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Preparation and characterization of urea-formaldehyde resin/reactive kaolinite composites

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ABSTRACT

Novel urea-formaldehyde resin/reactive kaolinite composites containing 20–40 wt% kaolinite were prepared by in situ polymerization. The kaolinite was modified with tetraethoxysilane and a silane coupling agent to introduce reactive groups. Fourier-transform infrared spectroscopy and X-ray diffraction confirmed preparation of the urea-formaldehyde resin/reactive kaolinite composites. The composite morphology was investigated using scanning electron microscopy; the composites consisted of uniform spherical particles. The surface chemical components of the composites were determined using energy-dispersive X-ray spectroscopy. The spectra showed that the reactive kaolinite was encapsulated by the urea-formaldehyde resin. The thermal properties of the composites were examined using differential scanning calorimetry and thermogravimetric analysis. The results showed that their thermal stability was much better than that of pure urea-formaldehyde resin. Reactive kaolinite addition greatly decreased formaldehyde emissions and improved the water resistance of the composites. A mechanism for urea-formaldehyde resin/reactive kaolinite composite synthesis is proposed.

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Introduction

Much research on polymer/inorganic composites has been performed, to improve the properties of traditional polymers and reduce the cost of polymeric materials. Many new materials with excellent properties such as high strain, high conductivity, and high water absorption have been successfully prepared (Ammala, Hill, Lawrence, & Tran, 2007; Chen et al., 2006; Podsiadlo et al., 2007; Şen, 2010; Tunc & Duman, 2010; Tunc, Duman, & Uysal, 2008; Villanueva, Cabedo, Lagaron, & Gimenez, 2010; Yoon, Hunter, & Paul, 2003; Zhang et al., 2014). However, some drawbacks, including weight increase, brittleness, and opacity, are also encountered (Fischer, 2003; Gorrasi, Tortora, Vittoria, Galli, & Chiellini, 2002; Varlot, Reynaud, Kloppfer, Vigier, & Varlet, 2001). Scientists have now begun to study composites that have superior properties but retain their original characteristics such as opacity (Chafidz, Ali,

Mohsin, Elleithy, & Al-Zahrani, 2012; Misra, Raney, De Nardo, Craig, & Daraio, 2011; Nakane, Kurita, Ogihara, & Ogata, 2004; Wheeler, Wang, & Mathias, 2006; Zhang et al., 2011; Zhao, Zhang, Huang, & Wei, 2012).

Urea-formaldehyde resins are widely used as adhesives, coating materials, synthetic woods, and mold powders, because of their panel properties, including fast adhesive curing, water solubility, and high reactivity. The main disadvantages of urea-formaldehyde resins are high formaldehyde emissions, low resistance to humid conditions, and poor thermal stability. Stricter control of the resin properties, especially in terms of thermal stability, formaldehyde emissions, and water resistance, is needed to enable expansion of their applications. Much research has been performed on enhancement of the thermal stability and mechanical properties. Effective methods for improving the properties of these resins include adding organic monomers such as melamine, trimethoxymethylmelamine, and dimethoxymethylmelamine, or inorganic additives such as carbon nanotubes and silicates (Roumeli et al., 2012; Samaržija-Jovanović, Jovanović, Konstantinović, Marković, & Marinović-Cincović, 2011; Xu, Tang, Gu, Fang, & Tong, 2007; Zorba, Papadopoulou, Hatjiissaak, Paraskevopoulos, & Chrissafis, 2008). However, to the best of our

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knowledge, there are no reports of thermal stability enhancement, formaldehyde emission reduction, and water resistance improvement of urea-formaldehyde resins using kaolinites.

Kaolinites are 1:1 layered silicates; they are readily available and widely used. The layers are held together via hydrogen bonds, dipolar interactions, and attractive van der Waals forces, resulting in small interlamellar spaces, in which it is hard for organic materials to intercalate. Although polymer/kaolinite composites have been prepared using ion-exchange reactions, industrialization is difficult and challenging (Chafidz, Ali, & Elleithy, 2011; Letaief & Detellier, 2007; Letaief, Diaco, Pell, Gorelsky, & Detellier, 2008; Tonle, Letaief, Ngameni, & Detellier, 2009). Another method for polymer/kaolinite composite synthesis is surface modification, in which a coupling agent is covalently bonded to the surface of the clay to make the kaolinite compatible with polymers. Typical examples are polyimide/kaolinite, poly(methyl methacrylate)/kaolinite, and polybutadiene/kaolinite (Piscitelli et al., 2010; Wheeler et al., 2006; Xu et al., 2007; Zhang, Liu, Zhang, Cheng, & Lu, 2012). Recently, many composites with outstanding properties have been synthesized; the amounts of clay in the composites were low, ranging from 1 wt% to 5 wt% (Khezri, Haddadi-Asl, Roghani-Mamaqani, & Salami-Kalajahi, 2012; Wang, Gao, Ma, & Agarwal, 2006).

This work focuses on the preparation of urea-formaldehyde resin/reactive kaolinite composites by in situ polymerization. This is the first time that reactive kaolinites have been used to improve the properties of urea-formaldehyde resin; the urea formaldehyde resin/reactive kaolinite composites consisted of uniform spherical particles; the kaolinite content of the composite was up to 40 wt%. The aims of the research were to prepare urea-formaldehyde resin/reactive kaolinite composites with high thermal stabilities, strong water resistance, and low formaldehyde emissions. In addition, we expect that a high kaolinite content will reduce the cost of the composites.

Experimental

Materials

Kaolinite particles of thickness 20–50 nm, average diameter 400 nm, and specific surface area 32 m²/g were supplied by the San Xing High-New Material Company (Zaozhuang, China). (3-Aminopropyl)triethoxysilane (KH550) and tetraethoxysilane (TEOS, chemical grade) were purchased from the Beijing Chemical Reagents Company (Beijing, China). Sodium hydroxide, urea, formalin (37 wt%), acetone, ammonia (28 wt%), and anhydrous ethanol (99.5%) were of analytical grade were purchased from Beijing Chemical Reagents Company (Beijing, China). Deionized water (18.2 MΩ cm) was used in all experiments.

Preparation of reactive kaolinite

The desired amount of dried kaolinite and anhydrous ethanol (100.0 mL) were placed in a ball grinding mill and ball-milled for 1 h. The mixture was transferred to a 250 mL three-necked round-bottomed flask containing ammonia (18.4 mL). After stirring at 60 °C for 1 h, TEOS (10.0 mL) was added, and the mixture was stirred for 6 h. The slurry was filtered and washed several times with ethanol. The solid residue was dispersed in ethanol and hydrolyzed KH550 (5.0 mL) was added, with continuous stirring for 1 h. The slurry was filtered, washed, dried at 110 °C for 12 h and crushed to a powder. The product yield was 96%. The product is denoted by KT-kaolinite.

For comparison, pristine kaolinite (denoted by P-kaolinite) was directly modified with hydrolyzed KH550; the product is denoted

by K-kaolinite. The product yield (actual output/theoretical output) was 90%.

Preparation of urea-formaldehyde resin/reactive kaolinite composites

Urea (3.0 g), formaldehyde (3.6 mL), and deionized water (25.0 mL) were placed in a three-necked round-bottomed flask. The pH of the solution was adjusted to 8–9 with sodium carbonate, followed by continuous stirring for 1 h at 75 °C. The product is denoted by pre-UF.

The desired amount of KT-kaolinite, deionized water (100.0 mL), and sodium dodecylbenzenesulfonate (2.0 g) were placed in a three-necked round-bottomed flask and the mixture was stirred for 2 h at room temperature. The pre-UF was transferred to the flask at 60 °C, followed by continuous stirring for 3 h. The slurry was filtered, washed several times with water, and dried at 120 °C for 12 h. The product yield was 92%. The product is denoted by KT-UF.

The products containing KT-kaolinite of 20, 30, and 40 wt% are denoted by KT-UF 20%, KT-UF 30%, and KT-UF 40%, respectively. Pure urea-formaldehyde resin and urea-formaldehyde resin/pristine kaolinite composite containing 30 wt% pristine kaolinite were also prepared; they are denoted by pure UF and P-UF 30%, respectively.

Preparation and testing of plywood

Three-layer plywood panels of dimensions 300 mm × 300 mm × 1.5 mm were prepared using eucalyptus veneers. The veneers were dried to a moisture content of approximately 3% before use. The urea-formaldehyde resin/reactive kaolinite composites were mixed with 5 wt% ammonium chloride solution and applied to both sides of the veneer at a spreading rate of 350 g/m² (to form two glue lines). The veneers were aged for 15 min to allow the composites to penetrate the veneers. The veneers were pre-pressed at room temperature and 1.0 MPa for 1 h, and then hot pressed at 120 °C and 1.5 MPa for 4.5 min (1 min/mm). After hot pressing, the panels were cooled and conditioned at 20 °C and (65 ± 2)% relative humidity until the weight was constant, to test the formaldehyde emissions and water resistance.

Characterization

Fourier-transform infrared (FTIR) spectroscopy was performed (Spectrum 1000, Perkin-Elmer, USA) in the region 400–4000 cm⁻¹ with a resolution of 4 cm⁻¹ and 32 scans. Specimens were prepared by grinding the sample with KBr.

The X-ray diffraction (XRD) patterns were acquired using a Siemens D-500 diffractometer operated at 40 kV and 30 mA (1200 W), with filtered Cu Kα radiation, at 2θ from 5° to 60° and a scanning speed of 20°/min.

The microstructures of the samples were examined using scanning electron microscopy (SEM, XL 30, FEI, USA; accelerating voltage 20 kV). The specimens were coated with a thin carbon film prior to SEM observations. Energy-dispersive X-ray spectroscopy (EDS) coupled with SEM, which enables qualitative analysis, was used to determine the elements present in the samples.

The thermal stabilities of the samples were determined by thermogravimetric analysis (TGA) under nitrogen at a heating rate of 20 °C/min from 25 to 700 °C using a TG-differential thermal analysis (DTA) instrument (STA449, Netzsch, Germany). The sample (more than 8 mg) was placed in an alumina crucible. An empty alumina crucible was used as a reference.

Three-layer plywood test specimens of dimensions 10 mm × 50 mm × 150 mm were placed in a 10 L glass desiccator together with a Petri dish filled with a specified amount of

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