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An Embedded PDMS Nanocomposite Strain Sensor toward Biomedical Applications

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***Abstract*—This paper presents a nanocomposite strain gauge composed of poly(dimethylsiloxane) and multi-walled carbon nanotubes. Possessing excellent mechanical and piezoresistive properties, the biocompatible nanocomposites could withstand large strains repeatedly, which is desirable for miniaturized implantable biomedical devices. Prototype strain sensor was fabricated with simplicity and efficiency via microcontact printing and cast molding. Experimental results revealed sensitive response of resistance with regard to change of tensile strains. Multiple cycles of stretching and relaxing of device revealed consistent and repeatable measurements. An interesting hysteresis phenomenon was also observed. With further investigation of the elastomeric mechanisms, this strain sensing technology could yield promising potentials in many biomedical applications.**

I. Introduction

The measurement of biomechanical strains within human body is vital for many biomedical applications. In the case of bone fracture treatment, for example, post-surgical therapies need to be adapted to the actual healing stage. In fact, this remains a challenge in today's medicine, since approximately 10% of bone fractures still do not heal properly due to abnormal strain profiles during the healing process [1-2]. Better understanding of strains induced in organic structures could also benefit the studying of other medical conditions such as osteoporosis, bone tumors, as well as prosthetic implants.

For biomedical applications, it would be desirable if the strain sensor could be attached, during surgery, to a certain location (e. g., on a fractured bone). After surgery, the sensor remains inside human body to continuously monitor local strain. Metallic strain gauges are commercially available and popular in many applications due to their low cost. However, their usage as implanted medical devices is limited because of their relatively large size and lack of biocompatibility to offer long-term implantable or wearable monitoring of biomechanical strains. On the other hand, elastomeric nanocomposite materials offer an alternative to be used as biocompatible strain sensors. Since elastomers are much more flexible compared to metals, they can sustain higher level of strains. Therefore, for a given displacement (e.g. interfragmentary motion during the healing of bone fracture could be up to a few millimeters [3]), the required dimensions for nanocomposite sensors could be relatively smaller—a desirable attribute for miniaturized implantable devices.

Elastomer nanocomposites are often composed of a polymer matrix and nano-scale filling materials. Among common fillers [4-7], carbon nanotubes present an ideal candidate for strain sensing applications. They possess superior mechanical properties, for instance, with tensile strength up to 63 GPa using multi-walled carbon nanotubes (MWCNTs) [8]. More importantly, MWCNTs have excel-lent piezoresistive property as well [9]. When exposed to tensile or compressive strain, the geometry and spacing of carbon nanotubes within the polymer matrices vary, which leads to a change in its resistance. Also, a good deal of research effort has been committed to select hosting matrix for the nanocomposites. Various polymers including poly(methyl methacrylate) (PMMA), polycarbonate (PC), poly(L-lactide) (PLLA), etc. have been reported to incorporate with MWCNTs to construct strain sensors, which could withstand relatively large tensile strains [9-14].

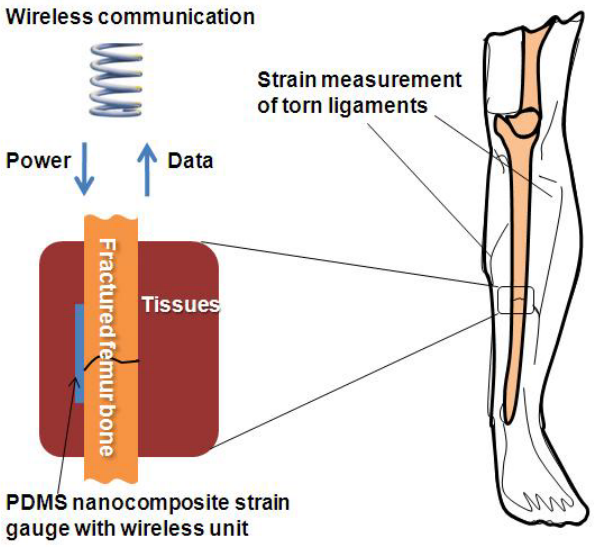

Fig. 1. A long-term goal of the implantable strain sensor with wireless sensing capability.

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In this work, we present a novel design of nanocomposite strain gauge, by utilizing elastomer poly(dimethylsiloxane) (PDMS) as the hosting matrix and MWCNTs as piezoresistive fillers. Compared with polymers mentioned above, PDMS owns unparalleled mechanical elasticity and is able to hold over 100% of tensile strain without any mechanical failure [15]. Its ability to withstand large deformation presents a clear advantage in downsizing the scale of strain sensors. Moreover, PDMS is known for its chemical inertness and exceptional biocompatibility, and is widely used in biomedical applications for implants, skin care, medical adhesives, etc. [16]. We also describe a simple and efficient approach of embedding PDMS-MWCNTs composite patterns into unmodified bulk PDMS by using microcontact printing and cast molding techniques. As a long-term goal of this work, the strain sensor could potentially be implanted into human body as illustrated in Figure 1, and integrated with a wireless communication unit [11-12, 14-15]. As power could be transmitted via the remote reader site, data generated by the strain gauge could also be processed on an external station, which allow continuous monitoring of biomechanical strains during our daily activities.

II. Device Fabrication

In the past few years, several approaches have been explored to address the issue of patterning PDMS-based nanocomposite materials [11, 17-20]. While promising feasibilities were demonstrated, the fabrication processes were labor-intensive and inefficient. In this study, we greatly improve the process by simply using a stamp to transfer PDMS composites from one surface to another. In this novel approach, the stamp, once made, could be repeatedly used to transfer nanocomposites, enhancing the efficiency and reliability of pattern generation. Also, an additional cast molding step follows to embed imprinted patterns into bulk PDMS.

To begin with, MWCNTs were mixed with PDMS to form conformal nanocomposites. This was facilitated by a strong organic solvent toluene as it dissolved PDMS base polymer easily and allowed monodispersion of MWCNTs [21]. Firstly, PDMS (Sylgard 184 from Dow Corning) base polymer was added into toluene (1:4 volume ratio). Dry MWCNTs (from Cheaptubes Inc, outer diameter: 20-40 nm) were separately dispersed in another toluene solution (1:20 weight ratio) and magnetically stirred for 2 hours before the two solutions were mixed together in an open container. While further magnetic stirring helped the mixing of PDMS and MWCNTs, volatile toluene slowly evaporated on the hot plate (50^{o}C) under a chemical hood. After toluene reached full evaporation overnight, PDMS curing agent (1:10 weight ratio to base polymer) was added into the mixture. Now, the conformal PDMS-MWCNTs composites were ready to work as stamping "ink." Figure 2 shows a scanning electron microcopy image of the cross section of a fractured nanocomposite sample showing distributed carbon nanotubes in the PDMS matrix.

For the purpose of prototyping, a simple stamp (glass) with pattern of a single straight line was used to create the strain sensor. First, the glass stamp was held onto the vertical beam of a wafer prober. Meanwhile, the prepared PDMS-MWCNTs composites were spin-coated into a thin layer (~ 60 μm) on a silicon wafer which was then placed underneath the stamp. Next, the stamp was carefully dipped into the ink and lifted up shortly in a few seconds. Then, all nanocomposite patterns were transferred by stamping the mold onto a slide glass substrate which had been previously treated with chlorotrimethylsilane (from Alfa Aesar)—a chemical release agent to facilitate the debonding of PDMS from glass. In a subsequent step, the imprinted conductive composites were partially cured in an oven at 60^{o}C for 30 minutes to solidify pattern. Additional bulk PDMS mixed with base polymer and curing agent was then poured atop to submerge the imprinted pattern. In the final step, whole PDMS block was degassed in a vacuum pump and fully cured at 60^{o}C for 4 hours before being debonded from the glass substrate. Eventually, an all-elastomer strain gauge was fabricated. Figure 3 demonstrates the superior mechanical flexibility of the manufactured strain gauge.

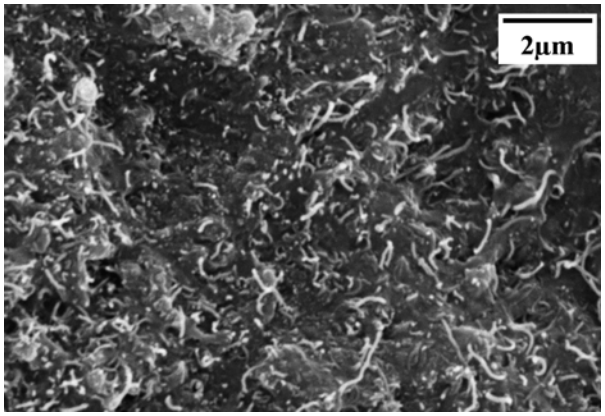

Fig. 2. SEM image of PDMS-MWCNTs nanocomposite cross section. Carbon nanotubes share a weight percentage of 5%, and they are quite uniformly distributed throughout the elastomer matrix, showing no sign of aggregation.

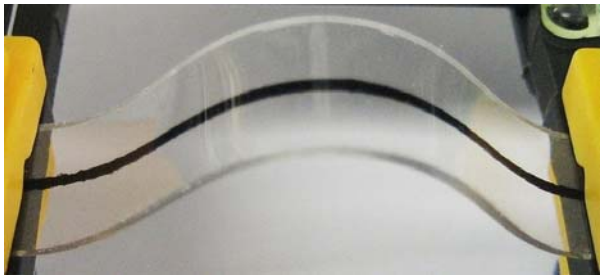

Fig. 3. PDMS Nanocomposite strain sensor embedded inside unmodified bulk PDMS through microcontact printing and casting techniques. Nanocomposites contain 9% of weight fraction of MWCNTs. The dimension of whole PDMS block is 43 mm-long, 10 mm-wide and 0.75 mm-thick, and embedded sensor is 43 mm-long, 0.9 mm-wide and around 50 μm in its thickness.

III. Results and Discussion

The principle of PDMS nanocomposite strain gauge operation is straight forward. Under tensile strain, the geometry and spacing of carbon nanotubes are altered causing a change in the resistance reading. In our experiments, as depicted in Figure 4, the PDMS block with embedded strain gauge was manually stretched and relaxed for multiple cycles while the change in its resistance was recorded with a digital multimeter. To ensure consistency of testing results and prevent possible buckling of device, tensile strain stayed at zero or positive throughout experiments.

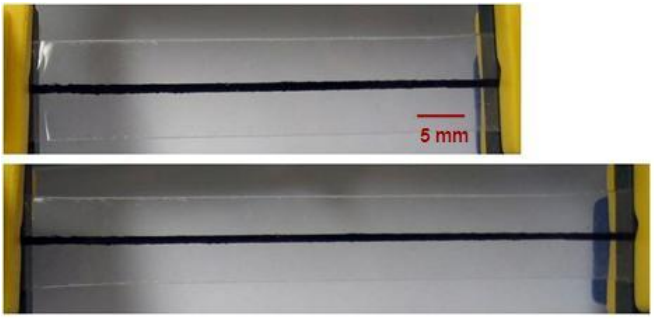

Fig. 4. Images showing manual stretching/relaxing of PDMS-MWCNTs nanocomposite strain sensor.

Under different levels of tensile strain, the sensor revealed significant change in its resistance. Maximum tensile strain of over 40% was applied to the device during tests resulting in a sensor resistance shift between 1.54 MΩ and 3.31 MΩ, as shown in Figure 5, which was a relative change of over 110%. As mentioned before, the ability of the PDMS composite strain sensor to endure large mechanical deformations repeatedly served as an advantage over strain gauges made of other materials. In addition, the strain gauge showed consistency in its resistance change over multiple cycles of measurements, which was a desirable attribute for sensing applications.

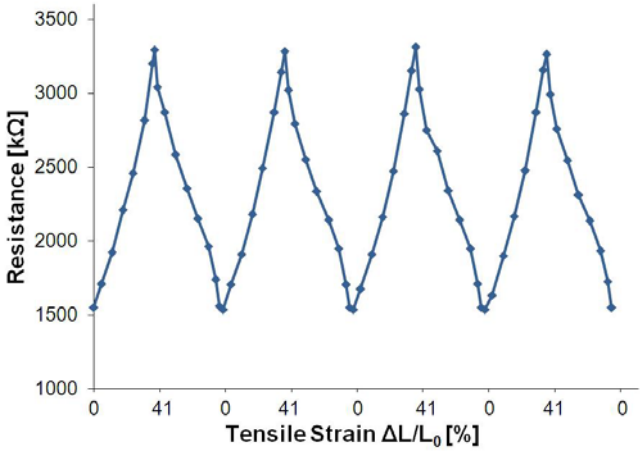

Fig. 5. Measured sensor resistance change to applied tensile strain. The demonstrated device was repeatedly stretched and relaxed. Resistance values were recorded when strains stabilized for 1~2 minutes after each change.

Conventionally, the gauge factor is an important parameter in estimating the quality of strain sensors. It is defined as the ratio of resistance change over the ratio of dimension change,

$$G = \frac{\Delta R / R_0}{\Delta L / L_0} = \frac{\Delta R / R_0}{\varepsilon} \quad (1)$$

where G is the gauge factor, L_0 and R_0 are original length and resistance of strain sensor, ΔL and ΔR are length change and resistance change, and ε is the tensile strain. For the sensor in our experiment, the gauge factor (G) was calculated to be around 2.7, which was comparable to previously reported works using MWCNTs-based nano-composite sensors [7-8].

An interesting phenomenon of the embedded strain sensor was the hysteresis effect of resistance change in regard to tensile strain. As indicated in Figure 6, one cycle of stretching and relaxing was extracted to reveal the relationship between resistance change and tensile strain. Starting from original length, when strain sensor was initially stretched, the resistance reading actually showed a slight decrease. While further studies are underway, one possible explanation was that since the nanocomposites were composed of carbon nanotube networks entangled amongst PDMS molecular chains, individual nanotubes could not settle down immediately after a change of strain, causing a time delay in its electric response [22]. Thus, when strain was just starting to increase from zero, inside the polymer matrix the spacing of carbon nanotubes was actually still decreasing from the previous cycle of strain dropping, resulting in a temporary decrease in its resistance reading. After strain rose above a few percent, the spacing of carbon nanotubes resumed increasing, so did the resistance reading.

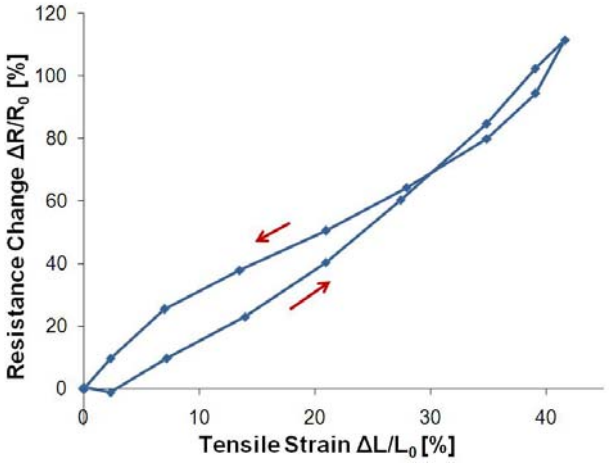

Fig. 6. Relative resistance change to tensile strain during one cycle of stretching and relaxing of strain gauge. Hysteresis effect was observed consistently during experiments.

On the other hand, although the resistance change with increasing strains was fairly linear for the most part, it was not the case when strain was reversed. Resistance tended to drop more quickly right after strain decreased from the maximum level, then after a certain cross point, it started to decline at a lower rate. While this "strange" pattern of hysteresis effect is

still being studied, according to a two-force node-node interaction model of polymer mechanism proposed by Hanson [23], the PDMS chain molecules were under taut condition with an increasing strain. When strain was reversed, the sudden change in external force momentum caused the polymeric chain force to decrease almost instantly, which led to an abrupt drop in the intrinsic stress. Tubule carbon nanotubes were similar to PDMS chain molecules structurally, therefore if its resistance was associated with the intrinsic stress of composites, a similar abrupt change in resistance could also occur. Overall, at the current stage further thorough investigation of elastomer composite mechanisms is still required for a better understanding of the resistance to strain response.

IV. Conclusions

Utilizing PDMS and MWCNTs as polymer matrix and nano-scale fillers, a novel design of nanocomposite strain sensor was introduced. Microcontact printing and cast molding were employed to realize simple and efficient embedment of strain gauge pattern inside PDMS block. During testing of prototype sensor, both significant and consistent resistance change was recorded in response to change of tensile strains. Owning superior mechanical flexibility and piezoresistivity, the biocompatible nanocomposites strain gauge could be integrated in a miniature system and implanted inside human body for monitoring of biomechanical strains. Provided that further understanding of the intrinsic mechanisms of the elastomeric composites related to its hysteresis effect could be obtained, this strain sensing technique yields promising potentials in many biomedical applications.

References

[1] K. V. Schuylenbergh, R. Puers, F. Rodes, F. Burny, M. Donkenvolcke, and F. Moulart, "Monitoring orthopaedic implants using active telemetry," in *Proc.14th Ann. Int. Conf. IEEE: EMBS,* vol. 6, Paris, France, 1992, pp. 2672–2673.

[2] T. A. Einhorn, "Enhancement of fracture healing," *J. Bone Joint Surg.,* vol. 77, pp. 940–956, 1995.

[3] H. McKellop, R. Hoffmann, A. Sarmiento, and E. Ebramzadeh, "Control of motion of tibial fractures with use of a functional brace or an external fixator. A study of cadavera with use of a magnetic motion sensor," *J. Bone Joint Surg.*, vol. 75, pp. 1019-1025, 1993.

[4] B. Lundberg and B. Sundqvist, "Resistivity of a composite conducting polymer as a function of temperature, pressure, and environment: applications as a pressure and gas concentration transducer," *J. Appl. Phy.*, vol. 60, issue 3, pp. 1074-1079, 1986.

[5] E. Segal, R. Tchoudakov, M. Narkis, and A. Siegmann, "Sensing of liquids by electrically conductive immiscible polypropylene/ thermoplastic polyurethane blends containing carbon black," *J. Polym. Sci., Part B*, vol. 41, issue 12, pp. 1428-40, 2003.

[6] J. Lu, M. Lu, A. Bermak, and Y.-K. Lee, "Study of piezoresistance effect of carbon nanotube-PDMS composite materials for nanosensors," in *Proc. 7th Int. Conf. IEEE Nanotechnology*, Hong Kong, China, 2007, pp. 1240-1243.

[7] X. Niu, S. Peng, L. Liu, W. Wen, and P. Sheng, "Characterizing and patterning of PDMS-based conducting composites," *Adv. Mater.*, vol. 19, issue 18, pp. 2682-2686, 2007.

[8] M.-F. Yu, O. Lourie, M. J. Dyer, K. Moloni, T. F. Kelly, and R. S. Ruoff , "Strength and breaking mechanism of multiwalled carbon nanotubes under tensile load," *Science,* vol. 287, no. 5453, pp. 637-640, 2000.

[9] I. Kang, M. J. Schulz, J. H. Kim, V. Shanov, and D. Shi, " A carbon nanotube strain sensor for structural health monitoring," *Smart Mater. Struct.,* vol. 15, issue 3, pp. 737-748, 2006.

[10] W. Zhang, J. Suhr, and N. Koratkar, "Carbon nanotube/polycarbonate composites as multifunctional strain sensors," *J. Nanosci. Nanotech.*, vol. 6, no. 4, pp.960-964, 2006.

[11] Y. Liu, S. Chakrabartty, D. S. Gkinosatis, A. K. Mohanty , and N. Lajnef, "Multi-walled carbon nanotubes_poly(l-lactide) nano-composite strain sensor for biomechanical implants," in *Proc. IEEE BCSC*, Montreal, Canada, 2007, pp. 119-122.

[12] F. Umbrecht, M. Wendlandt, D. Juncker, and C. Hierold, "A wireless implantable passive strain sensor system," in *Proc. IEEE Sensors*, Irvine, USA, 2005, pp. 20-23.

[13] G. T. Pham, Y.-B. Park, Z. Liang, C. Zhang, and B. Wang, "Processing and modeling of conductive thermoplastic/carbon nanotube films for strain sensing," *Composites: B,* vol. 39, issue 1, pp. 209-216, 2008.

[14] R. Melik, N. K. Perkgoz, E. Unal, C. Puttlitz, and H. V. Demir, "Bio-implantable passive on-chip RF-MEMS strain sensing resonators for orthopaedic applications," *J. Micromech. Microeng*., vol. 18, no. 11, 115017 (9pp), 2008.

[15] G. Y. Yang, V. J. Bailey, Y.-H. Wen, G. Lin, W. C. Tang, and J. H. Keyak, "Fabrication and characterization of microscale sensors for bone surface strain measurement," in *Proc. IEEE Sensors*, Vienna, Austria, 2004, pp. 1355-1358.

[16] J. P. Heggers, N. Kossovsky, R. W. Parsons, M. C. Robson, R. P. Pelley, and T. J. Raine, "Biocompatibility of silicone implants," *Annals Plastic Surgery*, vol. 11, issue 1, pp. 38-45, 1983.

[17] V. Wong, A. Huang, and C.-M. Ho, "SU-8 lift-off patterned silicone chemical vapor sensing arrays," in *Proc. 18th Int. Conf. IEEE MEMS*, Miami, FL, USA, 2005, pp. 754-757.

[18] J. M. Engel, N. Chen, K. Ryu, S. Pandya, C. Tucker, Y. Yang, and C. Liu, "Multi-layer embedment of conductive and non-conductive PDMS for all-elastomer MEMS," in *Tech. Dig. 12th Hilton Head*, Hilton Head Island, SC, USA, 2006, pp. 316-319.

[19] T. Fujigaya, S. Haraguchi, T. Fukumaru, and N. Nakashima, "Development of novel carbon nanotube/photopolymer nanocomposites with high conductivity and their application to nanoimprint photolithography," *Adv. Mater.*, vol. 20, issue 11, pp. 2151-2155, 2008.

[20] H. Cong and T. Pan, "Photopatternable conductive PDMS materials for microfabrication," *Adv. Funct. Mater.*, vol. 18, issue 13, pp. 1912-1921, 2008.

[21] D. Qian, E. C. Dickey, R. Andrews, and T. Rantell, "Load transfer and deformation mechanisms in carbon nanotube-polystyrene composites," *Appl. Phy.s Lett.*, vol. 76, issue 20, pp. 2868-2870, 2000.

[22] J. S. Bergstrom and M. C. Boyce, "Large strain time-dependent behavior of filled elastomers," *Mech. Mater.*, vol. 32, issue 11, pp. 627-644, 2000.

[23] D. E. Hanson, "An explicit polymer and node network model to compute micromechanical properties of silica-filled polydimethyl-siloxane," *Polymer*, vol. 45, no. 3, pp. 1055-1062, 2004.