

Research paper

Effect of clay modifier on morphology, thermal properties and flammability of newly synthesized poly(sulfide–sulfone–amide)



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ABSTRACT

A novel polyamide bearing sulfide and sulfone units was synthesized through a direct polycondensation reaction between 2,2'-(phenylmethylene)bis(sulfanediy)diacetic acid and 4,4'-diaminodiphenylsulfone. The resulting poly(sulfide–sulfone–amide) (PSA) exhibited high solubility in organic solvents such as tetrahydrofuran. A series of nanocomposites based on the synthesized functional polyamide loaded with a reactive organoclay containing triazine ring (Mt-Traz) and also with Cloisite 15A (Mt-15A) as reinforcing agents were prepared through a solution technique. Flammability and thermal stability of the neat PSA and corresponding nanocomposites were studied by microscale combustion calorimetry (MCC) and thermogravimetric analysis (TGA) techniques. Thermal properties of the nanocomposites were strongly influenced by the nature of the modifiers. Use of Mt-Traz led to a more improved flame retardancy and higher thermal stability as compared to Mt-15A. According to the MCC data, by introducing 5 mass% of Mt-Traz into PSA matrix a reduction of about 25% in the peak heat release rate and 47% in total heat release values was observed, while incorporation of Mt-15A led to a reduction of about 13% and 27% in those values.

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1. Introduction

The synthesis of ordered high performance clay polymer nanocomposites (CPNs) is of high interest because the interactions between components can significantly improve the properties such as mechanical and barrier properties, thermal stability and flame retardancy of polymers (Pavlidou and Paspaspyrides, 2008). In the preparation of CPNs, the dispersion of clay layers in a polymer matrix is important for good performance of nanocomposites. The affinity between polymer matrix and organoclay is one of the most important factors in achieving good dispersion. To a certain extent, the affinity can be enhanced by optimizing the structure of the organoclay for a given polymer matrix (Kim and Guymon, 2011; Shabanian et al., 2014a). The chemical modification process leads to a better interaction between organoclay layers and polymer matrix and improves its properties. Clays can be modified by the interlayer ion exchange with surfactant cations and/or the covalent bond formation involving the functional edge-hydroxyls (Acosta et al., 2003; Leone et al., 2010; Shabanian et al., 2014b). Fornes et al. (2002)

investigated the properties of Nylon 6/montmorillonite by using different organic modifiers. They found that the surfactant structure affects the morphology and properties of nanocomposites. The modification of clay by triazine compounds, having inherent excellent flame retardancy, can significantly improve thermal and fire properties of clay polymer nanocomposites (Gilman, 1999).

Aromatic polyamides are classified as high-performance polymers with unique properties such as high conductivity, chemical and solvent resistance, thermooxidative stability, outstanding flame resistance, low dielectric constant, retained properties at very high temperatures and superior thermal resistance (Caouther et al., 2007; Fink, 2008; Zulfiqar et al., 2008; Shabanian et al., 2013b, 2014a). Aromatic polyamides are finding increasing demands as useful replacements for metals or ceramics, or even as new materials in new technological applications (Cassidy, 1980; Fan et al., 2002; Chang and Liou, 2008; García et al., 2010; Shabanian et al., 2014c). However, poor solubility in common organic solvents and their extremely high transition temperatures are the limiting factors for processing and application of these materials. Recent basic and applied research has focused on improving their processability and solubility to widen the scope of the technological applications of these materials (Shabanian and Basaki, 2013). It seems that insertion of sulfide in the polymer structure enhances the processability and

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solubility of the final polymer besides improving some other properties (Kondo et al., 1982). Since SO_2 is a polarized bond and easily interacts with other functional groups, the polymer with a sulfone bond is expected to show improved solubility and miscibility with other polymers (Shockravi et al., 2006; Shabanian et al., 2013a; Shao et al., 2013). As a structurally modified aromatic polyamide, poly(sulfide–sulfone–amide) (PSA) possesses the desirable dimensional stability and chemical resistance of polysulfides and polysulfones as well as the high strength of polyamide (PA). PSA is soluble in some polar solvents due to its improved solubility and higher chemical resistance compared to PA. The superior characteristics of PSA make it a valuable candidate for possible applications in polymer electrolyte membranes, composite bipolar plates, conductive materials, surface mounted devices, vehicle sensors and so on (Xia et al., 2008; Barique et al., 2009; Yang et al., 2011; Javadi et al., 2013).

In order to synthesize novel high performance materials with improved processability and solubility with respect to aromatic polyamide, PSA nanocomposites were prepared through a solution intercalation method using two different organoclays namely Mt-15A and Mt-Traz. The effects of nanofiller type on morphology, thermal properties and flammability of PSA were explored in details.

2. Experimental

2.1. Materials

Benzaldehyde, thioglycolic acid, L-proline, 4,4'-diamino diphenylsulfone, N-methyl-2-pyrrolidone (NMP), tetrahydrofuran (THF), palladium charcoal, hydrazine hydrate, pyridine, 2,4-diamino-6-phenyl-1,3,5-triazine, methanol and triphenyl phosphite (TPP) (all purchased from Merck) were used without further purification. Commercially available calcium chloride (CaCl_2 , Aldrich) was dried under vacuum at 150°C for 6 h. Cloisite® 15A and Cloisite Na^+ (Mt-Na) (Southern Clay Products, Inc.) were used without further treatment.

2.2. Preparation of the nanocomposites

2.2.1. Synthesis of 2,2'-(phenylmethylene)bis(sulfanediyl)diacetic

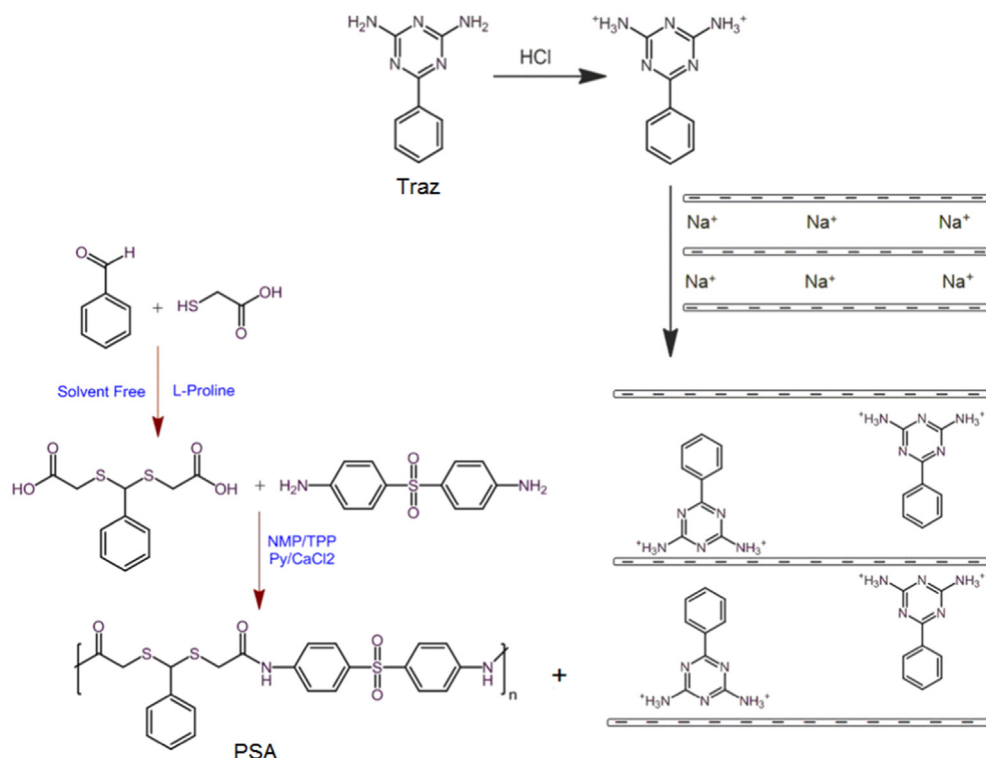
2,2'-(Phenylmethylene)bis(sulfanediyl)diacetic was synthesized according to the procedure reported by Ebrahimi and Sayadi (2012).

2.2.2. Synthesis of PSA

The PSA sample was prepared under already optimized conditions as follows: in a 50 mL round-bottom flask fitted with a stirring bar, 2,2'-(phenylmethylene)bis(sulfanediyl)diacetic (4.0 g, 14.7 mmol), 4,4'-diamino diphenylsulfone (3.6 g, 14.7 mmol), calcium chloride (0.5 g, 4.5 mmol), triphenyl phosphite (8.7 mL, 28 mmol), pyridine (1 mL) and N-methyl-2-pyrrolidone (9 mL) were mixed and heated at 120°C in an oil bath for 10 h. Subsequently, the reaction mixture was poured into 200 mL methanol. The precipitated product was collected by filtration and rinsed thoroughly with hot methanol. Finally, the product was dried at 70°C for 10 h inside a vacuum oven (Yield = 93%) (Scheme 1).

2.2.3. Preparation of Mt-Traz

The surface modification of montmorillonite was carried out by a cation exchange reaction between Mt-Na and an ammonium salt of 2,4-diamino-6-phenyl-1,3,5-triazine to yield Mt-Traz (Scheme 1). 5 g Mt-Na was suspended in 700 mL distilled water at room temperature and stirred overnight. In another flask, 2 g 2,4-diamino-6-phenyl-1,3,5-triazine was dissolved in 40 mL distilled water under magnetic stirring and the pH was adjusted to 3–4 using 1.0 M HCl aqueous solution. After stirring for 3 h, the second solution was added to the Mt-Na suspension at a rate of approximately 15 mL min^{-1} while vigorously stirring. The mixture was stirred overnight at room temperature. The precipitates were filtered using a Buchner funnel and washed at least four times to remove any excess ammonium ions. The product was



Scheme 1. Synthesis route of PSA and its nanocomposites.

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