

## Carbon nanotubes-polymer nanocomposites for controlled heating materials

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**ABSTRACT:** Nanocomposites of modified carbon nanotubes (CNT) and either polyurethane (PU), poly(vinyl acetate) (PVAc), or silicone materials were synthesized and characterized for thermal mat application. The obtained results revealed that the polymer used as a matrix had an impact on the electrical resistance of the mats. The lowest results of 32  $\Omega$  of resistance was registered with silicone-based mats containing 5 wt % of CNT. For the same CNT content the mats based on PVAc and PU displayed values of 55 and 60  $\Omega$ , respectively. The low resistance properties of silicone-based materials were due principally to the good compatibility of both polymer and functionalized CNT. Because of the low resistance values, this mat was subjected to thermovision analysis revealing that the samples reached temperature of about 60 °C in 9 min and 70 °C after 27 min of 27 V of applied potential. The results showed an almost uniform temperature distribution in the samples' surface with some high and low temperature spots, which were attributed to nonuniform distribution of CNT in the polymer matrix. In summary, all the obtained results confirm that silicone-CNT are very promising materials that can be used as low-voltage heating mats. © 2016 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* **2016**, *133*, 44194.

**KEYWORDS:** composites; properties and characterization; thermal properties

Received 5 January 2016; accepted 17 July 2016

DOI: 10.1002/app.44194

### INTRODUCTION

Composite materials including those with nanostructures are developed aiming to combine properties that single materials do not have to attend modern technologies requirements.<sup>1</sup> Since early 1990, new composite materials containing carbon nanotubes (CNT) that are graphene layers rolled into hollow cylinders have been proposed.<sup>2</sup> These structures are typically single layer carbon atoms forming single-walled nanotubes (SWCN).<sup>3</sup> Because of their structure, they are considered as one-dimensional objects because the aspect of their length to diameter ratio is in the order of 100–1000.<sup>4</sup> Nanotubes have unusual properties such as very high tensile strength, unique electrical properties, and very good thermal conductivity.<sup>5</sup> With these unique characteristics, they have a high application potential. Another variety of CNT are multiwalled carbon nanotubes (MWCN) made of concentric graphene layers.<sup>4</sup> Because of the high chemical resistance their properties can be modified by breaking some of the bonds between carbon atoms.<sup>6</sup> The main challenge when using CNT is the method of dispersing or bonding them with the carrier material that may be subjected to functionalization<sup>7</sup> such as polymer

matrices.<sup>8,9</sup> Solutions based on the use of CNT as polymer fillers have been known for many years.<sup>1</sup> However, a fundamental problem is to obtain a homogenous dispersion of CNT in the polymer matrix that can result in improved physical and chemical properties of the nanocomposite. Among different applications, CNT-based materials are investigated in the field of thermal management, which focuses mainly on thermal conductivity.<sup>10</sup> Despite the differences between theoretical predictions and experimental results of thermal conductivity of polymer-CNT nanocomposites,<sup>10,11</sup> some promising results were shown for CNT uniformly mixed with silicone elastomer resulting in materials with improved thermal conductivity.<sup>12</sup> The increased values of thermal conductivity were also reported for aligned CNT composites with silicone matrix.<sup>13</sup> The electrical conductivity properties for CNT-polymer nanocomposites were reported for several CNT composites.<sup>14,15</sup> An increase of *dc* conductivity with increase of CNT content as for example from 10<sup>-5</sup> to 10 S/cm, after addition of 0.17 vol % of CNT,<sup>16</sup> and from 10<sup>-14</sup> to 10<sup>-6</sup> or 10<sup>-2</sup> S/cm depending on the CNT,<sup>17</sup> was observed in CNT-PU samples. Modified CNT-methylvinyl silicone rubber nanocomposites also

displayed an increase of electrical conductivity from  $10^{-9}$  to  $10^{-2}$  S/m after addition of 0.03 vol % fillers.<sup>18</sup> Samples of polypropylene (PP)-CNT composites with maleic anhydride grafted styrene-(ethylene-co-butylene)-styrene copolymer (MA-SEBS) as additive were prepared by Xie *et al.*<sup>9</sup> These samples revealed a decrease of resistance from  $10^{17}$  to  $10^6 \Omega$  for 0 and 10 wt % of CNT, respectively. It was also shown that even nonuniformly distributed CNTs in poly(vinyl chloride) (PVC) promoted an increase of electric conductivity from nonconducting ( $10^{-16}$  S/cm) to conducting ( $10^{-5}$  S/cm) state at already 0.05 vol %.<sup>14</sup> CNT-poly(vinyl acetate) (PVAc) composites beside slightly improved mechanical properties revealed remarkably reduced resistivity from  $2.0 \times 10^{13}$  to  $3.44 \times 10^7 \Omega \text{ cm}$  with increase of MWCN content in the samples from 0.1 and 0.5 wt %.<sup>8</sup>

As mentioned above, several papers describe the thermal and electrical conductivity properties of the polymer-CNT composites, but in our knowledge no one presented results of heating properties of such materials induced by direct current or potential application. Aiming to develop low-voltage heating mat material PVAc, polyurethane (PU), and modified silicone elastomer with MWCN were obtained and analyzed. The electric resistances of the polymer-CNT nanocomposite mats were measured, and the polymer-CNT mat with lowest resistance was chosen to microscopic and thermal analyses.

## EXPERIMENTAL

Carboxyl group modified CNT were obtained by adding 50 g of raw CNT (MWCN; 10  $\mu\text{m}$  long and 20 nm large; TCNT Taiwan), 199 mL of nitric acid ( $\text{HNO}_3$ ), and 27 mL of sulphuric acid ( $\text{H}_2\text{SO}_4$ ) in a round bottom flask that was placed in an ultrasonic bath for 12 h. After that, the flask was heated under reflux for 2 h. The resulting product was filtered on Buchner filter, washed with 2 L of distilled water, and dried.<sup>19,20</sup>

Amide modified CNT were obtained by dispersing dry soluble CNT in 1:3 distilled water/ethanol and  $\text{NH}_4\text{OH}$  under vigorous mechanical stirring. Chemicals such as  $\text{HNO}_3$ ,  $\text{H}_2\text{SO}_4$ ,  $\text{NH}_4\text{OH}$ , THF, ethanol, and polymers (PU, PVAc, and silicone) were purchased from Avantor Performance Materials Poland S.A.

CNT modified with nonpolar groups were obtained by basic esterification reaction of carboxyl group modified CNT with either *iso*-pentyl or -hexyl alcohols (Sigma-Aldrich). Alternatively, the transesterification reaction could be performed with amide modified CNT and either hexyl acetate or formate (Sigma-Aldrich). These reactions were conducted under reflux for 8 hours.

Ethyl ester modified CNT were obtained by reacting carboxyl group modified CNT with 1:3 sulfuric:nitric acid solution. The resulting product was washed with distilled water or 10:1 distilled water:ethyl alcohol solution, dried, and dispersed in ethanol with  $\text{H}_2\text{SO}_4$  as catalyst. Then, this solution was heated for 10 hours under reflux, filtered, and dried.

Amide modified CNT were mixed with commercial poly(urethane) (PU;  $M_w = 2000$  to  $6000$  g/mol; ELSTOFLEX, BASF) dissolved in tetrahydrofuran (THF). Carboxyl modified CNT were dispersed in THF/ethyl acetate (EAc) with PVAc



**Figure 1.** Image of 307 mm  $\times$  300 mm silicone-CNT mat with connected electrodes. [Color figure can be viewed in the online issue, which is available at [wileyonlinelibrary.com](http://wileyonlinelibrary.com).]

(PVAc;  $M_w = 72,000$  g/mol). Similar results were obtained with nonpolar groups modified CNT mixed with PVAc in EAc and/or acetone. Ester modified CNT were dissolved with chemically inert FD80 silicone ( $M_w = >1500$  g/mol; WACKER Silicones) enriched with ethoxy groups in either THF or THF/EAc (Sigma-Aldrich) with 1 : 1 ratio. The MWCN content was between 5 and 90 wt % of polymer matrix.

The mats were obtained by mixing CNT:polymer solution for few minutes, and then poured it on the flat and clean surface. The thicknesses of 2 mm composite mats were controlled by doctor blade method. After drying at room temperature, the resistance of the mats (Figure 1) was measured.

The electrical measurements were performed by placing the electrodes at a distance of about 30 cm each other (Figure 1). The electric properties of the samples were studied by applying 22, 24, and 27 V of *ac* potential at a frequency of 50 Hz with KIKUSUI PWR400L and measuring current flow with METEX M-3850 apparatus.

The changes of sample's temperature were captured with thermovision camera VIGOcam v50 placed on suitable support. The composites were analyzed in the range from 20  $^\circ\text{C}$  to 80  $^\circ\text{C}$ .

The surface morphology of silicone-CNT mats were studied with LEO model 440 (Cambridge, England) scanning electron microscope (SEM).

Atomic force microscopy (AFM) analyses were performed with Bruker in intermittent-contact mode by using silicon AFM probes, a constant force of 48 N/m, and a resonance frequency of 190 kHz.

Thermogravimetric analyses (TGA/DTG) were performed with TA Instruments TGA Q50 equipment in the temperature range from 25  $^\circ\text{C}$  to 900  $^\circ\text{C}$  in a nitrogen atmosphere (50 mL/min) and at a heating rate of 10  $^\circ\text{C}/\text{min}$ . DMA analyses were performed with Perkin Elmer D8000 Analyzer in the temperature range of -100  $^\circ\text{C}$  to 150  $^\circ\text{C}$ , deformation rate of 5  $^\circ\text{C}/\text{min}$  under  $\text{N}_2$  atmosphere, and frequency of 1 Hz.

**Table I.** Resistance Values of the Silicone-Based Mats Obtained Using Either THF or THF/EAC Solvents and Different CNT Contents

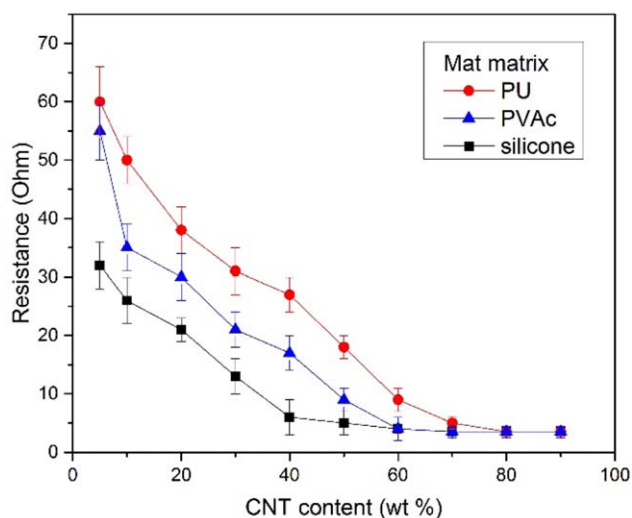
CNT content in mat (wt %)	Resistance ( $\Omega$ ) for the samples casted from solutions of	
	THF	THF/EAC
5	32 $\pm$ 4	32 $\pm$ 4
10	27 $\pm$ 3	26 $\pm$ 3
15	24 $\pm$ 2	24 $\pm$ 2

## RESULTS AND DISCUSSION

Aiming to obtain heating mat, three different composite mats composed by either PU, PVAc, or silicone and CNT were prepared and characterized. As an example of the obtained samples, Figure 1 shows the image of the silicone-CNT mat with connected electrodes. Because of the CNT, the mats were black. Moreover, they were flexible and very uniform in shape.

The silicone-based mats with 5–15 wt % of CNT were prepared using either THF or mixture of THF and EAC, and their electrical properties were measured. Table I displays the results of resistances, where one can see that almost no difference in the obtained values were found between two kinds of samples, indicating that the solvent does not influence the material electrical properties. Moreover, it was observed that the electrical resistance of the samples decreased with increase of the CNT content from 32  $\Omega$  for 5 wt % of CNT in the polymer matrix to 24  $\Omega$  for 15 wt % of CNT. Similar results of increased conductivity of the methylvinyl silicone rubber with chemically modified CNT nanocomposite were already observed and attributed to the strong interaction between modified CNT and silicone matrix.<sup>18</sup>

All the mats with 5–90 wt % of CNT:polymer composition were tested by their electrical properties. Figure 2 shows that the resistance of three samples decreases as a function of CNT

**Figure 2.** Resistance of composite mats containing silicone (■), PU (●), and PVAc (▲) with different CNT contents. [Color figure can be viewed in the online issue, which is available at [wileyonlinelibrary.com](http://wileyonlinelibrary.com).]**Table II.** Resistance Values of the PVAc-, PU-, and Silicone-Based Mats as a Function of Different CNT Content

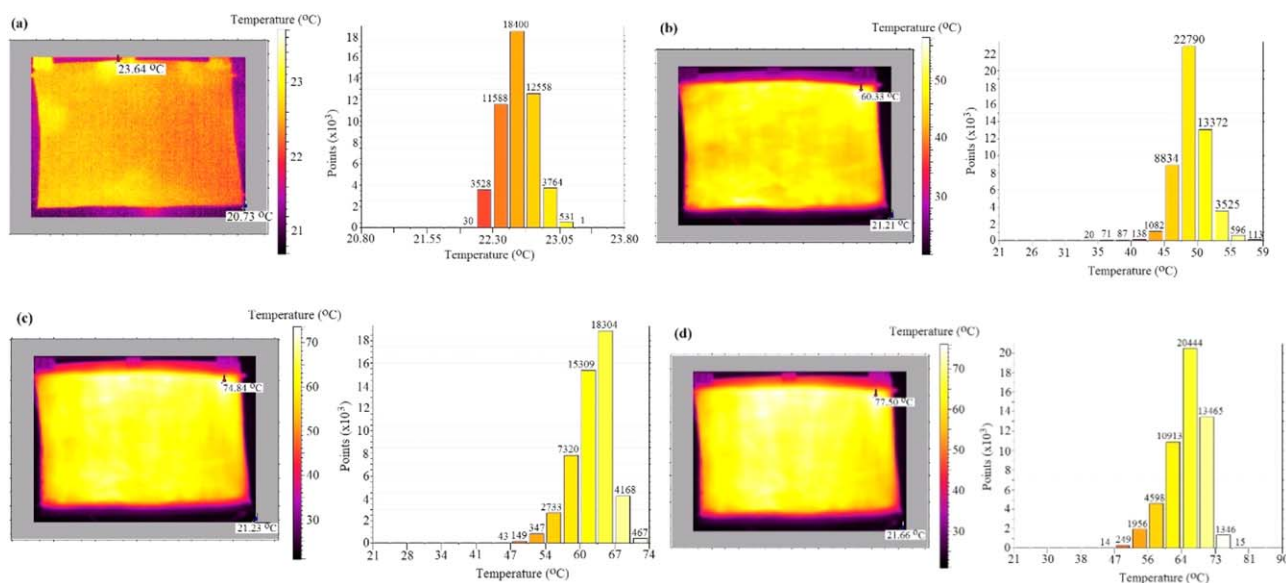
CNT content (wt %)	Resistance of the polymer mat ( $\Omega$ )		
	PVAc	PU	Silicone
5	55.0 ( $\pm$ 1.4)	60.0 ( $\pm$ 1.5)	32.0 ( $\pm$ 0.8)
10	35.0 ( $\pm$ 0.9)	50.0 ( $\pm$ 1.3)	26.0 ( $\pm$ 0.7)
20	30.0 ( $\pm$ 0.8)	38.0 ( $\pm$ 1.0)	21.0 ( $\pm$ 0.5)
30	21.0 ( $\pm$ 0.5)	31.0 ( $\pm$ 0.8)	13.0 ( $\pm$ 0.3)
40	17.0 ( $\pm$ 0.4)	27.0 ( $\pm$ 0.7)	6.0 ( $\pm$ 0.2)
50	9.0 ( $\pm$ 0.2)	18.0 ( $\pm$ 0.5)	5.0 ( $\pm$ 0.1)
60	4.0 ( $\pm$ 0.1)	9.0 ( $\pm$ 0.2)	3.5 ( $\pm$ 0.1)
70	3.5 ( $\pm$ 0.1)	5.0 ( $\pm$ 0.1)	3.5 ( $\pm$ 0.1)
80	3.5 ( $\pm$ 0.1)	3.5 ( $\pm$ 0.1)	3.5 ( $\pm$ 0.1)
90	3.5 ( $\pm$ 0.1)	3.5 ( $\pm$ 0.1)	3.5 ( $\pm$ 0.1)

content in the polymeric matrix, so the conductivity of the samples increases.<sup>14,17</sup> Comparing all the three mats, the highest resistance of 60  $\Omega$  was registered for the sample based on PU and 5 wt % of CNT. The sample of PVAc with the same quantity of CNT displayed the resistance of 55  $\Omega$  and the resistance of the silicone-based sample was 32  $\Omega$ . Similar to other studies, an increase of the CNT in the samples promoted a decrease of the resistivity and, consequently, an increase of the conductivity of the mats.<sup>8,16,18</sup> Addition of 60 wt % of CNT to the silicone, PVAc, and PU-based samples resulted in resistance values of 3.5, 4, and 9  $\Omega$ , respectively. The same resistance value of 3.5  $\Omega$  was measured in all samples only after the addition of 80 wt % of CNT. The detailed resistance values for all CNT-polymer compositions are displayed in Table II. The reported results indicate that the samples based on silicone probably have lower interfacial resistance between CNT and polymer matrix when compared with others, which results in lower resistance values of the mats.<sup>10</sup> The three samples were prepared with differently modified CNT aiming to enhance the compatibility between a matrix and a filler. However, from obtained results it seems that ester modified CNTs have a better compatibility and dispersion in silicone matrix than amide modified CNT in PU or carboxyl group modified CNT in PVAc. This good compatibility and dispersion resulted probably in good percolation network,<sup>9</sup> evidenced by lowest resistance values when compared with other samples.

The high resistance, above 40  $\Omega$  at low CNT content (5 wt %), of the samples of PVAc and PU combined with their high final cost showed that these ones are less suitable for heating purpose. Therefore, only the silicone-based samples containing 5 wt % of CNT were subjected to further analyses. The first one was the temperature analysis as a function of time after 0, 3, 9, and 27 min of potential application. The measurement set and the resistance values before and after this analysis with thermovision camera, for three different silicone-CNT samples, are listed in Table III. From this table one can observe that the resistance of two first samples decreased after potential application for 27 min. This can be because of the formation of percolating network.<sup>21</sup>

**Table III.** Electric Measurements Parameters and Resistance Results of Three Silicone-CNT Mats with 5 wt % of CNT

Silicone-CNT sample	Electric measurement parameters and results			
	Size (mm x mm)	Distance between the electrodes (mm)	Sample resistance before thermovision analysis ( $\Omega$ )	Sample resistance after thermovision analysis ( $\Omega$ )
1	300 x 297	265-270	18.5 ( $\pm 0.4$ )	8.8 ( $\pm 0.2$ )
2	300 x 295	265	22.6 ( $\pm 0.6$ )	12.3 ( $\pm 0.3$ )
3	307 x 300	275	26.3 ( $\pm 0.7$ )	Loss of electric contact

**Figure 3.** Thermograms and histograms of silicone-CNT composite mat (sample 2) with 5 wt % of CNT at (a) 0, (b) 3, (c) 9, and (d) 27 min of 27 V of applied potential. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

The electrical analysis revealed that the samples can be heated by applying potential ranging from 11 to 30 V or by applying current from 3 to 7 A. Consequently, simple car battery that provides 12 V can be used as a power supplier. However, in present study the samples were subjected to applied potentials varying from 22 to 27 V resulting in currents flow varying from 5.35 to 6.97 A (Table IV).

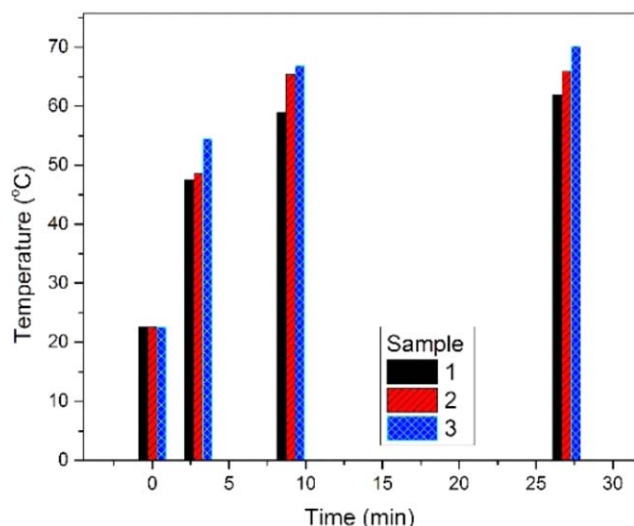
The silicone-CNT samples (Table III) were characterized by thermovision camera, and the representative pictures with respective thermal distribution graphs at 0, 3, 9, and 27 min of applied potentials, depicted in Table IV, are presented in Figure

3 for sample 2. One can observe that the sample starts heat quickly, and after 3 min of 27 V of applied potential the mat reaches about 50 °C [Figure 3(b)]. This can be an evidence of relatively high efficiency in transferring thermal energy from CNT to the polymer matrix.<sup>10</sup> Longer time of applied potential resulted in higher temperature of the sample reaching 60 °C in 9 min of applied potential [Figure 3(d)]. Twenty seven min of applied potential promoted an increase to 70 °C of sample temperature. Moreover, the obtained thermal images revealed that the samples have almost uniform thermal distribution of the analyzed surface. Some lighter and darker spots, visible on the

**Table IV.** Current flow after Applying Potentials Varying from 22 to 27 V to the Samples of CNT-Silicone with 5 wt % of CNT

Applied potential time (min)	Measured current of the sample (A)		
	1 (27 V; 23.9 °C; 49.4%) <sup>a</sup>	2 (22 V; 23.8 °C; 51.6%) <sup>a</sup>	3 (24 V; 22.5 °C; 49.0%) <sup>a</sup>
0	5.34 ( $\pm 0.05$ )	6.45 ( $\pm 0.06$ )	6.10 ( $\pm 0.06$ )
3	5.42 ( $\pm 0.05$ )	6.65 ( $\pm 0.07$ )	6.60 ( $\pm 0.07$ )
9	5.62 ( $\pm 0.06$ )	6.81 ( $\pm 0.07$ )	6.75 ( $\pm 0.07$ )
27	5.80 ( $\pm 0.06$ )	6.97 ( $\pm 0.07$ )	6.94 ( $\pm 0.07$ )

<sup>a</sup>Measurement conditions of applied potential, temperature, and relative humidity.

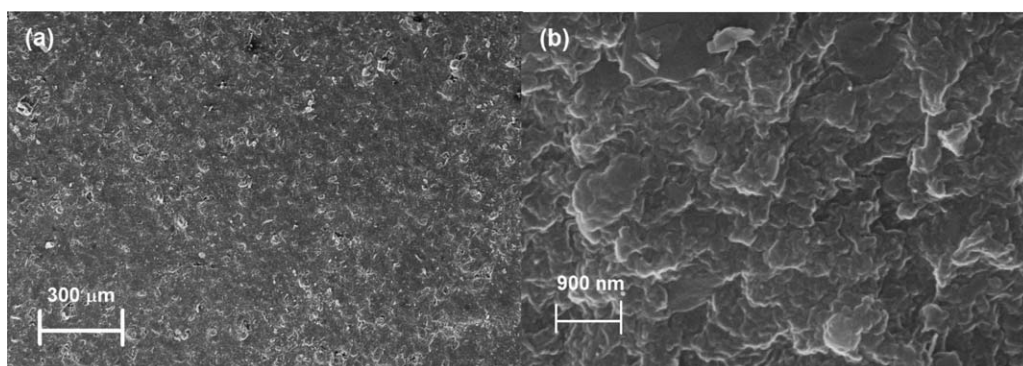


**Figure 4.** Temperature as a function of time of applied potentials in three silicone-CNT nanocomposite mats with 5 wt % of CNT. [Color figure can be viewed in the online issue, which is available at [wileyonlinelibrary.com](http://wileyonlinelibrary.com).]

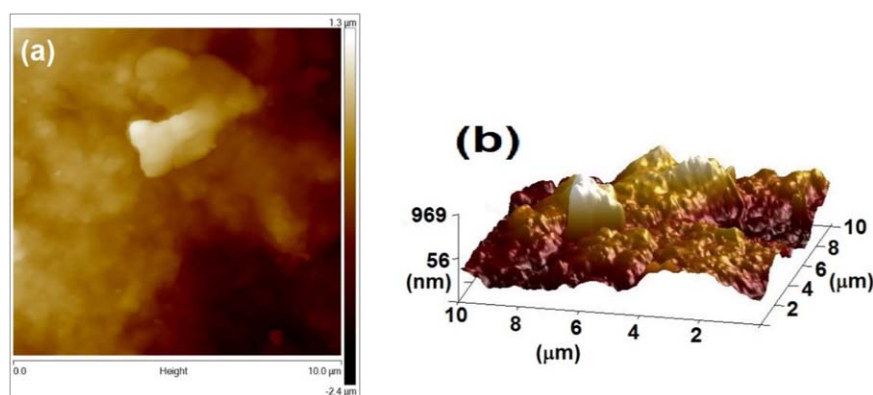
presented images, are because of higher and lower temperatures, respectively. These features can be because of the anisotropic CNT distribution in polymeric matrix.

The data obtained and showed in Figure 3 were resumed in Figure 4 that show the temperature as a function of time of applied potential in three silicone-CNT nanocomposite mats. As one can observe in this Figure 4, the time of applied potential promoted an increase of temperature from 22 °C to 48 °C after 3 min of applied potential of 27 V. After 9 min of potential application the samples reached about 60 °C, and after that an increase to about 70 °C was observed. Additionally, Figure 4 shows that analyzed samples 1, 2, and 3 (Table III) revealed the same thermal behavior under potential application, and small differences between the mats' temperature values can be because of the CNT distribution, dispersion, and alignment.<sup>10</sup>

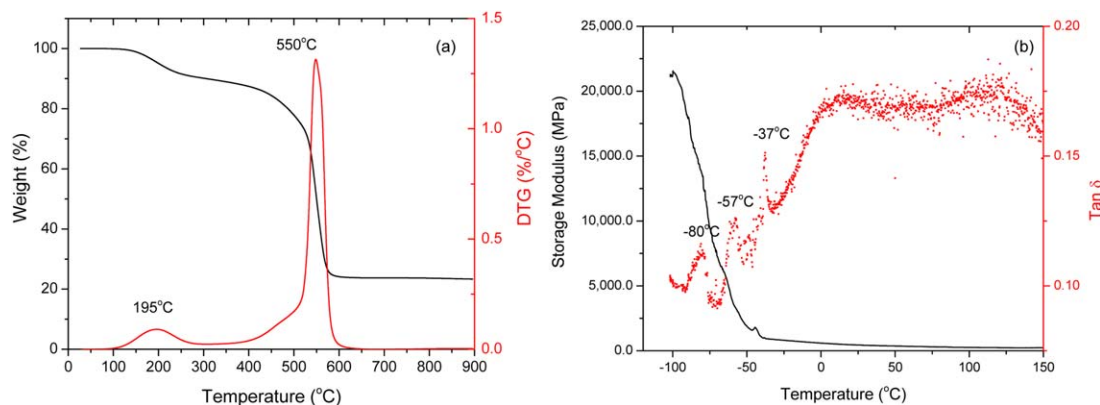
The silicone-CNT sample was then subjected to scanning electron microscopy (SEM) analysis. The result of this measurement is shown in Figure 5. At 200× magnification [Figure 5(a)], one can observe a very uniform surface of the sample with homogeneously distributed clearer features. A closer view, performed at 50,000× magnification, is shown in Figure 5(b). In this picture, the surface is composed by homogeneous plates connected to each other and of size ranging from 400 to 900 nm. The surface is uniform and no separation of phase between polymer and CNT is observed. Very similar results were obtained by Liu *et al.*,<sup>12</sup> who suggested that the CNT were uniformly dispersed in silicone matrix. About 1–5 wt % MWCNT-silicone rubbers were also prepared and characterized by Li *et al.*<sup>22</sup> Moreover, these authors confirmed uniform



**Figure 5.** SEM pictures of silicone-CNT nanocomposite mat with 5 wt % of CNT at (a) 200× and (b) 50,000× magnification.



**Figure 6.** AFM pictures of silicone-CNT nanocomposite mat in (a) 2D and (b) 3D high mode. [Color figure can be viewed in the online issue, which is available at [wileyonlinelibrary.com](http://wileyonlinelibrary.com).]



**Figure 7.** (a) TGA/DTGA and (b) DMA of silicone-CNT nanocomposite mat with 5 wt % of CNT. [Color figure can be viewed in the online issue, which is available at [wileyonlinelibrary.com](http://wileyonlinelibrary.com).]

distribution of CNT in the silicone matrix; however, their samples' SEM pictures suggested a microscopically visible less than 1 mm long CNT.

Figure 6(a) shows AFM pictures of silicone-CNT mat in 2D high mode. A picture reveals a nonuniform surface with lighter and darker areas. The total picture area is  $124 \mu\text{m}^2$  and has a roughness of 330 nm. A 3D image, shown in Figure 6(b), reveals a topography of the sample's surface that probably is affected by interaction of modified CNT with polymer matrix. These images also corroborate with SEM pictures shown in Figure 5.

Figure 7 shows TGA/DTG and DMA analyses of silicone-CNT mat. No weight loss up to  $150^\circ\text{C}$  is observed in TGA graph [Figure 7(a)]. After that, 10% of weight loss is seen up to  $250^\circ\text{C}$ , which is higher than that observed for silicone-10% of either  $\text{Al}_2\text{O}_3$  or ZnO fillers.<sup>23</sup> The main, 60% of initial weight is lost between  $430^\circ\text{C}$  and  $590^\circ\text{C}$ . After that, almost no weight is lost, and at  $900^\circ\text{C}$  the sample remains 23% of its original weight. The thermal transitions of the sample were detected throughout DMA analysis, shown in Figure 7(b), in the temperature range from  $-100^\circ\text{C}$  to  $150^\circ\text{C}$ . The storage modulus at  $-100^\circ\text{C}$  is about 22,000 MPa and decreases as the temperature increases, reaching almost zero values above  $50^\circ\text{C}$ . Tan delta reveals three peaks at  $-80^\circ\text{C}$ ,  $-57^\circ\text{C}$ , and  $-37^\circ\text{C}$ , which can be attributed to alfa, beta, and gamma transitions probably because of interaction of polymer matrix with CNTs. No other visible transition is seen up to  $150^\circ\text{C}$ ; however, above  $120^\circ\text{C}$  the tan delta starts to decrease, suggesting a loss of elastic properties and corroborating with TGA results.

## CONCLUSIONS

Three different polymer-CNT thermal mats were obtained and characterized: PU-, PVAc, and silicone-CNT. It was observed that the polymer used as a matrix had an impact on the electrical resistance of the mats. The lowest results of  $32 \Omega$  of resistance were obtained with silicone-based mats with 5 wt % of CNT. For the same CNT content the mats based on PVAc and based on PU displayed values of 55 and  $60 \Omega$ , respectively. The low resistance properties of silicone-based materials were due principally to the good compatibility of both polymer and functionalized CNT and probably to percolation network formation. Because of the low resistance values, this mat was subjected to

thermovision analysis, which revealed that the samples reached temperature of about  $60^\circ\text{C}$  in 9 min and  $70^\circ\text{C}$  after 27 min of applied potential. The obtained results showed an almost uniform temperature distribution in the samples' surface; however, some high and low temperature spots in thermal pictures were observed. These spots were attributed to nonuniform distribution of CNT in the polymer matrix. The silicone-CNT mats were also subjected to SEM, AFM, and thermal analyses. From microscopic measurements it was stated a surface composed by plates distributed uniformly and surface roughness of 330 nm. Thermal analyses revealed 10% of weight loss at  $120^\circ\text{C}$  and 60% at  $430^\circ\text{C}$ ; the remaining weight was 23% at  $900^\circ\text{C}$ . Tan delta of DMA analyses revealed three small transitions at  $-80^\circ\text{C}$ ,  $-57^\circ\text{C}$ , and  $-37^\circ\text{C}$ , which were attributed to alfa, beta, and gamma transitions, and confirmed a thermal event at  $120^\circ\text{C}$ . In summary, all the presented results confirm that silicone-CNT are very promising materials that can be used as low-voltage heating mats.

## ACKNOWLEDGMENTS

The work was supported by Wroclaw Research Center EIT+ with the project "The Application of Nanotechnology in Advanced Materials" – NanoMat (POIG. 01.01.02-02-002/08) co-financed by the European Regional Development Fund (Operational Program Innovation Economy, 1.1.2). The authors also are grateful to Patrycja Zgon, Ricardo Gomes, Dr. Rodrigo C. Sabadini, and Prof. Dr. Marcelo A. Chinelatto for their help in materials analyses.

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