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Influence of strain rate on the piezoresistive behavior of conductive polyamide composites

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ABSTRACT

Due to the matrix viscosity, polyamide composites take on the obvious strain rate sensitivity that affects their conductivities during compression. In this paper, the influence of strain rate on the piezoresistive behavior of polyamide filled by stainless steel fibers and carbon nanotubes (SSFs/CNTs/PA6) is investigated. Based on experimental observations under compression, the resistivity of SSFs/CNTs/PA6 composites is dominated by two kinds of competing mechanisms: the spacing decrease between conductive fillers induced by compressive strain and the spacing increase due to micro-damage. A piezoresistivity model is proposed, which is coupled with the strain rate effect of SSFs/CNTs/PA6 composites. It is shown that there is a power-law relationship between the tunneling barrier and strain rate.

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1. Introduction

Conductive polymer composites have been developed with multiple functionality, excellent performance and high intelligence. They are usually based on polymeric matrix filled by stainless steel fibers (SSFs) and carbon nanotubes (CNTs). Due to their piezoresistive effects (i.e., electrical properties are sensitive to pressure), conductive polymer composites have had many applications in electromagnetic shielding devices, pressure sensors, microelectrodes and electro-conductive rubbers [\[1,2\].](#page--1-0)

To explain the conductive mechanism of conductive polymers, McLachlan et al. [\[3\]](#page--1-0) introduced a general equation that combines percolation and an effective medium theory. As for their sensitivity to pressure, the stress-strain behaviors of conductive polymer composites have been widely studied. For example, Hussain et al. [\[4\]](#page--1-0) discovered that there is an obvious piezoresistive behavior in rubber-based pressure sensitive materials filled by conductive carbon fillers. The strain sensitivities of CNT/polycarbonate and multi-walled CNT/polyvinylidenefluoride nanocomposites were experimentally observed under tension [\[5,6\]](#page--1-0). Qu et al. [\[7\]](#page--1-0) investigated the piezoresistive properties of expanded graphitereinforced polypropylene modified by maleic anhydride. It is shown that the percolation threshold is as low as 0.1 vol% in CNTreinforced composites and 0.4 wt% in graphite-reinforced polymers [\[7,8\],](#page--1-0) and under compression (with stress less than 10 MPa), the electrical resistivity rapidly decreases with the increase of stress, and then reaches a steady state [\[7\]](#page--1-0). On the basis of experiments, a statistical model based on the Weibull distribution was introduced to explain the changing trend of electrical resistance in carbon fiber polymer-matrix composites, where three stages of the electrical resistance versus tensile strain were identified [\[9\].](#page--1-0) However, due to the viscous properties of polymer matrix, deformation of composites is time-related. Wang et al. [\[10\]](#page--1-0) studied the piezoresistive effect and resistivity creep of carbon black-filled polymers. The electrical and dynamic mechanical responses of CNT/E-glass/epoxy cycloaliphatic amine composites were explored by using the Hopkinson bar [\[11\].](#page--1-0) The conductivity of carbon blackbased polymer composites was investigated under creep in the molten state [\[12\]](#page--1-0), and it was found out that the mechanical deformation of conductive composites during melting considerably affects their final electrical properties.

In addition, mechanical properties of conductive polymers at different strain rates or under ramp and creep conditions were

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Table 1

Specimens with different filler contents in SSFs/CNTs/PA6 composites.

Specimens	Filler contents (wt%)	
	CNTs	SSFs
$CO - 10$	0	10
$CO - 12$	U	12
$C1 - 10$		10
$C1 - 12$		12
$C3 - 10$	ς	10
$C3 - 12$	κ	12

experimentally studied [\[13,14\]](#page--1-0). By using molecular dynamics simulations, stress-strain curves were generated on a single-walled CNT embedded in polyethylene [\[15\].](#page--1-0) Based on the studies of PA6/ elastomer composites, Huang et al. [\[16\]](#page--1-0) proposed a micromechanical model to predict the elasto-plastic response under uniaxial tensile loading.

Although numerous advances have been made on creep, piezoresistive, and strain sensitive behaviors of conductive polymers, there are few reports on the time-dependent piezoresistivities, especially on their strain rate effects. In this paper, the effect of strain rate is investigated on the conductivity of SSFs/CNTs/PA6 conductive polymers under compression. Based on the strainresistivity behaviors at different strain rates, the controlling mechanism of resistivity in polymer composites is revealed, and a resistivity model coupled with the strain rate is proposed.

2. Experimental

2.1. Material and specimens

SSFs/CNTs/PA6 conductive composites were prepared by meltblending and injection molding. Conductive fillers and PA6 were mixed in a plastic refining (10D6 mixer) machine at 260 \degree C for 10 min and then injected into a mould to form a plate with the thickness of 3 mm. PA6 (with a density of 1.1 g cm⁻³, melting

temperature of 230 \degree C, and elongation of 35%) was obtained from Nanjing Dellon Plastics Alloy Co. Ltd. The SSFs (with the diameter of 12 mm and length of 5 mm) were supplied by Hunan Huitong New Material Co. Ltd. Multi-walled CNTs from Chengdu Organic Chemistry Co. Ltd have an average dimension of 30 nm in diameter and $10-30$ um in length. The specimens were prepared with the CNT fractions of 0, 1.0 and 3.0 wt%, and the SSF fractions of 10.0 and 12.0 wt%. These specimens as listed in Table 1 were cut into cylinders with 10 mm in diameter for mechanical and electrical tests.

The scanning electron microscope micrographs of composites with different SSF contents are shown in Fig. 1. It is clearly observed that SSFs are in good dispersion throughout the matrix with less agglomeration (see Fig. $1(a)$ and (b)). The fillers constitute a conductive network in the specimen, and thus its conductivity is relatively stable. These well-bonded SSFs with matrix can be seen in Fig. 1(c), which ensures an effective transmission of stress. The random distribution of CNTs in PA6 matrix is shown in Fig. 1(d).

[Fig. 2](#page--1-0) shows the dependence of direct current electrical resistivity on the mass fraction of SSFs and CNTs at 25 \degree C [\[17\]](#page--1-0). The resistivity rapidly decreases with increasing the content of SSFs. The electrical percolation threshold of composites without CNTs is between 5 and 7 wt%, and the value with 3 wt% CNTs is between 2 and 4 wt%. Obviously, due to the addition of CNTs, the percolation threshold of SSFs decreases. However, it is worth noting that the filler contents selected here are high enough to ensure a stable conductivity.

2.2. Measure of electrical resistivity

As shown in [Fig. 3\(](#page--1-0)a), specimens were subjected to compression by using the material test system (MTS 810) with strain rates of 10^{-2} , 10^{-1} , 1, and 10 s⁻¹. The circuit diagram is illustrated in [Fig. 3\(](#page--1-0)b). During tests, the upper and lower indenters were kept insulated, and the two parallel surfaces of a specimen were coated with conductive silver paste acting as electrodes. The oscilloscope (TDS3014C, Tektronix) was used to record voltage in the specimen.

Fig. 1. Scanning electron microscope micrographs of SSFs/CNTs/PA6 composites: (a) the distribution of SSFs in the specimen C1-10, (b) the distribution of SSFs in C1-12, (c) wellbonded SSFs with matrix and the size comparison of SSFs and CNTs in C1-10, and (d) the random distribution of CNTs in C1-10.

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