Epoxy/MWCNT Nanocomposites as Thermal Sensors and Actuators

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1. Introduction

Besides the improvement of the mechanical properties and of the enhancement of the electrical conductivity, the addition of carbon nanotubes (CNTs) to organic films is also strongly improving the thermal conductivity [1]. The combination of good thermal and electrical conductivity makes them an attractive material for electrical heating applications.
Furthermore the same material, used as electrical heater, can be used for sensing purposes, because the electrical conductance of CNTs is temperature dependent. Positive and negative temperature coefficients have been reported, depending on the electronic bandstructure of the material (semiconducting or metallic). The situation is more complex in carbon-nanotube-networks (CNN), where often the contact resistance between nanotubes or the resistance between the metallic contacts- and the nanotubes is dominating the overall conductance.
Another important question, regarding temperature sensing and actuating applications is of course the thermal stability. In the case of polyethylene/CNT nanocomposites, we observed a rather complex behaviour with a conductivity increase when remaining below the glass transition temperature, but a irreversible loss of conductivity when exceeding this temperature by more than 30°C [2]. In literature, we find a temperature sensor based on carbon nanotubes, where the polymer layer is just used as an encapsulant [3].
Here we report on the temperature sensing and heating properties of an Epoxy/CNT composite, working stable over a wide temperature range.

2. Experimental

2.1 Material preparation

In this work, a diglycidyl ether of bisphenol A-epoxy resin (DGEBA) and 4,4’ diamine-dibenzyl-sulfone hardener (DDS) both supplied by Sigma-Aldrich Chemicals, were used. The Multiwalled Carbon Nanotubes (MWNTs) were obtained from Nanocyl S.A., synthesized by catalytic carbon vapor deposition (CCVD) process and consisted of 95 vol% carbon.
Regarding the production of the composites, 0.5wt% of carbon nanotubes were sonically dispersed for 20 min in the liquid epoxy resin before curing with DDS. The composite was then used to completely fill a rectangular mold cavity of 100x10x2 mm in dimensions.
The cure reaction was carried out in an oven with a standard temperature profile. It consists of a ramp from room temperature to 150°C at the heating rate of 10 °C/min, then a 60 min of isothermal stage at this temperature followed by a second heating ramp up to 220°C and finally an isothermal stage at 220°C for 180 min.

2.2 Sample geometry and measurement setup

Coplanar electrical contacts have been prepared by the evaporation of 100nm thick gold contacts on the top of the Epoxy/MWCNT samples.
The electrical characteristics have been measured using a KEITHLEY 2400 source-measurement-unit. Perfect ohmic behaviour has been observed for a wide range of voltages. For voltages exceeding 50V a superlinear current-voltage characteristics has been observed. This can be explained by the negative temperature coefficient (NTC) behaviour and the self-heating of the samples for high applied voltages.
This self-heating has been measured by determining the sample surface temperature with a FLUKE 576 pyranometer.

3. Results and Discussion

In order to determine the self-heating effect, we applied to the sample a series of voltage step-functions with an initial low voltage period at 1V, a successive high voltage period at either 85V, 140V and 170V, and finally a third period again at 1V applied voltage. During the first period, the stability of the electrical conductivity in the absence of self-heating has been tested, during the second period the sample has been heated and during the third period the sensing properties of the composite sample have been tested during the slow cooling.
In Fig. 1 the resulting current and the measured surface temperature are shown, as an example during the experiment with a high voltage during the second period of 140V.

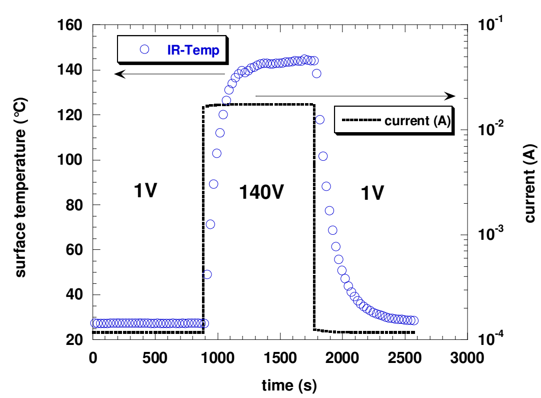

Fig. 1: Current and surface temperature, as measured on a Epoxy/MWCNT composite sample during a voltage cycle, with the successive application of 1V, 140V and again 1V .

We observe a constant surface temperature and current during the first period, a strong temperature rise up to a saturation value of 145°C during the high voltage period and a slowly decreasing temperature during the last period with 1V applied voltage.
During the high temperature and the third period the resulting current is not constant, which is, however, not easily seen on the logarithmic scale over three orders of magnitude in Figure 1. Therefore we show in Figure 2 the surface temperature and the measured current again, but now on a linear scale and limited to the third period, where the composite acts only as sensor without influencing the sample temperature.

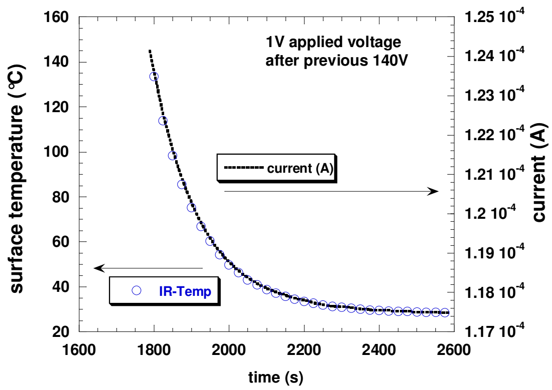

Fig. 2: Current and surface temperature, as measured on a Epoxy/MWCNT composite sample during cooling with an applied voltage of 1V, after a previous heating period, when applying 140V .

It can be seen, that the current and the surface temperature decay monotonically in the same manner over a wide temperature range between room temperature and 140°C. These results confirm the possibility to use the composite sample not only as heating element, but also as linear temperature sensor.

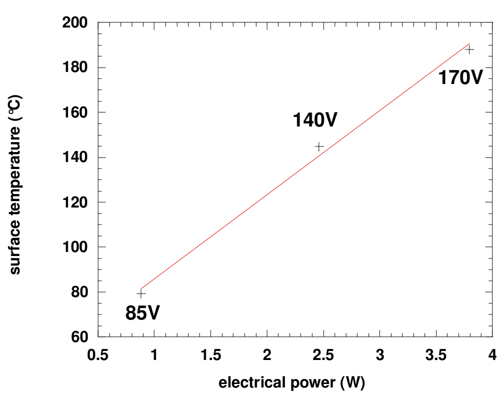

Fig. 3: Relationship between electrical power and surface temperature, as measured on a Epoxy/MWCNT composite sample for three values of the applied voltage.

It should be mentioned, that a linear relation between current and surface temperature has been observed also during the third period after applying 85V and 170V before.

In Figure 3 we see the relation between the applied electrical power and the resulting saturation value of the surface temperature for the different applied voltages. It can be seen that a wide range of temperatures up to 190°C can be covered and that also the relation between the applied electrical power and the resulting surface temperature is almost linear.

In conclusion, a new kind of electrical temperature sensor with linear conductivity-temperature characteristics was found, that due to the high conductivity can also be used as heating element.

7. References

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