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ScienceDirect
Chinese Chemical Letters 20 (2009) 877–880
CHINESE CHEMICAL LETTERS
www.elsevier.com/locate/cclet

Study of the structure and the mechanical properties of dynamically cured PP/MAH-g-SEBS/epoxy blends

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Received 27 October 2008

Abstract

A new method concerning with the simultaneous reinforcing and toughening of polypropylene (PP) was reported. Dynamical cure of the epoxy resin was successfully applied in the PP/maleic anhydride-grafted styrene–ethylene–butylene–styrene (SEBS) triblock copolymer, and the obtained blends named as dynamically cured PP/MAH-g-SEBS/epoxy blends**.** The stiffness and toughness of the blends are in a good balance, and MAH-g-SEBS was acted as not only an impact modifier but also a compatibilizer. The structure of the dynamically cured PP/MAH-g-SEBS/epoxy blends is the embedding of the epoxy particles by the MAH-g-SEBS.

Keywords: Dynamical cure; Epoxy resin; PP; MAH-g-SEBS

1. Introduction

Polypropylene (PP) has an important thermoplastic among the synthetic polymers because of its growing commercial applications. However, its application as a structural material is somewhat limited because of its relatively low impact resistance. The impact toughness of PP can be improved dramatically by the addition of elastomers such as ethylene–propylene rubber (EPR) [1], ethylene–propylene diene elastomer (EPDM) [2,3], ethylene–octene copolymer (POE) [4], styrene–ethylene–butylene–styrene (SEBS) triblock copolymer [5] and poly(ethylene vinyl acetate) (EVA) [6]. But, the incorporation of elastomers into PP leads to a drastic reduction in the modulus and strength, Therefore, issues concerning with the simultaneous reinforcing and toughening of PP have attracted considerable attention [7,8]. A functional group such as maleic anhydride (MAH) is commonly grafted to PP to enhance the interfacial adhesion of the composites. MAH grafted styrene–ethylene–butylene–styrene (SEBS) triblock copolymer has been used increasingly to improve the compatibility and toughness of phase components of the composites and polymer blends [9,10]. We have applied dynamical vulcanization to prepare a new type of PP/epoxy blends [11,12]. Dynamical cure of the epoxy resin led to an improvement in the modulus and stiffness of the PP/epoxy blends compared with pure PP.

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1001-8417/$ – see front matter
doi:10.1016/j.cclet.2008.12.054

In this paper, the dynamical cure of the epoxy resin in the PP/MAH-g-SEBS blends gives rise to the increases in the toughness and stiffness of the blends, and the obtained blends named as dynamically cured PP/MAH-g-SEBS/epoxy blends. The toughness and stiffness of the dynamically cured PP/MAH-g-SEBS/epoxy blends are in a good balance.

2. Experimental

Dynamically cured PP/MAH-g-SEBS/epoxy blends were prepared in the mixing chamber of a Haake Rheometer RC90 at 190 °C and 50 rpm. MAH-g-SEBS (MAH content of 1%, Shanghai Sunny New Technology Development Co., Ltd.) and PP (F401, a melt flow rate (MFR) of 1.9 g/10 min (230 °C, 2.16 kg), Yangzi Petrochemical Co., Ltd.) were first mixed for 2 min, then the epoxy resin (DGEBA, M_w: 4500 g/mol, Shanghai Resin Co., Ltd.) was added. 2 min later, the curing reagent was added with the mixing continuing. The total mixing process lasted about 10 min. The composition was moved out and compression-molded in a press at 195 °C for 10 min, then cold pressed to give samples for testing.

The torque of the samples was measured using of a Haake Rheometer RC90 at 190 °C and 50 rpm. The tensile properties were measured using an Instron 4465 Tester at a crosshead speed of 20 mm/min according to ASTM D638. Notched Izod impact strengths were tested using a Ray-Ran Universal Pendulum Impact Tester at an impacting pendulum speed of 3.5 m/s according to ASTM D256. Flexural properties were measured using the Instron 4465 Tester. A three-point-loading rig and the central head were loaded on the specimen at a speed of 1.7 mm/min according to ASTM D790. In each of the tests, at least five samples were tested, and the average results were reported.

A scanning electron microscopy (SEM, HITACHI-S-2150) was used to examine the morphology of the samples. All samples were fractured in liquid nitrogen. The samples of the PP/MAH-g-SEBS, PP/MAH-g-SEBS/epoxy and dynamically cured PP/MAH-g-SEBS/epoxy blends were etched by toluene at 40 °C for 10 min, and then covered with a thin gold layer for further observation.

3. Results and discussion

Torque measurements can be used to give qualitative information concerning the chemical reactivity and the extent of reaction in the reactively compatibilized blends [13]. Fig. 1 gives the relationship between torque and time for the PP/MAH-g-SEBS (75/25) and PP/MAH-g-SEBS/epoxy (75/25/10) blends with or without the curing reagent (EMI-2,4) at 190 °C. The addition of the epoxy resin into the PP/MAH-g-SEBS (75/25) blend results in a decrease in the torque, which is attributed to the lower viscosity of the epoxy resin compared to MAH-g-SEBS. It can be seen that the addition of the curing reagent at the mixing time of 4 min leads to an obvious rise of the torque of the PP/MAH-g-SEBS/epoxy (75/25/10) blend. This indicates that the epoxy resin has been cured during mixing with the molten PP and MAH-g-SEBS, and the obtained blends are named as dynamically cured PP/MAH-g-SEBS/epoxy blends.

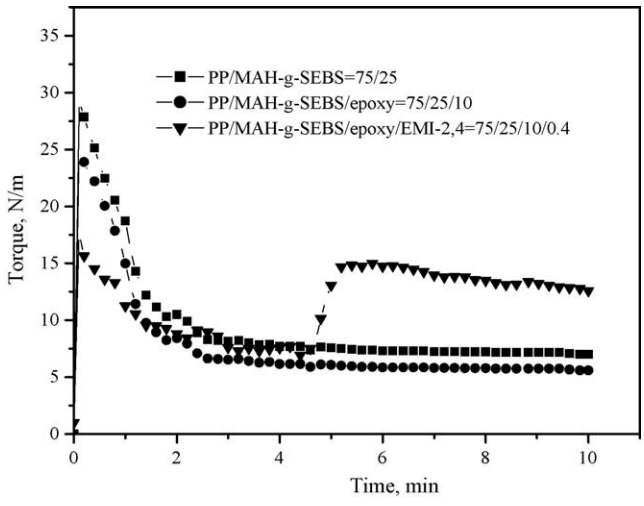

Fig. 1. Plot of torque versus time for the PP/MAH-g-SEBS and PP/MAH-g-SEBS/epoxy blends at 190 °C.

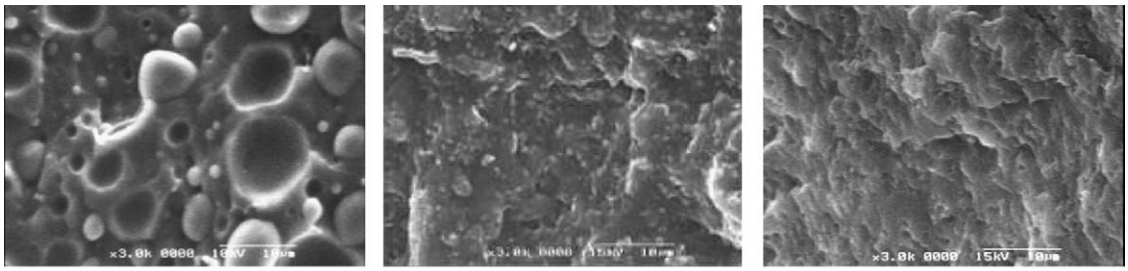

Fig. 2. SEM photographs of different blends. (a) PP/epoxy = 75/10, (b) PP/MAH-g-SEBS/epoxy = 75/25/10, (c) dynamically cured PP/MAH-g-SEBS/epoxy = 75/25/10/0.4 (etched).

Fig. 2(a) and (b) shows that SEM micrographs of PP/epoxy (75/10) and PP/MAH-g-SEBS/epoxy (75/25/10) blend, respectively. For the PP/epoxy (75/10) blend (Fig. 2(a)), the epoxy resin is dispersed as spherical particles with an average diameter of 2–6 μm in the PP matrix. But, fine epoxy particles with the average diameter of about 0.3–0.8 μm are distributed in the PP matrix (Fig. 2(b)). It shows that MAH-g-SEBS can improve the compatibility between PP and epoxy resin. The maleic anhydride groups of MAH-g-SEBS can react with the hydroxyl or epoxy groups of the epoxy resin to form a graft copolymer to act as a compatibilizer for promoting a fine dispersion of epoxy resin phase.

Fig. 2(c) shows the etched SEM micrographs of dynamically cured PP/MAH-g-SEBS/epoxy (75/25/10) blend, a few smaller epoxy particles with diameters of about 0.20 μm and a number of smaller holes representing etched MAH-

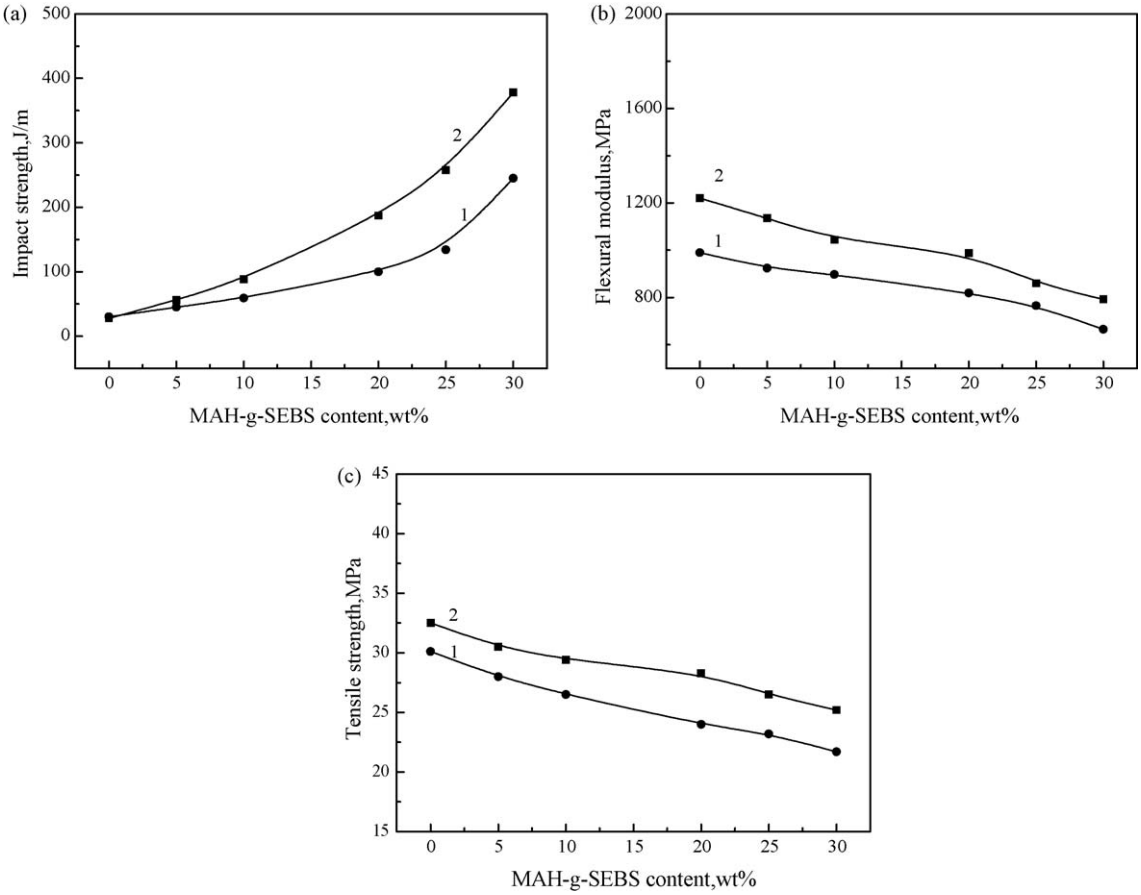

Fig. 3. Effect of MAH-g-SEBS content on the mechanical properties of: (1) PP/MAH-g-SEBS and (2) dynamically cured PP/MAH-g-SEBS/epoxy blends (formulation: (PP+ MAH-g-SEBS) 100, epoxy resin 10, EMI-2,4 0.4, MAH-g-SEBS variable).

g-SEBS were observed in the blend. It shows that the structure of the dynamically cured PP/MAH-g-SEBS/epoxy blends is the embedding of the epoxy particles by the MAH-g-SEBS. The epoxy particles could be embedded in MAH-g-SEBS, and form the structure of core–shell complex phase and PP continuous phase in the blend. Similar structures of three-component polymer/elastomer/fillers have been reported by other authors [14].

Fig. 3(a)–(c) shows the effect of MAH-g-SEBS content on the mechanical properties of the PP/MAH-g-SEBS and dynamically cured PP/MAH-g-SEBS/epoxy blends, respectively. When the MAH-g-SEBS is added into the PP matrix, the impact strength increases greatly with the increase of the MAH-g-SEBS content. But, with the increase of the MAH-g-SEBS content, the tensile strength and flexural modulus decrease. For the dynamically cured PP/MAH-g-SEBS/epoxy (75/25/10) blend, when 10 wt% epoxy resin was dynamical cured in the PP/MAH-g-SEBS blends, the impact strength is 264 J/m at the MAH-g-SEBS content of 25 wt%, increases by 100% compared to PP/MAH-g-SEBS (75/25) blend.

In the condition of the same content of the MAH-g-SEBS, the tensile strength, impact strength and flexural modulus of dynamically cured PP/MAH-g-SEBS/epoxy blends are higher than that of the PP/MAH-g-SEBS blends (shown in Fig. 3). The results show that the dynamical cure of the epoxy resin in the PP/MAH-g-SEBS blends improves not only the stiffness of the blends, but also their toughness. The stiffness and toughness of the dynamically cured PP/MAH-g-SEBS/epoxy blends are in a good balance. We thought that the cured epoxy particles as organic filler increases the stiffness of the PP/MAH-g-SEBS blends, and the improvement in the toughness is attributed to the embedded structure in the dynamically cured PP/MAH-g-SEBS/epoxy blends. Faulkner [15] studied the impact behavior of PP/mica/elastomer composites and tried to optimize stiffness and toughness, he thought that the embedded structure had a better impact resistance.

In conclusion, an obvious rise of the torques of the PP/MAH-g-SEBS/epoxy blends by the addition of the curing reagent shows the epoxy resin was dynamical cured in the PP/MAH-g-SEBS blends, and the obtained blends named as dynamically cured PP/MAH-g-SEBS/epoxy blends**.** SEM analysis shows that the dynamically cured PP/MAH-g-SEBS/epoxy blends with the structure of the embedding of the epoxy particles by the MAH-g-SEBS. The cured epoxy particles in the PP/MAH-g-SEBS blends improve not only the stiffness of the blends, but also the toughness. The stiffness and toughness of dynamically cured PP/MAH-g-SEBS/epoxy blends are in a good balance.

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