

Comparative study on penetration characteristics of modern wood coatings

M. de Meijer, K. Thurich, H. Militz

347

Summary The penetration characteristics of five modern wood coatings (three waterborne, one high solid and one solvent borne) into pine sapwood, spruce and dark red meranti have been systematically compared. The degree of coating penetration is mainly determined by the ability of the coating to flow into wood capillaries. Binder type, pigmentation, solid matter content and drying speed appeared to influence this ability. In softwoods the following different coating penetration routes are observed: the flow into open ends of longitudinal early- and latewood tracheids, the flow into ray cells and the transport from rays through the cross-field into longitudinal tracheids adjacent to rays. The possibility for the coating to follow the latter route is strongly influenced by the existing type of cross field pitting and to a lesser degree by the pigmentation of the paint. Clear differences between pine and spruce have been found with respect to the flow into ray parenchym and ray tracheids. The flow into open ends of longitudinal tracheids is strongly influenced by the grain angle of tracheids. Penetration into dark red meranti is mainly limited to vessels and rays. Tylose membranes can prevent the complete filling of vessels. The impact on penetration of the removal of extractives and of sanding of the surface has also been studied but appears to be of only minor importance.

Introduction

In the joinery industry of Western Europe a combination of two new developments has become increasingly important. The first is the protection of the wood

Received: 23 June 1997

M. de Meijer, K. Thurich
Foundation for Timber Research SHR, P.O. Box 497,
NL-6700 AL Wageningen, The Netherlands

H. Militz
Wageningen Agricultural University, Department of Forestry,
Section of Wood Science, P.O. Box 342 NL-6700 AH Wageningen,
The Netherlands

Correspondence to: M. de Meijer

The authors want to thank Akzo Nobel Coatings, DSM Resins and Sigma Coatings for their technical support. This research was financed by the the Dutch Innovative Research Program on Coatings under contract number IVE 93-812. Authors also thank the University of Hamburg (BFH) for the use of their electron microscopy facilities.

by different types of waterborne and high solid coatings instead of the traditionally used solvent borne alkyd paints and stains. This development is mainly encouraged by the need to avoid the emission of air polluting volatile organic compounds used as solvents. The second development taking place is the replacement of tropical hardwoods by softwoods, which are more sensitive to water, less durable and less dimensionally stable. For these reasons a more fundamental understanding of the interaction between wood and modern coatings is needed to improve their durability and performance. The penetration of a primer into the wood can be important for several reasons. Firstly the penetration of coatings can improve water repellency and dimensional stability and reduce wood cracking, as was shown in several studies on the impregnation of wood with waterborne resins comparable to those used in coatings (Militz and Peek 1993, Kabir et al. 1992, Smulski and Côté 1984). Secondly it seems to be very likely that the adhesion of the coating to the wood will benefit from a certain degree of penetration as well. Modern theories on adhesion (Allen 1987, Pizzi 1992) have distinguished several mechanisms like mechanical entanglement or interlocking, adsorption interactions by polar and dispersive forces and intermolecular diffusion. Mechanical entanglement is directly related to penetration, but the other mechanisms will benefit from penetration as well due to the increase in contact area between coatings and the cell walls. Thirdly coatings containing fungicides might have improved penetration of the fungicide because it penetrates together with the binder vehicles (Rødstrud and Sutcliffe 1994).

In the past the microscopic penetration patterns of solvent borne alkyd and linseed oil paints into wood have been intensively studied by several authors (Haslam and Werthan 1931, Wallenfang 1964, van Loon 1966, Schneider and Côté 1967, Côté and Robison 1968, Schneider 1970, Schneider 1979, Schneider 1980). More recently studies have been made on the penetration of waterborne alkyd emulsions (Smulski and Côté 1984, Nussbaum 1994). Rødstrud and Sutcliffe (1994) have compared the penetration of coatings based on waterborne acrylic dispersions, waterborne alkyd emulsions and solvent borne alkyds. In these studies the most commonly used techniques to study the microscopic distribution of binders and pigmented paints within the wood cells are: scanning electron microscopy of normal (Schneider 1979) and replica samples (Schneider 1970, Smulski and Côté 1984), fluorescence microscopy (van Loon 1966, Schneider and Côté 1967, Côté and Robison 1968), autoradiography with ^{14}C labelled coatings (Nussbaum 1994) and scanning electron microscopy with X-ray analysis (SEM-EDAX) of brominated alkyds (Nussbaum 1994).

The observed gross penetration patterns in softwoods are: filling of the outer layers of tracheids and radial penetration into the rays. In hardwoods the penetration mainly consists of filling the capillaries of the vessels and ray cells. Penetration of the cell walls by the coating is subject to many discussions in literature but have not been proven unambiguously. Penetration of the coating into the cell wall might be limited to the solvent (Côté and Robison 1968) or to small amounts of linseed oil (Schneider 1979, Schneider and Sharp 1982).

The objective of the present study is to systematically compare the penetration of five different types of coatings (three waterborne, one high solid and one reference solvent borne alkyd) with and without the addition of pigments. The mechanisms of penetration are studied qualitatively by fluorescence and electron microscopy on spruce, pine sapwood and dark red meranti. The influence of drying speed, surface preparation and extractive removal have been included in this study as well.

Materials and methods

Coating materials

Five types of binders were used in this study: two waterborne acrylic dispersions with different particle sizes (WAD100 and WAD300), one alkydemulsion (WBA), one alkyd-based high solid (HSA) and one traditional solventborne alkyd resin (SBA). The different types of binders were used in opaque model paints and as unpigmented solutions of binders. The opaque model paints were pigmented white with titanium-dioxide at a pigment volume concentration of 17% and formulated in the simplest possible manner. The binder solutions contained only the binder, water or organic solvent and about 1 weight % coalescent which improves film formation of the the acrylic dispersions or about 2 to 4 weight % cobalt-siccative which catalyses the oxidative drying of the alkyd resin. The most relevant properties of the coatings are given in Table 1.

Additionally binder solutions without coalescent or drier were tested to study the influence of the drying-speed. Part of the binder solution was also tested at a solid matter content of 10–20% below the standard amount of solids. All binders contained about one weight-percent anthracene dye to allow detection by fluorescence microscopy. The dye is chemically bonded to the binder molecules and is equally distributed over the molecular weight distribution of the binder polymer which was proven by gel-permeation-chromatographic measurements where binder and dye could be detected separately.

Wood species and sample preparation

Industrially pre-dried pine sapwood (*Pinus sylvestris*), spruce heartwood (*Picea abies*) and dark red meranti heartwood (*Shorea spp.*) were used. All the wood was straight grained, free from defects and was stored at 65% RH and 22 °C before coating application. To study the penetration into radial and tangential direction for every combination of coating and wood species, three planed blocks of 70 × 70 mm (height × width) were used for spruce and pine, and three blocks of 45 × 45 mm for dark red meranti. Spruce and pine had one pure radial and one pure tangential surface, on the other two surfaces the growth-ring angle was between 20° and 40°. Growth-ring width was between 0.5 and 2.5 mm for pine and around 1 mm for spruce. The dark red meranti had growth-rings purely parallel to the surface on two sides, radial to the surface on the other two sides.

The influence of extractives was studied by comparing the penetration in normal and extractive-free cubic wood blocks of 10 mm with the coating applied on the radial and tangential surface, taking care to avoid longitudinal penetration. Extractive-free wood was prepared by 16 hours of soxhlet-extraction with successively cyclohexane-ethanol, ethanol and water (according to ASTM-standard D1105).

Samples with a controlled angle between the length-axis of the tracheids and the surface were prepared as follows: pine panels were cut and hand-planed with the grain exactly parallel to the outer surface. Parts of the panels were further planed to an angle of 5 and 10°, see also Fig. 1. The angle between surface and tracheids was checked on microscope slides taken from the samples. In this case only the pigmented solvent borne alkyd paint (SBA) was applied because it has the highest penetrating capacity.

All coatings were applied in one or two layers by brush without prior sanding. The average spreading rate was approximately between 120 and 200 g/m². Additionally the influence of surface preparation was studied by comparing the penetration into spruce panels with the following pre-treatments: only planing,

Table 1. Properties of binders and coatings used in the experiments

Type of binder	Solvent type	Molecular weight		Particle size nm	Solid content weight %		
		Min g/mol (number average)	Mw g/mol (weight average)		Pigmented coating	Not pigmented	
WADI00 acrylic dispersion	water	approximately 500.000		100	56	31	23
WAD300 acrylic dispersion	water	approximately 500.000		300	61	32	19
WBA alkyd emulsion	water	1.800	50.000	500	55	37	21
HSA high solid alkyd	white spirit + reactive diluent	3.000	7.000	solution	90	85	61
SBA solvent borne alkyd	white spirit	4.000	109.000	solution	59	45	29

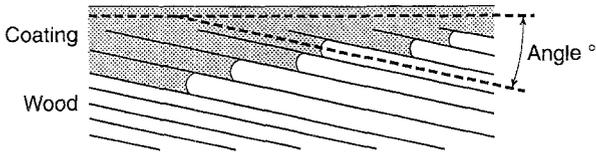


Fig. 1. Angle between length axis of the longitudinal tracheids and the surface

planing followed by sanding with coarse (120) sandpaper, planing followed by sanding with coarse and fine (180) sandpaper and coarse and fine sanding followed by intensive cleaning by air-blowing. These four treatments were studied in combination with the pigmented coatings WAD300, WBA and SBA.

Microscopic analysis

For every combination of coating and wood seven radial and seven axial samples were cut out of the blocks and prepared to study the penetration into the wood. All samples were flattened on a microtome and examined with incident light fluorescence microscopy. The fluorescent light-source was a 50 W Mercury or 100 W Halogen lamp with a 300–400 nm exciting filter. Additionally, transmitted light microscopy was used to study penetration into sliced samples of coated wood. Scanning electron microscopic studies were made with a Hitachi S-520 SEM on gold-coated samples. The SEM was equipped with a Kevex 7100 X-ray analyser (EDAX) operated at 4.35–4.68 keV to detect titanium presence in the pigment.

Results and discussion

Penetration of pigmented paint into pine and spruce

The planed softwood surfaces all basically showed the following structure as it was observed on axial cross-sectional surfaces. On the wood surface the thick-walled latewood cells are mainly intact although sometimes slightly compressed whereas the thin-walled earlywood cells are cut open during planing. The latter implies that there is an open connection between the lumina and the surface on which the coating is applied. When rays end on the surface there is also an open connection with the ray cells, both to ray-tracheids and ray-parenchyma. The extent of coating penetration is firstly controlled by the ability of the liquid coating to flow into the open ends of the cells and secondly by the possibility of coating transport from cell to cell through the interconnecting pits. In the following text the penetration patterns observed are described in more detail for the coatings studied on both pine sapwood and spruce heartwood. An overview of the penetration patterns is given in Table 2.

The pigmented WAD300 and WAD100 hardly show any penetration into neither pine or spruce. Only the outer earlywood cells which are cut open over the full length are filled. Occasionally paint is found one cell layer below the surface in the earlywood. Here the paint flows over a short distance into the open end of a longitudinal tracheid (see Fig. 2). The degree of this type of axial penetration of WAD100 is slightly less than with the WAD300. If the paint flows axially into the lumen only a thin layer covering the cell wall is present. Penetration in ray cells is limited to 10 μm with a preference to the ray parenchym cells. Although penetration in terms of transport from cell to cell is absent the increase in contact area

Table 2. Overview of the observed penetration patterns

	Type of binder					SBA solventborne alkyd
	WAD300 acrylic dispersion	WAD100 acrylic dispersion	WBA alkyd emulsion	HSA high solid alkyd		
Pigmented coating						
pine	outer tracheids in earlywood	only filling outer cells seldom one cell row below surface	only filling outer cells very seldom one cell row below surface	filling outer cells and often first cell row below the surface	filling outer cells and up to three cell rows below surface	filling outer cells and first (sometimes second) cell row below surface
	outer tracheids in latewood	no filling lumina coating only on surface	no filling lumina coating only on surface	no filling lumina coating only on surface	occasionally also some filling of latewood cells	no filling lumina coating only on surface
	ray parenchyma	limited to depth of 10 µm	no penetration	penetration up to 150 µm	deep penetration to depth of 1000 µm	deep penetration to depth of 1000 µm
	tracheids	limited to depth of 10 µm	no penetration	limited penetration	penetration up to length of 1 tracheid	penetration up to length of 1 tracheid
	longitudinal tracheids adjacent to rays	no coating present	no coating present	some filling of latewood cells	frequent filling of latewood tracheids	frequent filling of latewood tracheids
spruce	outer tracheids in earlywood	only filling outer cells seldom one cell row below surface	only filling outer cells very seldom one cell row below surface	filling outer cells and often first cell row below surface	filling outer cells and up to three cell rows below surface	filling outer cells and first (sometimes second) cell row below surface
	outer tracheids in latewood	no filling lumina coating only on surface	no filling lumina coating only on surface	no filling lumina coating only on surface	occasionally also some filling of latewood cells	no filling lumina coating only on surface
	ray parenchyma	limited to depth of 10 µm	no penetration	penetration up to depth of 40 µm	penetration of the first cell	penetration of the first cell

tracheids	limited to depth of 10 μm	no penetration	limited penetration	penetration up to length of 1 tracheid	penetration up to length of 1 tracheid
longitudinal tracheids adjacent to rays	no coating present	no coating present	no coating present	no coating present	no coating present
dark red meranti	filling of vessels by tyloses	yes, if not limited by tyloses	yes, if not limited by tyloses	yes, also under tyloses	yes, if not limited by tyloses
	rays	no coating present	very limited penetration	penetration up to length of 1 ray cell	penetration up to length of 1 ray cell
	axial parenchyma and sklerenchym fibres	no coating present	no coating present	penetration of cells close to surface	no coating present
Unpigmented coating					
pine	outer tracheids in earlywood	filling outer cells and 2-3 cell rows below surface	filling outer cells and 1-5 cell rows below surface	filling outer cells and 1-3 cell rows below surface	filling outer cells and 1-2 cell rows below surface
	outer tracheids in latewood	no coating present	filling outer cells and limited filling 1-3 cell rows below surface	filling outer cells and 1-6 cell rows below surface	only filling outer cells very seldom one cell row below surface
	ray parenchyma	no coating present	penetration up to depth of 150-500 μm	very deep penetration up to 2000 μm	very deep penetration up to 2000 μm
	tracheids	no coating present	penetration up to length of 1 tracheid	penetration up to length of 1 tracheid	penetration up to length of 1 tracheid
	longitudinal tracheids adjacent to rays	no coating present	frequent filling of latewood tracheids	completely filling latewood up to 2 mm away from surface	frequent filling of latewood up to 2 mm away from surface

Table 2 (Continuation)

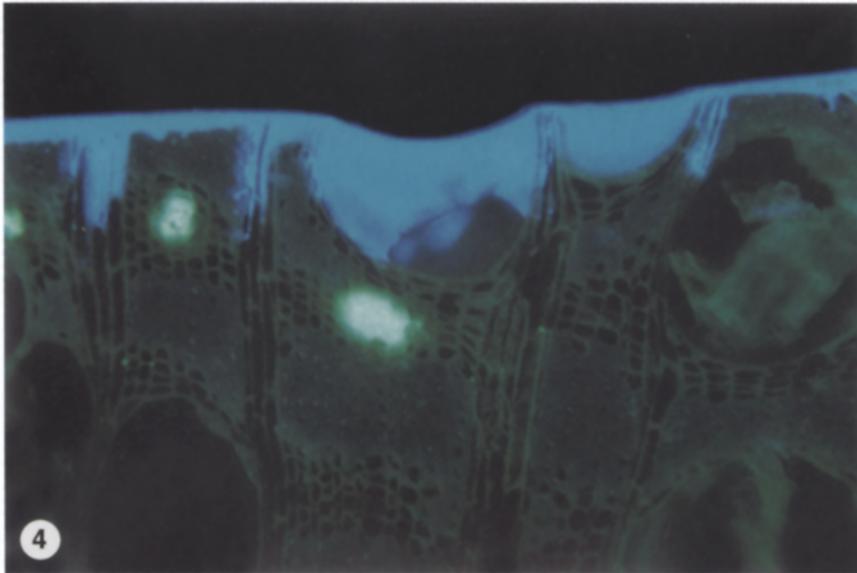
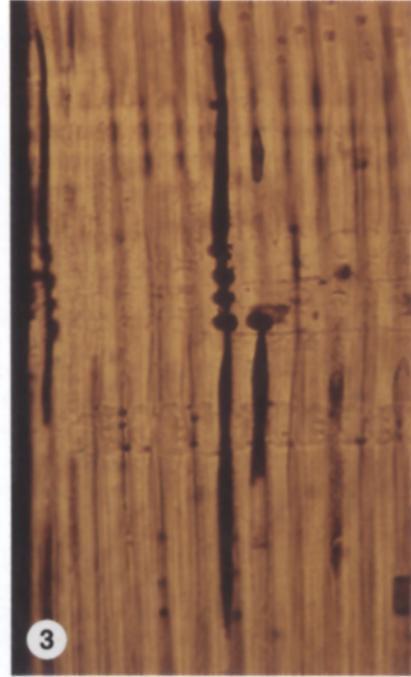
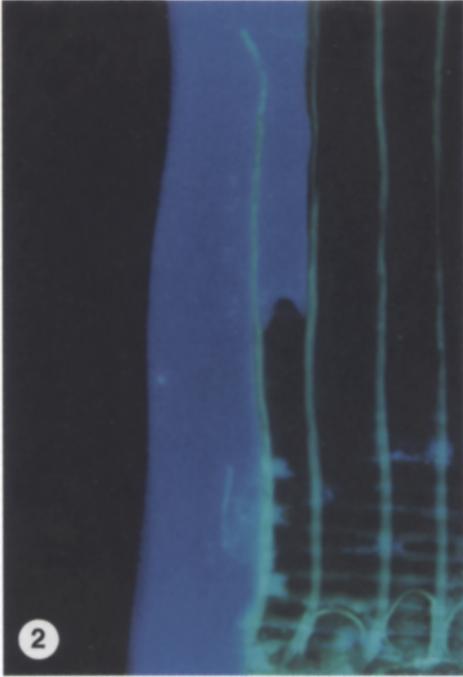
		Type of binder					
		WAD300 acrylic dispersion	WAD100 acrylic dispersion	WBA alkyd emulsion	HSA high solid alkyd	SBA solventborne alkyd	
Unpigmented coating							
spruce	outer tracheids in earlywood	filling 2-3 cell rows below surface	filling outer cells and often first cell row below surface	filling outer cells and 1-3 cell rows below surface	filling outer cells and 1-3 cell rows below surface	filling outer cells and 1-2 cell rows below surface	
	outer tracheids in latewood	no coating present	no filling lumina coating only on surface	limited filling 1-3 cell rows below surface	filling 1-6 cell rows below surface	no filling lumina coating only on surface	
	ray parenchyma	no coating present	no coating present	penetration up to depth of 40 μ m	penetration up to depth of 140-200 μ m	penetration of the first cell	
	tracheids	no coating present	no coating present	limited penetration	penetration up to depth of 1000 μ m	penetration up to length of 1 tracheid	
	longitudinal tracheids adjacent to rays	no coating present	no coating present	no coating present	frequent filling of latewood tracheids	no coating present	

between coating and cell wall in the early wood is considerable. Air bubbles entrapped in the coating were observed at the interface with the wood. The very limited penetration of the two acrylic coatings is in reasonable agreement with the findings of Rødsrud and Sutcliffe (1994) who reported no penetration for the acrylic coatings used in their study.

The pigmented alkyd emulsion WBA fills a major part of the first cell layer under the surface by paint flow into the lumina of the earlywood; latewood cells are not filled with paint. The WBA penetrates into the rays with a limited penetration into the ray tracheids. Penetration into the parenchyma cells is limited to about 40 μm for spruce but much deeper for pine where the coating penetrates to a depth of 150 μm . In pine some longitudinal earlywood tracheids adjacent to the rays are filled with paints showing that the paint flows from rays to longitudinal tracheids. The HSA and SBA paints fill the outer first and second layer of earlywood cells in spruce and the first three layers in pine although often empty cells are found in between (see Fig. 7). Penetration into the rays can be divided into penetration into ray tracheids which is limited to the length of a single tracheid and into penetration into ray parenchyma cells. The latter depends very much on wood species. In spruce the coating never penetrates from one parenchyma cell into another. This however does happen frequently in pine where the coating can penetrate to a depth of 1000 μm .

The HSA and SBA paints were also found in pine latewood cells adjacent to rays (Fig. 8) up to a distance of one growth ring away from the surface. In a radial cross-section (Fig. 3) it can be seen that the paint flows from the ray parenchym through the fenestriform pits into the longitudinal tracheids. A similar type of penetration from rays into latewood tracheids was found by Schulze and Theden (1942) for wood-preservatives applied on pine by brush and by Nussbaum (1994) for alkyd paints. That this phenomenon is only found in pine and not in spruce might be explained by differences in cross field pitting and the degree of pit aspiration. The fenestriform pine pits are much larger (pit diameter of 10 to 25 μm) than the piceoid and cupressoid pits in spruce (pit diameter of about 2–5 μm). The latter can be more easily clogged with agglomerates of pigment particles which have a single size of about 0.25 μm in case of the titanium dioxide used. Studies on alkyd resins penetrating Eastern white pine (Smulski and Côté 1984) also showed that fenestriform pits can be filled with resin. Why the paints only penetrate the longitudinal latewood tracheids is not fully clear. Although it is known that bordered pits in normal tracheids are less frequently aspirated in latewood compared to earlywood (Liese and Bauch 1967) it is not certain whether this is a good explanation here, because the half bordered pits between rays and longitudinal tracheids might not be aspirated in both early and latewood. At least pits between rays tracheids were found not to be aspirated (Liese and Bauch 1967). Another explanation for the differences in longitudinal penetration of early- and latewood adjacent to rays might be, that there are differences in capillary forces due to different sizes of the lumina in the longitudinal tracheids.

A remarkable observation in case of the HSA and SBA paint is the difference in colour of the paint penetrated into the wood. On the surface the colour is dark blue (typical for pigment and binder), whereas in the wood different shades of blue and white (typical for pure pigment) were observed. SEM-EDAX analysis of paint penetrated into the rays (see Fig. 10) proved that the pigment was present at depths of 180 μm but with fluorescence microscopy pigment was even observed at a depth of 1000 μm . A possible reason for the separation of pigment and binder could be the variation in molecular weight with a higher mobility of the lower



weight fractions, or the fact that binder not adsorbed to the pigment surface can penetrate more deeply into the wood than the binder that is used to wet the pigment particles.

As the flow from open ends into the longitudinal tracheids seems to be a very important factor in filling the outer cells, the following experiment was performed to prove the importance of this mechanism in more detail. For the combination of

◀
Fig. 2. Flow of pigmented WAD300 coating (dark blue) from the surface layer in open end of longitudinal tracheid in spruce. Microphotograph of radial cross section with incident fluorescent light (200×)

Fig. 3. Transmitted normal light microphotograph showing how the HSA coating (dark area) penetrates from the ray parenchyma cells into the longitudinal tracheids through the fenestriform pits (200×). The actual ray filled with the coating is out of the plane of the slide
Fig. 4. Axial cross-section showing penetration of SBA coating into dark red meranti (incident fluorescent light 100×). Note the incomplete filling of the vessel in the middle because of tylose membrane and completely filled vessel at the left, ray cell are also penetrated. Bright white spots are natural resin channels

pine sapwood and SBA coating the number of cell layers filled with coating was measured as a function of the angle between the surface and the length-axis of the longitudinal tracheids. As shown by the results presented in Table 3 no tracheids are filled at an angle of zero degrees. At a 5° angle already 5 cells or more are filled, whereas at a 10° angle a very large number of cells are filled in both early- and latewood (see Fig. 6). From these data it can be estimated that in normally prepared samples the angle was between zero and five degrees. The variation in angle between surface and length-axis of a tracheid can also explain why the penetration for one type of coating often varies from sample to sample.

Penetration of unpigmented binders into pine and spruce

In general the unpigmented binder solutions show the same basic penetration pattern as the pigmented coatings however, in general penetration is deeper. On the surface often no clear film is present, in particular if the coating has penetrated deeply. Table 2 gives an overview of the penetration pattern observed for each coating on both pine and spruce. In the following text the penetration is discussed in more detail including the influence of dilution and the presence of dryer or coalescent. The acrylic dispersion WAD300 forms a thin film on the latewood of pine and spruce and partially fills lumina in the earlywood up to 2 or 3 cell rows away from the surface. No differences were found between binder with and without coalescent. The diluted binder is very difficult to detect but is occasionally found in the lumina of earlywood tracheids. The acrylic dispersion WAD100 is found on the surface, in the first row of earlywood tracheids of pine and spruce and very seldom in latewood tracheids of pine, but not in spruce. The binder without the coalescent has a more irregular film formation on the surface which is consistent with the fact that the coalescent improves the film formation. For both the WAD100 and WAD300 no binder was found in the rays, neither in ray parenchyma or in ray tracheids. The diluted WAD100 clearly shows a deeper

Table 3. Number of tracheid-rows filled with coating as a function angle between surface and length-axis of the longitudinal tracheids in pine sapwood measured perpendicular to the surface from axial cross-section. Coating type: pigmented solvent borne alkyd (SBA)

Angle between surface and length axis of tracheid	Number of cell rows filled with coating	
	earlywood	latewood
0°	0-1	0-1
5°	6-9	5-6
10°	12-20	12-15

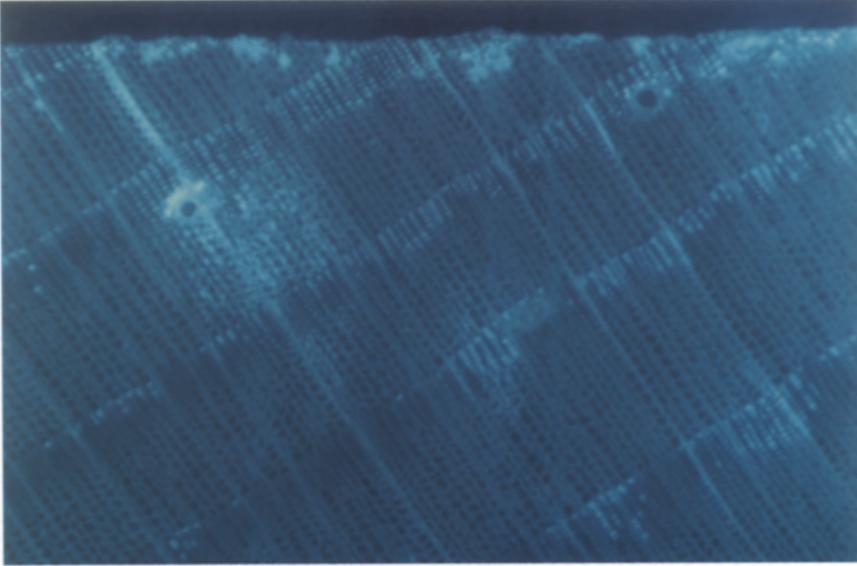


Fig. 5. Fluorescence microphotograph of axial cross-section (40 \times) showing the penetration of HSA binder (white colour) in pine. Note the filling of latewood bands four growth-rings away from the surface and the penetration into rays

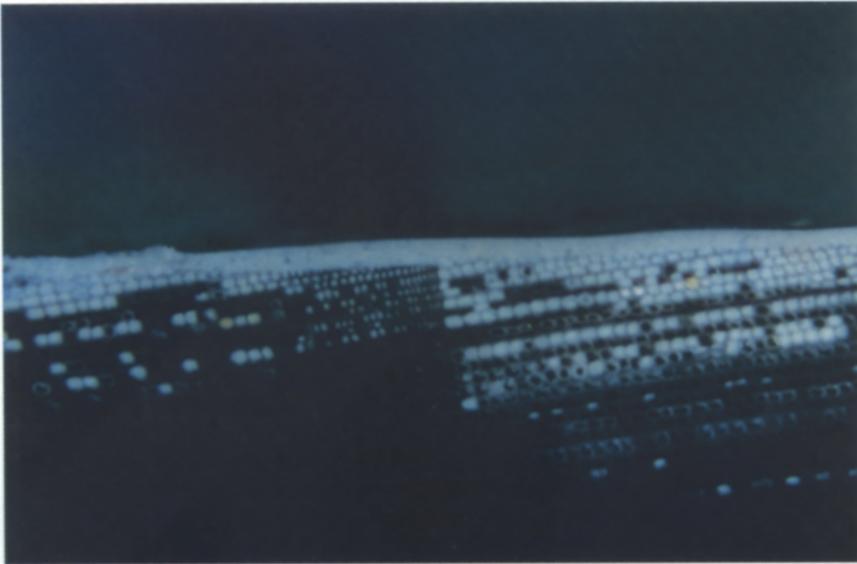


Fig. 6. Fluorescence microphotograph of axial cross-section (100 \times) showing the penetration of SBA coating (white colour) in pine with a controlled 10 $^\circ$ angle between surface and length axis of longitudinal tracheids

penetration compared to the diluted WAD300. The diluted binder WAD100 fills up to 5 cell rows away from the surface, also in rays binder is present and in pine latewood sometimes even longitudinal tracheids adjacent to rays are filled with binder.

The unpigmented waterborne alkyd emulsion (WBA) fills the outer tracheids several cell rows below the surface. In the earlywood cells, the binder is often present as a thin layer covering the inner walls of the lumina, whereas in the latewood cells the whole lumen is filled with binder. The rays in pine are filled to a depth of at least 150 μm but frequently even up to 500 μm . Penetration predominantly occurs through the parenchyma cells. The longitudinal pine latewood cells adjacent to a ray are often also filled with binder. In spruce the penetration into the rays is limited to the length of a single ray tracheid or ray parenchyma cell which has a direct connection to the surface. The WBA binder without dryer or with a higher degree of dilution shows deeper penetration, in particular in the rays and tracheids adjacent to rays, the latter only in case of pine. Increased penetration in the absence of dryer was also reported by Hofland (1994).

The unpigmented HSA binder fills the outer 1–3 earlywood and 1–6 latewood tracheids in both pine and spruce. The penetration in the rays of pine is extremely deep with the HSA. The binder penetrates through the ray parenchyma cells to a depth of 2 mm, in case of the diluted binder even up to 6 mm. From the rays a massive penetration into the longitudinal pine latewood cells takes place, even four growth-rings away from the surface binder could clearly be found in the lumina, as can be seen in Fig. 5. The earlywood bands in between are not filled with binder. Binder could also be detected in areas not directly adjacent to a ray. This however does not mean that the binder was not transported through the rays, because not all rays are located in the visible plane of intersection. From measurements of pit numbers pro tracheid in pine, Courtois (1964) calculated that each tracheid is on average in contact with 2.4 rays, which means that the longitudinal tracheids can even be filled by several rays. By studies with transmitted light microscopy it was found that in addition to the penetration through the ray parenchyma cells the binder also penetrates the intercellular spaces of the rays, often even much deeper than in the ray parenchyma. Similar findings were also reported by Laming (1974) for spruce and by Erickson and Balatinez (1964) for pressure impregnated Douglas fir. In the rays of spruce the HSA penetrates to a depth of 140 to 200 μm through the parenchyma cells. If no dryer is added the depth of penetration through the ray tracheids increases to a length of 1 mm. From the ray tracheids the binder also fills longitudinal latewood cells adjacent to the rays. In pine there is not much difference in penetration of the HSA with and without dryer. At a higher dilution the penetration of the HSA binder in pine and spruce is increased as well.

The SBA binder penetrates the outer cells and the first cell rows under the surface of the earlywood of pine and spruce. Occasionally some binder is found in latewood tracheids of pine. The SBA binder penetrates the rays of pine to a depth of 2 mm through the parenchyma cells. Penetration of the rays in spruce is very limited if dryer is added, but clearly increases if dryer is absent. In the latter situation the binder is present in both parenchyma and in the outer ray tracheids. Penetration of latewood cells adjacent to rays is only found in pine to a distance of 2 mm away from the surface. The filling of the latewood is comparable to that of the HSA but not as frequent. At a higher dilution the penetration of the SBA binder is only slightly improved; the absence of a dryer has a much stronger effect on the increase in penetration. In general the penetration patterns observed for both the SBA and WBA are very much comparable to the results described by Nussbaum (1994).

Penetration of pigmented paint into dark red meranti

The basic paint penetration pattern in dark red meranti consists in complete or partial filling the vessels directly connected with the surface and penetration into rays ending on the surface. The extent of penetration into the vessels is strongly influenced by the presence of tyloses. With the exception of the HSA the coatings do not penetrate the axial parenchyma cells. The acrylic dispersion paints WAD100 and WAD300 do not penetrate the rays. The vessels which have a direct connection to the surface are completely filled if tyloses are absent. If the vessel is filled with tyloses the paint rests on the membranes of the tyloses. The waterborne alkyd WBA also fills the vessel completely unless this is prevented by tylose membranes. At the edges of the vessels and the surface the film thickness of the coating is very low. The rays are only penetrated to a very limited extent. The HSA paint fills the vessels completely, also if tyloses are present. However in the filled vessels colour differences are seen with brighter colours, typical for unpigmented binder, between the membranes of the tyloses. The HSA penetrates the rays to a depth of about 100 to 200 μm with a relatively uniform distribution over the individual ray-cells. The deeper penetrating, brighter part of the HSA is also found in axial parenchym and sklerenchym fibres close to the surface. Most likely the paint has flowed into these cells from openings ending on the surface by the same mechanism more frequently found for the softwoods studied. The SBA paint fills the vessels but rests on the membranes of tyloses if these are present as can be seen in Fig. 4 and Fig. 9. The coating penetrates the rays to the length of one ray parenchyma cell which still has a direct connection to the surface (see Fig. 4). This means that the maximum depth of penetration is about 300 μm . Transport from one ray parenchyma cell to another does not seem to be possible in the case of dark red meranti.

Influence of extractives

In general the normal and extract-free samples do not show very significant differences in penetration. In the case of all three waterborne coatings (WAD100, WAD300 and WBA) no differences were found at all, neither in spruce, pine or dark red meranti. The tyloses in the dark red meranti vessels were not removed or seriously damaged during the extraction, which means that they are still effective in preventing penetration into the vessels. For the HSA and SBA the penetration in pine is somewhat improved in the extractive free samples. The removal of extractives improves the penetration from the rays into the adjacent longitudinal latewood tracheids. This might be explained by the fact that resinous material in the rays reduces permeability of the rays and therefore the transport of coating from the surface to the latewood through the rays. Another explanation could be an increased permeability of the fenestriform pits because of damaging the pit membranes during the extraction process. Depth of penetration into the rays themselves is also slightly improved after extraction, in particular for the SBA on spruce. The only influence of extraction on dark red meranti is found in combination with the HSA coating where the penetration into the rays is somewhat improved and vessels are more often completely filled with coating.

Impact of surface preparation

As could be expected the main effect of sanding is a change in structure of the outer wood cells. The cell walls of the spruce earlywood tracheids are compressed and cell walls are damaged by the sanding. This effect is not limited to the outer cells, changes in cell wall structure were observed up to distances of 500–1000 μm

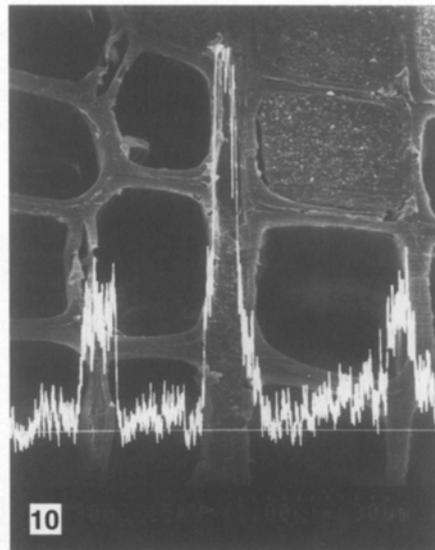
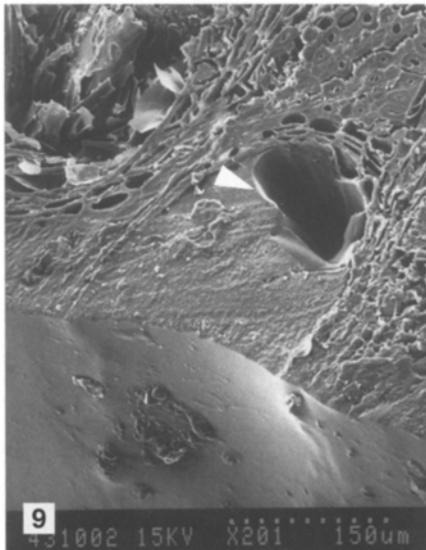
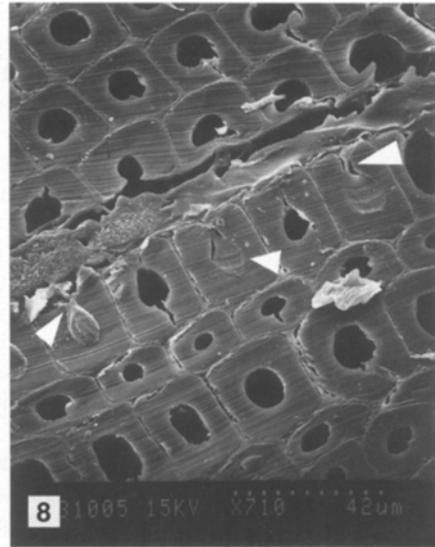
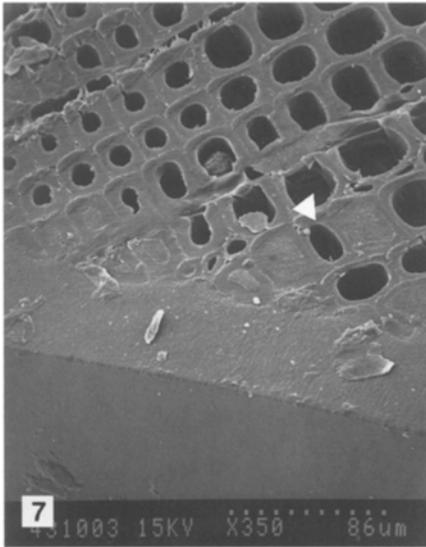


Fig. 7. SEM-microphotograph (350 \times) of pigmented HSA coating filling the outer early-wood tracheids. Note the fact that first, second and fourth tracheid are filled but that third one is empty (see arrow)

Fig. 8. SEM-microphotograph (710 \times) of pigmented HSA coating in a ray and in longitudinal tracheids adjacent to the ray which are filled through the half bordered pits (indicated by arrows)

Fig. 9. SEM-microphotograph (200 \times) showing how the SBA coating is resting on the membrane of a tylose (arrow) in a half open cut vessel in *meranti*

Fig. 10. SEM-microphotograph (1000 \times) with EDAX line-scan on titanium of HSA coating showing that the pigment penetrates into a ray cell. X-ray peak is strongly increased at the ray, smaller peaks at left and right are artefacts due to crossing of a cell wall

away from the surface. The structure of the latewood cells is basically unchanged. The strong damage of the earlywood cells and the less severe damage of the latewood cells are comparable to findings of Murmanis et al. (1986) for abrasive planing of Douglas-fir. Sanding also causes a large amount of dust particles which is accumulated in the cell lumina. This effect is particularly strong in the case of sanding with fine sand paper. The effect of cleaning the surface is only effective on the outer surface but not inside the contaminated lumina. The coating fills the outer compressed tracheids in the same order as for the surfaces planed only. The SBA paint fills 1 to 2 rows of compressed tracheids, whereas the WBA fills one row and the WAD300 coating is only present in the outer layer of half opened and compressed cells (see Fig. 11). No significant differences in penetration between coarse and fine sanded wood were found. Sanding the surface almost completely prevents penetration into the rays as was shown by comparing surfaces only planed and planed plus sanded coated with the SBA paint. Most likely the ends of ray cells are deformed and clogged with dust which prevents paint from flowing into the rays. The reduced coating penetration after sanding supports the finding of Richter et al. (1995) that sanded surfaces require less paint to obtain full coverage of the surface compared to planed ones.

Conclusions

Penetration of a coating into wood is first of all determined by the ability of the paint to flow into the open ends of the cell capillaries. The coatings studied showed very strong differences in penetration capacity. The type of binder has the biggest influence with the following increasing order in penetration: acrylic dispersions, alkyd emulsions, solventborne alkyds and high solid alkyds. Addi-

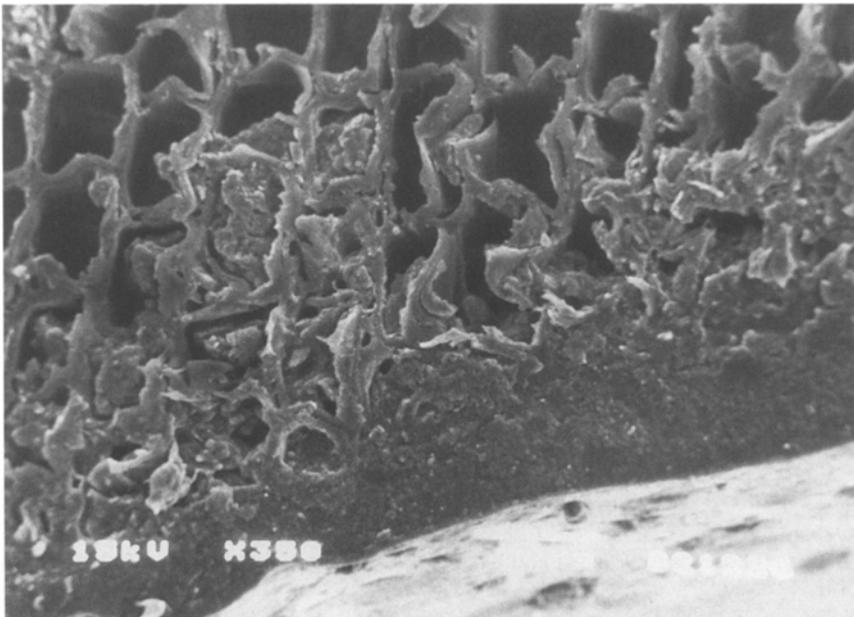


Fig. 11. SEM-microphotograph (350 \times) showing the WAD300 coating on spruce planed and sanded with fine (180) sandpaper. Note that the outer cell walls are deformed by the sanding process

tionally the penetration is influenced by: drying speed of the coating, the ratio of solid to liquid material and the presence of pigments. The latter was also found by Nussbaum (1994). Very likely, paint properties like viscosity, surface tension and the rate of transfer of solvent from liquid coating to cell wall determine the penetrating capacity of a coating. This will be studied in future research where the maximum depth of penetration in the cells will be measured as a function of the paint properties mentioned.

If the paint is able to flow into the wood cells, three different ways of penetration in softwood can be distinguished as is schematically shown in Fig. 12. Firstly the outer longitudinal tracheids are filled directly by coating flowing from the open ends on the surface. This predominantly occurs in the earlywood. The angle between length axis of the tracheid and the surface has a strong influence on the importance of this mechanism. A second way of penetration is through the rays, starting also at the open cut ends of the ray cells. In which way transport in the rays proceeds is strongly dependent on the wood species. In pine the major part of the coating flows through the parenchyma cells, transport from cell to cell must therefore be possible. In spruce the coating almost solely penetrates the ray tracheids. Studies on other wood species showed similar differences. Wardrop and Davies (1961) reported preferential penetration in ray parenchym in the case of *Pinus radiata*, whereas Erickson and Balatinecz (1964) found the ray tracheids to be most important for penetration in the case of Douglas fir. A third way of penetration is from ray cells to adjacent longitudinal tracheids in the latewood. By

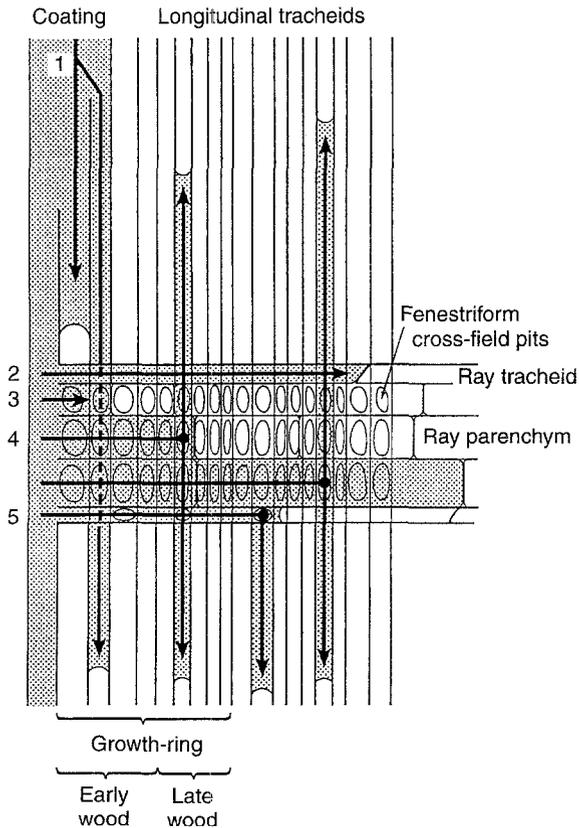


Fig. 12. Schematic presentation of the different ways of possible coating penetration in softwoods seen from a radial cross-section (1) flow into open end of longitudinal tracheid; (2) flow into ray tracheid; (3) flow into ray parenchym; (4) flow from ray parenchym into longitudinal latewood tracheid; (5) flow from ray tracheid into longitudinal tracheid

means of this mechanism unpigmented binder can reach longitudinal tracheids up to a distance of 2 mm away from the surface. The extent of transport from rays to tracheids is strongly dependent on permeability of the cross-field pits and within the scope of this study it was found to be almost totally limited to pine sapwood. The importance of the three penetration mechanisms mentioned above implicates that penetration of the coating can strongly be influenced by the way in which boards are sawn out of a log. This because of the impact on differences in flat and standing growth rings, orientation of grain to the surface, width of early and latewood bands and the number of rays ending in radial and tangential surfaces. The origin of the wood might influence penetration because of differences in early- and latewood portions, conditions of the pits, number of rays and length of longitudinal tracheids. Drying conditions of the wood might also have some influence on coating penetration because of its impact on pit aspiration.

Penetration in dark red meranti is mainly restricted to the filling of vessels and the first cells of rays and very occasionally axial parenchym and sklerenchym. The filling of a vessel by the coating is strongly reduced if tyloses are present. Extractives appeared to have none or only a very minor influence on the penetration in all tree wood species studied. Therefore negative influences of high extractive content on adhesion of a coating are probably not caused by reduced penetration, but are more likely the result of differences in chemical composition of the surface. Surface preparation can have some influence on coating penetration because the number of open cell capillaries in which paint can flow is reduced by sanding. This means that the contact area between coating and wood and the possibilities for mechanical entanglement are reduced by sanding. This might reduce the adhesion of the coating if it is applied on a surface which is sanded before the application of the first coating layer.

References

- Allen KW (1987) A review of contemporary views of theories of adhesion. *Journal of Adhesion* 21: 261-277
- Côté WA, Robison RG (1968) A comparative study of wood. *Journal of Paint Technology* 40(525): 427-432
- Courtois H (1964) Über den Einfluss einiger holzanatomischer Unterschiede auf die Tränkbarkeit von Mittelgebirgs- und Küstenfichtenholz. *Holzforschung Holzverwertung* 16: 61-66
- Erickson HD, Balatinecz JJ (1964) Liquid flow paths into wood using polymerization techniques Douglas-Fir and styrene. *Forest Prod. J.* 293-299
- Haslam JH, Werthan S (1931) Studies in the painting of wood. *Industrial and Engineering Chemistry* 23(2): 226-233
- Hofland A (1994) Water-borne coatings for decorative and protective coatings: a comparative survey. *Surface Coatings International* 7: 270-281
- Kabir FR, Nicholas DD, Vasishth RC, Barnes HM (1992) Laboratory methods to predict the weathering characteristics of wood. *Holzforschung* 46(5): 395-401
- Laming PB (1974) On intercellular spaces in the xylem ray parenchyma of *Picea abies*. *Acta Botanica Neerlandica* 23(3): 217-223
- Liese W, Bauch J (1967) On the closure of bordered pits in conifers. *Wood Sci. Technol.* 1: 1-13
- Loon J van (1966) The interaction between paint and substrate. *Journal of the Oil and Color Chemists Association* (49): 844-867
- Militz H, Peek R-D (1994) Möglichkeiten der Verbesserung einiger Eigenschaften von Pappelholz durch Tränken mit wasserlöslichen Harzen. *Material und Organismen* 28.Bd. 1993/1994 Heft 1: 55-73
- Murmanis L, River BH, Stewart HA (1986) Surface and subsurface characteristics related to abrasive-planing conditions. *Wood Fiber Sci.* 18(1): 107-117

- Nussbaum RM** (1994) Penetration of water-borne alkyd emulsions and solvent-borne alkyds into wood. *Holz Roh-Werkstoff* 52: 389–393
- Pizzi A** (1992) Non-mathematical review of adhesion theories as regards their applicability to wood. *Holzforschung Holzverwertung* 44: 62–67
- Rødsrud G, Sutcliffe JE** (1993) Alkyd emulsions – properties and application. Results from comparative investigations of penetration and ageing of alkyds, alkyd emulsions and acrylic dispersions. Presentation held at the 14th Congress of the Federation of Scandinavian Paint & Varnish Technologists – Copenhagen
- Richter K, Feist WC, Knaebe MT** (1995) The effect of surface roughness on the performance of finishes. Part 1. Roughness characterization and stain performance. *Forest Prod. J.* 45 (7/8): 91–97
- Schneider MH** (1970) Coating penetration into wood substance studied with electron microscopy using replica techniques. *J. Paint Technol.* 42(547): 457–460
- Schneider MH** (1979) Scanning electron microscope study of a coating component deposited from solution into wood. *Journal of the Oil and Color Chemists Association* 62: 441–444
- Schneider MH** (1980) Microscopic distribution of linseed oil after application to wood surface. *J. Coatings Technol.* 52(665): 64–67
- Schneider MH, Côté WA** (1967) Studies of wood and coating interactions using fluorescence microscopy and pyrolysis gas-liquid chromatography. *J. Paint Technol.* 39(511): 465–471
- Schneider MH, Sharp AR** (1982) A model for the uptake of linseed oil by wood. *J. Coatings Technol.* 54(693): 91–96
- Schulze B, Theden G** (1942) Das Eindringen aufgestrichener Holzschutzmittel in Kiefernspinnholz. *Holz Roh-Werkstoff* 5(7): 239–247
- Smulski S, Côté WA** (1984) Penetration of wood by a water-borne alkyd resin. *Wood Sci. Technol.* 18: 59–75
- Wallenfang WO** (1964) Zur Frage der Eindringtiefe von Ölen und Harzlösungen. *Farbe und Lack* 70(8): 258–263
- Wardrop AB, Davies GW** (1961) Morphological factors relating to the penetration of liquids into wood. *Holzforschung* 15(5): 129–141