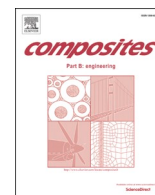




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Synthesis of urethane-modified aliphatic epoxy using a greenhouse gas for epoxy composites with tunable properties: Toughened polymer, elastomer, and pressure-sensitive adhesive

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ABSTRACT

Epoxy, which exhibits tunable mechanical properties, has been investigated to overcome its inherent brittleness and extend its application range. Herein, various epoxy composites, including a toughened polymer, elastomer, and pressure-sensitive adhesive (PSA), with tunable mechanical properties were prepared by adjusting the ratio of a rigid aromatic epoxy and flexible aliphatic epoxy. A flexible aliphatic epoxy that incorporated urethane linkages was used to extend the tuning range; it was synthesized via an eco-friendly route utilizing CO₂ without isocyanate. The prepared toughened polymer-like and elastomeric epoxy composites can be used in structural epoxy adhesive (lap shear strength = 27.8 MPa) and flexible electronics (plastic strain < 10%), as confirmed through single lap shear, cyclic tensile, and Izod impact tests, respectively. The most flexible among the prepared epoxy composites exhibited PSA properties (peel strength = 6.8 N·cm⁻¹) that were comparable to those of conventional tapes. A viscoelastic window also supported that the prepared PSA can be utilized as a general-purpose PSA and a removable PSA. This work proposes a new application of eco-friendly epoxies and a strategy to facilitate the wide tuning range of epoxy materials, including PSAs.

1. Introduction

Epoxy resins are some of the most useful polymers and exhibit excellent properties, including mechanical strength, chemical resistance, heat resistance, and bonding capability [1–7]. These resins are utilized in various forms such as coatings, composite matrices, and adhesives but have limitations including inherent brittleness and poor elongation [1,4]. To overcome the limitations, several strategies have been employed to improve elongation with minimal loss of strength, i.e., toughening of the epoxy, using reactive additives (e.g., rubber-modified epoxy, epoxy-functionalized particles, and epoxy-terminated resin) [8–16], and non-reactive additives (e.g., nanoparticles, elastomers, and block copolymers) [17–25]. In some cases, both elongation and strength are freely adjusted at the same time to prepare versatile epoxies. Until now, the mechanical properties of versatile

epoxies have been tuned between brittleness and elastomerism, which enabled the epoxy to be utilized in various applications [26–31].

Epoxies exhibiting tunable mechanical properties are generally prepared via the following methods: 1) employing an epoxy with different functionalities [26,27], 2) varying the stoichiometry between the epoxy and hardener [28,29], and 3) changing the ratio of rigid moiety to flexible moiety [30,31]. The key strategy for achieving tunable properties is to control the ratio between the flexibility and rigidity of epoxies, which is prevalent in all of the aforementioned methods. The fraction of rigidity increases upon incorporating high-functionality epoxy or aromatic epoxy resin, e.g., bisphenol A diglycidyl ether (DGEBA), which makes the epoxy more brittle. Meanwhile, the flexibility fraction increases upon employing mono-functional epoxy or aliphatic epoxy resin, which grants an elastomeric nature to an epoxy.

Aliphatic epoxy resins, e.g., poly(propylene glycol) diglycidyl ether

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