



AMERICAN
SCIENTIFIC
PUBLISHERS

Copyright © 2016 American Scientific Publishers
All rights reserved
Printed in the United States of America

**Journal of
Nanoscience and Nanotechnology**
Vol. 16, 1607–1611, 2016
www.aspbs.com/jnn

Strain Sensing Characteristics of Rubbery Carbon Nanotube Composite for Flexible Sensors

Gyong Rak Choi¹, Hyung-Ki Park¹, Hoon Huh¹, Young-Ju Kim², Heon Ham³, Hyoun Woo Kim³, Kwon Taek Lim⁴, Sung Yong Kim⁵, and Inpil Kang^{5,*}

¹Manufacturing Automation R&BD Group, Korea Institute of Industrial Technology, Chonan, Korea

²Mineral Resources Research Division, Korea Institute of Geoscience and Mineral Resources, Daejeon, Korea

³Division of Materials Science and Engineering, Hanyang University, Seoul, Korea

⁴Department of Image Science and Engineering, Pukyong National University, Busan, Korea

⁵Department of Mechanical Design, Engineering Pukyong National University, Busan, Korea

In this study, the piezoresistive properties of CNT (Carbon Nanotube)/EPDM composite are characterized for the applications of a flexible sensor. The CNT/EPDM composites were prepared by using a Brabender mixer with MWCNT (Multi-walled Carbon Nanotube) and organo-clay. The static and quasi-dynamic voltage output responses of the composite sensor were also experimentally studied and were compared with those of a conventional foil strain gage. The voltage output by using a signal processing system was fairly stable and it shows somehow linear responses at both of loading and unloading cases with hysteresis. The voltage output was distorted under a quasi-dynamic test due to its unsymmetrical piezoresistive characteristics. The CNT/EPDM sensor showed quite tardy response to its settling time test under static deflections and that would be a hurdle for its real time applications. Furthermore, since the CNT/EPDM sensor does not have directional voltage output to tension and compression, it only could be utilized as a mono-directional force sensor such as a compressive touch sensor.

Keywords: Flexible Sensor, Piezoresistive Sensor, Carbon Nanotubes, Rubbery Nanocomposite.

1. INTRODUCTION

The flexible and bendable characteristics of sensor have been a newly emerging area to make them ubiquitous and to overcome conventional sensing systems. In order to achieve flexible sensors, micro-structures have been molded by using MEMS process.^{1–3} New fabrication technology for flexible sensors should be inexpensive and simple for mass production with reliability and sustainability. The flexible sensors made by composite process may be one of the most promising technologies for this purpose, combining simple design and low cost preparation with high manufacturing flexibilities. To detect large-size flexible pressure and stretch, various filler based electro-conductive polymers or their based tactile sensors have been studied.^{4–9} A carbon black based composite has a limitation for accurate strain sensing due to its nonlinear

piezoresistivity with respect to force or strain.^{10–12} Nano filler based rubbery materials showed fairly good sensing properties, especially carbon nanotubes have been studied as promising filler thank to their excellent mechanical, electrical and other sensing properties. Having piezoresistivity, the nano-carbon based smart nanocomposite can be a sensory material for a novel flexible sensor.¹³ Sensor performance should be estimated for real world applications. However, the previous studies mostly have assured change of resistance of the polymers under loading conditions and, as far as we know, none of them exploits sensing characteristics with signal processing system for their engineering applications. A description of the flexible sensor from the point of view of real time control, the piezoresistive characterizations and sensing performances of CNT/EPDM composites are experimentally studied with a perspective outlook on the main promising applications. This study conducted as part of the feasibility test of nano-carbon based flexible sensory materials.

*Author to whom correspondence should be addressed.

2. EXPERIMENTAL DETAILS

2.1. Fabrication of EPDM/Carbon Nanotube Composites and Flexible Strain Sensors

Flexible CNT composites were fabricated with the EPDM matrix by using a traditional compounding technique. The compounding was performed in Brabender internal mixer with CAM rotor configuration. The MWCNT (Aldrich, 677248) and the nano-clay (Southern Clay Products, Cloisite 15A) were added to fabricate the composites. The nano-clay concentration was kept as a constant weight to EPDM phase, 8.8 parts of the MWCNT weight concentration ratio (wt%) was varied from 10 to 30 parts. The compounds were cured with α , β -bis (*t*-butylperoxy) di-isopropylbenzene at 170 °C in a compression mold.

Electrical conduction property is fundamentally required for the CNT-based composite sensory material because it makes the piezoresistivity of the composite. In this study, the conductivity was achieved over 10 parts of the CNT in the composites. It has been reported that the conductivity of the CNT rubber achieved rather higher fraction ratio than other CNT based composites.^{14,15} Since the nanotube-matrix interface is a critical parameter controlling the mechanical and electrical properties of the CNT composites, it is carefully observed based on compounding techniques. However, the physics of the intercalation of the CNT to the EPDM has yet to be clarified, and controlling the dispersion is still a challenge.

Flexible strain sensors were fabricated by cutting the conductive EPDM/CNT composites, and electrodes were connected to the sensors with silver conducting epoxy to reduce the contact resistance. A sample of fabricated flexible strain sensor made of EPDM/CNT composite is presented in Figure 1(a). Figure 1(b) also displays Transmission Electron Microscopy (TEM) image of fabricated composite sample prepared from the above procedure. The fairly dispersion of MWCNT as a filler can be found in the rubber matrix from the TEM image.

2.2. Piezoresistivity and Strain Sensing Experiment

The strain sensor electrodes made of CNT/EPDM composites were tightly attached on the surface of a flexible

beam to test their piezoresistive sensing characteristics. After connecting their wires to a signal processing system, the sensors were characterized based on their voltage outputs. A schematic illustration of experimental procedure in this study appears in Figure 2.

The piezoresistivity of the composites was found according to their deformations and their changes of electrical resistance also were measured after the deformations. The strains of the composites (ε) can be found from the beam's strain by using cantilever deflection theory as following:

$$\varepsilon = \frac{3c(L-a)}{L^3} \delta \quad (1)$$

Where, c is the half of the beam thickness, L is the beam length, δ is the end of beam deflection and a is the distance from the fixed point to the middle of the composites respectively.

The change of resistance of each composite (ΔR) was simply measured under beam deflection with a multi-meter (Tektronix DMM916). The measured values were converted to normalized changes of resistance (R_N):

$$R_N = \frac{R_s - R_0}{R_0} \quad (2)$$

Where, R_0 is the resistance without any displacement or strain and R_s is the measured the composite resistance when the beam is deflected.

For static and dynamic strain sensing characteristic, the CNT/EPDM 20 part composite sensor was tested and its responses was compared with a conventional foil strain gauge (CAS, AP-11S30N-120-EC). The resistance changes of 2 individual sensors were converted into voltage outputs by means of a signal processing system consisting of 2 Wheatstone bridge circuits, 2 DC amplifiers (Park Electronics DSA301A), a power supplier (GW Instek GPS-4303), a oscilloscope (Tektronix TDS 2014B) and a LabView data acquisition system.

3. RESULTS AND DISCUSSION

In micro scale, piezoresistivity of individual CNT was reported by strain which changes the band structure to

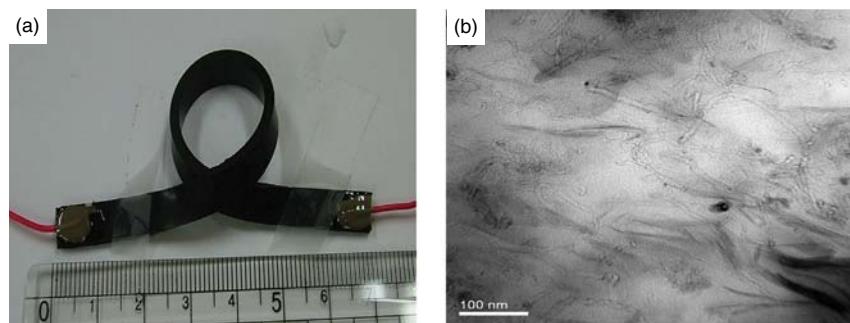


Figure 1. The CNT-EPDM rubber composite (10 wt%): (a) Flexible CNT/EPDM sensor electrode (150 mm × 5 mm × 2 mm, 160 kΩ); (b) TEM image.

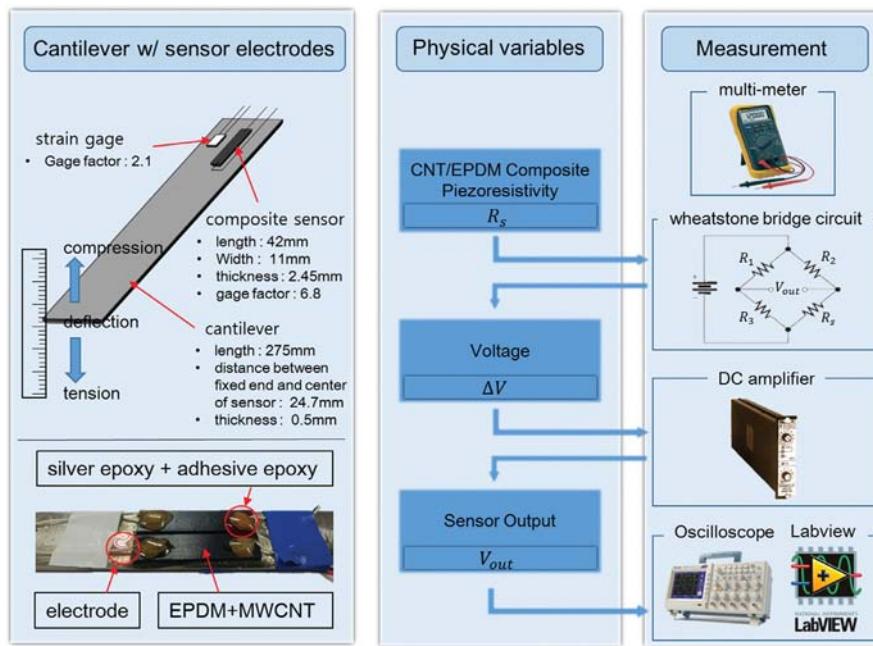


Figure 2. Schematic illustration of CNT-EPDM rubber composites to measure their piezoresistivity and sensing characteristics by using a signal processing with data acquisition system.

alter the electrical properties of the nanotube more or less conductive.¹⁷ While in macro scale, piezoresistivity of carbon nano fillers in a matrix results from the contact resistance of adjacent nano fillers.^{16,18} Figure 3(a) shows piezoresistive property of the CNT/EPDM composites with respect to the strain changes and slopes of the curves represent their strain sensitivities. The piezoresistive response shows somehow linear sensitivity in large deflection range, but it shows less sensitive behavior in small range of the deflection, probably, due to the softness of the rubber matrix. In the case of hard composite strain sensor, it shows a quite linear bidirectional piezoresistive response to strain and its piezoresistivity tends to increase to tension and to decrease to compression.¹⁶ However, the CNT rubbery composite has not only non-symmetric responses but also shows incrementally unidirectional piezoresistivity

under both of tension and compression. The change of resistance variation of CNT/EPDM composite is only increased and that might be caused to soft polymer bonding. According to literature survey, in the case of high aspect nanofiller such as carbon nanotubes in a soft matrix, the resistivity of the composite increases even compressive force condition due to the destruction of the existing conductivity of the filler.⁵ The non-directional piezoresistivity of CNT rubbery composite may hamper to achieve a fine flexible strain sensor and that also induces signal distortion under dynamic loads as shown in Figure 5(b) later. The CNT/EPDM composite might be available only to build a unidirectional force or deformation sensor due to its non-direction piezoresistivity.

Figure 3(b) shows the voltage output of 20 part CNT/EPDM sensor induced by tensile and compressive

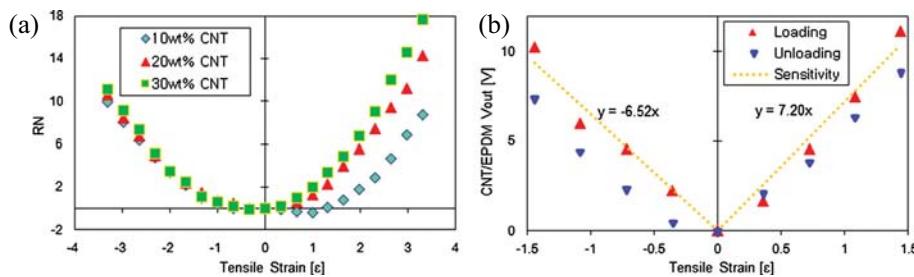


Figure 3. Strain sensing characteristics of the CNT/EPDM composite: (a) Piezoresistive property of the composites (10, 20, 30 wt%); (b) voltage output of the sensor (20 wt%) with hysteresis under static loading condition.

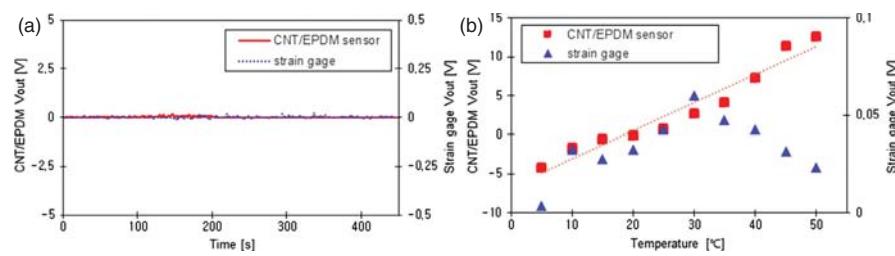


Figure 4. Voltage output responses of the CNT/EPDM (20 wt%) sensor system without load or deflection: (a) Drift test; (b) temperature test.

strain and its profile is similar to Figure 3(a). The sensor presents somehow linear responses at both of loading and unloading cases with hysteresis. From this result, it may be able to obtain better linear responses in extensional force or deflection rather than compressive cases for its further applications. The resistance of sensor definitely depends on the size of its electrode and its voltage output can be controlled with the resistance of it consequently. However, the sensor characteristic does not much related with its resistance but it is dominated by its sensitivity which is controlled by nanofiller contents.¹⁶

Figure 4 illustrates voltage output responses of the CNT/EPDM (20 wt%) sensor system without any load or deflection. Fairly stable voltage outputs can be achieved from the sensor system as shown in Figure 4(a). The sensor output was varied due to the change of environmental temperature as shown in Figure 4(b) and the changing tendency is similar to the reference. The resistivity of polymer based composites increase with temperature due to positive temperature coefficient of resistance effect.⁴ Since the sensor voltage output variation due to temperature change is quite severe, a dummy gauge should be required to compose the Wheatstone bridge circuit to compensate the temperature effect. This characteristic

may hamper its applications under wide temperature range.

Figure 5 shows voltage output responses of the sensor system under static or quasi-dynamic deflections. Figure 5(a) illustrates a step response under extensional deflection. The response is almost identical with the output of the conventional foil strain gage and that means the sensor can measure tensile force or strain from a structure without much distortion. However, the sensor may have limitations to use for a dynamic sensor as shown in Figure 5(b). Since the sensor has unsymmetrical strain responses previously described, it shows the distorted voltage responses when it shacked with a hand. The rising time of the sensor was tested and was compared with the strain gage. There was not much difference between the two results as shown in Figure 5(c). The CNT/EPDM sensor showed quite tardy response to its settling time test under static deflections shown in Figure 5(d). It is speculated that the time delay caused by relocation of electrical conduction paths of nanofillers in the rubber matrix. In general, a control system requires prompt response from sensors and the time delay characteristics of the CNT/EPDM sensor could be a potential huddle from a point of view in real time control.

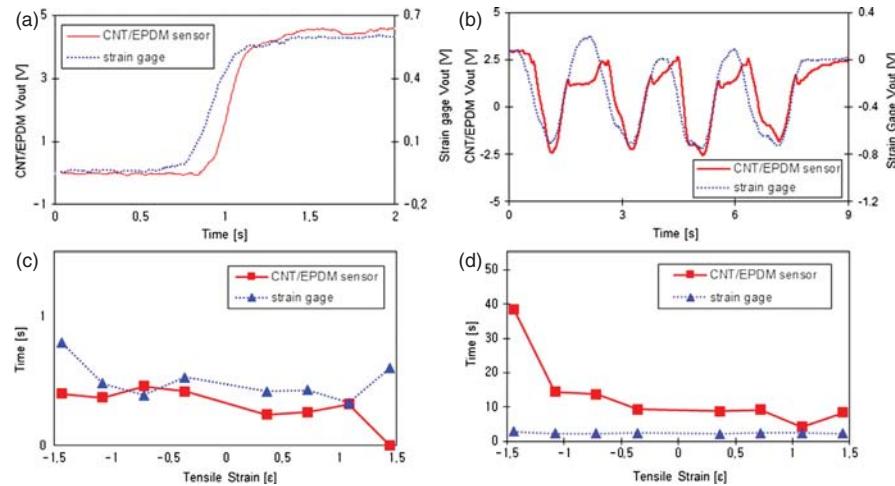


Figure 5. Voltage output responses of the CNT/EPDM (20 wt%) sensor system under deflections: (a) Static deflection test; (b) quasi-dynamic test with hand shaking; (c) rising time test; (d) settling time test.

4. CONCLUSION

In this study, the piezoresistive properties of CNT/EPDM composite are characterized for the applications of a flexible sensor. The CNT rubbery composites show non-symmetric responses unlike hard composites cases and that might be caused to soft polymer bonding. The piezoresistive response of CNT/EPDM shows fairly linear sensitivity in large deflection range, but it shows less sensitive behavior in small range of the deflection due to the softness of the rubber matrix.

The sensor was characterized by its voltage outputs by using a signal processing system and fairly stable voltage outputs can be achieved from the sensor. The sensor shows somehow linear responses at both of loading and unloading conditions with hysteresis. However, the sensor shows distorted voltage responses under a quasi-dynamic test due to unsymmetrical piezoresistive characteristics. The CNT/EPDM sensor showed quite tardy response to its settling time test under static deflections.

Thanks to its flexibility, the CNT/EPDM composite may be expected to measure large deformation of flexible structures and that may overcome the limitation of traditional strain sensors. However it does not have directional output it could be utilized one directional force sensor such as a compressive touch sensor. The tardy responses would be a hurdle for its real time applications as well.

Acknowledgment: This work was supported by the Pukyong National University Research Abroad Fund in 2012(PS-2012-0720).

References and Notes

1. D.-Y. Khang, J. Jiang, Y. Huang, and J. A. Rogers, *Science* 311, 208 (2006).
2. T. Sekitani, Y. Noguchi, K. Hata, T. Fukushima, T. Aida, and T. Someya, *Science* 321, 1468 (2008).
3. S. Gong, W. Schwallb, Y. Wang, Y. Chen, Y. Tang, J. Si, B. Shirinzadeh, and W. Cheng, *Nat. Commun.* 5, 3132 (2014).
4. M.-J. Jiang, Z.-M. Dang, and H.-P. Xu, *Appl. Phys. Lett.* 89, 182902 (2006).
5. L. Chen, G. Chen, and L. Lu, *Adv. Commun.* 17, 898 (2007).
6. J. Hwang, J. Jang, K. Hong, K. N. Kim, J. H. Han, K. Shin, and C. E. Park, *Carbon* 49, 106 (2011).
7. M. Segev-Bar, A. Landman, M. Nir-Shapira, G. Shuster, and H. Haick, *ACS Appl. Mater. Interf.* 5, 5531 (2013).
8. S. Tadakaluru, W. Thongsawan, and P. Singhai, *Sensors* 14, 868 (2014).
9. L. Wang and L. Cheng, *Carbon* 71, 319 (2014).
10. B. B. S. T. Boonstra and E. M. Dannenberg, *Indust. Engineer. Chem.* 46, 218 (1954).
11. A. E. Job, F. A. Liveira, N. Alves, J. A. Giacometti, and L. H. C. Mattoso, *Synth. Met.* 135–136, 99 (2003).
12. M. Knite, V. Teteris, A. Kiploka, and J. Kaupuzs, *Sens. Actuators A* 110, 142 (2004).
13. I. Kang, M. A. Khaleque, Y. Yoo, P. J. Yoon, S. Y. Kim, and K. T. Lim, *Compos. Part A: Appl. Sci. Manuf.* 42, 623 (2011).
14. C. L. Kane, E. J. Mele, R. S. Lee, J. E. Fischer, P. Petit, H. Dai, A. Thess, R. E. Smalley, A. R. M. Verschueren, S. J. Tans, and C. Dekker, *Europhys. Lett.* 41, 683 (1998).
15. K. H. An, S. Y. Jeong, H. R. Hwang, and Y. H. Lee, *Adv. Mater.* 16, 1005 (2004).
16. I. Kang, M. J. Schulz, J. H. Kim, V. Shanov, and D. Shi, *Smart Mater. Struct.* 15, 737 (2006).
17. T. W. Tombler, C. Zhou, L. Alexseyev, J. Kong, H. Dai, L. Liu, C. S. Jayanthi, M. Tang, and S.-Y. Wu, *Nature* 405, 769 (2000).
18. D. D. Chung, *Carbon* 39, 1119 (2001).

Received: 13 May 2015. Accepted: 29 May 2015.