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Research paper

Organoclay maleated natural rubber nanocomposite. Prediction of abrasion and mechanical properties by artificial neural network and adaptive neuro-fuzzy inference

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ABSTRACT

Maleated natural rubber (MNR) was produced and employed in different amounts as compatibilizer in preparation of organoclay/natural rubber (OC/NR) nanocomposite. Oscillating disk rheometry, scanning electron microscopy (SEM), X-ray diffraction (XRD) analysis, rheomechanical spectroscopy, dynamic mechanical analysis, tensile and abrasion tests were used to evaluate the prepared samples. The obtained results showed that the use of MNR caused an increase in the maximum torque of rheometry, curing rate index and crosslink density. XRD analysis indicated that using of MNR, especially in the amount of 8 parts, resulted in a greater intercalation of OC layers. SEM showed a good dispersion of the OC as well as higher interfacial interaction between NR molecules and OC. These lead to improvement of modulus, tensile strength and abrasion resistance. Then the obtained results were modeled by artificial intelligence techniques. Comparison made between experimental and predicted values, indicated that the modeling results were in good agreement with the corresponding experimental amounts.

1. Introduction

In recent years, great attention has been given to the nanoparticle/polymer nanocomposites which are a new class of materials wherein particles having at least one dimension lower than 100 nm are dispersed. In this regard, using different nano scale particles such as organoclay (OC), expanded graphite (George and Bhowmick, 2008; Mirzazadeh et al., 2011), silica (Bandyopadhyay et al., 2006; Rattanasom et al., 2007), carbon nanotubes (Das et al., 2008), calcium carbonate (Wang et al., 2011) and graphene (Hu et al., 2010) in polymeric matrices and their effects on the improvement of some properties of polymers were reported by the researchers. Among these nano particles, nanoclay due to its high aspect ratio, high specific surface area (SSA), and easier accessibility was used more commonly than the other ones (Alexandre and Dubois, 2000; Bandyopadhyay et al., 2006; Garces et al., 2000; Karger-Kocsis and Wu, 2004; Kato et al., 2011; Khanlari and Kokabi, 2011; Kooshki and Jalali-Arani, 2009; Maiti et al., 2008; Mirzazadeh and Katbab, 2006; Mohammadpour and Katbab, 2007; Monfared et al., 2014; Okada and Usuki, 2006; Sengupta et al., 2007; Sinha Ray and Bousmina, 2005; Sinha Ray and Okamoto, 2003; Utracki et al., 2007).

Clay is "a naturally occurring material composed primarily of fine-grained minerals, which is generally plastic at appropriate water contents and will harden when dried or fired." Therefore, natural clay will consist of a/several clay minerals mixed with additional minerals as impurities (Bergaya and Lagaly, 2013; Bergaya et al., 2011). The hydrophilic clay mineral needs to be modified with amphiphilic compensating cations to improve its compatibility with the hydrocarbon matrices. Therefore the clays are modified with various alkylammonium salts to expand the interlayer space and also reduce surface energy of the layers (Bergaya et al., 2011). Even distribution and dispersion is achieved thanks to the organophilic cations, the interaction of the OC with the hydrocarbon polymer chain can be faint indeed. In this respect a polar group on the chains can be helpful and can be accessed by using a compatibilizer (Carli et al., 2011; Carretero-González et al., 2008; Das et al., 2008; Doğan et al., 2011; Hakim and Ismail, 2009a; Maji et al., 2010; Mirzazadeh et al., 2011; Sahakaro and Beraheng, 2008; Stephen et al., 2006; Tavakoli et al., 2011; Teh et al., 2004, 2006; Wang et al., 2011).

Natural rubber (NR) is an elastomeric polymer which exhibits high strength as a gum vulcanizate. NR can be considered as one of the most important polymers and is widely used in preparation of rubber products. To improve its mechanical properties, reinforcing fillers such as carbon black and silica are commonly added to it (Rattanasom et al., 2007). In particular, the incorporation of nanoparticles such as OC leads to a significant improvement in the mechanical properties of

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the NR composites (Ali et al., 2010; Carli et al., 2011; Khanlari and Kokabi, 2011). Since NR is known as a hydrophobic substance, therefore the main problem with using OC to reinforce it lies in the incompatibility between them. The interfacial adhesion between OC and NR can be improved by various methods (Ali et al., 2010; George and Bhowmick, 2008; Hu et al., 2010). For example some researchers (Arroyo et al., 2007; Doğan et al., 2011; Teh et al., 2004) used the epoxidized natural rubber (ENR) as a compatibilizer to enhance the adhesion between NR and OC. Moreover Hakim and Ismail (2009a) compared the ENR with the NR as the matrix used in preparation of OC/polymer nanocomposites (CPN). They used montmorillonite intercalated with octadecylamine as the OC. According to their report, OC-filled ENR nanocomposites showed better mechanical properties than the NR ones, because of homogenous dispersion of the silicate layers in them, which was attributed to the reaction of epoxy group in the ENR with amine functionality in the OC structure.

Teh et al. (2004) used ENR50 and ENR25 as the compatibilizers in preparation of OC/NR nanocomposites and obtained a higher improvement of properties by using ENR50 in this nanocomposite.

Tavakoli et al. (2011) used maleic anhydride grafted ethylene propylene diene monomer rubber (EPDM-g-MA) and ENR as the compatibilizers for the preparation of OC/NR nanocomposites. Their obtained results showed that EPDM-g-MA was a better interfacial compatibilizer compared to ENR, and the NR compatibilized with this compatibilizer compared with the NR compatibilized with the ENR50, showed a greater thermo-dynamical affinity to diffuse into the OC interlayer spaces.

Maleic anhydride grafted natural rubber (MNR), was also found to be an effective compatibilizer (Hakim and Ismail, 2009b) because the NR segment of this compatibilizer is miscible with NR matrix and its maleic anhydride segments may form hydrogen bonds with the hydroxyl groups of the OC.

On the other hand, because of considerable improvements in using the computational calculations such as artificial intelligence techniques (artificial neural network (ANN) and adaptive neuro-fuzzy inference system (ANFIS)), some researchers have tried to employ these two techniques to predict the essential parameters which are hard, time consuming and costly to measure. For example, Zhang et al. (2003) employed the ANN method to predict the erosive wear of three polymers; polyethylene (PE), polyurethane (PUR) and an epoxy modified by hygrothermally decomposed polyurethane (EP-PUR).

Khajeh and Modarress (2010) applied the ANFIS method to predict flash point of the common esters by using the quantitative structure property relationship (QSPR) principle. By considering the rubber content, rubber particle size, duration and temperature of mixing as the inputs of ANN and then performing statistical analysis, Specht et al. (2007) predicted the viscosity of the asphalt-rubber binders. They compared the results with the experimental data and reported the better performance of ANN than the statistical analysis. Fazilat et al. (2012) predicted thermal degradation kinetics (TDK) of nylon6 (NY6)/feather keratin (FK) blend films by using the artificial intelligence techniques including artificial neural network, adaptive-neuro-fuzzy-interference system and radial basis function. Kok et al. (2010) modeled the complex modulus of the base and styrene–butadiene–styrene (SBS) modified bitumens by using ANN and concluded that the ANN was a suitable mathematical method to predict the complex modulus. In a similar research Yilmaz et al. (2011) modeled and predicted the complex modulus of the base and ethylene-vinyl-acetate modified bitumen by applying ANFIS as the modeling tool. ANN method was also used in our previous work to predict physical-mechanical properties of the base and polymer-modified bitumen (Golzar et al., 2012).

This work was divided into two main parts including experimental and mathematical analysis. In the first part, OC/NR nanocomposites were prepared by melt-mixing method. Different amounts of MNR produced by melt-mixing method were used as the interfacial compatibilizer. Then morphology, curing characteristics and crosslink density, mechanical and abrasion properties, dynamic mechanical and rheological behavior of the compounds in relation to their structure were investigated. In the second part of the current survey, by employing the experimental data obtained in the first part, the characteristics and properties of the prepared OC/NR nanocomposites were modeled by using artificial intelligent techniques such as artificial neural network (ANN) and adaptive neuro-fuzzy inference system (ANFIS). Moreover the entire modeled results were compared with the experimental obtained data. Remember that according to our investigation on published papers, using mathematical methods to model the preparing process and properties of OC/rubber nanocomposites has not been reported yet.

2. Experimental

2.1. Materials

NR (SMR GP grade) with a Mooney viscosity of ML (1 + 4) 100 °C = 61 was obtained from Malaysia. Organically modified montmorillonite (OC) was purchased from Southern Clay Products (Gonzales, TX, USA) under the trade name of cloisite 15A. This OC is modified with dimethyl dihydrogenated tallow quaternary ammonium with a concentration of 125 mequiv/100 g of clay. Maleic anhydride (MA) and dicumyl peroxide (DCP) were used to prepare maleic anhydride grafted natural rubber (MNR). Sulfur was supplied by Tesodak (Iran), ZnO by Sanaye Rangineh Pars (Iran), stearic acid by Unichema, TBBS (N-tert-butyl-2-benzothiazole sulfenamide) by Bayer Company and analytical-grade of toluene (Merck) was used as the solvent.

2.2. Grafting procedure

To prepare MNR, NR was mixed with maleic anhydride (12 phr) in an internal mixer (Brabender Plasticator, Germany) at temperature of 140 °C and rotor speed of 60 rpm for 12 min using two different procedures: a) in presence of 2 phr dicumyl peroxide (DCP) as an initiator and b) in the absence of DCP. It was expected that the heating and shearing applied during melt-mixing led to formation of free radicals on NR chains and thereby NR could react with maleic anhydride. The used method and recipes were similar in some aspects to those proposed by (Nakason et al., 2006).

2.3. Preparation of OC/NR nanocomposites

The compound recipes are listed in Table 1. To prepare OC/NR nanocomposites, the compound ingredients, except the curing materials, were mixed in the internal mixer at temperature of 100 °C and a rotor speed of 60 rpm. For this purpose, natural rubber was first fed into the mixer and after 3 min, OC was added to it. Finally compatibilizer was incorporated and mixing was continued for 15 min (as the overall time). Curing agent and curatives were added into the uncured compound on a two-roll mill (PM300 model, Germany) at temperature of 40–50 °C.

Table 1
Composition of prepared compounds.

No.	Ingredients	NR	OC/NR	OC/NR/M4	OC/NR/M8	OC/NR/M12
1	Natural rubber[a]	100	100	100	100	100
2	OC	0	5	5	5	5
3	MNR[b]	0	0	4[c]	8	12
4	Zinc oxide	5	5	5	5	5
5	Sulfur + Acc[d]	3.2	3.2	3.2	3.2	3.2
6	Stearic acid	2	2	2	2	2

[a] Total amounts of Natural rubber and existed Natural rubber in MNR is equal to 100.
[b] Maleated natural rubber.
[c] In modeling section the MA content of the used MNR was determined and applied as one of the inputs.
[d] N-tert-butyl-2-benzothiazolesulfenamide.