Microcapsule induced toughening in a self-healing polymer composite

E. N. BROWN
Department of Theoretical and Applied Mechanics and the Beckman Institute for Advanced Science and Technology, University of Illinois at Urbana-Champaign, Urbana, IL 61801, USA; Los Alamos National Laboratory, MS-E544, Los Alamos, NM 87545, USA
E-mail: en_brown@lanl.gov

S. R. WHITE
Department of Aerospace Engineering and the Beckman Institute for Advanced Science and Technology, University of Illinois at Urbana-Champaign, 306 Talbot Laboratory, 104 S. Wright St., Urbana, IL 61801, USA
E-mail: swhite@uiuc.edu

N. R. SOTTOS
Department of Theoretical and Applied Mechanics and the Beckman Institute for Advanced Science and Technology, University of Illinois at Urbana-Champaign, 216 Talbot Laboratory, 104 S. Wright St., Urbana, IL 61801, USA
E-mail: n-sottos@uiuc.edu

Microencapsulated dicyclopentadiene (DCPD) healing agent and Grubbs' Ru catalyst are incorporated into an epoxy matrix to produce a polymer composite capable of self-healing. The fracture toughness and healing efficiency of this composite are measured using a tapered double-cantilever beam (TDCB) specimen. Both the virgin and healed fracture toughness depend strongly on the size and concentration of microcapsules added to the epoxy. Fracture of the neat epoxy is brittle, exhibiting a mirror fracture surface. Addition of DCPD-filled urea-formaldehyde (UF) microcapsules yields up to 127% increase in fracture toughness and induces a change in the fracture plane morphology to hackle markings. The fracture toughness of epoxy with embedded microcapsules is much greater than epoxy samples with similar concentrations of silica microspheres or solid UF polymer particles. The increased toughening associated with fluid-filled microcapsules is attributed to increased hackle markings as well as subsurface microcracking not observed for solid particle fillers. Overall the embedded microcapsules provide two independent effects: the increase in virgin fracture toughness from general toughening and the ability to self-heal the virgin fracture event. © *2004 Kluwer Academic Publishers*

1. Introduction

A novel approach to recover the fracture properties of thermosetting polymers has been introduced by White *et al*. [1]. Healing is achieved by incorporating a microencapsulated healing agent and a catalytic chemical trigger within a polymer matrix. A propagating crack ruptures the microcapsules and exposes catalyst particles. Crack opening draws the healing agent into the crack plane, where contact with the catalyst phase initiates polymerization. The polymerized healing agent reestablishes structural integrity across the crack plane.

Conclusive demonstration of self-healing was obtained with a healing agent based on the ring-opening metathesis polymerization (ROMP) reaction [1]. Dicyclopentadiene (DCPD), a highly stable monomer with excellent shelf life, was encapsulated in urea-formaldehyde (UF) microcapsules. A small volume fraction of microcapsules was dispersed in an epoxy matrix along with Grubbs' transition metal catalyst [2]. This self-healing epoxy was able to recover over 90% of its virgin fracture toughness [3]. In addition to providing an efficient mechanism for self-healing, the presence of DCPD-filled UF microcapsules also significantly increased the inherent fracture toughness of the epoxy.

In the present work, we investigate toughening mechanisms induced by embedded microcapsules in a self-healing epoxy and the corresponding effect on healing efficiency. Several investigations of microcapsule-filled polymers have appeared in the literature using alternate terms such as hollow spheres [4], cenospheres [5], microballons [6, 7], hollow particles [8, 9], hollow microspheres [8, 10], and bubbles [11]. Most of the literature examines the behavior of glass (rather than polymer) microcapsules, with mixed conclusions regarding the effect on fracture behavior. Azimi *et al*. [4]

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