THE MECHANISM OF FAILURE OF CLEAR COATED WOODEN BOARDS AS REVEALED BY MICROSCOPY

by

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SUMMARY

Regions of wood-coating interface from clear coated Pinus radiata boards were examined by a combination of light and transmission electron microscopy techniques after six months exposure outdoors in order to understand the mechanism of clear coating failure. The primary cause of coating failure was the separation of cells in the surface layers of wood underlying the coating that was caused mainly by the photodegradation of the middle lamella. These results are discussed in the light of known effects of solar radiation on lignin and other components of wood cell walls.

Key words: Pinus radiata wood, varnish, clear coatings, ultraviolet radiation, delignification, middle lamella, light microscopy, transmission electron microscopy.

INTRODUCTION

Protection coatings applied to wood surfaces vary in their performance in outdoor situations. Among the weathering factors that may affect wood and coatings, solar radiation and water are considered very important (Chang et al. 1982). The performance of opaque or pigmented coatings is superior to that of clear coatings (Cassens & Feist 1991).

Considerable information is available on various aspects of weathering of chemically modified and unmodified wood, including the factors which affect the rate and extent of weathering, and the damaging effect of solar radiation on wood and wood components (Hon 1981; Rowell et al. 1981; Hon 1983; Feist et al. 1991a, b; Cassens & Feist 1991; Kuo & Hu 1991; Dunningham et al. 1992; Evans et al. 1996; Plackett et al. 1996; Turkulin et al. 2001; Williams et al. 2001a, b, c). However, detailed microscopic information on the changes that occur due to weathering at the wood-coating interface are limited (Singh et al. 1995; Turkulin et al. 2001). It is important to differentiate between the effect of weathering on wood from the effect on coatings in order to clearly understand the mechanism of coating failure; this warrants obtaining additional information from high resolution electron microscopy. Such information may form the basis of future developments to prolong the service life of clear coatings.

This paper describes the observations made on the wood-coating interface using correlated light microscopy and transmission electron microscopy (TEM) after six months of outdoor exposure.
Fig. 1. Coated boards (380 × 100 × 20 mm) after six months exterior exposure showing discoloration (upper part of photograph). The coated boards in the lower part of the photograph were not exposed.

Fig. 2–5. Transverse sections through coated-wood interface regions from unexposed (Fig. 2) and exposed (Fig. 3–5) boards. – 2: The coating is intact and is in close contact with the wood surface. – 3 & 4: Early stages of coating failure. – 3: Arrowheads indicate small gaps between the coating and wood surface. Microorganisms (arrow) are present in a region where the coating has cracked or detached from the wood (asterisk). – 4: The coating has detached in some parts due to separation of weathered surface cells. The arrowhead points to a region where a ray is fully penetrated by the coating and the coating is in close contact with the wood surface. – 5: Separated cells on the wood surface appear to be only loosely connected (asterisks). The coating is no longer present in this region. — Scale bar = 40 µm for all figures. Light micrographs.
MATERIALS AND METHODS

Samples
Radiata pine (*Pinus radiata* D.Don) timber, conventionally kiln-dried (90°C / 60°C dry bulb/wet bulb) to an average moisture content of about 12%, was spot tested for heartwood (Cummins 1971). All heartwood was discarded as other parts of the trial required liquid treatments which are too variable if heartwood is present. Sample boards, with dimensions 380 × 120 × 20 mm (longitudinal × tangential × radial), were machine surfaced, sanded and end-sealed with epoxy resin. Three coats of polyurethane, acrylic or polyester based clear varnishes were spray applied to one face and both edges, with light sanding between coats. Samples were then exposed outdoors in Rotorua, New Zealand over summer, for a period of six months on a rack facing north and inclined 45°. The coated face of each board pointed north.

Microscopy
For light microscopy, hand sections were taken in the region of the wood-coating interface. Microtoming of un-embedded blocks can produce considerable cell wall distortion, and the hand sectioning was intended to minimise this. A drop of water was placed on the wood surface to moisten it just prior to sectioning to facilitate sectioning because sectioning of hard dry surfaces can also cause severe distortion. Since the exposure to water was very brief (and the wood surface was only moistened and not soaked) it is considered unlikely that the water applied contributed to film detachment observed. This assumption is supported by the fact that the film appeared intact in those regions where cell separation had not occurred in the underlying tissue layers. The sections were sequentially stained with 0.1% Sudan IV (in 80% ethyl alcohol) and 0.1% aqueous toluidine blue to highlight the coating and the wood cell walls respectively. The sections were then examined and photographed using a Zeiss Photomicroscope II. For transmission electron microscopy (TEM) the hand sections taken for light microscopy were cut into smaller pieces. The samples were dehydrated in an acetone series prior to embedding in Spurr’s low viscosity resin (Spurr 1969). Ultrathin sections were cut with an ultramicrotome using a diamond knife, stained with 1% potassium permanganate (prepared in 0.1% sodium citrate) and then examined with a Philips 300 TEM.

RESULTS AND DISCUSSION
A range of clear coatings on exposed wooden surfaces was examined. The extent of clear coating failure was variable but the mechanism was the same. Consequently, no reference is made to a particular coating type when describing the results.

The boards sampled for microscopy were similar to those shown in Figure 1 and had checking and discoloration after six months exterior exposure.

Before examining the microscopic data of the present work, it would be useful to review how sunlight contributes to the general weathering process. The impact of solar radiation on the weathering of wood is well documented (Hon 1981, 1983; Fengel & Wegener 1984; MacLeod et al. 1995; Plackett et al. 1996). During weathering, the lignin component of wood cell walls is photodegraded by sunlight, and to a much lesser
extent, so is the cellulosic material (Evans et al. 1996). The middle lamella, which consists largely of lignin (Donaldson 1985, 2001) and which holds adjoining wood cells together, may be affected as degradation of lignin and the breakdown of surface wood cells occurs. This process occurs with both uncoated and clear coated wood. Opaque coatings, on the other hand, restrict this degradation by preventing solar UV radiation from reaching the underlying wood surface. In such situations, coating failure is caused by the shrinkage and swelling of the wood surface, as moisture levels change, leading to cracking of the wood surface and the overlying coating. Once there is a loss in coating integrity, moisture levels in timber can increase rapidly leading to rising microbial populations, blistering of the coating and loss in wood-coating adhesion.

In the unexposed boards of the present work, the coating was in close contact with the wood surface and appeared to adhere well to it (Fig. 2). Staining of sections with toluidine blue (a metachromatic dye which stains lignified cell walls green or greenish blue and the primary cell walls pinkish purple because of their polyuronide content; O’Brien & McCully 1969) indicated that tracheid walls at the wood surface had uniform lignin distribution since the cell walls stained greenish blue (Fig. 2). In the boards that had been exposed outdoors for six months, the coating had failed in places and had lifted off the wood surface. Also, these boards were discoloured (Fig. 1), due to the presence of microorganisms on coating/wood surfaces.

Light microscopy of toluidine blue-stained sections taken from the wood-coating interface revealed that the failure (delamination) of the coating resulted mainly from the separation of wood cells from each other at and near the wood surface. An early stage of coating failure is shown in Figure 3 where small gaps between the coating and the wood surface are visible (arrowheads). Figure 3 also shows presence of microorganisms in a region where the coating has cracked or detached from the wood. There is delamination of coating in some parts due to separation of weathered surface cells (Fig. 4). Groups of weathered cells are still attached to delaminated parts of the coating, suggesting that the delamination resulted from failure within the wood tissue. One to two layers of tracheids under the coating have not reacted to the toluidine blue stain (are not stained blue-green) indicating a loss of lignin from the cell walls. However, it has to be emphasised that the staining method used here for light microscopy is useful only for qualitative assessment, and the absence of staining of surface cell walls, as seen in Figure 4, cannot be taken as an indication of total loss of lignin. Furthermore, there does not have to be complete loss of lignin from all parts of the wall for the cell separation to occur, since loss of lignin from the middle lamella is more critical than loss of lignin from the secondary wall. Evidence of this can be seen in Figure 5 where the surface cells, which have either completely separated from each other or are only loosely connected, appear to still have an affinity for toluidine blue, as their walls are blue to purplish blue.

Further evidence that failure within the weathered wood surface may account to a large part for the delamination of the coating can be seen in Figures 6 and 7. Figure 6 shows a region where a large part of the coating has separated taking with it the surface cells from the wood underneath. A higher magnification view (Fig. 7) shows the cells from the outermost wood layer to be in close contact with the coating.
Fig. 6 & 7. Coating detachment. – 6: A region where the coating (arrow) has detached from the wood taking with it some cells which were originally part of the wood surface. – 7: Higher magnification view of the detached coating (arrow) shown in Fig. 6. — Scale bar in 6 = 100 μm, in 7 = 40 μm. Light micrographs.
Fig. 8 & 9. Transverse sections through weathered surface cells underlying the coating. – 8: The middle lamella region (asterisk) is almost completely delignified (only some residues remain together with microbial slime). The cell walls are unevenly stained and are swollen, causing the $S_3$ layer to be wrinkled (arrow). – 9: Cells have completely separated. The $S_2$ layer in one of the cells appears striated (asterisk) due to irregular loss in lignin. The $S_3$ layer is highly undulated (arrow). — Scale bar = 2 µm for both figures. Transmission electron micrographs.
Transmission electron microscopy (TEM) of ultrathin sections after staining with KMnO₄ (a stain widely used to contrast lignin in plant and wood cell walls; Hepler et al. 1970; Maurer & Fengel 1990) has provided clear evidence of loss in lignin from the middle lamella and indicates that this is the most important factor in cell separation. Figure 8 shows almost complete depletion of lignin from the middle lamella in the surface cells underlying the coating, indicated by the low electron density of this region. The secondary walls in these cells are ‘patchy’, showing variable electron density. This patchiness reflects wide variation in lignin concentration; dense regions correspond to lignin rich areas and lucent or clear regions correspond to areas depleted of lignin. The secondary walls appear to be swollen, judging from the irregular thickness of the S₂ layer and the presence of wrinkles in the S₃ layer. In Figure 9 there is an intercellular gap unlike in Figure 8, suggesting that cells in Figure 9 have completely separated. Although the intercellular region that would have once been occupied by the middle lamella is lucent, indicating complete loss of lignin, a large proportion of the secondary wall appears dense, indicating the presence of substantial amounts of lignin. However, the S₂ layer in Figure 9 is ‘patchy’, much as in Figure 8, suggesting uneven loss in lignin from this layer.

Fig. 10. Transverse section through parts of weathered surface cells. Microorganisms (asterisks) are present in the region of the corner middle lamella where cells have separated. Arrows indicate the regions of the S₂ layer where microfibrillar structures are wavy. — Scale bar = 250 nm. Transmission electron micrograph.
Ultraviolet radiation is responsible for the primary photochemical processes in the oxidative degradation of wood (Hon 1991). Since lignin is the dominant UV absorber in wood, with an absorption maximum at 280 nm and extending down to over 400 nm, it absorbs more UV radiation than cellulose resulting in more photochemical degradation reactions. These reactions are principally light-initiated free radical reactions. An important free radical source is the phenolic hydroxyl group that readily absorbs UV light to produce a phenolic radical that can react further to form stable and lower molecular weight o- and p-quininoid structures.

Microorganisms can be seen in the highly porous and delignified middle lamella region between cells (Fig. 10), as well as within the gaps between delaminated coating and wood (Fig. 3). Some dense granular and fibrillar materials are present in the middle lamella region shown in Figure 10, and may represent a mixture of residual lignin and the extracellular microbial slime. However, this residual cell wall material is sparse and is not likely to provide any physical resistance to invading microorganisms. Although these microorganisms are not likely to be wood degrading fungi and bacteria (Blanchette et al. 1990; Eriksson et al. 1990; Singh & Butcher 1991), it would be interesting to investigate whether the microorganisms that invade such middle lamella regions can accelerate cell separation by utilising the wood cell wall degradation products of this region.

The combined light and electron microscopy observations of the wood-coating interface in this study provide evidence that coating failure resulted primarily from the photodegradation of wood cell wall material due to insolation, in particular the destruction of lignin from the underlying surface cells. The light microscopic observations made using toluidine blue as a lignin-specific stain (O’Brien & McCully 1969), which suggested lignin loss from cell walls, were confirmed by TEM observations of KMnO$_4$ stained ultrathin sections. In addition, the much greater resolution of TEM enabled detailed information to be obtained on the pattern of lignin loss from various cell wall regions. TEM provided evidence of the irregular loss of lignin in the secondary wall and showed lignin removal from the middle lamella to be greater and to occur at a faster rate than lignin removal from the secondary wall. The observed complete or near-complete loss in lignin from the middle lamella from the cells that still had substantial amounts of lignin left in their secondary walls clearly indicates this. Thus, TEM observations confirmed observations made by light microscopy, which suggested a loss in the lignin from the middle lamella region may be the main reason for the separation of cells in the surface layers.

Collectively, the observations of cell separation in the surface layers and adherence of wood cells to the delaminating coating suggest that wood failure and not the loss in coating adhesion to wood to be the main cause of coating failure. Although microorganisms were present in the regions where the coating had cracked or detached and were in the middle lamella regions containing sparse residues of the degraded wall material, there was no indication of any wood cell wall degradation through microbial activity (Blanchette et al. 1990; Eriksson et al. 1990; Singh & Butcher 1991). Microorganisms are known to colonise coating surfaces in outdoor situations (Singh et al. 1995), with indications that some microorganisms can utilise coating components (Ross
and Weinert 1962) and can cause or promote coating failure (Duncan 1963). In the present work there was no evidence that the microorganisms present had degraded the coating.

Solar radiation penetrates through the clear coatings on the surfaces of radiata pine panels and degrades the cell walls in the surface layers underneath the coating. Although such coatings can firmly attach to wood surfaces and can also resist degradation by microorganisms and solar radiation at least over relatively short periods of exposure, they are destined to fail in outdoor situations from wood failure. Ways have to be found to prolong the service life of clear coated wooden products, thus allowing the natural beauty of wood grain to be appreciated and enjoyed.

REFERENCES


