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Castor-oil-based waterborne acrylate/SiO_2 hybrid coatings prepared via sol–gel and thiol-ene reactions

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ABSTRACT

Castor-oil-based waterborne acrylate (CWA)/SiO_2 hybrid coatings with organic-inorganic covalent cross-linked network structures were prepared via sol–gel and thiol-ene reactions. The stabilities of the emulsions as well as the structures, thermal properties, and mechanical properties of the films were characterized. The results showed that the emulsions had good stability. All of emulsions remained stable after being stored for more than 6 months and the mean particle sizes of emulsions increased from 33.8 to 70.1 nm as the SiO_2 loading increased from 0 to 20 wt%. Furthermore, the thermal and mechanical properties of the films improved significantly. The residues at 600°C, glass transition temperature (T_g), crosslink density, Young's modulus, and pencil hardness increased from 4.6 %, 9.4°C, 23 mol/m^3, 2.1 MPa, and H to 16.2 %, 19.2°C, 736 mol/m^3, 30.0 MPa, and 3H, respectively. Among these coatings, the coating with 10 wt% of SiO_2 loading had best comprehensive performances for the application of wood coating.

1. Introduction

Nowadays, the national environmental protection policies are increasingly strict. Non-pollution, low cost, energy saving and consumption reduction have become the development trend of the coatings industry [1–3]. The vegetable-oil-based waterborne coatings (VWCs), which take into account the advantages of both vegetable-oil-based and waterborne coatings, have rapidly turn into the focus of the coatings [3–5].

VWCs are synthetized by a part of vegetable oils and their derivatives as raw materials to substitute the petroleum-based materials. In recent years, VWCs have received broadly attention and been frequently reported in literature due to their many advantages such as their wide range of raw materials, renewability, biodegradability, multiple active sites and low VOCs emissions [6–9]. However, the introduction of long fatty acid moieties of vegetable oils results in unsatisfactory mechanical properties (e.g., modulus, strength, and hardness), limiting their wider applications [10–12]. These issues can be effectively overcome by fabricating organic–inorganic nanocomposites, which combine the advantages of polymers and inorganic nanoparticles [13]. Organic–inorganic nanocomposites are mainly prepared by blending [14], sol-gel [15] and in situ polymerization [16].

In waterborne hybrid coating systems, good emulsion stability is key to the coatings. The introduction of inorganic nanoparticles reduces the stability of the emulsion to some extent. Therefore, the dispersion of inorganic nanoparticles in emulsions has become an urgent problem for preparation of organic-inorganic hybrid coatings [17,18]. Recently, some studies have shown that this problem can be overcome by a sol–gel synthesis with silane coupling agents as precursors [19–21]. For example, Larock et al. directly grafted a silane coupling agent (3-aminopropyl triethoxysilane) on a castor-oil-based polyurethane molecule to obtain a waterborne nanocomposite with stable storage properties [20]. Moreover, in nanocomposites, silica particles serve as an enforcement filler and improve the thermal properties, wear resistance, chemical resistance, hardness and strength of materials [22]. The interaction between polymer and the filler also play a significant role in the properties of materials. The strong interaction (such as covalent bonding) which is preferred can improve the compatibility [23]. But most of existing researches just loaded low content of SiO_2 (less than 5 wt%) and built weak interaction (such as hydrogen bonding) in hybrid coatings [13–19]. Sometimes, that was not enough for VWCs to meet the application requirement. In order to significantly improve the

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Abbreviations: CWA, castor-oil-based waterborne acrylate; VWCs, vegetable-oil-based waterborne coatings

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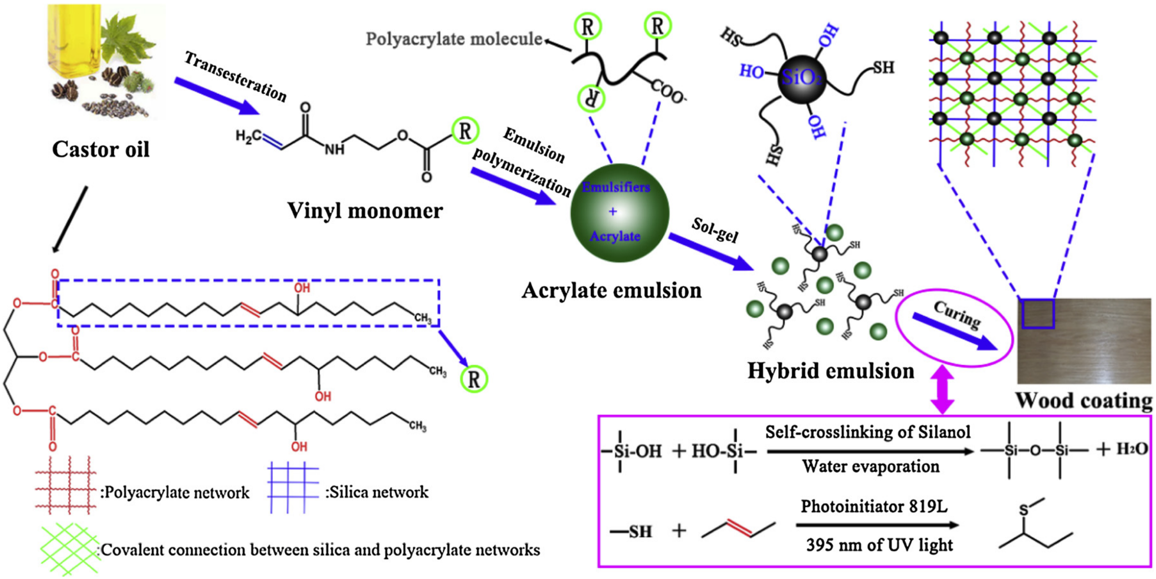

Fig. 1. Experimental process for preparation of the CWA/SiO_2 hybrid coatings.

thermal properties, mechanical properties and hardness of materials, the hugely increasing crosslink density and covalent bonding were needed. However, this issue has been rarely reported.

In this paper, a simple sol–gel method and thiol-ene reaction were developed to prepare CWA/SiO_2 hybrid coatings containing organic-inorganic covalent cross-linked network structures. The CWA/SiO_2 samples prepared herein exhibited good emulsion stabilities and significantly improved thermal properties, mechanical properties and hardness. In the sol–gel process, silica particles with large numbers of −SH and Si−OH groups were formed via hydrolysis and condensation reactions. In the curing process, the Si − OH groups of the nano-silica condensed readily to form SiO_2 network as water evaporated. The −-SH groups of the nano-silica generated covalent bridges between the silica and the polyacrylate networks via thiol-ene reactions under UV light. The goal of this paper is to enhance the thermal properties, mechanical properties and hardness of VWCs for wider application by the hybrid way of significantly increasing the crosslink density and building covalent bonding. The experimental process for preparation of CWA/SiO_2 hybrid coatings is shown in Fig. 1.

2. Experimental

2.1. Materials

Castor oil (Macklin, analytical reagent (AR)), N-(hydroxyethyl)acrylamide (Macklin, AR), methyl methacrylate (MMA, Macklin, AR), butyl acrylate (BA, Macklin, AR), acrylic acid (AA, Macklin, AR), ammonium persulphate (APS, Macklin, AR), sodium hydroxide (Macklin, AR), 2,6-dimethylphenol (Macklin, AR), anhydrous magnesium sulfate (Macklin, AR), sodium bicarbonate (Macklin, AR), sodium dodecyl sulfate (SDS, Macklin, AR), and (3-mercap-topropyl)trimethoxysilane (MPTMS, Macklin, AR) were used herein. Polyoxyethylene octyl phenol ether-10 (OP-10, AR), hexane (AR), methylene dichloride (AR), and tetrahydrofuran (THF, AR) were purchased from Tianli Chemical Reagent Inc. (Tianjin, China). Phenyl bis (2,4,6-trimethylbenzoyl)phosphine oxide (Photoinitiator 819 L) was purchased from Wraio Chemicals (Guangzhou, china). Deionized water and saturated brine were prepared in house.

2.2. Preparation of castor-oil-based vinyl monomer (CVM)

CVM was synthesized by following the method reported by Popadyuk with some modifications [24]. A four-neck 250-mL round-bottom flask provided with a mechanical stirrer, a condenser tube, and a kerosene thermometer was loaded with ca. 57.5 g of N-hydroxyethyl-acrylamide, 150 g of castor oil (acrylamide alcohol to triglyceride molar ratio of 3 to 1), 150 mL of tetrahydrofuran, 1.0 g of ground sodium hydroxide as a catalyst, and 0.1 g of 2,6-dimethylphenol upon continuous stirring. The reaction mixture was subsequently heated at 40°C in water bath for 3 h. The mixture was then cooled to room temperature and stored overnight. Specific amounts of CH_2Cl_2 were added to dilute the mixed solution, which was purified by washing it with saturated brine, treated with magnesium sulfate, and vacuum-dried at 60°C for 4 h. About 80 g of CVM was obtained. The transesterification reaction

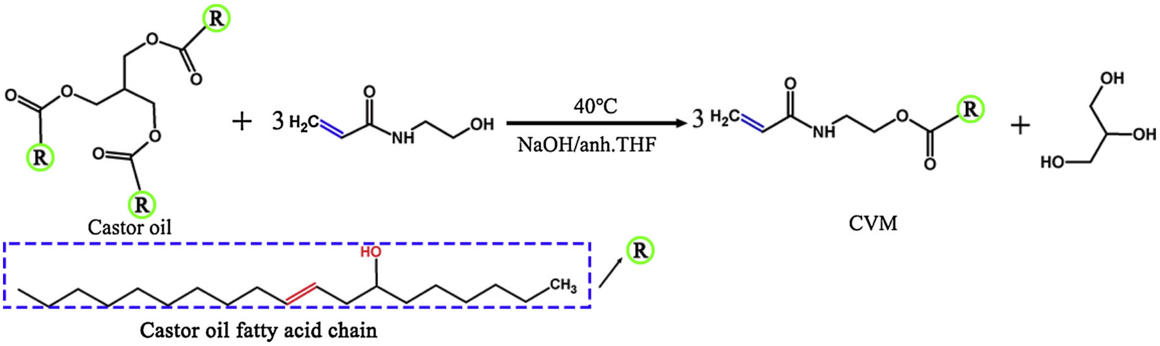

Fig. 2. The transesterification reaction for the synthesis of CVM.

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