Microautoradiographic Studies of the Penetration of Alkyd, Alkyd Emulsion and Linseed Oil Coatings Into Wood

Ralph M. Nussbaum—Trätek, Swedish Institute for Wood Technology Research*
E. John Sutcliffe—Dyno Industrier AS†
Ann-Charlotte Hellgren—Institute for Surface Chemistry**

The penetration of wood coating primer products into pine and spruce softwood was evaluated with microautoradiography. ^{14}*C-labeled binders of alkyd and linseed oil were synthesized and used in eight different products.*

The penetration front in wood is uneven due to a heterogeneous structure with different types of wood cells. It was demonstrated that conventional solventborne alkyd primers and waterborne alkyd emulsion primers have similar ability to penetrate into wood. Improved penetration was found for products with lower viscosity, such as an alkyd stain and a linseed oil coating. A priming oil had superior penetration. Generally better penetration was obtained for coatings applied onto sawn and rough surfaces as compared to planed and smooth surfaces.

INTRODUCTION

Wood is a challenging material when used as a substrate for coatings in exterior applications. The fact that wood is an anisotropic and hygroscopic material will inevitably lead to severe strain on any type of coating as well as the wood material itself. The key to extended wood/coating durability can be attributed to (1) the freshness of the wood surface to be coated, i.e., a non-degraded, “high-energy” surface[1-3] and (2) favorable primer coating characteristics in terms of penetrability and moisture dynamic behavior.[4,5] An effective wood primer should thus present optimal anchorage and act as a hydrophobic protective layer.

Solventborne alkyd coatings are known as excellent exterior wood primers and have for years dominated in the Scandinavian market. Due to the environmentally unfavorable organic solvents used in these coatings, the trend today is towards waterborne systems such as alkyd emulsions, which have now gained widespread use in several areas of application including decorative exterior paints and industrial factory priming.

The fact that the binder in an alkyd emulsion coating is made up of rather large emulsion droplets (0.5 - 1.0 μm in diameter) in contrast to the free molecules present in a solventborne alkyd coating, has caused the penetration ability of these alkyd emulsion coatings to be questioned.

Penetration of wood by a fluid can be separated into gross penetration and cell-wall penetration. The former relates to liquid flow into the gross openings in the wood structure, e.g., the cell lumina, and is mostly governed by capillary action. The latter relates to penetration into interstitial voids of the cell-wall ultrastructure. The three-dimensional structure of softwood with its main cell-types is shown in *Figure* 1.

Several methods have been used for detecting substances penetrating into wood, such as scanning electron microscopy combined with energy dispersive X-ray analysis (SEM-EDS), fluorescence microscopy and microautoradiography. A recent study by Nussbaum, including references to most of the techniques used, demonstrated that microautoradiography is a sensitive technique well suited for gross penetration studies of binders into wood.[7] By using microautoradiography, where a radioactive tracer isotope is introduced in the molecule to be studied, the physical and chemical properties of the molecule are not changed as opposed to most other methods. The study showed that 50% solutions of solventborne alkyd and waterborne alkyd emulsion binders had similar penetration patterns when applied onto softwood surfaces.

The present study evaluates and compares the gross penetration of fully formulated solventborne alkyd coat-

---

*Box 5609, S-114 86 Stockholm, Sweden.
†P.O. Box 160, 2001 Lillestrøm, Norway.
**Box 5607, S-114 86 Stockholm, Sweden.

ings, alkyd emulsion coatings, and a linseed oil coating by using microautoradiography. The coating viscosity and particularly the pigment volume concentration (PVC) are factors which may affect the penetration. Alkyd coating formulations are therefore varied regarding the PVC or the amount of thickening agent.

Different surface roughness due to varying wood machining is an important factor known to affect the adhesive strength between a coating and a wood substrate.[1] It may also affect the penetration. A comparison between sawn and planed surfaces is therefore included in this study.

EXPERIMENTAL

Radioactivity Labeled 80% Oil Length Alkyd Resin

To a 100 ml two-necked, round-bottomed flask was added pentaerythritol (13.6 g) and a solution of 59.0 g tall oil fatty acid containing 4.5 mCi [1-^{14}C] linoleic acid (55 mCi/mmole) in toluene (Amersham Life Sci.). The round-bottomed flask was equipped with a scrubber tube, thermometer, and magnetic stirrer. Nitrogen was bubbled through the reaction mixture throughout the duration of the experiment.

The reaction mixture was heated to 260-265°C for a period of two hours. During this time all the toluene was removed from the reaction mixture. After two hours the mixture was cooled to below 165°C and 11.0 g of isophtalic acid was added. The mixture was heated to 260-265°C for an additional 1.5 hr at which time the acid value had fallen to 13-14 mg KOH/g. The reaction mixture was then cooled and was found to have the following properties: acid value = 13.7 mg KOH/g, molecular weight (Mw) = 5702, molecular weight number average (Mn) = 1853. The molecular weight of the alkyd was determined by gel permeation chromatography. The columns used were calibrated with polystyrene standards with known molecular weight. A stock of 66 g alkyd resin with a final ^{14}C-activity of 58 μCi/g was produced.

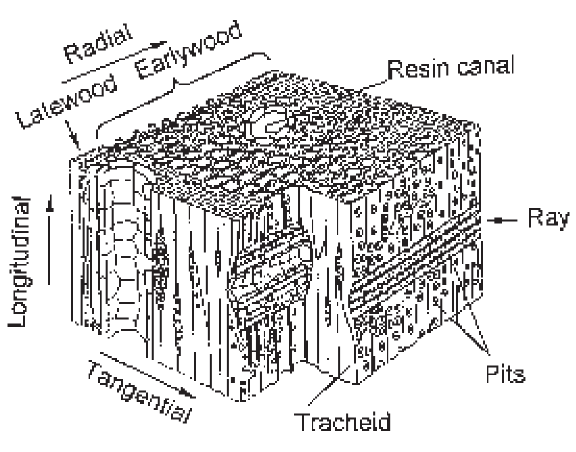

Figure 1—A schematic three-dimensional structure of softwood with its main cell-types. The hollow interior of a tracheid is called lumen. The length of a side is roughly 0.75 mm (from reference 6).

Radioactivity Labeled Linseed Oil

A diglyceride of linseed oil was obtained from the corresponding triglyceride by reacting one gram of cold pressed linseed oil with the enzyme *Rhizo mucor miehi lipase*, supplied as lyophilized powder, and a small drop of water at room temperature.[8] The reaction was monitored by thin layer chromatography (TLC). Duration of reaction was typically eight hours. The reaction mixture was purified by gradient chromatography in hexane/ethyl acetate/acetic acid. The diglyceride product was identified with nuclear magnetic resonance (^{1}H-NMR) and was found to be >95% pure (reactive OH).

[1-^{14}C]linoleic acid, 55 mCi/mmole, was dissolved in CH_2Cl_2 and converted to the acid chloride with an excess of oxallyl chloride in the presence of catalytic amount of dimethyl formamide. The product was evaporated to dryness and 2.2 mg of the acid chloride was redissolved in dry toluene. Ten milligrams of the diglyceride was also dissolved in dry toluene. To this mixture was added 0.15 mg dimethyl amino pyridine (DMAP) as catalyst. A two-fold excess of diglyceride to acid chloride was used in order to avoid unreacted acid chloride in the product mixture. The acid chloride solution was injected into the sample flask together with 1.2 μl triethyl amine and the reaction was continued for 24 hr at room temperature under a nitrogen atmosphere. After this time, no free acid or acid chloride could be detected by TLC or ^{1}H-NMR in the reaction mixture. The product was evaporated to dryness and used without further purification.

Coating Formulation

Eight coating products were made from the ^{14}C-labeled material: four of solventborne alkyd, three of waterborne alkyd emulsions, and one of linseed oil. All of the formulations were composed of representative standard raw materials commonly used in exterior wood coatings. The main components of all products are given in *Table* 1 along with some important physical properties.

The solventborne alkyd products were all prepared by mixing the ^{14}C-labeled alkyd stock directly with the other components to final quantities ranging from 7 to 17 g.

The alkyd emulsions were prepared in the following way: the ^{14}C-labeled alkyd (51.0 g) was transferred to a beaker and heated to 80°C and 3.0 g of a nonionic emulsifier (Berol 542) was added under stirring. To this solution 53.1 g tap water, with a temperature of 80°C, was slowly added while stirring with a propeller at 600 rpm. After the addition of water, the coarse emulsion was stirred for an additional 12 min. The coarse emulsion was heated to 80°C and homogenized, using a microfluidizer homogenizer, at a pressure of three bar until constant droplet size was obtained. The resulting ^{14}C-labeled alkyd emulsion was characterized as having an average droplet diameter of 2.71 μm (Malvern Master-

Table 1—Main Coating Components and Physical Properties

	Pigmented Solventborne Alkyd Primer	Pigmented Solventborne Alkyd Primer	Low-Pigmented Solventborne Alkyd Stain	Unpigmented Solventborne "Priming Oil"	Pigmented Solventborne Linseed Oil Primer	Pigmented Waterborne Alkyd Emulsion Primer	Pigmented Waterborne Alkyd Emulsion Primer	Pigmented Waterborne Alkyd Emulsion Primer
Abbreviation	**A1**	**A2**	**AS**	**PO**	**L**	**AEM1**	**AEM2**	**AEM3**
Alkyd, % (Bergvik S 78)	34.6	27.0	56.3	15.0	—	—	—	—
Alkyd emulsion (50%), %	—	—	—	—	—	52.5	52.0	41.7
Linseed oil, %	—	—	—	—	31.1[a]	—	—	—
Pigment, % (Bayertitan R-KB-2)	37.5	49.0	13.0	—	56.6[b]	25.3	25.0	36.1
Thickening agent, % (Acrysol RM 8)	—	—	—	—	—	2.0	3.0	2.7
White spirit, %	25.8	22.3	27.3	84.9	10.0	—	—	—
Water, %	—	—	—	—	—	18.2	18.0	17.8
PVC	21	30	5	—	29	20	20	31
Dry matter, weight-%	73.2	76.9	70.9	15.0	88.6	53.1	53.2	59.5
Dry matter, volume-%	57.4	58.2	63.0	12.4	76.4	39.8	39.9	40.6
Density, g/cm^3	1.25	1.42	1.00	0.80	1.45	1.24	1.24	1.39
Viscosity, mPa·s (Brookfield)	—	—	—	—	440	2100	5800	13400
^{14}C-activity, µCi/g	20	16	33	9	18	15	15	12
Applied amount, g/m^2	173	223	106	200-300	193	242	242	303

Component content in weight-%.
(a) 20.7% raw linseed oil + 10.4% standoil 20.
(b) 52.1% Bayertitan R-KB-2 + 4.5% zinc oxide.

sizer) and a nonvolatile solids content of 50%. The alkyd emulsion was then mixed with the other components to prepare three alkyd emulsion coatings of 50 g each.

The linseed oil coating was prepared by dissolving 15 mg of the ^{14}C-labeled linseed oil residue in 1.5 g of white spirit and mixing it with 13.5 g of unlabeled (i.e., no ^{14}C isotope) solvent-free linseed oil coating.

In addition to the components listed in *Table* 1, the solventborne alkyd primers and the alkyd stain contained a drier and antiskinning agent, the priming oil a drier and the waterborne alkyd emulsion primers a drier, a dispersing agent, a defoamer, an antiskinning agent and a fungicide. Pigment was added as pigment paste. Alkyd primers A1 and A2 differ from each other mainly by the PVC and alkyd emulsion primers AEM1 and AEM2 by the thickening agent content. Because of the small amounts of material involved, direct viscosity measurements of the solventborne alkyd coatings were not possible. It was apparent though that primer A1 had lower viscosity than A2 and that the stain AS had substantially lower viscosity than A1 and A2. It should be pointed out that the consistency of these three coatings corresponded to that of representative commercial products. A set of all eight coating products were prepared with unlabeled binders as a reference material.

The amount of binder applied to the wood specimens (see the following) was kept constant at 60 g/m^2 for all coatings except the priming oil, where 30-45g/m^2 of binder was applied. Therefore the total amounts of coating applied were varied according to *Table* 1.

Wood Material

Since wood is a heterogeneous material which may exhibit large variability even within the same tree, a total knowledge and control of the selected test material is indispensable. Also, tests should be repeated with specimens of different origin.

The wood material was selected from straight-grained, defect-free, thick boards of pine sapwood (*Pinus silvestris*) and spruce (*Picea abies*), which had earlier been carefully stored and dried. Unlike pine, sapwood and heartwood of spruce are not markedly different from each other and therefore were not separated during the sample preparation. Test specimens free from visible cracks were prepared in three groups regarding the upper horizontal surface to be coated:

(a) tangential with the annual rings parallel to the surface;

(b) radial with the annual rings perpendicular to the surface; and

(c) tangential/radial with the annual rings in 45° to the surface.

Sketches and dimensions of the test specimens are shown in *Table* 2 together with the number of specimens treated with the different labeled coating products. The surface of specimens having a tangential surface always consisted of earlywood of at least 1 mm in thickness. The spruce material used in the tests was of seven different origins and the annual ring densities were in the range of one to six rings per 10 mm. Corresponding data for pine were six origins and two to seven annual rings per 10 mm. All direct comparisons between different coating products were made with specimens with positions adjacent to each other in the original board.

Most test surfaces had a finely sawn structure. For the examination of the impact of the surface roughness, tangential specimen surfaces with a planed structure were also included. The surface roughness comparison was

Table 2—Test Specimen Dimensions (in mm) and Number of Replicates with the Same Coating Product. Annual Rings are Marked Out on End-Grain Surfaces. T = Tangential Surface; R = Radial Surface (For abbreviations see *Table* 1)

		Coating Product							
		A1	A2	AS	PO	L	AEM1	AEM2	AEM3
	Spruce	11	3	3	7	3	8	2	2
	Pine sapwood	10	3	3	4	3	6	2	2
	Spruce	5	2	2	2	3	3	1	1
	Pine sapwood	4	2	2	1	3	1	1	1
	Spruce	1	—	—	1	—	1	—	—
	Pine sapwood	1	—	—	1	—	1	—	—

performed with all coating products except the linseed oil coating.

Penetration was restricted to one direction by sealing the four vertical sides with a silicone rubber compound (Shin-Etsu, KE 45 RTV). In order to protect the top surface from any silicone contamination the sealing procedure was performed with a clean glass slide tightly pressed to the surface. The specimens were stored from 3 to 60 days at 20°C and 65% RH before the coatings were applied with new, small and rather stiff, paint brushes. The coatings were allowed to dry for at least three weeks at room temperature.

Microautoradiography

Detection of binder penetration into wood was made according to basic microautoradiographic procedures,[9] which involves several steps. After removal of the sili-

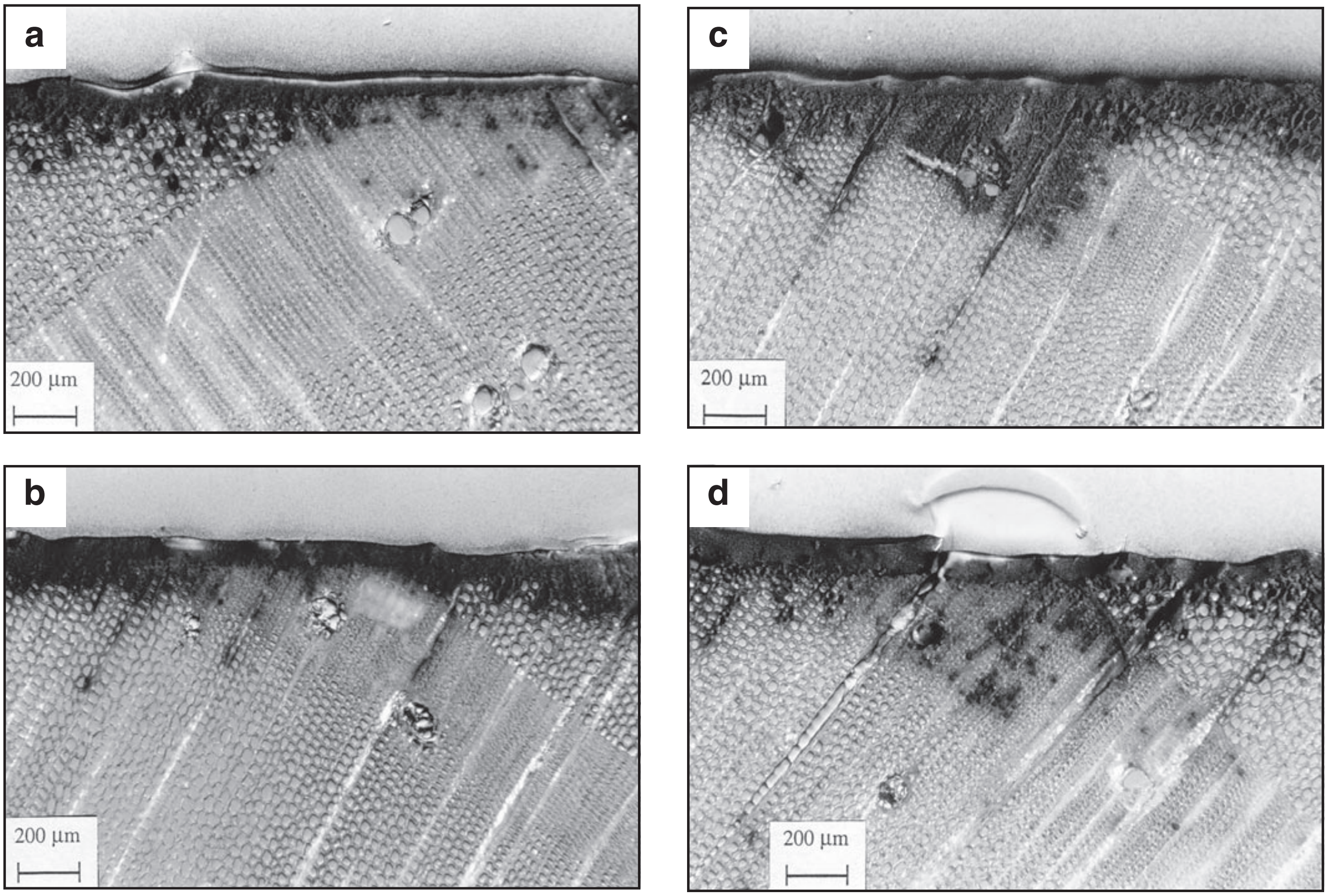

Figure 2—Autoradiomicrographs showing cross sections, including one annual ring border, with the radial/tangential penetration of solventborne coating products into sawn pine sapwood. (a) = alkyd primer A1, PVC=21; (b) = alkyd primer A2, PVC=30; (c) = alkyd stain AS; and (d) = linseed oil coating L.

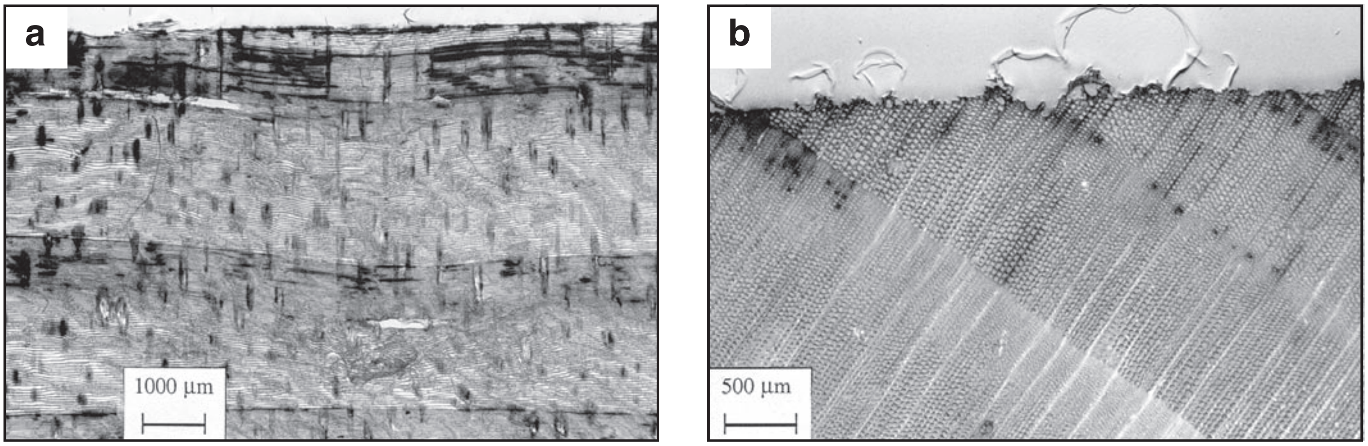

Figure 3—Autoradiomicrographs showing the penetration of priming oil PO into (a) sawn pine sapwood, radial section showing three annual ring borders; (b) sawn spruce, cross section (three ring borders). Note that the ray cells, running perpendicular to the top surface in 3a, often appear as short fragments.

cone sealer, the coated specimens were cut in 25-40 µm thick sections by a microtome knife moved at an angle of 45° to the coated surface. The sections were then mounted on glass slides with a gelatine/chrome alum solution. From each specimen, 6 to 10 replicate sections were made. Air trapped in the wood cell lumen was removed by impregnating the dry sections with water under vacuum. The slides were then dipped into a light-sensitive photographic nuclear emulsion coating (Ilford, K5), dried, and stored in the dark at 4°C. Any beta-particles emitted from the radioisotope then strike the nuclear emulsion coating and lead to silver grain formation. This grain formation increases with increasing storage time, i.e., increasing exposure. After a standard photographic development process, the localization of the radioisotope and thus of the binder was revealed by the dark areas caused by grain formation in the otherwise transparent nuclear emulsion. After initial tests on sections of several coating products, where the exposed nuclear emulsion was developed after 14, 35, and 70 days, a period of 35 days was chosen as an appropriate exposure time. Sections with unlabeled coating products were consistently included in the tests. The sections were finally photographed in a light microscope.

RESULTS AND DISCUSSION

The results of the penetration study are shown as autoradiomicrographs for some representative wood sections in *Figures* 2-6, where the binder penetration is evident as black areas. A section with an unlabeled reference coating is shown as comparison in *Figure* 7.

A general and expected result is the unevenness of the penetration front due to the differentiated cell structure in the wood. A typical example is a deeper penetration into the radial ray cell structure, which in itself consists of two to three different cell types, when compared with the rather superficial penetration into the longitudinal (axial) tracheids as shown most clearly from the radial sections (see *Figures* 4 and 6). The structure of the ray cells, in particular the so called ray tracheids, permits an easier radial penetration than the longitudinal tracheids. Whereas the interconnecting pits in the longitudinal tracheids are mostly irreversibly closed in dried wood, the pits in the ray cells are not closed to the same extent and therefore permit penetration more easily.

The preference for penetration into the radial ray cells was also apparent for coating products applied onto radial surfaces. Analyzing the surface prependicular to the painted radial surface, i.e., a tangential surface, where the ray cell cross sections are outlined as small ellipses, revealed several ellipses with dark spots where penetration accordingly had occurred (not shown in any figure).

The following direct comparisons are all based on the relevant overall results for each coating product, including all replicates of all wood sample origin.

Coating Viscosity and PVC

No significant differences in penetration patterns were found between the two solventborne alkyd primers A1 and A2 in spite of their different PVC (21 and 30, respectively). Comparing these alkyd primers with the two other solventborne products, i.e., the alkyd stain, AS, and the linseed oil primer, L, showed a substantially better penetration for the latter ones. AS and L had similar penetration patterns. The comparisons are shown for cross sections of pine sapwood in *Figure* 2.

The binder penetration in the longitudinal tracheids is generally restricted to the 2 to 10 outermost tracheids for most products, which corresponds to a depth of about 0.08-0.5 mm, whereas the penetration into the radial ray cells may well exceed a depth twice of that. The considerably lower PVC and viscosity of the alkyd stain and the lower viscosity of the linseed oil coating may well explain the better penetration of these two products (see *Figure* 2).

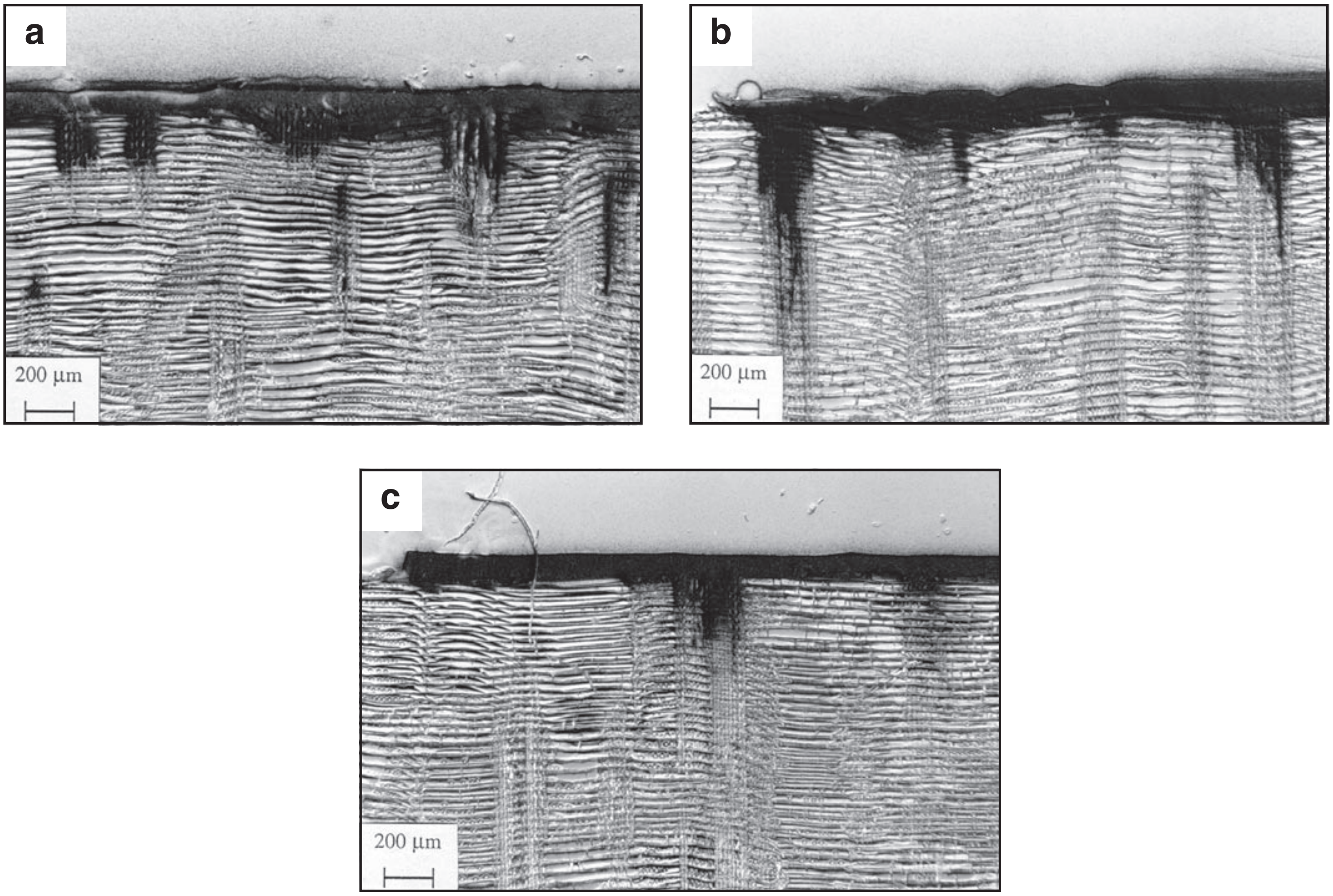

Figure 4—Autoradiomicrographs showing radial sections with the penetration of waterborne alkyd emulsion primers into planed pine sapwood. (a) = alkyd emulsion AEM1, η=2 100 mPas; (b) = alkyd emulsion AEM2, η = 5 800 mPas; and (c) = alkyd emulsion AEM3, η = 13 400 mPas.

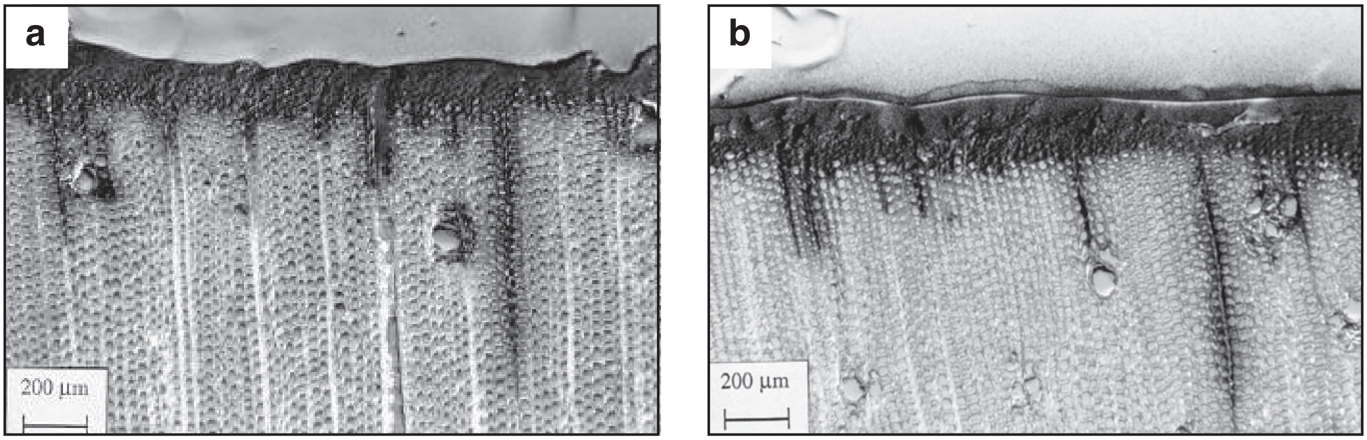

Figure 5—Autoradiomicrographs showing cross sections with the penetration of (a) waterborne alkyd emulsion AEM1; (b) solventborne alkyd A1, into sawn pine sapwood.

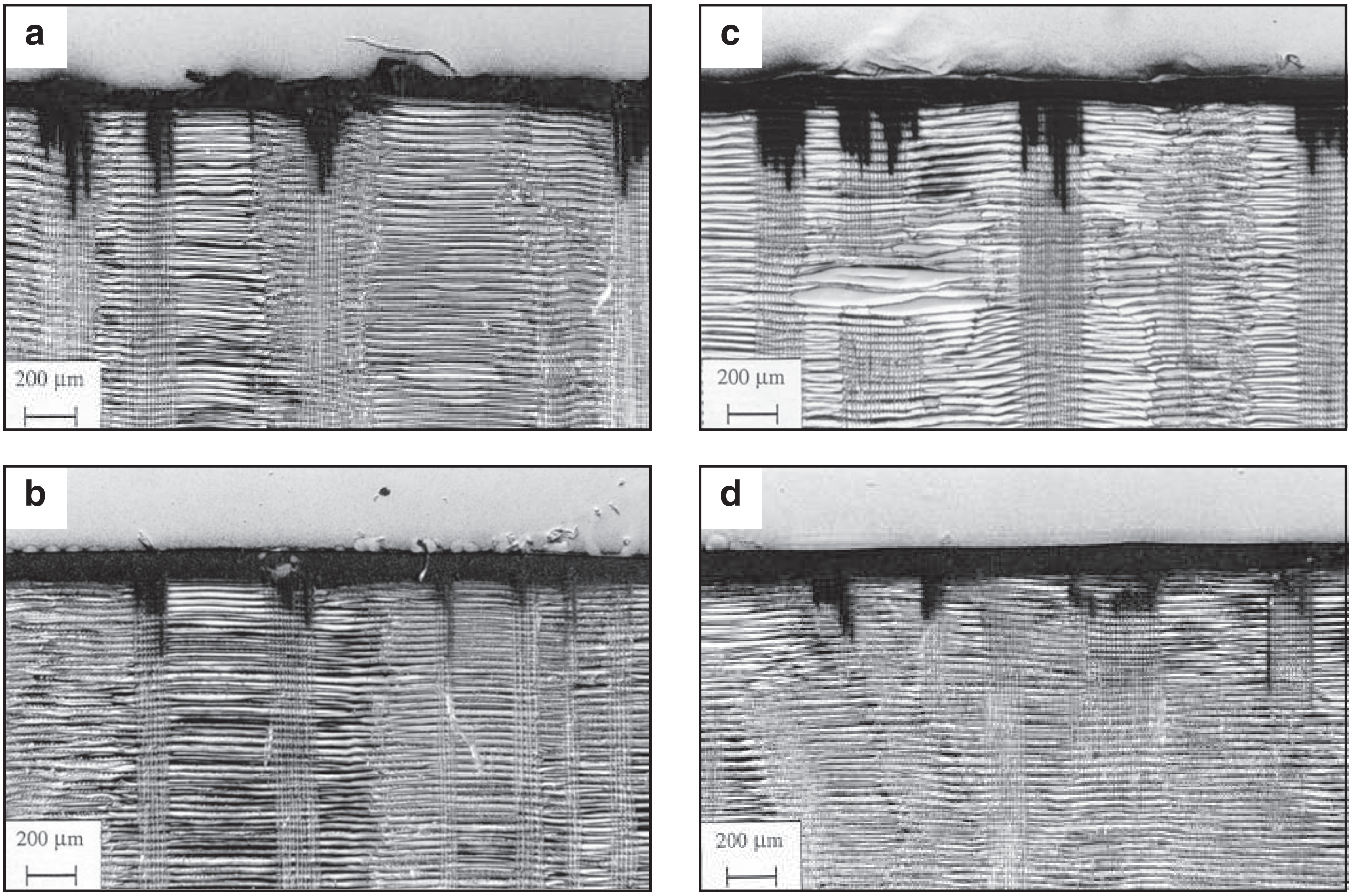

Figure 6—Autoradiomicrographs showing radial sections with penetration into planed and sawn surfaces. (a) = sawn spruce, alkyd emulsion AEM1; (b) = planed spruce, alkyd emulsion AEM1; (c) = sawn spruce, solventborne alkyd A1; and (d) = planed spruce, solventborne alkyd A1.

Generally lower alkyd coating penetration, restricted to merely one to three tracheids, has recently been reported by de Meijer et al.[10] The fact that the molecular weights of their alkyds were higher than 50,000 may partly explain the reduced penetration.

A comparison of the penetration into the thin-walled earlywood versus the thick-walled latewood show some interesting differences between the two categories of solventborne coatings. While the penetration of the two alkyd binders A1 and A2 is better in the earlywood than in the latewood, the opposite is the fact for the alkyd stain (AS) and the linseed oil coating (L), (see *Figure* 2). This may be explained by the fact that the latewood generally contains more open (unaspirated) pits than the earlywood. Fluids of relatively low viscosity, like AS and L, may well utilize the higher frequency of unaspirated pits and thus penetrate deeper into the latewood whereas for more viscous fluids, like A1 and A2, the unaspirated pits are only of secondary importance. Instead the thicker latewood cell-wall and the smaller tracheid cavity (lumen) seem to be determining factors preventing the penetration into the latewood. The fact that low-viscosity products predominantly penetrate into the latewood is also verified by the penetration pattern of the priming oil shown in *Figure* 3. The priming oil penetrates several annual rings into pine sapwood. This is far deeper than any of the pigmented products. The priming oil is present in both latewood ray cells and tracheids as compared to earlywood where presence in the ray cells clearly dominates.

A direct comparison between the three alkyd emulsion products reveals similar penetration for the two emulsions with the lowest viscosity, AEM1 (η=2,100 mPas) and AEM2 (η=5,800 mPas). The microautoradiographs in *Figure* 4 show that these two alkyd emulsions penetrate into three to four of the outermost longitudinal tracheids and that they are also found in the ray cells. The alkyd emulsion with the highest viscosity, AEM3 (η=13,400 mPas), has a generally lower penetration ability and is merely found in the ray cells. Hence, minor changes in the normal viscosity range of alkyd emulsion primers seem to have little effect on the penetration. A more marked increase in viscosity will, however, undoubtedly result in reduced penetration.

The preferential location of coating binders in the ray cells, besides the cell structural differences already mentioned, may also be attributed to differences in hydrophilic/hydrophobic character between different wood cells. Whereas the pure lignocellulosic longitudinal tracheids may be characterized as highly hydrophilic, the

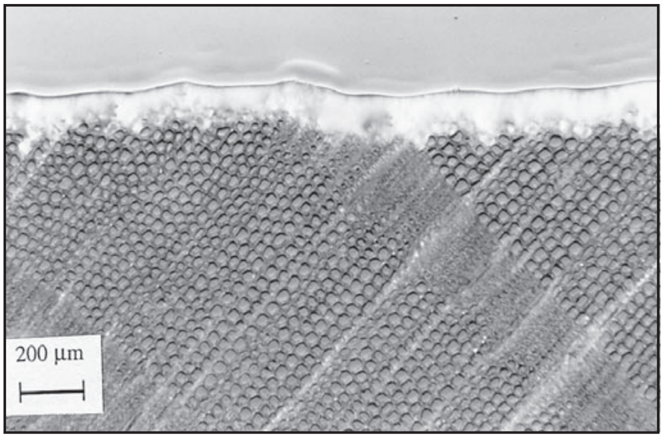

Figure 7—Micrograph of an unlabeled alkyd emulsion reference (cross section of sawn pine sapwood).

ray cells, which also contain resins and fats, can be described as more hydrophobic. Thus the hydrophobic character of an alkyd, a linseed oil binder, or white spirit may well be a factor of some importance when considering their preferential location in the ray cells.[11] The deep penetration of the priming oil, which predominantly occurs via the ray cells, may be attributed to pure solvent property effects such as polarity, viscosity, and surface tension. In the light of the surprisingly small penetration into the surface tracheid cells of such a priming oil (see *Figure* 3a), its function and contribution to the moisture barrier of multicoat wood coating systems should therefore be questioned.

Solventborne Alkyd vs. Waterborne Alkyd Emulsion

Solventborne alkyd primers and waterborne alkyd emulsion primers have similar penetration when primers within the same viscosity range are compared (see *Figures* 5 and 6). This result is in line with earlier findings where 50% binder "solutions" were compared.[7] It is not shown how in fact the penetration of the alkyd emulsion products proceeds. On one hand, the relatively large sized emulsion droplets, which in this study were even larger than in normal commercial alkyd emulsions, will not easily penetrate the openings in the pits in the ray cells, which are roughly of the same size as the droplets themselves. On the other hand, if the penetration would not start until some stage of the "breaking" of the alkyd emulsion, i.e., after evaporation of water when coalescence of the emulsion droplets takes place, the viscosity would then inevitably become markedly increased making penetration more difficult.

A good penetration ability is assuredly an essential wood primer property. From a penetration point of view, waterborne alkyd emulsion primers may be recommended as an environmentally more favorable alternative to solventborne primers. However, when the important moisture dynamic behavior factor of a wood/coating system is taken into account, the alkyd emulsion primers appear slightly less favorable than the solventborne primers. This is probably due to the content of hydrophilic surface active agents in alkyd emulsions.[5]

Wood Material and Wood Machining

All coating products have significantly better penetration into pine sapwood than into spruce. For pine sapwood the penetration of a given product occurs more into the longitudinal tracheids as well as deeper into the ray cells. The fact that spruce wood is less penetrable than pine sapwood regarding liquid penetrability is well known and often explained by the much larger cross-section area of the ray tracheids and the less complete aspiration of the pits in pine.[12] A very clear difference between pine sapwood and spruce was found for samples treated with the low-viscosity priming oil (see *Figure* 3). While the penetration into pine sapwood is deep and distinguished, the penetration into spruce is more indistinct and on a surprisingly low level. Although the autoradiographic results in this study are mainly qualitative, the total presence of priming oil in spruce seems to be at an inexplicably low level.

A comparison between wood samples of different origin did not reveal that the annual ring width had any effect on the penetration. However, since annual ring width was not of primary interest in this study, further investigation is needed before any final conclusion can be drawn.

The effect of the machining method on the penetration may not appear decisive when planed and sawn surfaces are compared as in *Figure* 6. The fact that the penetration continues into a few additional tracheids may nevertheless be of great importance. It is likely that the improved penetration results in a better surface stabilization which may be a key to extended performance of exterior painted wood.[13] Penetration into the longitudinal tracheids is therefore probably more important for the wood/coating interaction and thus the coating anchorage than the generally deeper penetration into the ray cells. However, by also taking microbiological aspects into account, ray cell penetration is certainly important since in this way coating fungicides may be introduced deeper into the wood.

CONCLUSIONS

Microautoradiography has been proven to be a useful technique well suited for studying the gross penetration of different coating products into wood.

The radial penetration front was uneven due to a heterogeneous structure with different types of wood cells. The important penetration into the longitudinal tracheids varied between 0.08 and 0.5 mm for pigmented products. Penetration into pine sapwood was significantly better than into spruce.

Coating viscosity seemed to have no major effect on the penetration when the same type of alkyd primers representing a conventional viscosity range were compared. For products with lower viscosity, an alkyd stain with low PVC and a linseed oil coating, the penetration was improved. A priming oil type product had superior penetration into pine sapwood.

Solventborne alkyd primers and waterborne alkyd emulsion primers of comparable viscosity had similar ability to penetrate into wood.

The penetration was improved for coatings applied onto sawn and rough wood surfaces as compared to planed and smooth surfaces.

ACKNOWLEDGMENT

Appreciation is expressed to the Swedish Council for Forestry and Agricultural Research (SJFR) for financial support. The authors wish to thank Dr. Eva Malmström, Polymer Technology, Royal Institute of Technology for the enzymatic diglyceride preparation and Ms. Katarina Nordman-Edberg, Trätek and Ms. Karén Olsen, Dyno Industrier for skillful technical assistance. Mr. Gudbrand Rødsrud is acknowledged for his valuable contribution in the initial phase of this study.

References

(1) Williams, R.S. and Feist, W.C., "Effect of Preweathering, Surface Roughness and Wood Species on the Performance of Paint and Stains," JOURNAL OF COATINGS TECHNOLOGY, *66*, No. 728, 109 (1994).

(2) Hse, C-Y. and Kuo, M-I., "Influence of Extractives on Wood Gluing and Finishing–A Review," *Forest Prod. J.*, 38 (1), 52-56 (1988).

(3) Nussbaum, R.M., "The Time Limit to Avoid Natural Inactivation of Spruce Surfaces Intended for Painting or Gluing," *Holz als Roh- und Werkstoff*, 54 (1), 26 (1996).

(4) Rødsrud, G. and Sutcliffe, E.J., "Alkyd Emulsions–Properties and Application. Results from Comparative Investigations of Penetration and Aging of Alkyds, Alkyd Emulsions and Acrylic Dispersions," *Surf. Coat. Int.*, 77 (1), 7-16 (1994).

(5) Ekstedt, J., "Moisture Dynamic Assessment of Coatings for Exterior Wood" Licentiate Thesis, Royal Inst. of Techn., Stockholm, TRITA-BYMA 1995:12, 1995.

(6) Howard, E.T. and Manwiller, F.G., "Anatomical Characteristics of Southern Pine Stemwood," *Wood Sci.*, 2 (2), 77-86 (1969).

(7) Nussbaum, R.M., "Penetration of Waterborne Alkyd Emulsion and Solventborne Alkyds in Wood–Autoradiographic and SEM-EDXA Studies," *Holz als Roh- und Werkstoff*, 52 (6), 389-393 (1994).

(8) Hult, K., Dept of Biochemistry and Microbiology, Royal Institute of Techn., Stockholm, personal communication, 1996.

(9) Rogers, A.W., *Techniques of Autoradiography*, Elsevier/North Holland, Amsterdam, 3rd Edition, 1979.

(10) de Meijer, M., Militz, H., and Thurich, K., "Surface Interaction Between Low VOC-Coatings and Wooden Substrates," *Proc. of the XXIII FATIPEC Congress*, Brussels, Vol. C, 191-214, 1996.

(11) Svensson, I.G. et al., "Water-Based Water Repellents for Treatment of Wood," The Int. Res. Group on Wood Pres., Doc No. IRG/WP/3446, 1987.

(12) Liese, W. and Bauch, J., "On Anatomical Causes of the Refractory Behavior of Spruce and Douglas Fir," *J. Inst. Wood Sci.*, 4 (19), 3-14 (1967).

(13) Richter, K., Feist, W.C., and Knaebe, M.T., "The Effect of Surface Roughness on the Performance of Finishes, Part I. Roughness Characterization and Stain Performance," *Forest Prod. J.*, 45 (7-8), 91-97 (1995).