Prediction of wood tangential shrinkage from cellulose crystallite width and density in one 11-year-old tree of *Eucalyptus globulus* Labill.

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Summary

The relationship between variation in wood shrinkage and cellulose crystallite width determined by X-ray diffraction was assessed using SilviScan-2 (a system developed at CSIRO for rapid assessment of wood microstructure). Cellulose crystallite width, density and microfibril angle were determined for 600 µm wide zones on small wood samples with known tangential shrinkage. The tangential shrinkage measurements included shrinkage to 12% moisture content after reconditioning (MC AR); and a shrinkage differential calculated from tangential shrinkage to 12% MC AR recorded for each zone; and the minimum shrinkage recorded in adjacent wood of similar cambial age. Spearman correlations and forward stepwise regressions showed that the cellulose crystallite width was a good predictor of the shrinkage measures and that density was a minor predictor. Together, cellulose crystallite width and density could explain 75% of the variation in tangential shrinkage for randomly selected locations throughout the tree, and 87% of the variation in tangential shrinkage in samples selected from the lower 5% of tree height. The results suggest that SilviScan-2 can be used to predict tangential shrinkage during drying in increment cores, and may therefore be useful in developing non-destructive sampling strategies in tree improvement programs for *E. globulus.*

Introduction

Plantation expansion in southern Australia in recent years has been dominated by planting of *Eucalyptus globulus*. The primary objective of these plantations is to produce wood fibre for paper production. However, some tree owners are also interested in growing the plantations as a source of logs for production of high quality solid wood. One possible constraint to development of a resource that can be utilised efficiently by industry is the variation in shrinkage of solid wood during drying. Problems of excessive shrinkage leading to drying degrade in young plantation-grown trees have been reported by Northway and Blakemore (1996) and Washusen *et al.* (2000), and considerable variation in shrinkage in screening trials was reported by Washusen and Ilic (2001). The excessive shrinkage found in all of these studies was associated with tension wood.

Tension wood is usually found on the upper side of leaning trees and develops as a mechanism for realigning the stem and positioning the crown to maximise the capacity to receive light. However, it is not clear which factors may contribute to tension wood formation (and hence excessive shrinkage) in well-grown, straight, vertical trees. In order to understand why excessive shrinkage occurs, and to determine which trees are likely to cause problems during processing, rapid detection methods are needed.

Tension wood differs from normal wood in microstructure and topochemistry. In normal wood, cellulose forms crystallites in microfibrils that are aligned at an angle to the longitudinal axis of the fibre. The microfibrils are surrounded by lignin that is deposited soon after the fibre is formed and acts as a reinforcing agent. In tension wood, the cellulose is often reported to be more crystalline than normal wood, and lignin may be absent from part of the fibre wall. The absence of lignin is the primary cause of the characteristically high shrinkage in tension wood possibly because the cellulose microfibrils are relatively unrestrained and there is little resistance in the fibre wall to prevent the inward collapse of the cell wall (Wardrop and Dadswell 1955).

The differences in the nature of the cellulose in tension wood and normal wood suggest that it may be possible to relate shrinkage to the characteristics of the cellulose crystallites. One of these characteristics is the width (diameter) of the crystallites. Crystallite width can be estimated by X-ray diffraction using SilviScan-2, an instrument developed at CSIRO for rapid measurement of wood and fibre properties in increment core samples. In a preceding paper (Washusen and Evans 2001) crystallite width, estimated by SilviScan-2, was shown to be greater in tension wood than normal wood in *Eucalyptus globulus*. Because of the association between tension wood and shrinkage, cellulose crystallite width may be a useful predictor of shrinkage in solid wood. The objective of this work was therefore to determine if SilviScan-2 could be used to predict tangential shrinkage in solid wood.

Materials and methods

Drying and selection of wood samples

The samples used to investigate crystallite width were taken from a single 11-year-old tree having abundant tension wood in discrete bands surrounded by normal wood. The tree was cut into 10 discs representing a known fraction of tree height ranging from the base of the tree to 80% of tree height. Each disc was cut parallel with the grain into 16 radial pith-to-cambium strips 8x8 mm in tangential and longitudinal dimensions. Each radial strip was divided into three sections by positional reference marks at 75% and 50% of the radial distance from the pith. In the centre of the longitudinal face of each section another reference mark was added for shrinkage measurements. The strips were dried in a laboratory-scale kiln set to an equilibrium moisture content (EMC) of 12% (relative humidity 66.6% and temperature 22.5°C). Once at 12% EMC the strips were reconditioned in steam at 100°C for 40 minutes and placed in a 12% EMC constant condition room until they could be re-measured at all marked locations and at additional marked sites of excessive shrinkage. The strips were then oven dried and re-measured at all marked locations.

Moisture contents based on oven dry weight were determined after reconditioning. Tangential shrinkage was determined based on green dimensions and expressed as a percentage after adjustment to 12% using a calculated unit shrinkage value determined from the dimensions at 12% EMC and at Oven Dry[1].

In addition to tangential shrinkage a shrinkage differential was calculated for each measured location, based on the minimum tangential shrinkage recorded for each radial zone on each disc, and the maximum shrinkage recorded in each radial zone on each radial strip. This parameter has been found to account for normal shrinkage due to variation in cell wall properties. This variation results in normal shrinkage being higher near the stem periphery than near the pith. Above this normal shrinkage range, tension wood begins to appear (Washusen 2000). Shrinkage differentials above 3.1% in any zone generally indicated the presence of tension wood.

Samples were selected randomly by stratification and accrual methods based on the shrinkage differential so that the range in shrinkage differentials was selected from all parts of the tree. Once the shrinkage sites were selected, sections 10 mm long were cut from the radial strip with a bandsaw so that each shrinkage measurement was at the centre of the sample. A total of 66 samples were selected.

Sample preparation for SilviScan-2

The wood samples were re-saturated with water by alternating boiling with vacuum treatment over a period of 24 days. Once re-saturated (when the samples ceased to gain weight), anatomical sections were cut on a microtome for comparisons of crystallite width and wood anatomy as described by Washusen and Evans (2001). After the anatomical sections were taken, to eliminate collapse on drying, the water was replaced with ethanol in three stages, each stage taking approximately one week. During the first stage the samples were placed in 95% ethanol with two days in vacuum, followed by two stages with 99% ethanol. The samples were dried to a nominal 8% moisture content by equilibration in approximately 8% EMC conditions (temperature 20.5ºC and relative humidity 40.5%). They were then mounted on wooden samples holders (with PVA glue) and 2 mm strips running parallel to the grain were cut from the centre of each sample on a twin blade saw. The strips were renumbered on one freshly cut face with the sample number, and they were also marked on the opposite face with the location where shrinkage measurements had been taken. To ensure that the strips were oriented correctly, each of the samples was examined with an Olympus SZH10 stereo microscope to ensure that the anatomical sections matched the wood strips by matching vessels and the tension wood bands where they were present.

X-ray diffraction and X-ray densitometry

The samples were run in three batches on SilviScan-2 as described by Washusen and Evans (2001). Air-dry density profiles were obtained using X-ray densitometry at intervals of 0.01 mm, and of microfibril angle (MFA) at intervals of 200 μm. The density profiles were normalised to the gravimetric density that was determined by the tangential, longitudinal and radial dimensions and weights of the strips at 8% EMC. All diffraction patterns were saved to disk as conventional cartesian patterns and as images mapped onto spherical co-ordinates where the ϕ co-ordinate and the 2θ co-ordinate are straight and orthogonal (Fig. 1). These are here designated as phi/theta images and from these the width of the 002 peak in the 2θ direction was determined. The 002 peak in an X-ray diffraction pattern contains two kinds of information about the cellulose crystallites. The azimuthal or ϕ direction (Fig. 1) gives information relating to the microfibril angle and the 2θ direction (Fig. 1) information relating to cellulose crystallite widths and cellulose crystallinity.

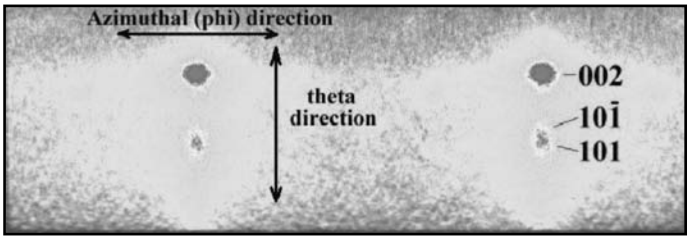

Figure 1. A phi/theta image showing the phi (ϕ) and 2θ directions and the location of the 002 peak

The width of the 002 peak in the 2θ direction at half-maximum intensity was determined by methods described by Washusen and Evans (2001). The width of the 002 peak was converted to crystallite width by the Sherrer formula (Cutter and Murphey 1972). The mean MFA and mean crystallite width at the point where the shrinkage measurement as taken was calculated for all 66 sample sections. In addition the mean density (at 8% EMC) was determined for the same location and the density profile used to ensure that all data were matched to within ±100 μm.

Statistical analysis

A Spearman correlation matrix was determined for the variables of crystallite width, MFA, density, tangential shrinkage to 12% MC AR and the shrinkage differentials. In the case of the tangential shrinkage and shrinkage differentials the data were for the original 8 mm wide radial strip.

Forward stepwise regressions were calculated to predict tangential shrinkage and the shrinkage differentials from crystallite width, density and MFA. The regressions were carried out on the complete data set and repeated for the discs at the base and 5% height (breast height). Because of heteroscedasticity and non-normality both the tangential shrinkage and shrinkage differentials were transformed using the square root.

Results and discussion

Spearman correlation results

The results of the Spearman correlation matrix are given in Table 1. The results showed that the crystallite width was strongly correlated with tangential shrinkage to 12% MC AR and shrinkage differentials and that microfibril angle was relatively poorly correlated with these parameters. This result shows that higher tangential shrinkage coincides with wider cellulose crystallites. It is also apparent that MFA is not a useful indicator of tension wood presence or shrinkage potential in *E. globulus.* Density was well correlated with all parameters.

Table 1. Spearman correlation matrix of shrinkage, density and crystallite width involving all data

Parameter	MFA	Shrinkage to 12% MC AR	Shrinkage differential	Density
	r_s	r_s	r_s	r_s
Shrinkage to 12% MC AR	-0.513			
Shrinkage differential	-0.376	0.885		
Density	-0.651	0.699	0.583	
Crystallite width	-0.499	0.801	0.761	0.592

Shrinkage prediction

Table 2 shows stepwise regression results of predicting tangential shrinkage to 12% MC AR (Model 1) and the shrinkage differential (Model 2). Judging by the size of beta coefficients and probability levels, crystallite width was a better predictor of both variables than density.

Table 2. Multiple regression results for prediction of tangential shrinkage to 12% MC AR (Model 1) and shrinkage differentials (Model 2) using crystallite width and density. All data were transformed by taking the square root.

Model 1 Dependent variable: tangential shrinkage to 12% MC AR			
All data N=66	**r^2 = 0.751**	**F(2,63) = 94.87**	**p <0.001**
	Beta	Intercept and β coefficients	p-value
Intercept		-2.79	<0.001
Crystallite width	0.607	+1.31	<0.001
Density	0.370	0.00158	<0.001
Lower discs N=22	**r^2 = 0.874**	**F(2,19) = 65.62**	**p <0.001**
	Beta	Intercept and β coefficients	p-value
Intercept		-2.89	<0.001
Crystallite width	0.724	+1.45	<0.001
Density	0.264	0.00121	<0.05
Model 2 Dependent variable: shrinkage differential			
All data N=66	**r^2 = 0.668**	**F(2,63) = 63.31 p <0.001**	
	Beta	Intercept and β coefficients	p-value
Intercept		-6.57	<0.001
Crystallite width	0.674	+2.12	<0.001
Density	0.224	+0.00139	<0.05
Lower discs N=22	**r^2 = 0.821**	**F(2,19) = 44.66**	**p <0.001**
	Beta	Intercept and β coefficients	p-value
Intercept		-5.77	<0.001
Crystallite width	0.722	+1.95	<0.001
Density	0.232	+0.00144	NS*

*NS = not significant at p<0.05

The regression results confirm the importance of crystallite width and density as predictors of tangential shrinkage in solid wood in the selected tree. Both measures of shrinkage are important indicators of the variation in shrinkage within the tree. Tangential shrinkage is a measure of the overall variation and between trees may be an important method for comparison and determining the uniformity of a number of trees from a given plantation or plantations. The shrinkage differentials will also give an indication of uniformity at a localised level within trees and may be useful for indicating trees having large shrinkage variation. The regression model for prediction of tangential shrinkage and the shrinkage differentials may be represented by equations (1) and (2) given below:

$$\textbf{(1)} \qquad Y^{1/2} = -2.79 + 1.31\,X_1 + 0.00158X_2$$

Where: Y = tangential shrinkage to 12% MC AR

X_1 = crystallite width (nm)

X_2 = density (kg m^{-3})

$$\textbf{(2)} \qquad Y^{1/2}_{SD} = -6.57 + 2.12\,X_1 + 0.00144X_2$$

Where: Y_{SD} = shrinkage differential

X_1 = crystallite width (nm)

X_2 = density (kg m^{-3})

The plots of the observed and predicted values for Model 1 for all data and for the subset are shown in Figures 2 and 3 (tangential shrinkage for all heights and for the two lowest discs). The regression results suggest that the first subset for Model 1 would explain 75% of the variation in tangential shrinkage and, for the samples from the base and 5% height, 87% of the variation. Given that sample selection was not weighted to give a whole tree prediction, this better result for the lowest two discs would suggest that improvement would be possible had selection been weighted to reflect proportional representation of the tree.

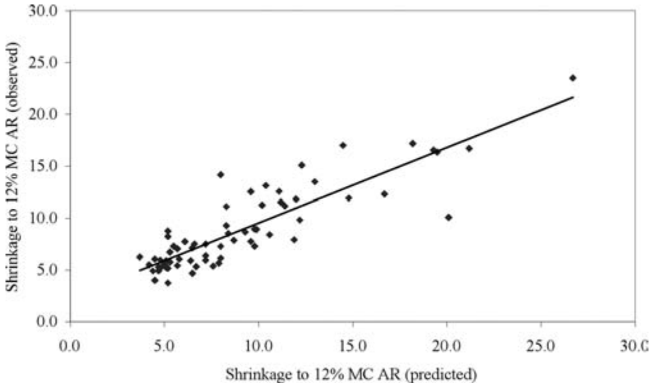

Figure 2. Plot of the observed and predicted values of tangential shrinkage to 12% MC AR for the regression Model (1)

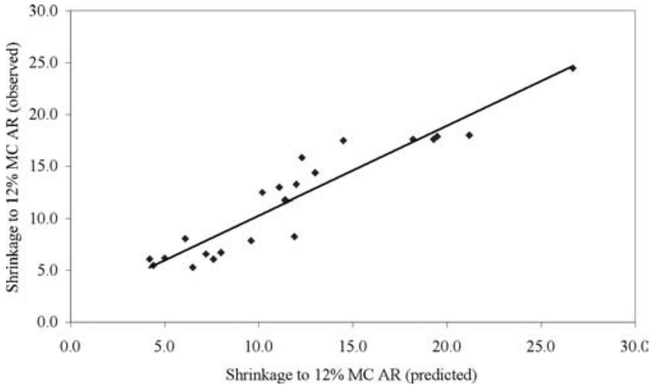

Figure 3. Plot of the observed and predicted values of tangential shrinkage to 12% MC AR for the two lower discs in regression Model (1)

Conclusions

Cellulose crystallite width was closely associated with tangential shrinkage to 12% MC AR in the tree examined and was found to be an important predictor of shrinkage. As crystallite width increased, shrinkage also increased. The correlation between tangential shrinkage and crystallite width was strongest in the lower 5% of tree height. Density was of less importance and MFA was not a significant predictor of shrinkage in this study. High density was found to be associated with high tangential shrinkage and high shrinkage differentials. The results indicate that SilviScan-2 may be used to predict variation in tangential shrinkage in increment core samples taken from trees with tension wood through its association with crystallite width. Given the high variability in shrinkage and other associated wood properties in plantation-grown *E. globulus*, the encouraging results suggest that further development of the methods is warranted. The eventual aim would be to develop a non-destructive sampling method to help understand the factors contributing to tension wood formation and to produce a more uniform resource that industry can use with confidence.

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