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Review

Polyaniline thermoset blends and composites

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ABSTRACT

The blending of polyaniline (PANI) with insulating polymers is an active area of research which has been driven by the possibility to combine the good mechanical properties and processability of the polymer matrix with low electrical resistance. This review will focus on thermosetting polymer matrix composites or blends. A good dispersion of PANI in the thermoset matrix is essential for the enhanced mechanical and electrical properties of the material. Much effort has been exerted in attempts to improve the compatibility of PANI with thermoset matrices. Attention will be given to describe some of the processing parameters that affect the morphology of PANI thermoset blends and composites. In recent years, there has been renewed interest in PANI thermoset composites with the emergence of multifunctional ternary composites. The different approaches for the design of ternary composites will be reviewed. Additionally, promising applications of PANI thermoset composites in different fields will be described such as electromagnetic shielding and microwave absorption, static electricity dissipation, flame-retardant materials, conductive adhesives, coatings for anticorrosion protection, sensor materials and electro-stimulated drug delivery systems.

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Abbreviations: DMSO, dimethylsulfoxide; DMF, dimethylformamide; NMP, *N*-methyl-2-pyrrolidone; DBSA, dodecylbenzenesulfonic acid; DSA, dodecylsulfonic acid; CSA, camphorsulfonic acid; TSA, *p*-toluene sulfonic acid; PA, phosphoric acid; DPA, *n*-decylphosphonic acid; DESSA, dialkoxyester of sulfosuccinic acid; DEHSSA, di(2-ethylhexyl)sulfosuccinic acid; TA, tartaric acid; CP, conductive polymer; PANI, polyaniline; EB, emeraldine base; ES, emeraldine salt; ACAT, amine-capped aniline trimer; TANI, tetraaniline; POTOE, copoly(*o*-toluidine/*o*-ethylaniline); POTOEPTS, copoly(*o*-toluidine/*o*-ethylaniline)-*p*-toluenesulfonate; OAN, oligoaniline; PAPTS, polyaniline-*p*-toluenesulfonate; EP, epoxy resin; DGEBA, diglycidyl ether of bisphenol A; PU, polyurethane; DPI, dopable polyimide; CFRP, carbon fiber-reinforced polymer; PAMPS, poly(2-acrylamido-2-methylpropane sulfonic acid); PAAm, polyacrylamide; DVB, divinylbenzene; EPDM, ethylene propylene diene monomer; NR, natural rubber; PVA, polyvinyl alcohol; PMMA, poly(methyl methacrylate); POE, polyoxyethylene; POA, poly(*ortho*-anisidine); TETA, triethylenetetramine; DAB-AM-4, *N,N,N′,N′*-tetrakis(3-aminopropyl)-1,4-butanediamine; TMDA, trimethylhexanediamine; DPA, 4-diphenylamine diazonium; B-DPA, 4-diphenylamine diazonium-modified bentonite; NC, nanocomposite; IPN, interpenetrating polymer network; Semi-IPN, semi-interpenetrating polymer network; HG, hydrogel; ISP, in situ polymerization; SIP, surface initiated polymerization; P, particles; W, wires; G, globules; NP(s), nanoparticle(s); NS, nanospheres; NR, nanorods; FL, flake-like; NF, nanofibers; GF, glass fiber; SCF, short carbon fibers; MWCNT, multiwall carbon nanotube; rGO, reduced graphene oxide; CN(s), cellulose nanowhisker(s); DMA, dynamic mechanical analysis; Tg, glass transition temperature; T_{CP}, cloud point temperature; RL, reflection loss; HRR, heat release rate; MAM, microwave absorbing materials; RAM, radar absorbing materials; LSP, lightning strike protection.

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1. Introduction

Among the intrinsically conducting polymers, polyaniline is one of the most widely studied because it combines ease of synthesis, low cost, stability, conductivity and a unique protonic doping process. Two major limitations of polyaniline are difficulties regarding its processing since the polymer is insoluble in its doped form in common organic solvents and its mechanical properties are poor.

The interest for composites or blends of PANI with common polymers started from the early 1990's with the discovery of solution processability of polyaniline. The continuously growing interest in the study of PANI polymer blends and composites over the years is driven by the need to replace traditional inorganic conductive fillers (metallic particles, carbon black) and to improve the mechanical properties and processability of PANI. The preparation method and properties of PANI blends and composites with organic polymers have been the subject of an excellent review by Pud et al. in 2003 [1], with a focus on polymer thermoplastic matrices materials. To the best of our knowledge, there is still no review on PANI blends and composites with thermoset polymer matrices. A thermosetting resin is a prepolymer in a soft solid or viscous state that changes irreversibly into an infusible, insoluble polymer network by curing. Curing is generally induced by the action of heat or a suitable radiation (UV, electron beam, etc.). Reactive combinations that cure at room temperature can be used in two-package thermoset systems, in which one package contains a resin with one of the reactive groups and the second package contains the component with the other reactive group or a catalyst for the reaction. The packages are mixed shortly before use. For thermoset coatings, several approaches have been developed to permit cure at room temperature. For example, by using a cross-linking reaction that requires an atmospheric component as a catalyst or reactant (such as oxygen or water vapor) or by using a volatile inhibitor that works when the coating is stored in a closed container but volatilizes after application as a thin film, permitting the cure to proceed. For the preparation of PANI thermoset, polyaniline (or aniline, when in situ polymerization method is used), is always added in the prepolymer stage, i.e. before curing. In other words, curing of the thermoset polymer matrix, is always the last step in the preparation of PANI thermoset composites. There are basically two approaches for the synthesis of PANI thermoset blends (1) *blending* methods which correspond to the mixing of a previously prepared PANI with the matrix polymer and (2) in-situ *polymerization* methods which correspond to the chemical in situ polymerization of aniline in the matrix polymer. Homogeneous dispersion of PANI in the polymer matrix is a prerequisite for obtaining a material with high conductivity and low percolation threshold. Using one of the two methods above, many efforts have been made to increase the compatibility of PANI with thermoset matrices. Recently, the design of PANI thermoset ternary composites has attracted much attention. The introduction of an organic component or inorganic filler into the thermoset matrix together with PANI can provide matrix with specific properties (magnetic [2,3], thermal sensitivity [4], etc.), decrease the percolation threshold by serving as a bio [5,6] - or hard [7] template or enhance mechanical [8,9], thermal [10] or barrier [11] properties. This review covers the design principles being applied to synthesize binary and ternary PANI thermoset composites. In addition, some of the emerging applications of these materials will be outlined. Chemically crosslinked hydrogels are covalently connected network which swell but do not dissolve in water. Smart conductive hydrogels have received increasing attention in recent years [12–14] but they will not be discussed further as the fundamentals and recent advances in electroconductive hydrogels design and applications have been already reviewed [15–17].

2. Preparation methods of binary blends and composites

2.1. Basic aspects of the physical blending method

PANI decomposes before melting and it is insoluble in common solvents. PANI is thus generally considered an intractable material. Different synthetic strategies are used to assist dispersion of the conductive polymer in the matrix polymer.

The most typical and simple method to prepare PANI thermoset is to blend PANI with polymer matrix in solutions. One way to improve the dispersion of PANI into polymer matrix is to include substituents (*N*- or ring substituents) on the polymer backbone of PANI that make it more soluble and/or sterically hinder interchain interactions. However, the chain torsion generally causes significant decreases in conductivity ($<10^{-5}$ S/cm for the *N*-alkylated PANIs [18]) and the ring-substituted PANIs have poor stability towards hydrolysis [19]. This is certainly why only a few papers have been published reporting the preparation of ring-substituted PANI thermoset composites. Copoly(*o*-toluidine/*o*-ethylaniline)-*p*-toluenesulfonate (POTOEPTS) were incorporated into a photocurable system consisting of poly(urethane-acrylate) resin [20]. The different components were dispersed in DMF. The authors noted that POTOEPTS dissolves in DMF while PANI-TSA does not dissolve to an appreciable degree. The absence of a percolation region for PU loaded with POTOEPTS was explained by the molecular dispersion of PANI into the PU matrix. The literature also reports the formation of poly(*ortho*-toluidine)/nano-ZnO/epoxy coating but the conductivity values of the composites are not given [11]. Alkoxy derivative of polyaniline, poly(ortho-anisidine), POA have also been recently used for the preparation of PANI thermoset coatings [21]. TA–DBSA doped POA nanofibers with high conductivity (~2.1 S/cm) were synthesized

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