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Cure mechanism of novel bismaleimide resins based on fluorene cardo moiety and their thermal properties

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ABSTRACT

In this paper, two novel bismaleimide resins based on 9, 9-bis[4-(4-maleimidophenoxy) phenyl] fluorene (PFBMI), 9, 9-bis[4-(4-maleimidophenoxy)-3-methylphenyl]fluorene (MFBMI), and 2, 2′-diallyl bisphenol A (DABPA) were prepared. Their curing mechanism and curing kinetic were carefully investigated by Differential scanning calorimetry (DSC) and Fourier transform infrared spectroscopy (FTIR). The thermal mechanical properties of the composites based on these BMI resins and the glass cloth were obtained by Dynamic mechanical analysis (DMA), displaying that the novel resins whose T_g were 296°C and 289°C had excellent thermal performance. In addition, Thermogravimetric analysis (TGA) results showed that both the cured PD and MD resins possessed good thermal stability, and their $T_{5\%}$ were all higher than 410°C.

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novel bismaleimide resins; fluorene cardo structure; cure mechanism; kinetic; thermal properties

1. Introduction

Bismaleimide resin (BMI) as a high performance thermosetting polymer possessed many excellent properties. It not only had high temperature resistance, radiation resistance, heat resistance, high modulus, low moisture absorption rate and low thermal expansion coefficient similarly to polyimide resin, but also had the same easy process ability as epoxy resin (EP), meeting the requirements of advanced polymer matrix composites in many ways (1–4). So far, BMI had become the best matrix resin for high performance resin, and replaced EP as the dominant matrix material for aerospace structural composites gradually (5). However, the unmodified BMI generally possess many disadvantages such as, high melting point and poor solubility of the monomer, high crosslinking density, brittle, poor impact resistance and crack resistance of the cured resin. These disadvantages had greatly limited their applications (6–8).

Extensive research had been undertaken for enhancing the toughness of BMIs. Methods included the copolymerization of two or more BMI monomers (9–11), the copolymerization of BMI monomer and other reactive monomer (12–18) or electron enrichment (19, 20). The regularity of the crystal structure of BMI molecule was destroyed by these above-mentioned copolymerization, leading to the disorder of the BMI crystal structure and reducing the interaction force and the crystallization ability between the two BMI molecules. Thereby, the crosslink density of the cured BMI resins was reduced and their processing performance was improved. Hitherto, 2, 2′-diallyl bisphenol A (DABPA) had been shown to be the most promising modifier for toughening BMI. It copolymerized with BMI monomer to form linear chain extension by Ene reaction and further crosslinked by Diels-Alder reaction at high temperature (21). After being modified by DABPA, the BMI resins offered easy processability, and the toughness of the cured resin was improved obviously with only minimum reductions in the thermal properties.

In previous research, two novel chain-extended BMI monomers containing fluorene cardo structure and ether bond (PFBMI and MPBMI) were designed and synthesized by our group (22). These monomers had lower melting point, broad thermal processing window, good solubility in low boiling point organic solvent and excellent heat resistance. Nevertheless, their high melt viscosity caused by the huge fluorene cardo moiety disfavored the preparation of good-quality resins. In order to solve this problem of manufacturing and obtain high performance bismaleimide resin, DABPA was selected to copolymerize with PFBMI and MFBMI for preparing two novel bismaleimide resins (PD and MD). The mechanism of curing and the kinetic for the novel resins were carefully investigated by nonisothermal DSC and FTIR. The thermal properties of the cured resins were characterized by DMA and TGA, and their water absorption behavior were also discussed.

2. Experimental

2.1. Materials

Commercially available 2,2′-diallyl bisphenol A (DABPA) was provided by Honghu City Shuangma Advanced Materials Tech Co., LTD. (Hubei, China). The glass fabric was supplied by Anhui Danfeng Group and had treated using KH-550. 9,9-bis [4-(4-maleimidophenoxy) phenyl] fluorene (PFBMI), 9,9-bis [4-(4-maleimidophenoxy)-3-methylphenyl]fluorene (MFBMI) were synthesized according to the reported method (22).

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