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ORIGINAL ARTICLE

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Measurement of the uptake of linseed oil in pine by the use of an X-ray microdensitometry technique

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Abstract Pine sapwood (*Pinus sylvestris*) was impregnated with linseed oil to three levels of uptake. The distribution of the penetrant was found by taking microdensity measurements of an impregnated sample and then using an ethanol extraction procedure to remove the linseed oil. A second set of X-ray measurements at identical locations in the same sample allowed the linseed oil to be indirectly mapped. An uneven distribution of linseed oil in the specimens with the lowest uptake (25% increase in weight) was seen as sharp gradients in the densitometry curves. With increased filling by the linseed oil, these gradients were gradually smoothed. Microstructural changes in specimens with high uptake were revealed using scanning electron microscopy. Through a combination of X-ray microdensitometry investigation and changes observed in the wood's mechanical properties and morphology, it was concluded that liquid flow during impregnation results in significant damage to the cell structure.

Key words Impregnation · X-ray microdensitometry · Linseed oil · Microstructure · Damage

Introduction

The use of impregnated wood is necessary in many building applications. Water in the form of rain or seasonal changes in humidity cause a rise in the moisture content of wood, even if it is treated with commercial surface-coating systems. Water is one of the fundamental requirements for biological degradation. Extending the life in service for wood products by inhibiting the penetration and effects of water is of great commercial interest.

At present, the most frequently used impregnation chemicals contain toxic inorganic compounds such as copper, chromium, and arsenic (CCA). Growing public concern about environmental contamination from treated wood has resulted in the need for alternatives to these preservatives. Linseed oil, traditionally used as a surface coating, is a natural, organic, hydrophobic chemical that can be used as a wood preservative. Given sufficiently good penetration, linseed oil may also act as a stabilizer and reduce hygroscopical movements in the wood. Linseed oil is, in this context, regarded as a nonswelling chemical, as its base molecules are considered too large and hydrophobic to enter the cell wall during the penetrating processes typical of commercial applications. Instead, during impregnation the linseed oil fills cavities such as tracheid lumens, rays, and cracks resulting from the initial drying process.

Knowledge of the microscopic distribution (within annual rings) is useful when evaluating an alternative process from technical and economic points of view. The uptake and distribution of linseed oil in wood has been studied earlier.[1] Scanning electron microscopy[2] and radiographic methods[3] provide qualitative information, and quantitative data can be obtained using nuclear magnetic resonance (NMR).[4]

In this study X-ray microdensitometry techniques were used to characterize the uptake of linseed oil in annual rings. The technique is well established and widely employed to measure density heterogeneity in nonimpregnated wood.[5–9] Absorption of electromagnetic radiation in a material depends on the X-ray energy and the density and chemical composition of the material.

When forcing a liquid into the porous structure of wood, the liquid follows the path requiring the lowest amount of energy. Earlier studies[10] on low-permeable species have shown the need to introduce damage in the microstructure to facilitate impregnation. The permeability of pine is relatively high, and the microstructural changes are expected to be less dramatic. The correlation between uptake of the

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linseed oil and the mechanical properties of the impregnated wood is reported separately,[11] although some generalities are noted in the present article. The impregnation procedure used is not discussed in this paper. Instead, the objective was to investigate the location of linseed oil at different levels of uptake in the wood using a microdensitometry technique. In addition, scanning electron microscopy (SEM) was used to characterize morphological changes due to filling of the cavities.

Materials and methods

Sample preparation

Samples of sapwood from pine (*Pinus sylvestris*) with dimensions of 500 × 35 × 24 mm (longitudinal, radial, tangential directions), were impregnated with linseed oil. The annual rings were oriented parallel to the tangential surface. The initial moisture content was 12%, and the specimens were free from visible defects.

Three batches were impregnated to provide a range of samples with significant differences in uptake. These were called low, intermediate, and high (Table 1). The uptake was measured by weighing the bulk sample before and after processing.

X-ray microdensitometry

From the center part of the impregnated bulk sample, thin specimens (1 mm) were sawn in the radial direction using a twin blade circular saw[12] (Fig. 1). The thin specimens were clearly cut (0.5 mm) at both edges in the radial direction. Each specimen was then mounted on a tray and moved through the X-rays. The transmitted energy was measured every 20 μm, and the individual data points were smoothed using a three-point moving average in computer software. The density of the linseed oil and the wood were obtained from these data. All density values were from positions approximately 5 mm from the transverse cut surfaces. The linseed oil was then extracted from the specimens by placing them in an environment of condensed ethanol for 24 h. The X-ray measurements were then repeated at the same positions. Quantitative analysis of the uptake was achieved by subtracting the density profiles obtained from the impregnated specimen with that from the specimens after the oil has been removed.

The absolute density was calibrated by macroscopic measurements. Small samples (weighing about 2 g) from each batch were immersed in mercury and weighed to an accuracy of 0.001 g. The density values obtained from these measurements were then compared with the integrated mean value of density obtained from the X-ray measurements. The extraction of an unimpregnated reference gave an average weight loss of 3%.

Table 1. Uptake of linseed oil

Uptake	Macroscopic weight increase (%)
Low	25
Intermediate	75
High	105

Microscopy

Morphological observations were performed with a Jeol 5200 scanning electron microscope (SEM). Samples from each batch were taken and the linseed oil removed using ethanol as described previously. To establish the presence of oil gradients, impregnated specimens were also studied. Radial and transverse fracture surfaces were created in both impregnated and de-oiled specimens using a Reichert sliding microtome using steel blades. All specimens were sputter-coated with platinum in a Denton Desk II sputter.

Results and discussion

Low level of linseed oil uptake

Results obtained for the samples with a low increase in weight are shown in Fig. 2. The radial density profiles for impregnated and de-oiled specimens are included. Three SEM micrographs show the characteristics of the impregnated specimen where the densitometer curve was recorded.

The horizontal axis in Fig. 2 represents the distance from the left-hand side toward the center of the sample (17.5 mm). The radial penetrating front (RPF) is also located toward the right. The vertical axis shows the measured density. Studying the density profile for the de-oiled specimen (dashed gray curve), the late wood can be seen as peaks located at 2.0, 3.5, 5.0 mm, and so on; between these peaks are the less dense earlywoods. The highest latewood

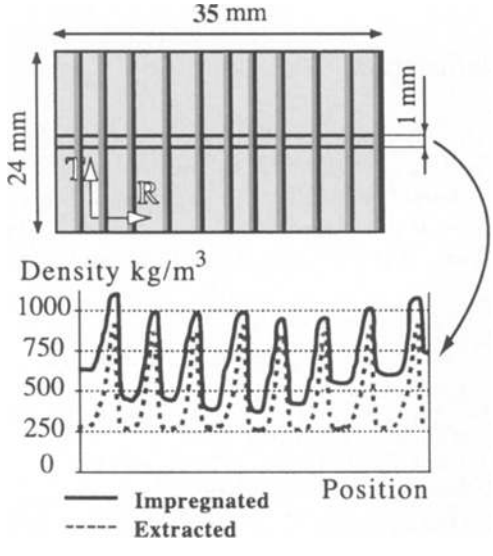

Fig. 1. Design of the experiment

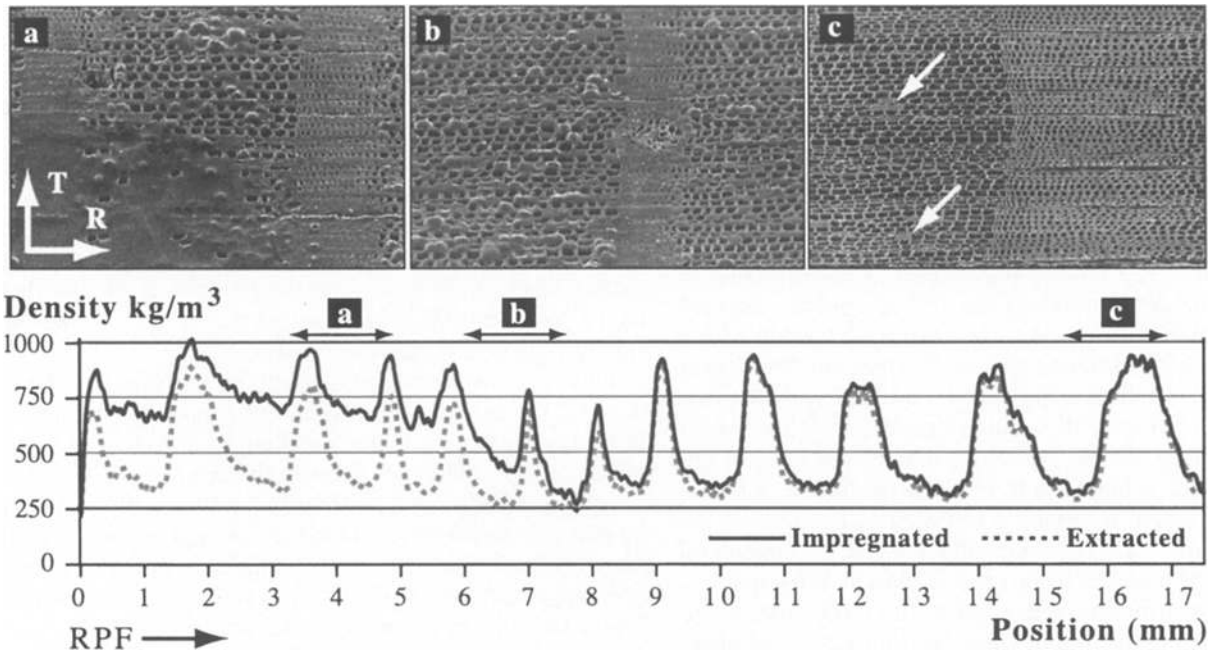

Fig. 2. Radial density profiles for a specimen with low weight increase. Impregnated and extracted profile are shown by *dark* and *dashed gray curves*, respectively. The decreasing content of linseed oil with increasing distance from the edge can be seen in the micrographs (**a–c**). The radial penetrating front (*RPF*) is from left to right. *T*, tangential; *R*, radial; *arrows*, tracheids

density for this particular specimen is close to 1000 kg/m^3 and the lowest earlywood density approximately 250 kg/m^3. The density profile of the de-oiled specimens is used as the reference against which the impregnation results are correlated.

The extent of penetration of the linseed oil can be seen by comparing the density profiles shown in Fig. 2, where the dark bold curve represents the density of the impregnated (unextracted) wood. The amount of linseed oil taken up is fairly constant up to a point about 6 mm into the specimen; it then decreases rapidly up to approximately 8 mm and is practically zero from about 11 mm to the center of the specimen. It is clear that the earlywood takes up more linseed oil than the latewood.

The SEM micrographs in Fig. 2 confirm the heterogeneous distribution of the impregnated linseed oil. By studying the transverse sections from several specimen, it is evident that the linseed oil is found predominantly in regions near the radial extending rays. The size of these regions varies with the distance from the penetrating front; they range in extent from a few tracheids to the larger areas seen in Fig. 2a,b.

The area covered by the SEM micrograph (Fig. 2c), virtually the full width of the specimen, shows a curved region in the otherwise linear interface between latewood and earlywood. Small imperfections such as this can cause the transition between latewood and earlywood to become blurred.

High level of linseed oil uptake

Density profiles obtained from the specimen with a 105% increase in weight are presented in Fig. 3. The density profiles are shown with the center of the specimen located at the left-hand side. The RPF is thus directed from right to left. The density reaches 1250 kg/m^3 for the impregnated latewood and approximately 900 kg/m^3 for earlywood. The integrated mean density for the impregnated specimen is 1013 kg/m^3.

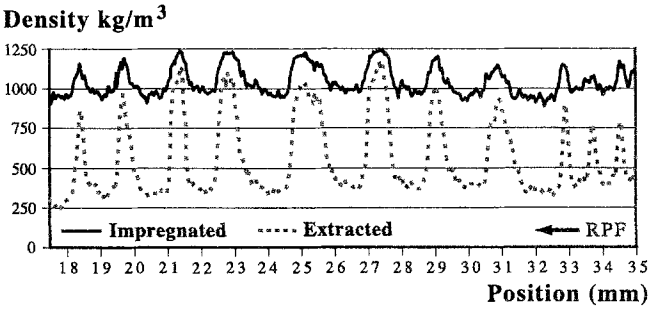

Fig. 3. Density profile for a specimen with 105% increase in weight following impregnation

By comparing the density profiles from impregnated and de-oiled specimens in Figs. 2 and 3, it can be seen that earlywood takes up more linseed oil than latewood. However, it is not possible to say to what extent the cavities are filled in the annual rings. Some researchers suggest that latewood is more easily filled than earlywood,[13,14] although the literature is not unanimous on this point.

Impregnation results are often analyzed using anatomical studies[15,16] of the bordered pit pairs, with the degree of aspiration being claimed to be lower in latewood than in earlywood. Similar explanations have been put forward using the pits that connect ray tracheids with longitudinal

tracheids. In general, the literature is not in agreement as to the occurrence and mechanisms related to selective uptake in the annual ring. Better analytical techniques are required to explore this issue.

Pore volume and relative filling of the lumen cavity

In an attempt to quantify the amount of linseed oil in the porous structure, a model was devised to cover uptake of linseed oil in one annual ring (Fig. 4). The cylinders (lumen) at the left of Fig. 4 represent the porous earlywood with a gradual decrease of lumen size to the right and the denser latewood. In the model, the degree of filling by the linseed oil is illustrated with the assumption that the linseed oil homogeneously fills the available volume. The ratio of filling to available volume is referred to as the degree of filling; and in this case it is assumed to be 50%. Because of the higher porosity of earlywood, the volume of linseed oil taken up is also higher in earlywood than in latewood.

For the distribution presented in the model, the wood porosity, volume fraction of linseed oil, and degree of filling are given in Fig. 4. The porosity (P), which is the available volume for linseed oil, can be calculated from Eq. (1).[17]

$$P = 1 - \rho_u\left(\frac{1}{\rho_{cw}} + \frac{u}{\rho_w}\right) \quad (1)$$

where ρ_u is the specific density of wood at moisture content u. The actual value of ρ_u is obtained from the density profile after removing the linseed oil after impregnation, ρ_{cw} is the density of the dry cell wall, u is the moisture content, and ρ_w is the density of the water. The values for ρ_{CW}, u, and ρ_W are taken as 1.5 g/cm^3, 0.12, and 1.0 g/cm^3, respectively.

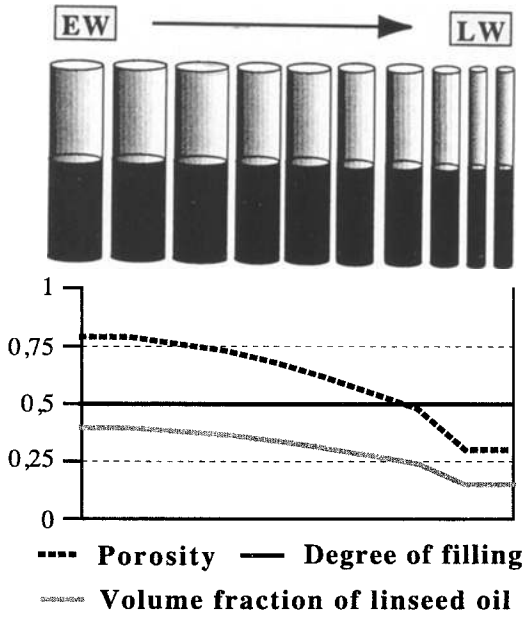

Fig. 4. Model for the uptake of linseed oil in one annual ring (**top**) and relative filling of the lumen cavities (**bottom**). *EW*, earlywood; *LW*, laterwood

Referring to Fig. 4, the volume fraction of linseed oil is obtained by comparing the density profiles before and after de-oiling the specimens, given that the linseed oil has a density of 0.92 g/cm^3. Finally, the degree of filling can be calculated from the porosity and fractional volume of linseed oil in each position.

A similar analysis is presented in the upper part of Fig. 5 based on data from the density profiles for impregnated samples yielding a 25% increase in weight (Fig. 2). As the dashed black curve represents the porosity, it follows that the peaks in this diagram represent the earlywood and the valleys represent the denser latewood.

Figure 5 covers a region from the surface, (left-hand side) of the sample and some 12 mm into the specimen. From the measured degree of filling it is evident that the cavities are not uniformly filled. In the first region (0–6 mm), the degree of filling in the earlywood averages 49% and that for the late wood 43%. Over the subsequent two annual rings (6–8 mm) the degree of filling falls drastically, and from approximately 8 mm and into the center the average degree of filling (with small variations) is 7%.

Results using the same analysis on the specimen with the largest increase in weight are presented in the lower part of Fig. 5. The degree of filling is close to the theoretical maximum, with the relative amount of linseed oil appearing to be higher in the latewood areas. However, it must be remembered that relatively small displacements between the density curves can cause errors in the calculations. This is probably the reason the degree of filling exceeds 100% in some places. Furthermore, the model assumes that no changes occur in the cell structure during impregnation.

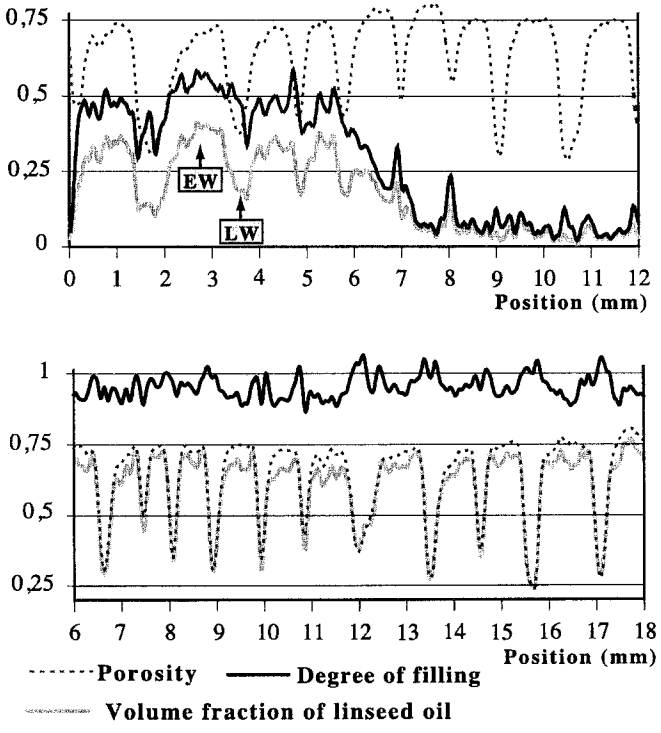

Fig. 5. Relative distribution of linseed oil. Upper and lower graphs represent low and high increases of weight, respectively

Intermediate level of linseed oil uptake

The density profiles and the degree of filling obtained from a 75% postimpregnation increase in weight are presented in Fig. 6. The RPF is from right to left. The density profiles, obtained by microdensitometry, from the impregnated and de-oiled specimen are included in the upper graph. The relative distribution of linseed oil, calculated in accordance with the scheme given in the previous section, is presented in the lower graph.

Evidence of a gradient between 24 and 28 mm can be seen, but it is not as pronounced as in the specimen with a low weight increase (Fig. 5). In the region between 28 and 35 mm the degree of filling averages 72%, and toward the center of the specimen (<24 mm) the equivalent value is 52%.

When selectively analyzing uptake in earlywood and latewood, no definite conclusions can be drawn as to which has a larger degree of filling. However, at low levels of macroscopic uptake there appears to be a tendency for more filling in the earlywood. The opposite is observed in samples with high uptake, where more filling appears to occur in latewood.

In the discussion above, it is assumed that the moisture content does not change between measurements of the impregnated samples and those of the de-oiled specimens. The fact that the densitometry profiles from impregnated and de-oiled specimens showed no significant displacement support this assumption. Some small relative displacements were found in the original data, but they were randomly distributed.

Although at least one author[2,18] suggested that linseed oil has a swelling effect, we assume that this is not the case, as the molecule is considered too large and hydrophobic to enter the cell wall during the short impregnation process in use commercially today. It is therefore believed that the uptake is by the first-order gross pore cavities such as the lumens of tracheids and ray parenchymal cells. This view is supported by the work of Wallström.[19]

Microstructural changes due to impregnation

It is obvious that the flow of liquid during impregnation follows the path of lowest energy demand. Discussions concerning flow in wood often focus on the anatomy of the bordered pits. The fibrillar network surrounding the torus has been claimed to offer an important flow path when the pit is in the unaspirated state. However, there remain a small number of unaspirated pits in our specimens as they are dried to 12% moisture content.

The pores in the fibrillar network are assumed to be too small to allow a high level of uptake of linseed oil within a reasonable time. To confirm this, a morphological study of specimens from each batch was undertaken.

A micrograph of a bordered pit from the specimen with high weight increase (105%) are shown in Fig. 7. The pits are recorded approximately 2 mm from the tangential penetrating surface. The damaged torus membranes present evidence that flow is possible through these membranes during impregnation. By studying a large number of bordered pits from impregnated and unimpregnated samples, the authors' conclusion is that the observed damage in the present study occurs in the specimens as a consequence of liquid flow. Flynn and Goodell[20] performed a similar investigation on spruce but observed no significant morphological changes following impregnation.

Although the present study suggests that flow can occur through distorted membranes in bordered pits, this alone does not explain the superior treatability of pine sapwood. The quantity and anatomy of bordered pits in spruce are comparable to those of pine. Based on a recent microscopic study[21] of transverse flows in pine (*Pinus sylvestris*) and

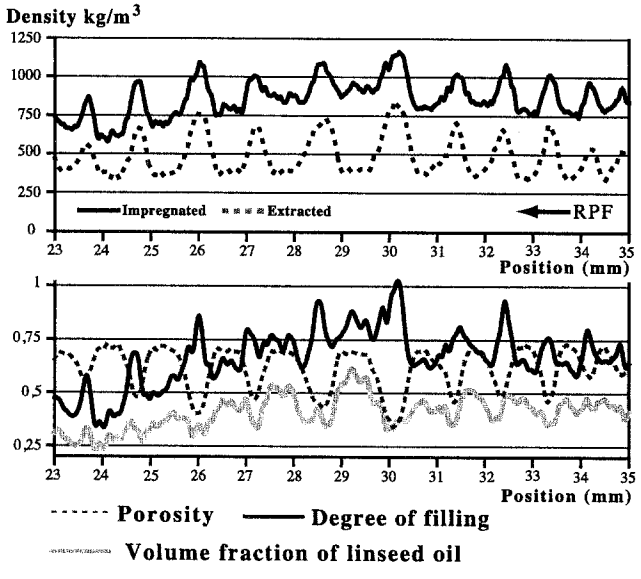

Fig. 6. Top Density profiles obtained from microdensitometry. **Bottom** Calculated relative distribution of linseed oil. RPF is from right to left

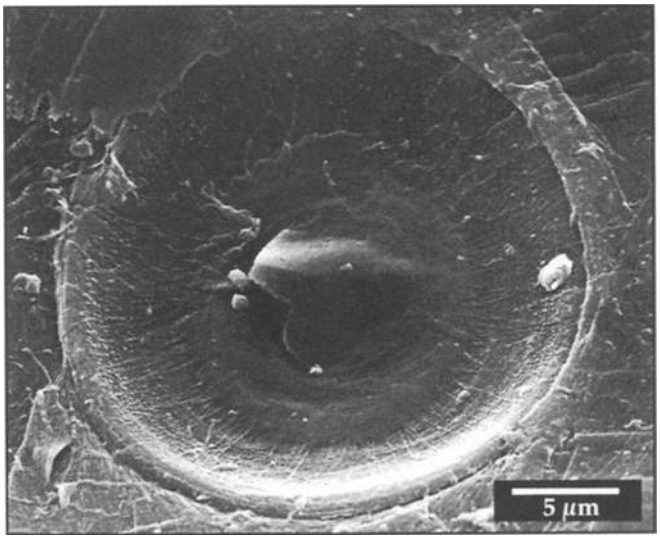

Fig. 7. Disrupted bordered pit membrane from a specimen with initially 105% increase in weight

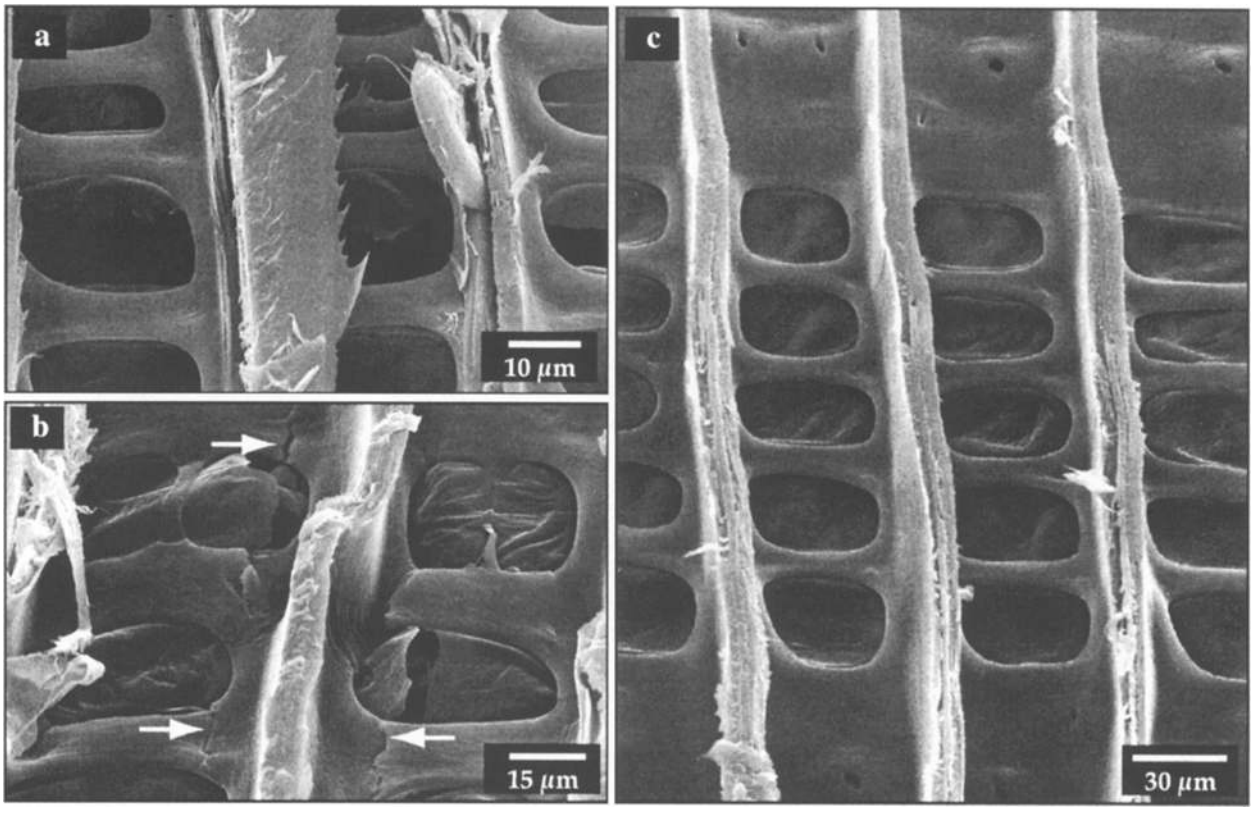

Fig. 8. a,b Cross-field pits between ray parenchyma and longitudinal tracheids initially impregnated to 105% increase in weight. Note the cracks in the cell walls of the tracheids (*arrows*). **c** Crossfield pits are from an unimpregnated specimen

spruce (*Picea abies*), the authors suggested that transverse flow through ray parenchyma cells in pine (sapwood) is of more significance.

Micrographs of de-oiled 105% weight gain specimens are shown in Fig. 8a,b. They reveal structural damage in the thin membrane between the parenchymatous cells and the longitudinal tracheids. Furthermore, the micrograph in Fig. 8b shows cracks in the cell walls of the tracheids. These structural changes are almost certainly caused by the impregnation process. Figure 8c shows a similar area from an unimpregnated specimen with virtually no damage. Although some damage was occasionally observed in the thin membrane of the parenchymatous cells in the unimpregnated specimens, no damage was ever observed in the cell wall.

Based on observations of the uptake of linseed oil, microscopy, and changes in the mechanical properties of the samples,[11] it must be concluded that at some critical level liquid flows resulting from impregnation initiates damage. This critical level is a function of the external pressure, the viscosity of the penetrant, and the anatomy of the specific wood. Pine, particularly sapwood, is known to be a highly permeable wood. Hence, the introduced damage and subsequent loss of mechanical properties are expected to be moderate at impregnation. However, because the damage described above is observed only in the ray region, it cannot be the sole reason for the observed changes of macroscopic mechanical behavior. It is therefore suggested that microscopic cracking takes place in cell wall layers during impregnation, much as occurs during drying.[22,23] Because of the pressure gradient at the oil front, localized mechanical loading is applied to cell walls that are superimposing on any preexisting residual stresses causing a change in the internal stress state in the cell walls. The resulting stresses cause an increase in microcrack density in cell wall layers. The micro damage in the S_1 layer, which is under combined tensile, transverse, and shear loading, seems to be most severe. Weakening of this layer results in reduced resistance to intercell splitting, which is one of the basic mechanisms of failure.

Microdensitometry data obtained from the samples with 25%, 75%, and 105% weight increase are shown in Fig. 9. The figures used are averages with no allowance for intraring variation. There are three impregnation levels. Figure 9 shows the degree of filling as a function of the distance from the sample edges. It is evident that the slope of the gradient is related to the weight increase. For higher increases in weight, the gradients are smoothed.

For samples with low (25%) increase in weight, the degree of filling is fairly low, even at the edges. The liquid flow is likely to be transverse through weaknesses such as the window pit membranes in the ray parenchymal cells. Mechanical properties are not reduced by this treatment[11] but are, in fact, slightly improved, which might be explained by the hydraulic resistance of linseed oil trapped in the tracheid lumen cavities.

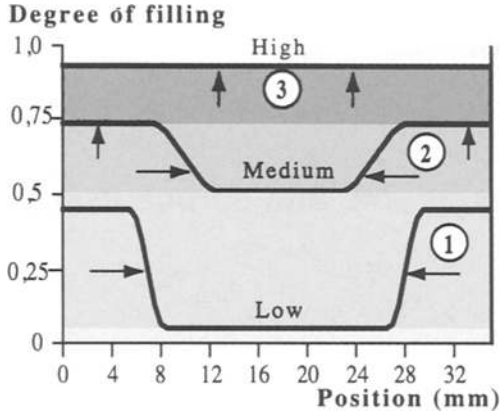

Fig. 9. Plot of the linseed oil distribution at different uptake levels. *1*, *2*, *3*, impregnation levels (*1*, low; *2*, intermediate; *3*, high)

Another gradient is seen for the samples with intermediate uptake (75%). At this level, further damage occurs, which can be seen in the reduced mechanical properties[11] of the samples. It is interesting to note that the least degree of filling at this level is approximately the same as the edge level for the low fill samples.

Finally, at the highest uptake (105%) there are no gradients present, and the degree of filling is close to the theoretical maximum. It is evident that microstructural changes occur at this level (Figs. 7, 8).

Conclusions

The distribution of linseed oil in impregnated pine sapwood (*Pinus sylvestris*) was measured using an X-ray microdensitometry technique. The results obtained can be summarized as follows: The method provided reliable, high-resolution (within annual rings) information about the localization of linseed oil following impregnation. Density gradients due to the heterogeneous distribution of linseed oil can be observed, with the most significant gradients occurring at low uptake levels. Density plateaus form at successively higher concentrations with a corresponding decrease in density gradient. Initial flow of the linseed oil follows existing weaknesses without causing any significant structural damage. At higher levels of uptake, microstructural damage in the form of cracks can be observed by SEM. They are predominantly found in regions near the rays.

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