



Study of electric heating effects on carbon nanotube polymer composites



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ABSTRACT

Carbon nanotube (CNT) composites are ideal materials for electric heating (E-heating) components owing to their lightweight, rapid heating ability, and easy processability for microscale patterns. However, careful consideration of their thermal stability and the effect of E-heating on CNT composites is necessary to ensure safety and long-term durability. Thermal degradation of CNT composites as a result of E-heating was found to be much more severe than air convection heating (C-heating). Enhanced mechanical properties of CNT composite were observed after 40 h C-heating, while E-heating composites became fragile that the matrix was severely damaged at the same temperature/hours. Consequently, the electrical resistance of the CNT composite for E-heating was dramatically increased. To avoid thermal degradation and a dramatic increase in resistance, the E-heating temperature should be much lower than the temperature limit of thermally stable polymer. In addition, thermal degradation of the polymer matrix could be relieved through the addition of CNTs, which can be understood as parallel resistor model, by spreading thermal concentration throughout the polymer matrix.

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1. Introduction

Carbon-based polymer composites have been extensively investigated in various applications such as electromagnetic interference shielding, biosensor, reinforcement units, thermal management, strain sensor, and high-charge storage capacitors owing to their light weight and processability [1–6]. In particular, they are a practical choice of material in heat-related applications such as patternable micro heaters, temperature sensors, heating glasses for vehicles, thermoelectric devices, water heaters, and flexible deicing units [7–9]. Through electric joule heating (resistive heating) of conducting composites, electrical energy can be converted into heat energy easily and quantitatively. In general, composites that exhibit high conductivity and

low heat capacity are ideal for rapid heating. While carbon-based materials such as carbon black and fibers have previously been used in conducting polymer composites [10], there is a limit to the impregnation of polymers with traditional filler materials due to degradation of the composite's mechanical integrity as the amount of filler is increased. Carbon nanotubes (CNTs) are therefore more attractive candidates for filler materials in composites, primarily due to their high aspect ratio coupled with electrical and thermal conductivities, which could be as high as 10^6 S/m and 6600 W/m K [11–13]. These properties enable electrical percolation with very small volumes of nanotubes [14]. In the case of CNT composites, the small thermal capacitance of the composite and the high thermal conductivity of the CNTs enable rapid heating, which is a remarkable advantage in light-weight, flexible, and printable electric-heating applications. Consequently, several researchers have examined the electric-heating behavior of CNT devices or CNT composites for sensor and

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heating-unit applications [15,16]. However, several challenges to the utilization of CNT–polymer composites remain, such as (a) thermal and chemical reliability and (b) aggregation and bundling of CNTs, which lead to non-uniform dispersion [17,18]. For example, CNTs synthesized with cobalt catalysts were found to severely decrease the thermal stability of silicone-based CNT composites, which is a critical flaw in high temperature environments [17]. In addition, while various studies on transparent thin-film heaters using CNTs, graphene, and nanowires have been conducted [19–21], there have been few studies on the relationship between electric joule heating and thermal degradation in a composite structure consisting of a polymer matrix and a conducting filler, which is a crucial design consideration for practical applications.

Unlike transparent CNTs or graphene films, each CNT in the composite is embedded in the polymer matrix, with potentially dramatic impact on the thermal degradation of the polymer during electric heating. For example, the prepared CNT networks were firmly fixed in polymer matrix, as shown Fig. 1(a), resulting in electrical pathways with multiple contact points. However, electric heating (E-heating) could initiate thermal degradation of the polymer matrix around the nanotubes surfaces and junctions, causing many void defects, partial disconnections between the nanotube junctions, and displacement of CNTs owing to the lack of a matrix support, as shown Fig. 1(c). Here, we present our analysis on the thermal degradation of CNT composites as a result of long-term E-heating which is much more severe than thermal degradation caused by long-term convection heating (C-heating) for long-term stability and optimal operating conditions of the heating

elements. The fabrication of composites comprising highly conducting CNTs and polydimethylsiloxane (PDMS) is presented with the aim of applying these composites in heating units. We discovered that the CNT composites after 40 h E-heating were seriously damaged or grew fragile, while they showed enhanced mechanical properties after C-heating possibly due to further chemical reactions in the polymer at the same temperature. Thermal degradation of the polymer matrix could be prevented through the addition of nanotubes, which can be understood as parallel resistor model, by spreading thermal concentration. Various analyses were conducted to determine the difference between the effects of long-term E-heating and C-heating on the matrix. Using the isothermal mode of a thermogravimetric analyzer (TGA), we discovered that the thermal degradation effect of E-heating at 200 °C was similar to the effect of C-heating at 350 °C. These results are expected to lead to useful design guidelines for the use of nanotube composites in the fabrication of heating elements.

2. Experimental

For fabrication of CNT composites, elastomer PDMS (DOW Corning, silicone elastomer base) was selected as a polymer matrix due to easy processability (e.g., patternable, flexible, etc.) and bio-compatibility. Non-functionalized multi-walled carbon nanotubes (MWNTs) with an outer diameter of 10 nm and a length of 10 μm were purchased from Nanocyl. In spite of superiority of single-walled carbon nanotubes (SWNTs) for thermal stability

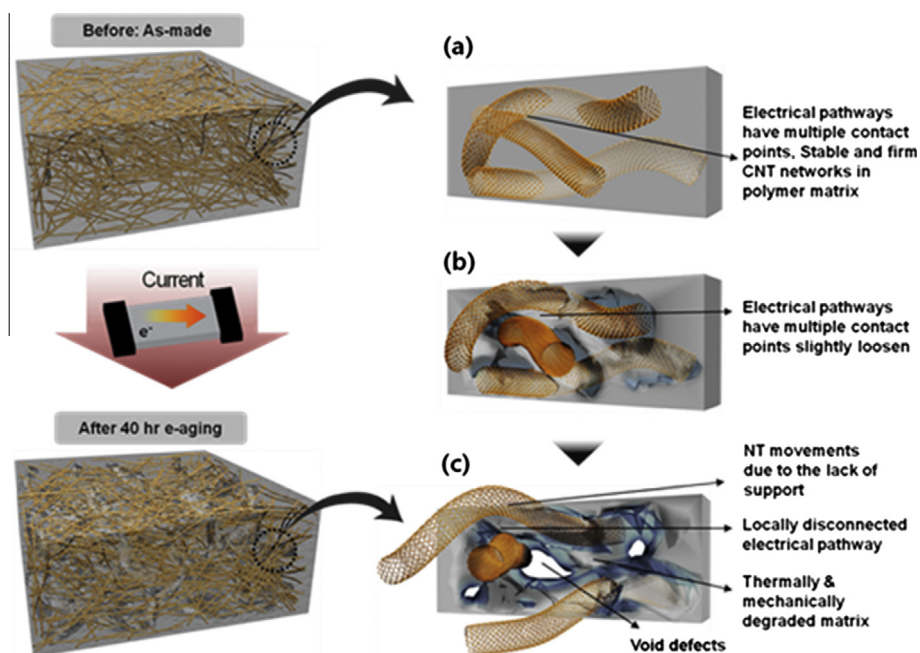


Fig. 1. A scheme of thermal degradation of a CNT composite during electric joule heating. (a) In an as-prepared CNT composite, which had numerous electrical pathways with multiple contact points, the CNT networks were firmly fixed in the polymer matrix. (b) However, during 40 h E-heating, thermal degradation of the polymer matrix around the nanotubes surfaces and junctions could be initiated. (c) Then, many void defects, a thermally degraded matrix, and disconnection of the electrical contacts were found in the E-heating sample. The long-term E-heating sample showed a significant increase in resistance as a result of thermal degradation.

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