



Physical and morphological properties of UV-cured cellulose nanocrystal (CNC) based nanocomposite coatings for wood furniture



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ABSTRACT

In this study, UV curable high solid content wood coating systems with high performance were developed by using cellulose nanocrystal (CNC). CNC modified by a cationic surfactant was added to the coating system at two loadings (1 and 3%). Water uptake and permeability, optical clarity and glass transition temperatures (T_g) of these new nanocomposite coatings were measured. The quality of CNC dispersion in the matrix was also studied. Results showed that CNC improved the barrier and optical properties of the nanocomposite coatings. T_g of the polymer matrix did not change significantly with the addition of CNC. The quality of CNC dispersion in the polymer matrix was a function of CNC loading. High CNC loading (3%) resulted in sizable agglomeration, but this agglomeration did not negatively impact coating performance.

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1. Introduction

Wood is a beautiful material that has been used for centuries in construction. Despite its natural beauty and other unique features, its durability and vulnerability toward fungus and insects is an ongoing source of concern. Wood is a challenging material to protect, as it is derived from nature and is therefore intrinsically variable. Traditionally, wood is protected by preservatives which are usually toxic and harmful to the environment. Replacing traditional wood preservatives is more urgent for indoor applications and there are regulations that forbid or restrict the usage of preservatives. Coatings are one of the alternatives that have been used by the wood industry to replace preservatives.

Wood coatings protect the beauty of the wood and increase its value, which is important in the wood processing industry. Most traditional wood coatings, including nitrocellulose lacquer, polyurethane, unsaturated polyester and amino acid curing coatings [1], use solvent-based paint, which contains numerous volatile organic compounds (VOCs). Some coatings contain

benzene, toluene, xylene, and substances of high carcinogenicity, which harm the environment and human health.

The furniture industry is one of the biggest consumers of wood coatings in the wood industry. Transparent coating systems are widely used to protect wood and to enhance its beauty. Solvent-borne coatings are the dominant coating systems in the furniture industry, because of their fast drying time, good properties and low cost. Environmentally friendly or compliant coatings, such as high-solid-content, waterborne, radiation-curable or powder coatings have been considered as substitutes for the traditional solvent-borne coatings [2,3].

In addition to replacing coating components, the introduction of UV-curable wood coatings has accelerated the development of environmentally friendly coatings. UV curing has now been established as an alternative curing mechanism to thermal hardening. This alternative technology uses photon energy from radiation sources in the short wavelength region of the electromagnetic spectrum in order to form reactive species, which trigger a fast chain growth curing reaction. UV-curable coatings are well known for their superior durability, chemical and stain resistance, as well as for their many benefits for production facilities such as faster production rates with fast short term handling and packaging, as well as significant reductions in, or complete elimination of VOCs [4–6].

In this study, a UV-curable high solid content coating system was chosen as an alternative for the classic solvent-borne coatings.

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Developing nanocomposite coatings is reported to be effective in enhancing the performance of wood coating systems [7–12]. In the past, nanocomposite coatings for wood were mostly developed using inorganic nanomaterials, although there are some reports of using organic nanomaterials. Cellulose nanocrystals (CNC), rendered hydrophobic by a cationic surfactant, were used in this study as a reinforcing agent for UV-curable high solid content coatings.

The first reports on CNC composites were published by Favier et al. [13,14]. The use of CNC has been targeted for a number of unique features, including reinforcement and barrier properties. CNCs are typically produced by the acid hydrolysis of native crystalline cellulose using hydrochloric, sulfuric, or phosphoric acid. The crystalline portion of cellulose has been characterized as having excellent mechanical properties. CNCs have been used as composite reinforcements in various applications [15–19].

The cornerstone of this work is to produce nanocomposite materials by radiation induced polymerization of multifunctional monomer and oligomer containing nano-scale particles. This type of nanocomposite material, when used in the furniture industry, will have a solvent free-formulation with essentially no emissions of VOCs and will be user friendly, improve productivity and increase product performance. The main objective of this research was to develop for the furniture industry, a UV curable coating system with superior performance. The physical and morphological properties of the CNC based nanocomposite coatings were studied.

2. Materials and methods

2.1. Materials

Cellulose nanocrystals (CNC), kindly provided by Forest Products Laboratory in Madison, WI USA, were used as the reinforcing material in 5.5% water suspension. The cationic surfactant, hexadecyltrimethylammonium (HDTMA), 99% purity, was purchased from Sigma-Aldrich. The wood coating system used in this study was a free radical photopolymerization system. The photoinitiator was Darocur 1173 (2-Hydroxy-2-methyl-phenyl-propanone) supplied by BASF, USA. A defoaming agent (Byk-1798 from Byk) was used to reduce the bubbles produced during mixing process. Properties for the oligomers and monomers are given in Table 1. Oligomers and monomers were supplied by Sartomer. Both oligomers, CN 104A80Z and CN 131B, were of the acrylate type. CN 104A80Z (epoxy acrylate) has a higher reactivity and good water resistance. CN 131B has lower viscosity than CN 104A80Z and facilitated fast curing and developed strong, flexible cured films. For monomers, the SR 350 and SR 9003B had methacrylate and acrylate double bonds in their chemical structure, respectively.

2.2. Methods

2.2.1. CNC surface modification

Before planning the experimental design, pre-tests were conducted to evaluate the compatibility of the matrix with unmodified CNC and CNC modified by adding two different concentrations (0.35

and 1.4 mmol/g) of HDTMA for 2 h. The results of pre-tests showed that unmodified CNC and CNC modified by using the 0.35 mmol/g concentration of HDTMA had no compatibility with the matrix. Inversely, CNC modified by 1.4 mmol/g of HDTMA demonstrated excellent compatibility with the matrix. As a result, the study was focused on the effects of adding CNC modified by 1.4 mmol/g of HDTMA.

2.2.1.1. Reaction. To prepare the HDTMA solution for the reactions, 3.06 g HDTMA was added progressively to 48 ml of deionized (DI) water heated at 45 °C. The solution was subjected to magnetic stirring until it reached a stabilization point. In a 300 ml triple-neck round-bottom flask, 35.09 g of the 5.5% CNC solution was weighed and 17.02 g of the HDTMA solution was then added very slowly while the solution was stirred using a propeller stirrer system. The concentration of HDMTA was adjusted by adding 8 ml of deionized water to the solution. At this point, two necks of the flask were covered by a natural rubber septum and a syringe was inserted to allow air circulation. The solution was then stirred for 2 h.

2.2.1.2. Washing. The modified CNC was washed with deionized water before being filtered. The suspensions were then centrifuged for 3 separate 15-minute periods at 15,000 rpm to remove any possible excess of quaternary ammonium salt that could have aggregated on the surface of the CNC. At the end of each round of centrifugation, the suspensions of CNC were again filtered and washed with deionized water.

2.2.1.3. Drying. The CNC samples were kept in a freezer at –80 °C, before being freeze-dried at –91 °C for at least four days. The results of modified CNC characterizations were reported in another publication [20].

2.2.2. Preparation of UV-curable formulations

Chemical compounds used for the coating formulations in this study are presented in Table 2. For the coating without CNC, the two oligomers were first mixed by using a high speed mixer (Disolver Dispermat® LC30 VMA-Getzmann GmbH). Afterwards, the two monomers were gradually added to the oligomers, followed by the addition of a defoaming agent. The photoinitiator was the last component that was added to the formulation. To prepare the nanocomposite coatings, the two oligomers were mixed by using the high speed mixer. Afterwards, the two monomers were slowly added to the oligomers, followed by the addition of a defoaming agent. Then, a given amount of modified CNC was added to the blends and the stirring continued for another 15 min. The photoinitiator was added last. The formulation was homogenized by ultrasonication to ensure that the CNC was well dispersed in the matrix. The ultrasonication was conducted using a high intensity ultrasonic horn (60 kHz, maximum amplitude of 100 nm from tip to tip, Branson PG). The temperature of the solution was kept below 80 °C to ensure that the coating did not overheat. To maintain the temperature of the mixture below 80 °C, the vessel with the mixture was placed in an ice bath. Ultrasonication was carried out with

Table 1
Properties of the oligomers and monomers used in this study.

	Commercial name	Chemical name	Viscosity (cps) at 25 °C	Refractive index
Oligomers	CN 104A80Z	Difunctional bisphenol A based epoxy acrylate blended with tripropylene glycol diacrylate (TPGDA)	22,500	1.5317
	CN 131B	Monoacrylate oligomer	250	1.5247
Monomers	SR 350	Trimethylolpropane trimethylacrylate	44	1.4701
	SR 9003B	Propoxylated neopentyl glycol diacrylate	15	1.4464

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