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SOME ANATOMICAL AND PHYSICAL ASPECTS OF WOOD-PLASTIC (pMMA) COMBINATION OF SPRUCE

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Summary. Test-pieces of spruce (*Picea abies* (L.) Karst.) were vacuum-impregnated with commercial grade methylmethacrylate which was then polymerized by the application of heat. The position of the polymer (pMMA) was identified light microscopically and with the aid of the scanning electron microscope; in the latter work macerated material was also used. In general, the lumina of longitudinal tracheids and ray-tracheids and of the pit cavities were well filled with the plastic; parenchymatous cells were only rarely filled. Although the polymer may be in contact with (attached to) the cell walls, there are no signs of a close interaction of cell wall and polymer. By means of interference microscopy a varying, but low-averaged, value of volume percentage of polymer in the cell wall was determined. Swelling tests have been carried out on sections of untreated material and of wood impregnated in the oven-dry state and at 14% moisture content. Results of these tests indicate that pMMA does not influence, especially reduce, the hydrophyllic nature of the cell wall substantially. The considerable reduction of swelling in radial and tangential direction of solid wood-plastic test pieces must be ascribed to mechanical hindrance by the polymer, in which the presence of non-aspirated bordered pit pairs must play a very important role. Additional data are presented for Scots pine.

INTRODUCTION

Timber is successfully employed as a structural and decorative material for a wide variety of applications. There are, however, certain situations where its inherent property of dimensional change under varying humidity conditions is a disadvantage. Much research aiming at the improvement of dimensional stabilization of wood has therefore been carried out in the past 15 years. One method to improve dimensional stability is to impregnate with vinyl monomers, e.g. methylmethacrylate, styrene, diisocyanates etc. (sometimes in combination with other liquids), or with mixtures of these, and subsequently to polymerize by heat or gamma radiation. Several authors presumed a graft copolymerization of the vinyl monomer with the cell wall constituents to take place, eliminating hygroscopic hydroxyl groups on adjacent cellulose chains, reducing at the same time shrinking and swelling capacity of the wood substance. Many different methods for modifying the dimensional stability of wood in this way have been reported (Stamm, 1964; Laidlaw *et al.*, 1967; Siau *et al.*, 1968; Loos, 1968; Erin'sh, 1970; Erin'sh & Alksne, 1970; Shiraishi *et al.*, 1972; Helinska-Raczokowska *et al.*, 1973; Burmester & Wille, 1973). Generally the decrease of moisture sorption or hygroscopicity of the wood was found to be small in the wood-plastic combination (WPC), which might be interpreted as supporting the view that little cross-linking between the plastic molecules and the hydroxyl groups of the cellulose has occurred, and that, where cell wall penetration occurs, the polymer forms an independent system in the amorphous regions of the cell wall. In other words, water accessibility of cell walls is largely unaffected by treatment with plastic monomers and subsequent polymerization. Other recent studies have reported on the presence of polymer in the cell walls of both softwood and hardwood species after impregnation with and subsequent polymerization of: methylmethacrylate (MMA) monomer (Timmons *et al.*, 1971; Shiraishi *et al.*, 1972; Jokel *et al.*, 1974; Baas, 1975; Furuno *et al.*, 1975); of MMA following solvent-exchange (Pullmann *et al.*, 1974; Furuno *et al.*, 1975); and of a mixture of butylmethacrylate and MMA (Timmons *et al.*, 1971).

This paper reports and interprets the results of a study of the location of pMMA in the wood and in the cell wall and of the influence of the polymer on structure and swelling properties of the most important species for timber construction in the Netherlands: *Picea abies* (L.) Karst.; spruce. This study formed part of a technological research program largely carried out from 1967–1970, in which economical factors prevailed. This implied that a desirably sophisticated approach to fundamental aspects was not always possible.

MATERIAL AND METHODS

Material and pretreatments

Wood. Heartwood of good commercial quality of spruce (*Picea abies* (L.) Karst.) was used. Additional studies were carried out on sapwood of Scots pine (*Pinus sylvestris* L.). Special precaution was taken to exclude material with compression wood. Test specimens of 65 (L) × 30 (R) × 30 (T) mm of both species were conditioned to 14% moisture content or oven-dried to 0% m.c. prior to impregnation.

Methylmethacrylate. The inhibitor (0.5% hydroquinone) was removed from commercial grade MMA (CH_2: $C(CH_3)COOH_3$) monomer by washing with 5% NaOH in water. The monomer was subsequently dried over $CaCl_2$. Finally 0.5 wt% azobisisobutyronitril catalyst was added.

Wood-plastic Combination (WPC). Oven-dried and air-dried blocks were evacuated for ½ hr and subsequently impregnated with MMA monomer at 5–6 mm Hg pressure. After introduction of the monomer and soaking in the liquid for 2 hr at atmospheric pressure, the impregnated blocks were wrapped in aluminium foil and placed in an oven at 90°C. for 6 hr for polymerization. With spruce a polymer content of the WPC of 60 to 90% (based on the oven-dry weight of the untreated wood) was achieved in this way. For Scots pine WPC the polymer content was not determined, but the wood was filled with polymer throughout as seen with the naked eye.

Microscopy

Normal light microscopical studies were carried out on transverse (25 μm), and radial and tangential (5–20 μm) sections cut with a Leitz sledge microtome. The introduction of fluorochrome dyes (Rhodamine B and 6G) into the monomer prior to polymerization in order to locate the polymer using fluorescence microscopy was not successful on account of degradation of the dyes at high temperatures. Staining the sections with Sudan III proved to be a suitable method to contrast the polymer with the cell walls. Sections were mounted in sewing machine oil and sealed. Transverse sections of spruce WPC, 20 and 60 μm thick, were also treated with 'Cuoxam' (copperammoniumtetrahydroxyde), resulting in complete maceration, leaving behind the polymer.

Interference microscopy. A tentative determination of the volume percentage of pMMA in the S_2 of longitudinal tracheids of spruce was carried out applying the interference microscopical method described and discussed by Baas (1968, 1975). For theory and equations used the reader is referred to Baas (1975). The method is based on the determination of refractive indices of microscopical objects of known dimension and optical properties (isotropic or birefringent). From the refractive indices the composition of the S_2 of the cell walls in the WPC can be calculated. The refractive index of the S_2 of the native cell wall was determined in freeze-dried transverse sections with an average thickness of 20.6 μm. Transverse sections of WPC (impregnated at 14% m.c.) with an average thickness of 21.5 μm were studied with a Zeiss interference microscope using the following reference liquids: light paraffin oil (N = 1.4672 at 20° C); heavy paraffin oil (N = 1.4817); Carl Zeiss immersion oil (N = 1.5142); A.L.P. immersion oil (N = 1.5238). In each section at least 10 measurements were made in normal light on late wood cell walls, of the last but 7th row of tracheids in the growth ring. In view of the limited number of observations the obtained values are unsuitable for statistical analysis.

Scanning electron microscopy. Transverse, radial, and tangential surfaces of the Scots pine and spruce WPC were studied with a Cambridge Scanning Electron Microscope (SEM). Final surface cuts were made using a new razor each time. The cubes were coated with gold. SEM studies were also carried out on carefully macerated WPC material in which the polymer still showed the cellular organization of the wood. In order to obtain this, lignin was first dissolved in a mixture of equal volumes of nitric acid and chromic acid (both 10% in water) for 72 hr at room temperature, followed by prolonged and careful rinsing in distilled water. Finally, cellulosic material was dissolved out in 'Cuoxam' over a period of 48 hr at room temperature.

Swelling tests

Swelling behaviour of the wood plastic composite was compared with that of untreated wood in sections as well as in test pieces. Transverse sections (6–8 μm) of untreated spruce and of spruce WPC (impregnated with MMA monomer either in the oven-dry state or at 14% m.c.) were irrigated on the slides and under cover slips with water,

water ethanol mixtures (30% and 75% ethanol), absolute alcohol and MMA monomer. Changes in radial and tangential dimensions were recorded after 15 minutes in each test liquid. Entire blocks of untreated and WPC material of spruce and Scots pine were impregnated with water under vacuum for 10 minutes, and kept in water at atmospheric pressure for at least 7 weeks. Dimensional changes were recorded at regular intervals.

RESULTS AND DISCUSSIONS

Anatomical aspects

In spite of the fact that spruce is known to be very refractory to the penetration of liquids, MMA appeared to impregnate the wood quite thoroughly. The histological distribution of the polymer appeared to follow a normal pattern. Longitudinal and ray-tracheids were fairly well filled with the plastic, but ray parenchyma cells were usually devoid of polymer (Plate 1A–D). Ray parenchyma cells were only filled with plastic at the periphery of the test blocks. Radial and longitudinal resin ducts were found to contain very little polymer, and the epithelium cells always remained 'empty'.

The light microscopical and SEM study revealed a diversity of shapes or forms of the polymer in the lumina of the tracheids. In transverse sections the plastic often follows the outline of the lumina, but rectangular shapes, deviating from the slightly rounded lumina were also found. In the case of an incomplete filling of the lumina, there may be cavities in polymer masses of these shapes, but the hollow polymer masses may also show collapse resulting in a gelatinous fibre-like image as seen in transverse section. The cavities are probably caused by local inequalities in the thermal conductivity of the composite being formed; these result in accelerated polymerization ('hot spots'). In the vicinity, a lower vapour pressure then arises, and the uneven evaporation of the monomer may cause bubble formation (Napjus, 1963). Bordered pit pairs play an important part in inter-tracheidal bubble formation: frequently two bubbles in the pMMA filling of adjacent tracheids were found to be connected via bordered pit pairs (Plate 2A). This indicates a good free passage of monomer vapour through at least part of the pit membrane. It was deduced from microscopical observations that no monomer transport occurs through aspirated pit pairs. Plate 2C illustrates a well-filled and an empty lumen of two adjacent tracheids with an aspirated pit in between. In the case of non-aspirated bordered pit pairs the polymer contents of adjacent tracheidal elements was always found to be connected by polymer 'replicas' of the pit cavities. Plate 2B shows such a polymer connection in longitudinal tracheids after maceration. Plate 2D shows a similar situation for ray-tracheids. If we consider that in normal industrially dried heartwood (*c.* 15% m.c.) of spruce, 1–9% of the radial bordered pit pairs are not aspirated in the early wood (Philips, 1934; ter Welle & Laming, 1972), and 5–30% of these pits are not aspirated in the late wood (Liese & Bauch, 1967; Laming & ter Welle, 1970), and that in

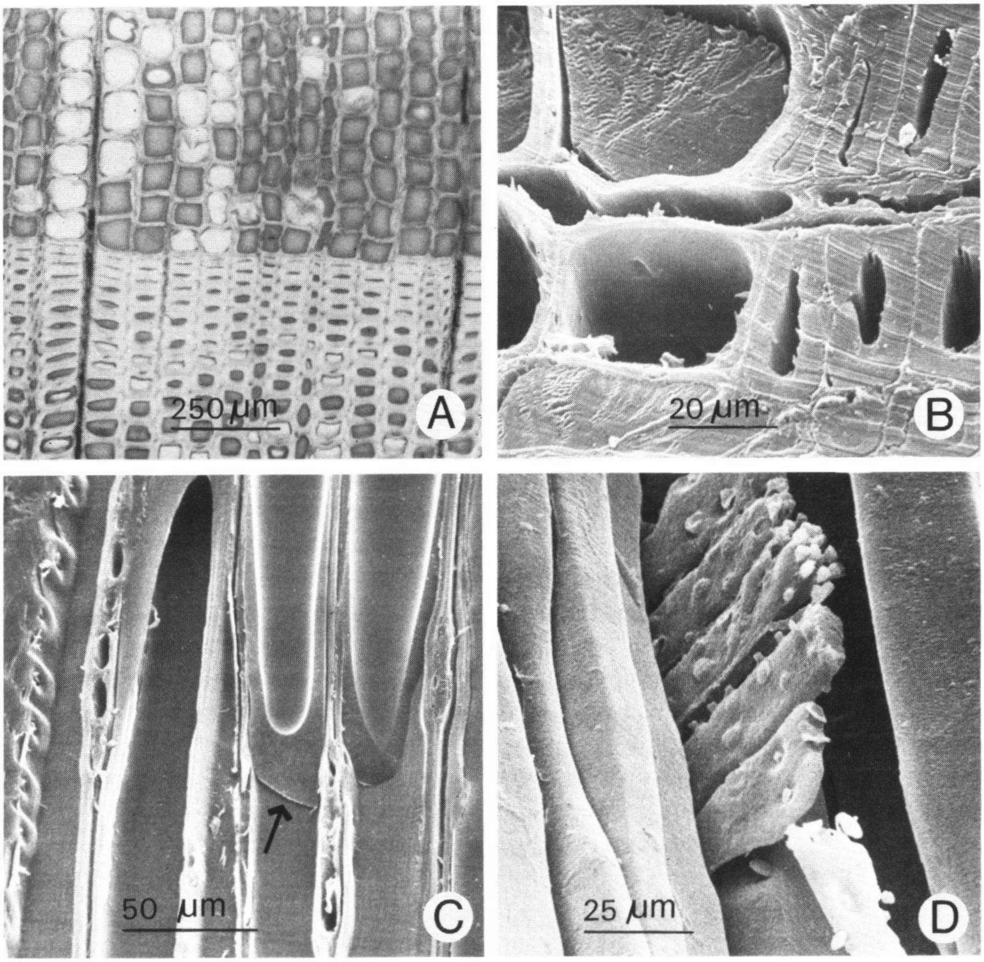

Plate 1. —A. Transverse section of spruce WPC at growth ring boundary. The greater part of the lumina are filled with pMMA (stained with Sudan III).—B. Transverse view (SEM) of spruce WPC at growth ring boundary. Both in latewood and in early wood there is some space between polymer plugs and the cell wall. The ray parenchyma cells are free from polymer.—C. Tangential view of spruce WPC(SEM). On the left a wall with bordered pits separating an empty lumen and a lumen filled with pMMA (left). Note the longitudinally sectioned pMMA plugs (upper middle) containing bubbles, and also the shrinkage of the polymer away from the cell wall (arrow).—D. SEM view of the outer-most area of spruce WPC, after the wood substance has been removed and the plastic skeleton left behind. From the skeleton it is clear that the ray parenchyma cells were here permeable to MMA monomer.

the specific late late-wood-zone (last 6–8 cell rows) 25–75% of these pits are not aspirated (ter Welle & Laming, 1972), one can imagine that in the major part of the wood only few possibilities are present for connections of the pMMA skeleton. This skeleton (see also Plate 3A) probably forms the basis for increased dimensional stability in the tangential direction (see also below). For radial dimensional stability the situation is

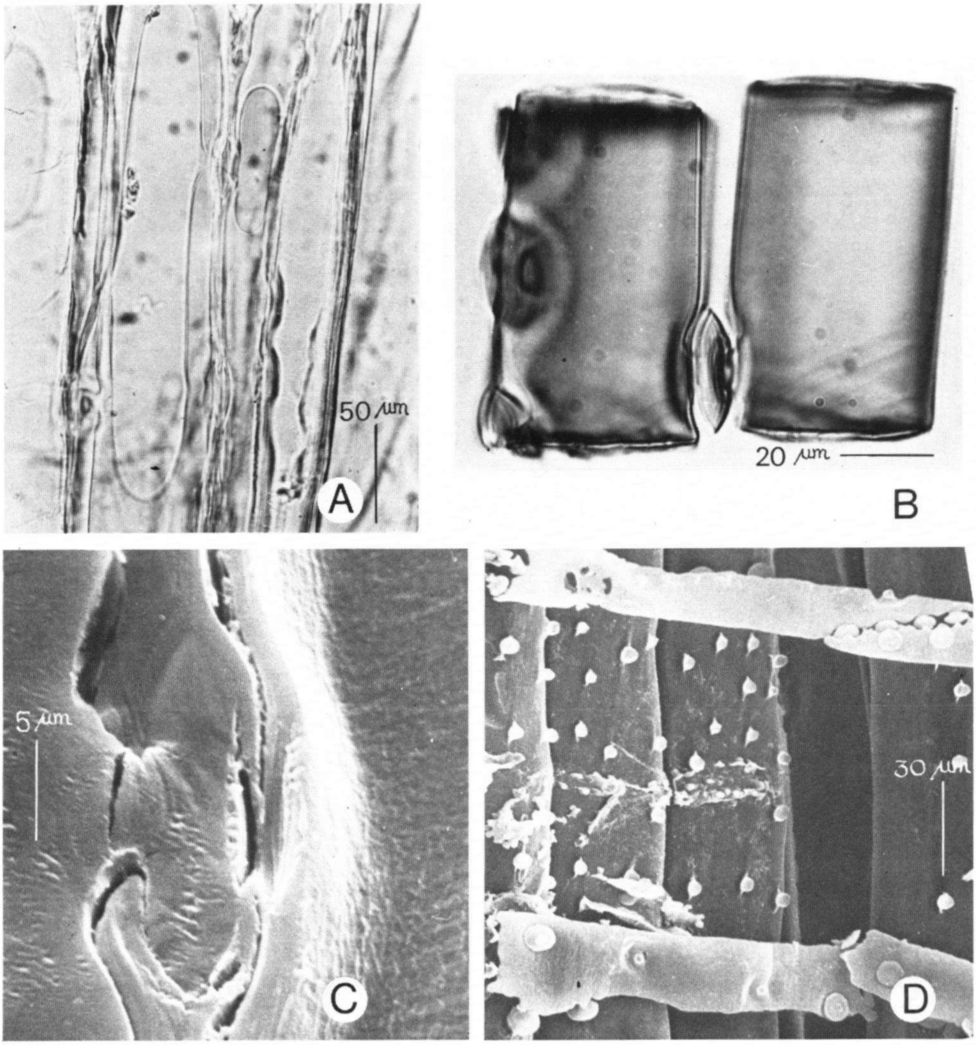

Plate 2.—A. Tangential longitudinal section of spruce WPC, demonstrating bubbles in the pMMA which are connected through bordered pit pairs.—B. Tracheidal lumina plugs of pMMA after removal of the cell wall material in spruce WPC, connected by the polymer filling of an unaspirated bordered pit pair.—C. Tangential view of spruce WPC (SEM) in the latewood zone. The polymer filling in the left lumen and in the pit cavity seem to be disrupted. The lumen on the right does not contain pMMA due to pit aspiration.—D. SEM view of ray area in spruce WPC after removal of cell wall material. The ray parenchyma cells have disappeared, but the polymer plugs of the ray-tracheids are held in position on account of the non-aspiration of most bordered pit pairs between longitudinal and ray-tracheids. Note also connections between the ray-tracheids (upper right).

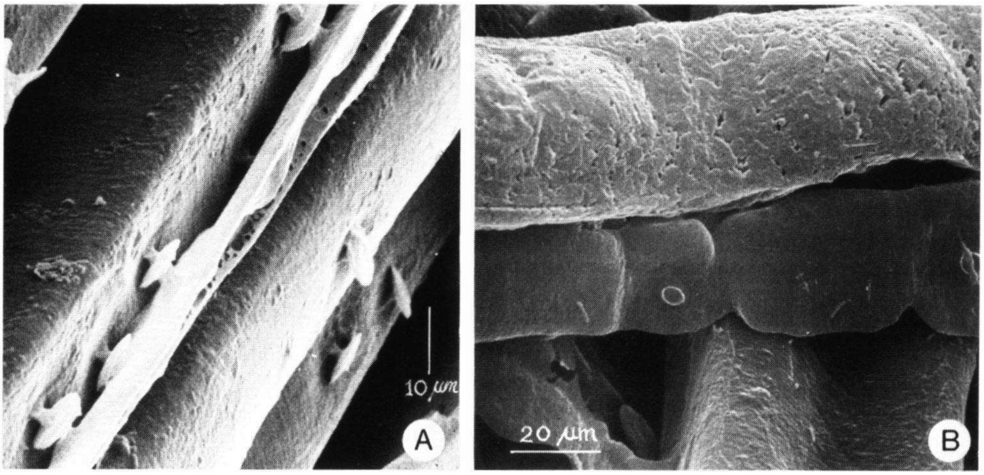

Plate 3.—A. SEM view of polymer plugs in tracheids in late latewood zone of spruce WPC after removal of cell wall material. The small tangential bordered pit pairs serve as a connection between the tracheidal pMMA fillings. The tracheid in the middle is only partially filled with a layer of polymer.—B. SEM view of a ray in the WPC of Scots pine sapwood after removal of cell wall material. The upper plug is of a ray parenchyma cell with abnormal texture of the polymer; the lower plug is of a ray-tracheid showing profile of denticles.

probably more favourable in this respect, because in industrially seasoned heartwood of spruce 80% of the bordered pits in the tangential walls of longitudinal tracheids, and 40–95% of the bordered pits between ray- and longitudinal tracheids are unaspirated (ter Welle & Laming, 1972).

The lumina of the tracheidal elements were practically never completely filled with polymer. A complete filling resulting in a close overall contact of cell wall and polymer is, however, not to be expected, because MMA shows a volume shrinkage of at least 20% during polymerization (Fengel, 1968; Nobashi & Yokota, 1974). Since pMMA is an isotropic substance the linear shrinkage will be *c.* 7% in all directions, including the longitudinal one. However, untreated spruce shows only a longitudinal shrinkage of 0.15–0.20% if dried from the green state to 10% m.c.: a very small fraction of the polymer shrinkage. It is to be anticipated that the considerable differences in directional as well as volumetric shrinkage between cell wall material and the polymer (regardless of the fact whether impregnation is effectuated at 14% m.c. or in the oven-dry state) will result in forces causing micro damage to the wood elements especially to the bordered pits between longitudinal tracheids and between ray- and longitudinal tracheids. No observations were, however, made to substantiate this hypothesis.

Normal light microscopical studies of stained WPC sections did not give any qualitative indication of the presence of polymer in the cell walls. The interference microscopical study gave calculated values for the volume percentage of polymer in the cell wall ranging between -7 and $+12\%$, with an average of 3.6% (see also Laming &

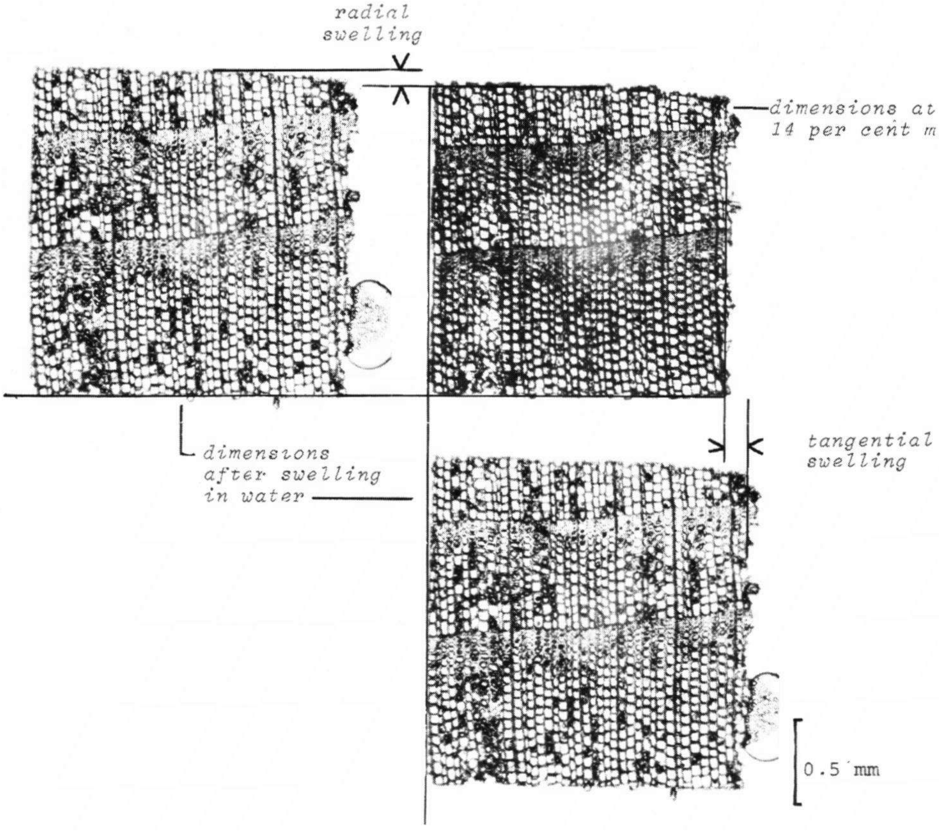

Plate 4. Transverse sections of spruce WPC, demonstrating method for measuring radial and tangential dimensional changes.

Keijzer, 1969). It should be stressed once more that these figures apply to the S_2 of the tracheid cell walls, and although the bulk of these cell walls (78%, *fide* Harada, 1965) is composed of this layer, our figures do not take into account any pMMA located in S_1, S_3 and compound middle lamella. The highly variable volume percentages reported here are probably greatly influenced by fluctuations in section thickness. Since, however, serial sections were used, the average value is more reliable than the great range of values would suggest.

In the literature no data are available for direct comparison with our results. The 2.9% volumetric cell wall impregnation reported by Timmons *et al.* (1971) for unextracted latewood of Eastern white pine (*Pinus strobus* L.), applies to the polymer of 2.5-dichlorostyrene and is therefore not comparable although the figure is of the same order of magnitude. Siau & Meyer (1966) calculated from swelling behaviour that in a WPC constituted of pMMA and yellow birch (*Betula alleghaniensis* Britt.) 3.5% of the total amount of polymer present is located in the cell wall. Assuming a complete filling of lumina, and densities of 0.65 and 1.50 of the timber and cell wall material respectively one can calculate a % vol. of polymer in the cell wall of 4.9%. Assuming more realistic incomplete filling of the lumina a somewhat lower % volume follows (*c.* 4%). This figure seems quite in agreement with the results for spruce, but it should be stressed that the value for yellow birch is entirely based on inference and therefore not wholly reliable. Contrasting with these values are the results for MMA and beech (*Fagus sylvatica* L.) obtained by Baas (1975) with exactly the same interference microscopical method as used in this study. He found a % vol. of 18 in the S_2 of fibre-tracheid walls, but considered the % vol. of 12, found after soaking sections in monomer a more realistic approximation, because of inherent sources of error due to changes of refractive index of substances in different systems. The differences between our low values and Baas' high percentage volumes cannot be fully explained. Two points should, however, be stressed to illustrate that Baas' data on beech and our results are not directly comparable. In the first place, Baas used very small test pieces, often sections, for determining the maximum accessibility of the cell wall to liquids. Our studies aimed at the estimation of polymer in technologically impregnated samples. Secondly, the interference microscopic calculations are applicable to optically isotropic situations only; for the S_2 the equations are therefore only valid if the microfibrillar angle is very steep and the sections are cut at right angles to the microfibrillar direction. This situation is well approximated in beech, but probably less so in spruce (Baas, personal communication). Wardrop & Preston (1951) recorded an average microfibrillar angle of 20° to the vertical in the S_2 of spruce tracheids.

Swelling behaviour

Swelling tests using transverse sections

Thin transverse sections (6–7 μm) were used in order to study the influence of polymer on the tangential and radial swelling of the cell walls of longitudinal tracheids (Plate 4). Rays are generally held responsible for much of the differences between tangential and radial swelling (Kelsey, 1963; Schniewind, 1959; Stamm, 1964), and by using thin sections their restraining influence on the ground tissue is minimized.

Table 1 gives the results of these swelling tests in different liquids. In contrast to wood blocks, the cell wall material in sections is readily accessible for liquids and swelling in water is for instance completed within a few seconds. Swelling in water-ethanol mixtures takes rather longer. Both in pure ethanol and in MMA monomer no swelling was observed in sections of untreated wood. This is highly remarkable, since Stamm (1964) mentioned that pure ethanol does swell wood to a considerable extent (83% of the swelling in water). Baas (1975) also found the cell wall of untreated beech fairly accessible to both ethanol and pure MMA. Beall & Witt (1974), Laidlaw (1967), and Laidlaw *et al.* (1967), moreover, reported that MMA does swell solid wood blocks.

TABLE 1. Swelling determinations of cross-sections of spruce and of spruce wood-plastic (pMMA) combination in several liquids

m.c. = moisture content; f.s.p. = fibre saturation point.
Number of sections tested is given between brackets.

	cross-sections of		dimensional change in % of dimensions at	average dimensional change caused by swelling after 15 minutes soaking in:				
				water	ethanol 30%	ethanol 75%	ethanol absolute	MMA monomer
swelling from 14 % m.c. to f.s.p.	untreated wood	rad. tang.	14 % m.c.	5.7 9.0 (12)	4.8 8.2 (5)	5.9 6.7 (5)	0 0 (4)	0 0 (4)
	impregnated with MMA monomer at 14 % m.c.	rad. tang.	14 % m.c.	6.0 8.4 (15)	4.5 7.2 (4)	4.9 7.2 (5)	0 0 (5)	
	impregnated with MMA monomer at oven-dry state	rad. tang.	14 % m.c.	5.4 7.2 (15)	5.0 7.0 (4)	7.4 8.7 (4)	0 0 (4)	
swelling from oven-dry state to 14 % m.c.	untreated wood	rad. tang.	oven-dry state	2.2 2.8 (10)				
	impregnated with MMA monomer at oven-dry state	rad. tang.	oven-dry state	1.7 2.2 (10)				
swelling from oven-dry state to f.s.p.	untreated wood	rad. tang.	oven-dry state	6.6 9.6 (10)	7.0 10.0 (4)	6.1 8.3 (5)	0 0 (4)	0 0 (4)
	impregnated with MMA monomer at 14 % m.c.	rad. tang.	oven-dry state	7.2 9.8 (15)	6.3 8.7 (4)	6.2 9.3 (4)	0 0 (4)	

In comparison with sections of untreated wood, tangential swelling in water from 14% m.c. is less in WPC sections of material impregnated at 14% m.c. and is least in material impregnated in the oven-dry state. However, radial swelling is much the same in these three different categories, and the differences in tangential swelling are only slight. Both radial and tangential swelling from the oven-dry state to 14% m.c. is slightly less in WPC material than it is in untreated wood. The dimensional changes from the oven-dry state to full saturation with water are again much the same in WPC material and untreated wood. The differences in swelling in the two water-ethanol mixtures are not consistent if one compares values for the two kinds of WPC and untreated material. Part of the differences in the above swelling values may also be due to different proportions of late wood in different sections used for testing. Although based on limited observations, the data from Table 1 are suggestive of hardly any change in water accessibility of the cell wall after impregnation with MMA and subsequent polymerization. This agrees with data on beech (Baas, 1975).

Swelling tests using wood blocks

In the last decade much research has been done on the influence of pMMA in wood on dimensional stability in conditions, more or less comparable to humidity fluctuations in service (van der Elburg, 1971; Gibson *et al.*, 1966; Jokel *et al.*, 1974; Laidlaw, 1967; Laidlaw *et al.*, 1967; Pullmann *et al.*, 1974; Villière, 1969). The results usually indicate decrease in rate of water or water vapour sorption, and the ultimate volumetric swelling is often reduced. In other words combinations of wood and pMMA usually have a better antishrink efficiency as compared to untreated wood. In order to gain information about the influence of the polymer on the dimensional stability of Scots pine and spruce under extreme conditions (vacuum impregnation with water, followed by prolonged soaking), a number of test pieces of WPC and untreated wood were compared.

In spruce, maximum radial and tangential swelling of untreated wood is reached within one day. WPC material impregnated at 14% m.c. reached maximum dimensions within one week and then showed a slight decrease in both tangential and radial dimensions until after three weeks the measurements remained constant. Spruce impregnated in the oven-dry state and afterwards conditioned at 65% relative humidity shows much the same swelling behaviour as untreated wood, with the exception that maximum tangential dimensions are only reached after 1½ weeks.

In Table 2 the data of these experiments are summarised. It appears that wood impregnated at 14% m.c. takes longest to reach maximum dimensions and that the ultimate tangential swelling is less than in the untreated wood or WPC impregnated in the oven-dry state. It is remarkable that sizes may decrease again after one week of soaking in water. This is shown in Table 2 for the material impregnated at 14% m.c. The ratio between tangential and radial swelling (T/R ratio) fluctuates more in material impregnated at 14% m.c. than in other material. Tangential antishrink efficiency is

TABLE 2. Swelling of untreated wood and WPC blocks of spruce and Scots pine in water, after vacuum impregnation and prolonged soaking

Swelling values expressed in percentages of dimensions at 14% moisture content (m.c.). T/R ratios are given between brackets.

		SPRUCE			SCOTS PINE		
Soaking period		untreated	impregnated with monomer		untreated	impregnated with monomer	
			in oven-dry state	at 14 % m.c.		in oven-dry state	at 14 % m.c.
1 day	rad. tang.	3.2 6.2 (1.9)	3.3 5.0 (1.5)	2.4 2.7 (1.1)	3.3 6.2 (1.9)	2.2 3.1 (1.4)	3.5 5.1 (1.5)
1 week	rad. tang.	3.2 6.3 (2.0)	3.3 5.4 (1.6)	3.1 4.6 (1.5)	3.1 6.4 (2.1)	4.2 5.8 (1.4)	4.5 6.1 (1.4)
4 weeks	rad. tang.	3.0 6.5 (2.1)	3.4 5.6 (1.8)	2.5 4.9 (2.0)	3.2 6.3 (2.0)	4.3 6.5 (1.5)	4.7 6.2 (1.3)
7 weeks	rad. tang.	3.1 6.5 (2.1)	3.3 5.6 (1.8)	2.3 3.8 (1.7)	3.1 6.3 (2.0)	4.2 6.5 (1.5)	4.6 6.2 (1.3)

highest in this material. Radial antishrink efficiency is also better in WPC impregnated at 14% m.c. than in material impregnated in the oven-dry state.

Similar tests on Scots pine sapwood gave different and unexpected results (see Table 2). Maximum radial and tangential swelling in wood impregnated at 14% m.c. is reached in one week. Wood impregnated in the oven-dry state takes $2\frac{1}{2}$ weeks to reach maximum tangential dimensions. Most striking, however, is the fact that ultimate tangential swelling is much the same in all three categories, and that radial swelling of WPC blocks even exceeds that of untreated wood. This is more or less in agreement with recent experimental data presented by Burmester & Wille (1976). They found that tangential swelling of the WPC of Scots pine sapwood is equal to or even supersedes the swelling of untreated wood considerably, depending on pMMA loading. The striking differences in this respect between spruce and Scots pine must probably be ascribed to differences in ray anatomy and in chemical composition. In the hard pines (including Scots pine) the lignification of the ray parenchyma cell walls does not take place until the transition from sapwood to heartwood (Balatinecz & Kennedy, 1967). In this transition zone phenolic substances migrate from the ray parenchyma through the pit membrane into the cavities of the bordered pit pairs and subsequently into the lumina of the tracheids (Fengel, 1970). Recent studies (Nobashi & Yokota, 1974) have demonstrated the inhibiting influence of phenolic compounds on polymerization of MMA. Plate 3B illustrates the deviating texture of polymer in a ray parenchyma cell of Scots pine. These considerations may lead to the hypothesis that in the WPC of Scots pine there is no interaction of pMMA and the parenchymatous cell walls and probably also none with the bordered pit pairs, so that water may enter the inner cell wall layers without any hindrance. This does, however, not explain the swelling behaviour of treated wood as compared to untreated wood.

The swelling tests reported here largely confirm previous studies on the influence of pMMA on dimensional stability. The polymer delays the swelling, which must be ascribed to slower water penetration in the wood-plastic composite due to the presence of polymer in the lumina. The results on spruce evidencing increased tangential dimensional stability indicate that the inter-tracheidal polymer connections through the radial unasperated bordered pit pairs play an important part in a mechanical hindrance to tangential swelling. Plastic connections through tangential bordered pit pairs and in ray tracheids probably also play a part, but a less important one, in establishing increased radial dimensional stability.

CONCLUSIONS

(1) The anatomical studies have demonstrated that there is no close contact between the pMMA and the cell wall in spite of a fairly good filling of tracheid lumina and of cavities of bordered pit pairs. This is most probably due to the isotropic shrinkage of the plastic during polymerization. The cell walls thus remain accessible to water. As-

pirated bordered pit pairs are impermeable to MMA monomer. The polymer skeleton is therefore of a loose character in regions with a high proportion of aspirated bordered pits. The only possibility of inter-tracheidal polymer connections is offered by unaspirated bordered pits.

(2) Determination of the fraction of MMA polymer in the cell wall of WPC of spruce, impregnated at 14% m.c., with an interference microscopical method indicated that *c.* 3.6% vol. of the S_2 of the longitudinal tracheids is composed of the polymer.

(3) In thin transverse sections, releasing the longitudinal tracheids from restraints by ray cells, swelling tests indicated that the presence of pMMA in the cell wall does not appreciably reduce lateral swelling if WPC and untreated wood are compared.

(4) In long term swelling tests of entire blocks, WPC of spruce shows increased dimensional stability, particularly in the tangential direction. This increased antishrink efficiency is stronger in spruce impregnated in the air-dry state (at 14% m.c.) than in spruce impregnated in the oven-dry state. In Scots pine tangential swelling after prolonged soaking in water is not at all affected by the presence of pMMA, and radial swelling of WPC is even appreciably greater than in untreated wood.

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