture change (Nanri 1993).

\[ \beta = \text{dimensional change (%)}/\text{MC change (%)} \quad (1) \]

In machine-made papers, hygroexpansion is different in the machine direction 
(MD) and cross direction (CD). The cross-machine direction is more sensitive to changes 
in fiber orientation and the degree of fiber-to-fiber bonding. As the hygroexpansion in the 
transverse direction of a single fiber far exceeds the hygroexpansion in the fiber axis 
direction (Page and Tydeman 1962), with increasing fiber orientation in the machine 
direction, the stress-transfer in the fiber axis direction decreases and the hygroexpansion 
in the cross direction increases. The magnitude of hygroexpansivity in the cross direction 
for anisotropic sheets produced in the paper mill (β_CD) is in the range of 0.1-0.15 (Kajanto 
and Niskanen 1996). This gives us confidence that this method can be used at least to 
rank pulp samples of different origin.
RESULTS AND DISCUSSION

The non-linear behavior can be seen after the first absorption stage (10% → 50% RH) (Fig. 2). Therefore, the hygroexpansion coefficient throughout this study was determined as the mean value of the gradient of the first desorption stage (from 50% → 10% RH) and the first absorption stage (10% → 50% RH). This is the region where there was no, or at least very little, non-linearity.

In Appendix A, the OPTIDIM results of all seven samples are presented. The hygroexpansivity of conventional sheets was generally greater with increased refining. The values were roughly of the same order of magnitude, with the major differences derived from fiber morphology.

Figure 3 shows hygroexpansivity coefficients of anisotropic sheets as a function of refining for samples studied in the MD and CD, respectively. As the figures illustrate,
refining generally had a small effect on the hygroexpansivity in the MD and a bigger effect on the hygroexpansivity in the CD (Salmén et al. 1987; Uesaka and Qi 1994).

Fig. 3. Hygroexpansivity coefficient as a function of sheet density in the MD (top) and CD (bottom) (error bars 95% confidence interval).

In anisotropic sheets, there are more fibers aligned towards the MD, and therefore there are more bonds along the fibers in the CD. In the cross-machine direction, the transverse shrinkage of fibers can pull the entire sheet together during drying. This leads to various deformations, such as microcompressions in the fiber wall at the bonding sites. Due to the higher number of bonds along the fibers in the CD, these fibers are straightened out more effectively than those in the MD. As stress transfer is effective in compressed fiber segments, the relative bonded area of the network should therefore have a large effect on the hygroexpansivity in the CD (Uesaka 2002), which is seen as higher.
values of CD hygroexpansion compared to MD hygroexpansion values. In order to characterize the activation of the handsheets in respect to fiber morphology, in addition to geometric mean value of hygroexpansion coefficients, also the MD and CD hygroexpansion coefficients were compared separately for all pulps.

There was no general trend for hygroexpansional behavior as a function of density for isotropic handsheets (Fig. 4, Appendix A). This may be due to the small differences in the formation of the pulp sheet due to fiber deformations (curl etc.).

![Fig. 4. Hygroexpansivity coefficient as a function of sheet density of isotropic handsheets (error bars 95% confidence interval).](image)

In the case of dynamic sheets, the highest hygroexpansivities were found for birch, acacia, E. urograndis/E.grandis, and E.grandis/E.dunnii. In theory, a higher amount of fiber bonding with a fiber network consisting of thin-walled fibers should result in a higher degree of fiber segment activation (straightening of free, unbonded fiber segments) if the same amount of restraint is applied during drying. Therefore we have decreased the potential for hygroexpansion of thin-walled samples when the samples are refined. This is shown by the hygroexpansivity values of Appendix A.

**The Effect of Fiber Wall Thickness**

If we take a look at how fiber wall thickness affects the hygroexpansivity coefficient of unrefined samples, it can be seen (Appendix A, Fig. 5) that the mean value and standard deviation of fiber wall thickness had negative correlations with the hygroexpansivity of anisotropic sheets. The mean value and the standard deviation follow each other closely, as expected. A better way to characterize the relative importance of fiber wall thickness would be to study fibers with a similar fiber wall thickness mean value, but with different moments of distribution data. In this study, thin-walled fibers with narrow fiber wall thickness distributions had higher hygroexpansivity values (Fig. 5), measured as the geometric mean of MD and CD hygroexpansion of the anisotropic sheets. A low fiber wall thickness can be seen especially in connection with high values
of the CD hygroexpansion coefficient (Appendix A). The outlier marked in red is the birch sample. The difference in the hygroexpansional behavior of birch samples could be a result of its very different chemical and structural nature and the high amount of fines present (Fig. 6 and Table 2). It is also noteworthy that birch had the highest variability in hygroexpansion measurements. The reason behind this is still unknown. The two groups seen in Fig. 5 could not be explained based on the differences between the chemical compositions of the samples. Even though the properties of fines differ considerably from the properties of the fiber fraction (Retulainen et al. 1998), the insignificance of the amount of fines as an explanation for the difference between the hygroexpansional behavior of these two groups was evident, based on Figure 6 and Table 2.

![Graph 1](image1.png)

**Fig. 5.** Geometric mean of hygroexpansivity of unrefined anisotropic sheets as a function of fiber wall thickness mean value (left) and standard deviation (right) (the outlying birch indicated as a square, acacia and eucalyptus samples as diamonds).
Based on the theory presented in this paper, a fiber network consisting of thin-walled fibers with homogeneous wall thickness distribution shrinks less during drying and swells less during rewetting than a fiber network consisting of thick-walled fibers with heterogeneous wall thickness distribution over a relative humidity range from 10% to 90%. This is probably due to the wider drying stress distribution of the thick-walled fibers, as there is more porous space between fibers in the fiber network.

Figure 6. The fines content (%) measured with FiberLab® as a function of the geometric mean hygroexpansion coefficient (%/%).

Figure 7 shows the FSP values measured, which indicate that fiber wall structure was not significantly modified during pulping or pulp bleaching. There were no huge differences between different pulp samples regardless of their origin. The FSP could not explain the difference seen in the hygroexpansional behavior of birch and eucalypt. The possible differences in drying of the pulps at their respective origins could also have an effect on the results of FSP measurements, as drying has an effect on the swelling ability of the fibers (Weise and Paulapuro 1999).

Figure 8 and Table 2 show that dynamic sheet samples with low fiber wall thickness index exhibited a lower overall change in sample dimensions (as the mean of CD and MD dimensional changes) than samples with thick-walled fibers when the relative humidity was changed from 10% to 90%. This was not a part of the study made by Uesaka and Moss (1997), who only concluded that hygroexpansion coefficient (in the range of recoverable transformation) was higher for thin-walled fibers, as supported by this study. When Nanko and Wu (1995) examined the mechanisms of fiber shrinkage during drying, they found out that the transverse shrinkage of thick-wall fibers was larger than that of thin-walled fibers regardless of the restraining conditions applied. These studies support the findings of this study (Figure 8, Table 2) that the extent of sheet hygroexpansion was higher for sheets consisting of thin-walled fibers in the region of low moisture content, and lower in the region of high moisture content.
Fig. 7. Fiber saturation point at different levels of refining as a function of fiber wall thickness.

Fig. 8. Overall change in sample size (dynamic sheet in the CD direction) when subjected to relative humidity between 10% to 90% as a function of fiber wall thickness (error bars 95% confidence interval).

The explanation for this may be that while the amount of water absorbed by paper samples is lower, the rate at which this happens is faster in the linear part of the hysteresis curves. After non-linear behavior occurs (over 50% RH), the amount of water absorbed is higher for sheets with more thick-walled fibers. It is however possible that irreversible hygroexpansion occurs even below 50% RH. The overall changes in the sample size were quite small. An increase of approximately 0.5% in size was obtained for the samples of 70mm * 70mm presented in Table 2. The CD of the anisotropic sheets was chosen, since it should best reflect the effect of fiber wall thickness on fiber network hygroexpansion.
The hemicellulose content (Table 3) was measured as the combined amount of xylose and mannose in the pulp samples. No correlation was found between the hygroexpansion coefficient and the hemicellulose content. E. urograndis had the lowest hemicellulose content of the samples studied.

The fiber orientation anisotropy was characterized as a ratio of MD and CD tensile stiffness values. As depicted in Table 3, the fiber orientations of all eucalypt pulp samples and acacia were similar at a similar refining level, while birch had a different fiber orientation. So we can ignore both the hemicellulose content and fiber orientation as variables affecting hygroexpansion of the eucalypt samples and acacia of this study, and consider only the role of fiber wall thickness in fiber network activation.

As the sheets were dried under restraint, there was no correlation between the MD/CD- ratio and the hygroexpansion coefficients (Table 3 and Appendix A). Assuming that the variation in restraining the drying of the sheets was negligible, the differences in the shrinking/collapse-potential of the fiber walls mainly determined the activation potential of the sheets. In the MD, the activation potential is limited for moderately straight fibers, since bonds along the fiber axis straighten them effectively. Therefore these values do not necessarily have to correlate for sheets dried in constant restraint (Uesaka 2002).

If the orientation of the fibers and fiber curl is to be constant for all eucalyptus samples and acacia, when examining the fiber network we can see that hygroexpansion is controlled by the transverse shrinkage of fibers (Uesaka 1994a, b).

### Table 2. Change in Sample Dimensions During a Change in Humidity from 10 → 90% RH. (Changes are measured for a sample of the size of 70mm*70mm.)

<table>
<thead>
<tr>
<th></th>
<th>change in sheet dimension (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>10% --→ 50RH%</td>
</tr>
<tr>
<td>Acacia mangium (Indonesia)</td>
<td>0.162</td>
</tr>
<tr>
<td><em>Birch, B. pendula</em> (Finland)</td>
<td>0.179</td>
</tr>
<tr>
<td><em>E. grandis</em>/<em>E. dunnii</em> (Uruguay)</td>
<td>0.208</td>
</tr>
<tr>
<td><em>E. globulus</em> (Portugal)</td>
<td>0.163</td>
</tr>
<tr>
<td><em>E. urograndis</em> (Brazil)</td>
<td>0.169</td>
</tr>
<tr>
<td><em>E. urograndis</em>/<em>E. grandis</em> (SA)</td>
<td>0.186</td>
</tr>
<tr>
<td><em>E. grandis</em>/<em>E. saligna</em> (Brazil)</td>
<td>0.155</td>
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Table 3. Chemical Composition, MD/CD Stiffness Ratio, Original Curl of the Fibers in the Samples and the Fines Content.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Xylose + mannose (%)</th>
<th>Refining (kWh/t)</th>
<th>MD/CD TSI</th>
<th>Curl (%)</th>
<th>Fines (n)</th>
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<tr>
<td>E.grandis/E.dunnii (Uruguay)</td>
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<td>0</td>
<td>2,47</td>
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<td>8,27</td>
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<td>21,4</td>
<td>11,15</td>
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<td>1,68</td>
<td>19,7</td>
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<td>E.urograndis (Brazil)</td>
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<td>18,2</td>
<td>8,91</td>
</tr>
<tr>
<td></td>
<td>50</td>
<td>2,34</td>
<td>17,5</td>
<td>9,59</td>
<td></td>
</tr>
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<td>150</td>
<td>1,97</td>
<td>19,8</td>
<td>11,03</td>
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<tr>
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<td>Acacia mangium (Indonesia)</td>
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<td>2,29</td>
<td>18,3</td>
<td>10,60</td>
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CONCLUSIONS

The objective of this study was to gain further understanding of the role of fiber wall thickness distribution in the hygroexpansion behavior of laboratory handsheets dried under restraint. The use of restraint-dried sheets was done in order to mimic the effects seen in the mill environment. However, one has to bear in mind that the conditions used (wet pressing, sheet making, refining, drying) were far from ideal. The results showed that:

1. For the same amount of fiber network activation potential (approximately at the same level of FSP values) at the beginning of drying, characterized as the initial curl of the fibers and the MD/CD tensile stiffness ratio of the sheet samples, fiber wall thickness was responsible for the differences seen in the dimensional changes of the in-plane sheet structure of anisotropic sheets in varying humidity conditions for restraint-dried samples.

2. Narrow fiber wall distribution results in higher hygroexpansion coefficients but in a smaller change in sample dimensions. With thick-walled fibers the more heterogeneous stress distribution will deform the whole sheet in the direction of the higher force, resulting in a higher hygroexpansion of the sheet within the plane of the paper over the whole humidity range.
3. For isotropic sheets, the effect of raw material was possibly concealed by the sheet making process and the imprecision of the hygroexpansion measurements.

Based on our experiments, the amount of water absorbed by the thin-walled fibers and thick-walled fibers differ at different stages of moisture absorption cycle. The results suggest that water uptake is higher for thin-walled fibers between relative humidity range of 10% to 50%, and lower between relative humidity range of 50% to 90%. The results of this study show some indication that there were differences between the hygroexpansional behavior of restraint-dried laboratory sheets of acacia and eucalyptus samples due to the fiber wall thickness. When considering the results, we have to remember that, for example, wet pressing in the laboratory environment can affect the results, as the dynamic nature of the procedure can lead to non-uniform densification of the sheets in the z-direction. This could also have an effect on the in-plane swelling properties of the samples, for example as an increased friction between the glass weight and the sample as a result of curling tendency of the sheets. This study confirmed what has been established about the effect of fiber wall thickness on hygroexpansivity of paper. The bonding and bond strength are affected by the fiber treatment (pulping, bleaching, refining), fines content, and process parameters such as wet pressing and drying.

Based on the results of our experiments, some new observations on hygroexpansivity can be made. For example, some differences in the water uptake capacity of thin-walled fibers under different relative humidity conditions were observed. There was no effect of the hemicellulose content on hygroexpansion, and no effect of fiber curl or fines content for that matter. Overall, there were clear indications that fiber wall thickness was a significant factor in determining the amount of hygroexpansion of the samples.

ACKNOWLEDGMENTS

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LITERATURE CITED


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APPENDIX A:

Hygroexpansivity Coefficients of Conventional and Dynamic Sheets Measured with OPTIDIM at Different Levels of Refining, Fiber Dimensions Measured with FiberLab®, and the Densities of the Samples.

<table>
<thead>
<tr>
<th>Sample</th>
<th>μW/m²</th>
<th>βiso</th>
<th>stdv (iso)</th>
<th>isotropic</th>
<th>fDYN</th>
<th>fDYN,MD</th>
<th>fDYN,CD</th>
<th>stdv (dyn)</th>
<th>Density (kg/m³)</th>
<th>Fiber Wall Thickness (μm)</th>
<th>Fiber Width (μm)</th>
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<tr>
<td>E.grandis/E.dunnii (Uruguay)</td>
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<td></td>
</tr>
<tr>
<td>0</td>
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<td>0.015</td>
<td>608</td>
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