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Understanding the influence of wood as a substrate on the permeability of coatings by NMR imaging and wet-cup



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ABSTRACT

An important reason to apply coatings on wood is to protect wood against moisture. As a result of regulations and ecological concerns, there has been a shift towards waterborne coatings, which make coatings intrinsically more sensitive to water. As a consequence of the higher sensitivity to water, the durability of both wood and coatings can be negatively affected. In order to use waterborne coatings for woodcare, the main factors influencing transport through these coatings have to be understood. The aim of this study is to elucidate the influence of the wooden substrate on the water permeability of the coating applied to it. Pine sapwood, oak and teak were selected as the wood types, covering a whole range of low to high density wood. Three types of coatings were formulated: a solventborne alkyd, a waterborne alkyd and a waterborne acrylic. NMR imaging was used to measure the moisture distribution and quantify the changes in bound and free water in coated wood during drying. For all wood-coating combinations that were investigated, water transport appeared to be externally (i.e. coating) limited. The loss of bound water started only after evacuation of free water, which showed a local thermodynamic equilibrium associated with bound and free water. We furthermore compared water permeability of free films and wood supported films to understand the influence of the wood-coating interactions. The permeability of free coating films has been determined by the wet-cup method, followed by investigating the solubility and diffusion of water in order to understand the differences in the permeability. We found that the interaction of the coating with the wood has no influence on the water permeability for the considered combinations. Furthermore, the permeability is largely determined by the water solubility.

1. Introduction

Wooden products are found everywhere, e.g. furniture, cladding on houses or floors. Wood is a hygroscopic and porous material, which in many situations undergoes fluctuations in moisture content due to periodic water absorption and desorption. A high moisture content, such as in condensing environments, may result in durability loss due to fungal growth. Consequently, coatings are widely applied on wooden substrates for protective and decorative reasons, and to prolong the service life if they are maintained correctly. Poorly maintained coatings can even decrease the service life. The regulations on volatile organic compounds (VOCs) have led to increased use of waterborne (WB) coatings, which are more sensitive to water compared to solventborne (SB) coatings [1]. Since protection is one of the main properties claimed for coatings, obtaining knowledge on how a coating resists water is helpful in predicting the service life of the substrate and the coating.

Previously, water transport properties through coatings have been investigated by measuring the average moisture content in bulk wood [1-3]. Many studies have demonstrated the inhomogeneous distribution of water close to the surface [4-7]. Therefore, the barrier properties of coatings should be re-evaluated by monitoring the moisture content with spatial and time resolution. The influence of two coating systems and their barrier effect against water diffusion in spruce wood have been investigated by Sonderegger et al. [8] with the neutron imaging method, where the moisture gradient through the wood specimens was determined at different times during water uptake. However, the neutron imaging method does not give information on the state of moisture as bound (in cell wall) or free (in lumen or other void spaces) water. It is recognised that such identification of the state of water within wood [9,10], during uptake or drying by common experimental techniques like weighing [11,12], X-ray computer tomography (CT) [13] or neutron radiography [14] remains a challenge.

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Nuclear Magnetic Resonance (NMR) imaging has proven to be an excellent tool for determining the moisture content of wood samples during water sorption [10,15]. Combining NMR imaging with relaxometry allows for characterizing the state of moisture as bound or free water [10]. It is a non-invasive method that provides temporally and spatially resolved moisture profiles [16]. NMR imaging was also applied to study the distribution and penetration of oil in wood, in which the oil was used as water repellent for wood preservation [17]. It has been used to determine the distribution of water in coated wood mostly during absorption/uptake [6,7,18]. Van Meel et al. [6] showed that the moisture permeability depends on the specific combination of wood and coating, since the coating influences the moisture sorption of wood in different ways. They found that the water absorption of a hardwood dark red meranti only occurs via diffusion through the solid matrix. In their study, application of a WB acrylic coating had no influence on this absorption process, which was attributed to the large water uptake of the coating. On the other hand, a WB coating does reduce the water uptake of pine by preventing capillary water uptake where pine - in contrast to meranti [6] - contains relatively large and highly permeable pores. A coating may also reduce the drying rate of wood, but there is very limited work done on coated wood during desorption/drying by NMR [4]. In situ determination of local moisture content for uncoated wood has been achieved by portable NMR devices [19,20]. Moreover, the protective properties of wood coatings against moisture migration were evaluated and compared with the use of a portable NMR sensor [5]. However, the penetration depth with portable NMR devices is limited up to few millimetres.

The performance of coatings depends on many factors, of which the wood-coating interface is a crucial one. Microscopic tools, which differ in imaging capabilities, have been most widely applied to examine wood-coating interfaces [21,22]. A comparative study on confocal laser scanning microscopy (CLSM) and light microscopy (LM) to image the interface between *P. radiata* wood and clear coating showed that CLSM is superior to LM [23]. CLSM can resolve wood-coating interfaces more clearly and provides more information on the physical nature of interaction between the coating and the wood surface [23]. In this work, we used LM to determine the effective thickness of the dried coating layer, and get information on which depths the applied coatings penetrated the wood samples. Instead of focusing on the physical aspects of wood-coating interactions, we will investigate the influence of wood-coating interactions on the permeability of coatings by comparing the free films and wood supported films.

The aim of this study is to elucidate the influence of the wooden substrate on the water permeability of the coating applied to it. NMR imaging was used to measure the moisture content distribution and quantify the changes in bound and free water in wood (pine sapwood, teak and oak) as a function of coating permeability (SB alkyd, WB alkyd and WB acrylic) during drying of completely water saturated samples. NMR moisture profiles were used to calculate the decrease in average moisture content in order to determine the permeability of coatings on wood. The permeability of free coating films was determined by the wet-cup method, followed by comparison with the permeability of wood supported films.

2. Materials and methods

2.1. Wood and coating types

Three types of wood were studied: pine sapwood, oak and teak, which are commonly used in Central Europe for outdoor applications, such as furniture, cladding on houses or floors. The properties of these wood types were given in detail in a previous study [10] and summarized in Table 1.

Due to increased use of WB coatings, as a result of the regulations on volatile organic compounds, two types of waterborne (WB) coating and one solventborne (SB) coating as a reference were studied to gain

 Table 1

 Selected wood types and their general characteristics.

Wood type	Softwood/ hardwood	Density (kg/dm³)	Structure	Ring width (mm)
Pine sapwood (Pinus sylvestris)	Softwood	~0.54	No vessels	~1.4
Oak (Quercus petraea)	Hardwood	~0.64	Ring porous	~1.4
Teak (Tectona grandis)	Hardwood	~0.64	Semi-ring porous	~1.2

knowledge of how coatings resist water. Three transparent coating formulations, WB acrylic, WB alkyd, and SB alkyd, were prepared by AkzoNobel Decorative Paints, Sassenheim, the Netherlands, specifically for this work. The WB acrylic dispersion was based on butyl acrylate and methyl methacrylate, with a glass transition temperature (T_g) of 2.4 °C and a minimum film formation temperature of 7 °C. The final WB acrylic composition had a solid content of 35 wt.(weight)%, and the surfactant amount of 6.9 wt.% on solid content. The alkyd binder for both WB and SB compositions was based on tall-oil fatty acids with a long oil length and low molecular weight. The WB alkyd emulsion was prepared using a 2% load of non-ionic surfactant to obtain an average particle size of 200 nm. The final WB alkyd composition had a solid content of 35 wt.%, and the surfactant amount of 5.1 wt.% on solid content. The final SB alkyd formulation had a solid content of 65 wt.%.

2.2. Coatings on wood

The coatings (WB Acrylic, WB alkyd, SB alkyd) were applied by brush on wood (pine sapwood, oak, and teak). The overview of studied wood-coating combinations is given in Table 2.

In all combinations, three layers were applied to achieve a final effective thickness of around 50 μm on wood. The first layer was sanded to eliminate surface roughness. Two more subsequent layers were applied. The time between the applications of consecutive layers was one day. The coated wood samples were dried at room condition, i.e. 21 $^{\circ}C$ and 40% relative humidity, for at least four months before the measurements were performed. Note that the effect of aging the coated wood samples on the water transport properties was not studied, which is beyond the scope of this study. All measurements were performed on unaged coatings.

The coatings were applied on the radial plane for pine sapwood and on the tangential plane for oak and teak. The direction influences the coating penetration characteristics [21]. In the case of coated pine sapwood for all 3 types of coating, the rays are parallel and the growth rings are perpendicular to the coated surface. In the case of coated teak for all 3 types of coating, the growth rings and rays are oriented diagonally (about 45° angle) to the coated surface, but perpendicular to each other. In the case of coated oak, the orientation of the growth rings and rays is different between samples. In the early-wood rings of oak, there are big pores, i.e. vessels (\sim 300 µm), which is shown with the CT image in Fig. 1, as also explained in a previous study [10].

For SB alkyd coated oak, the growth rings and rays are oriented diagonally (about 45° angle) to the coated surface, i.e. there are vessels on the surface aligning into the wood with about 45° angle. For WB acrylic and WB alkyd coated oak, the rays are perpendicular and the growth rings are parallel to the coated surface, i.e. vessels are oriented parallel to the surface.

The dry film thickness of coatings on wood was determined from the cross-sections of the samples by using light microscope (Leica DMRX) according to ISO 2808:2007 [24] and ISO 1463:2003 [25]. Firstly, 20 mm diameter cylindrical samples were drilled from 10 mm thick coated wood panels. The samples were fixed on a small piece of wood with epoxy glue for better handling. The cross-sections were made with

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