

Nonisocyanate Poly(Hydroxyl Urethane)-Based Green Polymer Hybrid Coating Systems: Tailoring of Biomacromolecular Compound Architecture Using APTMS-ZnO/TEMPO-Oxidized Cellulose Nanoparticles

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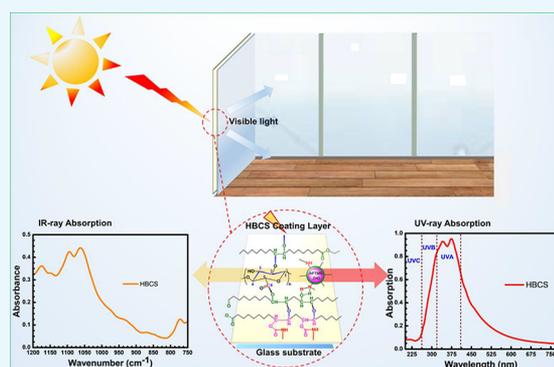


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Supporting Information

ABSTRACT: Hybrid bionanocomposite coating systems (HBCSs) are green polymer materials consisting of an interface between a coating matrix and nanoparticles. The coating matrix was prepared by using a nonisocyanate poly(hydroxyl urethane) (NIPHU) prepolymer crosslinked via 1,3-diaminopropane and epoxidized *Jatropha curcas* oil. TEMPO-oxidized cellulose nanoparticles (TARC) were prepared from microcrystalline cellulose, and (3-aminopropyl)trimethoxysilane (APTMS)-coated ZnO nanoparticles (APTMS-ZnO) and their suspensions were synthesized separately. The suspensions at different weight ratios were incorporated into the coating matrix to prepare a series of HBCSs. FT-IR, ¹H-NMR, ¹³C-NMR, XRD, SEM, and TEM were used to confirm the chemical structures, morphology, and elements of the coating matrix, nanomaterials, and HBCSs. The thermomechanical properties of the HBCSs were investigated by TGA-DTG and pencil hardness analyses. The UV and IR absorption spectra of the HBCSs were obtained using UV-vis spectroscopy and FTIR spectroscopy, respectively. The HBCSs exhibited good thermal stability at about 200 °C. The degradation temperature at 5% mass loss of all samples was over around 280 °C. The HBCSs exhibited excellent UV block and IR active properties with a stoichiometric ratio of the NIPHU prepolymer and EJCO of 1:1 (wt/wt) containing 5 wt % TARC and 15 wt % APTMS-ZnO nanoparticles. It was observed that the sample with 5 wt % TARC and 15 wt % APTMS-ZnO (HBCS-2) exhibited a uniform crosslinking and reinforcement network with a T_{onset} of 282 °C. This sample has successfully achieved good coating hardness and excellent UV and IR absorption.



INTRODUCTION

The present perspective of the development of sustainable biobased chemistry and greener components of polymer materials has attracted both industrial and research-oriented applications.^{1–3} Hybrid nanocomposite coatings are a novel class of advance materials containing both a coating matrix and nanoparticles. A potential combination of the advantages of inorganic materials including high thermal and chemical stability, high hardness, high refractive index, etc. with those of the coating matrix such as processability, flexibility, low weight, etc. can enable a wide range of applications for these nanocomposites. The applications range from conventional plastics reinforcement to abrasion-resistant coatings,^{4,5} optical devices,^{6,7} catalysis,^{8,9} memory devices,¹⁰ electrical insulation in microelectronics and nanodielectrics,^{11,12} integral capacitors,¹³ fuel cells,¹⁴ flame-retardant materials,^{15,16} etc. However, in terms of bio-based hybrid nanocomposite coating, vegetable oils and nonisocyanate poly(hydroxyl urethane) (NIPHU)-based fabrication methods have widely attracted researchers

and academics during the past few decades. NIPHU is an alternative green chemistry approach that can replace toxic isocyanate-based polyurethane production. However, the hydroxyl groups positioned at the α - and β -carbon in the NIPHU moiety promote adhesiveness and steric hindrance among polyurethane macromolecules.¹⁷ Furthermore, the NIPHU networks become weaker owing to the steric hindrance of their hydroxyl moieties. Therefore, a casting temperature above 100 °C and a longer curing time are required^{18–20} to prepare NIPHU-based polymer materials. A recent study has reported a temperature of about 120 °C for the curing process to obtain NIPHU thick film materials.²¹

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