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Development of high-performance resin nanocomposites by resin cellulation using multi-walled carbon nanotubes

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ABSTRACT

This work aims to enhance resin reinforcing performance and thermal resistance through the innovative use of multi-walled carbon nanotubes (MWCNTs) for advanced material applications. The new method, which relied on MWCNT disentanglement under high shear stress, produced resin matrix nanocomposites. The MWCNT/resin nanocomposites showed improved stiffness without significant flexibility loss and did not flow above 160 °C. These dramatic improvements may result from the formation of a three-dimensional structure at the MWCNT/resin interface.

1. Introduction

Carbon nanotubes (CNTs) were synthesized by catalytic vapour growth for the first time in 1976 [1,2] but were only analysed in 1991 [3]. In addition to numerous synthetic processes exploiting various vacuum technologies, their unique properties have attracted considerable attention for the development of advanced materials.

Initial polymer-based composite materials using CNTs as fillers involved epoxy resin [4–18], polypropylene [19–34], polycarbonate [35–38] and polyamide [39–42]. These CNT fillers have remarkably enhanced polymer electrical properties [4,5,14,25–28], thermal conductivity [6,29,42,43], and crystallinity [30–32,39,40]. They have also boosted resin mechanical characteristics [4,7–18,21,22,27–34,36–38,40–42]. However, these mechanical improvements have been predominantly restricted to CNT filling rates lower than 10 vol% and, thus, remain limited compared to electrical and thermal conductivity enhancements. The rarity of high-filled CNT/polymer composites stems from existing challenges in generating uniform dispersions at high CNT concentrations. Despite the existence of low-filled composites featuring improved mechanical properties, few studies have achieved CNT dispersions at the macro-nanoscale [30,32,37,41,42] instead of the nanoscale. In addition, the composites exhibited rather poor flexibility, even at filling concentrations of wt% [4,9,17,33,40], which indicates that uniform CNT dispersions remain problematic.

Attempts at generating uniform dispersions include CNT surface chemical treatment [10,11,16,19,34], masterbatch application [22,29] and mechanical treatment by biaxial milling using high shear force [8,11,18,20,21]. However, chemical treatments encounter problems regarding their industrial applicability, such as post-processing liquid-waste disposal under strict conditions. Biaxial milling using a high shear force produces CNT clumps at the microscale. Therefore, neither of these methods provides uniform dispersions at the nanoscale, suggesting their inability to facilitate the exploitation of CNTs as nanofillers.

An elasto-milling method utilizing the polarity and free radical effect in addition to the elastomer-specific viscosity and elasticity has been developed to process rubber and elastomer materials. This method produced a composite in which multiwalled carbon nanotubes (MWCNTs) were disentangled in an elastomer matrix, resulting in excellent thermal and mechanical properties [44–46].

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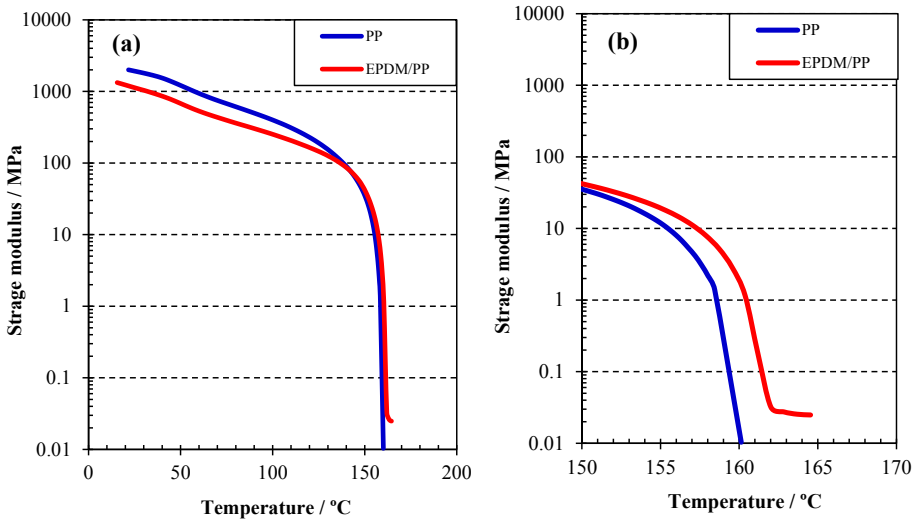

Fig. 1. Storage elastic moduli of PP and EPDM/PP as a function of temperature. (a) Full temperature range and (b) 150–170 °C range.

This disentanglement relies on a cellulation mechanism, in which MWCNTs and matrix robustly form a three-dimensional nano-continuous structure [45–47]. This cellulation technology has generated several composites and has found application in an increasing number of areas [48–50].

The elasto-milling process was developed to enhance elasticity and shear force at low temperature, resulting in extremely high-performance rubber reinforced with MWCNTs [48]. An effective process hinges on several matrix-related requirements. (1) Elastomer molecules forming the matrix need to fill the voids created by the physically intermingled nanotubes, effectively breaking their inherent agglomeration during mixing. (2) The matrix has to exhibit good 'wettability' with carbon nanotubes and (3) its elasticity facilitates kneading. However, many resins do not meet this third processing condition because of their lack of elasticity.

Because of its dependence on elastomer-specific elasticity, the elasto-milling method has proven difficult to apply to resins. Nonetheless, this problem was addressed by blending a resin with an elastomer to reveal an elastic region. In addition, a elasto-milling method that facilitates MWCNT disentanglement in a resin matrix was developed. Along with a description of this resin-specific method, the physical properties of the obtained MWCNT/resin composites and resin cellulation were evaluated.

2. Experimental

2.1. Materials

Polypropylene (PP, Admer QE800) and ethylene propylene rubber (EPDM, Tafmer MH7020) were purchased from Mitsui

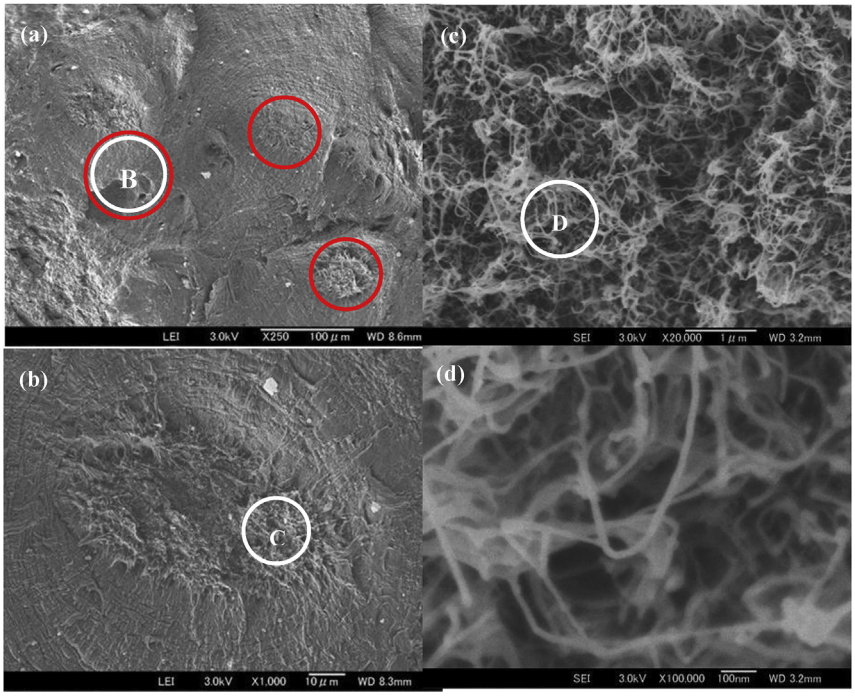

Fig. 2. Cross-sectional SEM images of 10 wt% Ref: (a) Full image, (b) magnification of part B, (c) magnification of part C, and (d) magnification of part D.