REACTIVE TELECHELIC POLYETHERIMIDE TOUGHENED TETRAFUNCTIONAL EPOXY

Hengxi Chen^{1*}, Devendra Bajaj², Dadasaheb Patil², Nikhil Verghese³, and Hung-Jue Sue¹

¹ Department of Materials Science and Engineering, Texas A&M University, College Station, TX, USA ² SABIC's Specialties Business, Technology and Innovation, Mt. Vernon, IN ³ SABIC, Corporate, Technology and Innovation, Houston, TX * Presenting Author email: hengxichen@tamu.edu

Abstract

Highly crosslinked multifunctional epoxy resins possess superior properties, like high Tg, modulus, chemical resistance, etc, yet they are brittle due to their high crosslink density. It is desired that toughness improvements should not compromise other properties, including mechanical, thermal properties, and processability. Here, reactive polyetherimide containing amine functional end groups with two different molecular weights and loading levels were incorporated in tetraglycidyl diamino diphenyl methane tetrafunctional epoxy resin to study their structure-property relationship.

1. Introduction

Epoxies are extensively used as structural adhesives, protective coatings, electronic encapsulants, and composite matrices for various engineering applications. Due to their highly crosslinked network structure, they are known to be brittle. Therefore, efforts to decrease the brittleness of epoxies through introducing toughening agents have been pursued.

Toughening in tetrafunctional epoxies, such as tetraglycidyl diamino diphenyl methane (TGDDM), becomes rather difficult due to the fact that the shear banding mechanism in highly crosslinked epoxy network is significantly restricted. Therefore, rubber-toughening has been shown to be ineffective in improving the fracture toughness of epoxy matrices. Incorporation of rubber particles also reduces the mechanical and thermal properties.

Thermoplastic toughening through particle bridging process, is not affected by the ductility of the polymer matrix. The particle bridging effect depends on the particle size, the inter-particle distance. the amount of the particles introduced, and the interfacial adhesion between the particle and the matrix. In this work, reactive polyetherimide (rPEI) with -NH₂ end-group functionality was utilized as a toughening agent to toughen tetrafunctional TGDDM epoxy. The relationship between phase morphology and mechanical behavior of rPEI-toughened TGDDM were investigated. The effects of rPEI functional group and molecular weight (MW) on structure-property relationship is established for potential use in aerospace and other structural applications.

2. Results

Different phase morphologies (particulate, sea-island, and phase inverted,) based on low and high MW (LMW and HMW) of rPEI-modified TGDDM blends were observed (Fig.1a). The thermomechanical behavior and network architecture of rPEI-modified TGDDM systems were probed via dynamic mechanical analysis (DMA), Fig. 1b. It is found that rPEI-modified TGDDM systems can retain the T_g and G' to those of the neat TGDDM. The tan δ curves show evidence of a raised shoulder hump indicating a partial phase separation morphology (Fig.1b), which is consistent with our TEM and SEM observations. For mechanical properties, rPEI-modified TGDDM show a 30% increase in tensile strength and 100% increase in elongation at break, while the Young's modulus decreased by about 15%.

To investigate the toughening mechanisms, the double-notch four point bend (DN-4PB) technique was employed to examine the subcritical crack tip regions of the epoxy blends. In TGDDM/PEI blend, it is evident that pullout of PEI particles occurred due to the poor interfacial adhesion, evident from crack propagation around the PEI domains. While in the TGDDM/rPEI systems, the crack propagated through the rPEI-rich phase with evidence of crack bridging mechanism (Fig. 1c). The mode-I fracture toughness

is increased by as high as 140% through addition of 30 wt% rPEI with a sea-island morphology, while both high modulus and high T_g of the epoxy is well maintained.

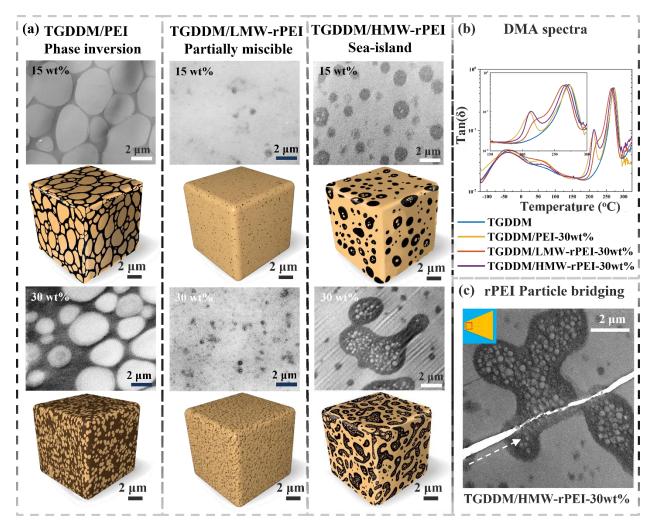


Fig.1 – (a) The phase morphologies of TGDDM/PEI and TGDDM/rPEI blends based on different MWs. (b) DMA spectra of TGDDM, PEI and rPEI modified TGDDM blends. (c) TEM micrograph showing possible crack bridging mechanism at the crack wake region of TGDDM/HMW-rPEI-30% blend.

3. Conclusions

It is evident that high performance thermosets based on TGDDM epoxy can be significantly toughened using telechelic rPEI with $-NH_2$ end-group functionality. The $-NH_2$ functionality on PEI telechelics can participate in the epoxy crosslinking process to achieve better interfacial adhesion over its unfunctionalized counterparts. Different MW of rPEI can also influence miscibility and phase morphology in epoxy. Ductile drawing of rPEI phase via crack bridging mechanism found in TGDDM/rPEI are responsible for the observed toughening effect.