NANOCOMPOSITE RUBBER ELASTOMER WITH PIEZORESISTIVE DETECTION FOR FLEXIBLE TACTILE SENSE APPLICATION

J.-J. Wang¹, C.-E. Lu², J.-L. Huang¹, R. Chen^{1, 2}, and W. Fang^{1, 2}

¹Department of Power Mechanical Engineering, National Tsing Hua University, Hsinchu, Taiwan ²Institute of NanoEngineering and MicroSystems, National Tsing Hua University, Hsinchu, Taiwan

ABSTRACT

This work presents flexible tactile sensor using piezoresistive nanocomposite rubber elastomers (C-PDMS): Carbon-black (C) powders filled into polymethylsiloxane (PDMS) using solvent-wetting methods with n-Hexane to produce well-dispersed conductive polymeric nanocomposites. The C-PDMS nanocomposites are utilized to measure resistance changes. In application, the C-PDMS is casted and stacked into PDMS substrate and gold films deposited with 3-mercaptopropyltrimethoxysilane (MPTMS) adhesion layers as bond pads to implement the flexible polymer-based devices. Typical normal load tests show resistance changes of C-PDMS piezoresistors on flat surface are ranging 2.92%~4.45% (ΔR/R₀/mN). Moreover, resistance changes of C-PDMS piezoresistors on curved surface are ranging $0.92\% \sim 0.98\%$ ($\Delta R/R_0/mN$).

INTRODUCTION

Various types of tactile sensors have been developed for the applications of robotic and prosthetic over the past few years. For this aim, flexible and skin-attachable piezoresistive electronics have been highly needed and rapidly utilized for deformable, bendable, stretchable, foldable, and wearable devices [1].

Tactile sensors have been employed in a variety of robotic applications to manipulate and detect objects. It is important to measure and control with an optimal force between targets and robot-hands in the next generation [2]. While the objects touch tactile sensors along the interface between the target and the robotic hand, these sensors can detect not only force-induced strains resulting from pressures applied, but also strengths and positions by analyzing the distributions of the loads [3-4].

In this regard, the large-area flexible piezoresistive sensors are rapidly developed and integrated into artificial e-skins to detect the strength and position of forces applied on the surface of objects [5]. Piezoresistive force sensor can detect the applying loads by the variations of electrical resistance. Piezoresistive force sensors (strain gauge) have been widely utilized due to various advantages such as feasible preparation, low cost, and easy signal collection [6]. Polymer filled with conductive particles can be employed to act as the sensing materials for the applications of force detections. Such polymer-based composites exhibits the piezoresistor characteristics [7], and thus could convert the mechanical load to electrical signal for force detection.

The feasibility of a stretchable sensors using C-PDMS is reported in [8]. Thus, this study extends the concept to achieve the well dispersion and adhesion of C-PDMS, metal films, and polymer substrates. In addition, the 3D-microstructures are further embedded into stretchable substrates. Moreover, the wafer-level polymer molding

and bonding processes are exploited to implement piezoresistive nanocomposite rubber elastomers for flexible tactile sense applications.

DESIGN AND PRINCIPLE Design Concept

Fig. 1 illustrates the schematic design and typical construction of the proposed flexible tactile sensor. In Fig. 1(a), the piezoresistive nanocomposite rubber elastomer which comprises four individual C-PDMS piezoresistors with meander-like patterns (\sim 19000 \times 500 \times 75 μ m³) as the sensing layer, metal films (Au/MPTMS \sim 200 nm), the PDMS tactile bump (\sim 7.5 \times 7.5 \times 1 mm³), and a stretchable substrates as flexible supports (\sim 18 \times 18 \times 1 mm³). The cross-sectional view of the embedded piezoresistors of this device for the force-induced strain sensing are designed in this study, as shown in Fig. 1(b).

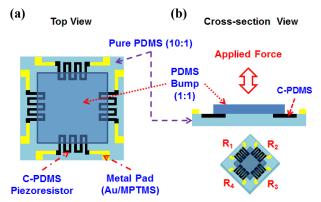


Figure 1: Schematic illustrations and design concepts of the proposed flexible tactile sensor using the piezoresistive nanocomposite rubber elastomers.

Operating Principle

Fig. 2 depicts working principles of piezoresistive characteristics and tactile sensing mechanisms. The C-PDMS nanocomposite is utilized to measure resistance changes while substrates deformed (e.g. bending, tension, etc.) by external forces due to the conductive path changed inside the polymeric matrix, as shown in Fig. 2(a). In short, the C-PDMS acts as the electrical resistors of the sensing layer. The merit of this device is to extend the multi-casting and multi-stacking processes to implement piezoresistive nanocomposites into seamless homogeneous stretchable polymer substrates, and thus leverage the advantages of PDMS such as good homogeneous interface of adhesion, bonding and cross-linking of each molecules, and monomers between solid and liquid phases [9]. In Fig. 2(b), the electrical resistances of C-PDMS will be changed as the proposed sensor deformed by external loads (i.e. piezoresistive effect of C-PDMS piezoresistors). Note the load is applied on the tactile bump.

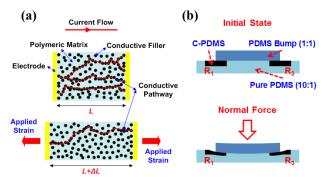


Figure 2: Illustrations of working principles, (a) piezoresistive behaviors, (b) tactile sensing mechanisms.

In this study, the processes to implement highly sensitive and flexible piezoresistive materials have been developed. As such, the proposed piezoresistive C-PDMS rubber elastomers in this study can be readily integrated to serve as soft, flexible, and highly sensitive electromechanical transducers, which are desired for a variety of MEMS sensors and applications. The proposed flexible tactile sensors are fabricated and measured to demonstrate their feasibility subsequently.

FABRICATION PROCESSES

Material Preparation

Fig. 3 illustrates the preparation of solvent-wetting C-PDMS nanocomposites. In Fig. 3(a)-(b), the desired 30 wt% of carbon black powders and PDMS base polymers were respectively mixed into the specific weight ratio of n-Hexane [10] for 2 hours. In Fig. 3(c), after mixing the C/n-Hexane and PDMS/n-Hexane for 1 hour, the n-Hexane was then heated up and fully evaporated overnight. In Fig. 3(d)-(e), the C-PDMS nanocomposite was prepared after adding curing agents (10:1 base polymer to curing agent).

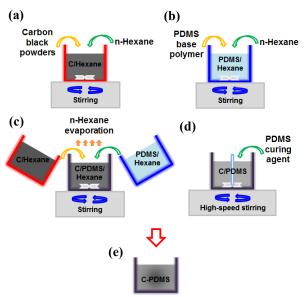


Figure 3: Fabrication processes of the solvent-wetting C-PDMS nanocomposite blends.

Device Fabrication

Fig. 4 illustrates micro-fabrication and assembly processes to implement the device. In Fig. 4(a)-(c), the 1H,

1H, 2H, 2H-perfluorooctyltrichlorosilane (PFOTS) anti-adhesion monolayer was deposited onto Si substrate [9], and the C-PDMS was then patterned on the substrate through the shadow mask using screen-printing processes [11]. In Fig. 4(d)-(e), a thicker PDMS layer was then casted on the Si substrate with patterned C-PDMS layer. After removed the Si substrate, the patterned C-PDMS layer was transferred into PDMS substrate. In Fig. 4(f)-(g), the evaporated gold thin film and MPTMS monolayer [12] were deposited onto PDMS substrates (patterned by shadow mask) to define the bond pads. Finally, PDMS tactile bumps were added onto PDMS substrates by plasma bonding processes to accomplish the tactile device, as in Fig. 4(h). Note that other 3D structures can be achieved by using the multi-casting and multi-stacking processes.

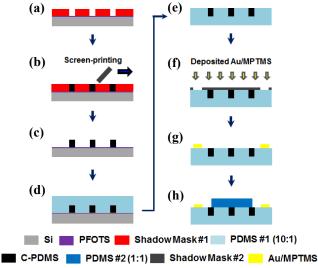


Figure 4: Schematic fabrication processes of the piezoresistive nanocomposite rubber elastomers.

RESULTS AND DISCUSSION

Morphological Feature

The photographs in Fig. 5(a)-(c) respectively show the fabrication results corresponding to Fig. 4(c), 4(e), and 4(g). Fig. 5(a) shows the C-PDMS nanocomposites after screen-printing processes to deposit and pattern onto the surface of the treated Si substrate.

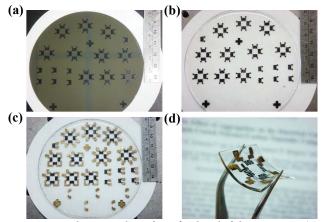


Figure 5: Photographs of wafer-level fabrications, (a) C-PDMS patterned on Si wafer, (b) multi-stacking, molding and transferring to flexible-supports, (c) metal films deposition, and (d) a fabricated flexible device.

In Fig. 5(b), the PDMS substrate with patterned C-PDMS was peeled off from the silicon mold. The wafer-level metal deposition on the PDMS substrate with patterned C-PDMS is shown in Fig. 5(c). The evaporated gold bond pads and the patterned C-PDMS are clearly observed. The photograph in Fig. 5(d) demonstrates the final fabricated flexible sensing device which is ready for the following load-deflection tests.

The SEM micrographs in Fig. 6 further display the zoom-in of the C-PDMS piezoresistor embedded in PDMS substrate. By utilizing the multi-casting and multi-stacking processes to transfer the nanocomposites onto the flexible polymer substrates, the fabricated C-PDMS piezoresistors are embedded into the PDMS substrates consequently (as in Fig. 6(a)), and finally polymerized each other after curing, as shown in Fig. 6(b).

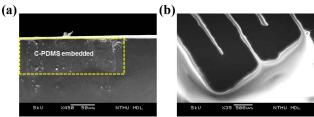


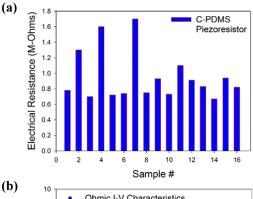
Figure 6: SEM photographs of embedded C-PDMS piezoresistors, (a) cross-sectional, and (b) top view.

Electrical Characterization

Fig. 7 shows electrical properties and ohmic I-V characteristics C-PDMS nanocomposites. Measurements in Fig. 7(a) show the resistance of C-PDMS piezoresistors varying with different fabricated samples. The average electrical resistance of C-PDMS piezoresistor was approximately ~ 0.95 M Ω (the correspondent resistivity (p) was approximately $\sim 1.88 \ \Omega$ -m). Fig. 7(b) shows the measured current flow after applying a DC-bias on C-PDMS nanocomposites. Due to the conductivity of C-PDMS nanocomposites, the current was measured when applying a DC-bias voltage on the sample through a source meter (Keithley 2400). Moreover, the slop depicts that the relative ohmic I-V characteristics of the C-PDMS piezoresistor was approximately $\sim 1.53 \, \mu A/V$.

Sensor Performance

The tactile loading tests and the measured corresponding fractional changes in electrical resistances $(\Delta R/R_0)$ are shown in Fig. 8. The force gauge (FSH-5N, YOTEC) to specify as well as monitor tactile loads has a probe with a ~5mm diameter tip to contact the tested sensors. Moreover, the loads were applied on the tested sensors through the tactile bump, and the output signals were recorded by a commercial source meter (Keithlev 2400). Firstly, this study performed the tactile loading tests on a flat flexible sensor. As depicted by the photo in Fig. 8(a), the flexible sensor attached to a rigid flat surface during the loading tests. Measurements in Fig. 8(a) depict the resistance change $\Delta R/R_0$ varying with the applied normal loads. The tactile response of C-PDMS piezoresistors (R₁ ~ R₄) versus normal loads were respectively ~4.3%, ~2.92%, ~4.36% and ~4.45% $(\Delta R/R_0/mN)$ in average within the effective sensing range of $0 \sim 15$ mN.



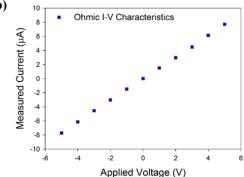
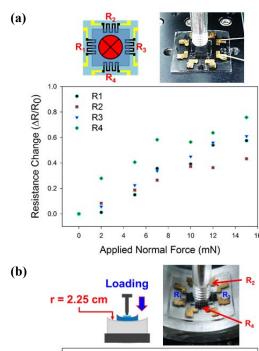


Figure 7: Measured electrical properties of fabricated devices, (a) electrical resistances, (b) I-V characteristics.



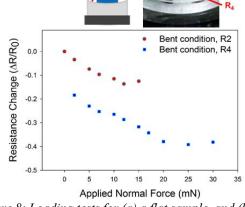


Figure 8: Loading tests for (a) a flat sample, and (b) a bent sample on a curved surface (r = 2.25 cm).

This study also performed the tactile loading tests on a bent flexible sensor. During the tests, the flexible sensor was attached to a curved surface to specify its radius of bending curvature (r = 2.25 cm). The measurements in Fig. 8(b) show the fractional changes in electrical resistances of R₂ and R₄ piezoresistors with applied normal loads. Measured results depict that the sensitivities of R₂ and R₄ piezoresistors respectively dropped to $\sim 0.98\%$ and $\sim 0.92\%$ $(\Delta R/R_0/mN)$ within the effective sensing range in average of $0 \sim 15$ mN. Thus, the sensitivity of the presented flexible sensor may significantly decrease for several folds when attached to a curved surface. Moreover, the piezoresistors of R₂/R₄ and R₁/R₃ are aligned in different directions, as depicted in Fig. 1. The pre-bent of the piezoresistors in Fig. 8(b) will cause different influence on the R_2/R_4 and R_1/R_3 piezoresistors. These issues will be further investigated and solved in the future. In summary, the deformation which resulted from normal loads through the tactile bump is mainly the longitudinal elongation of each C-PDMS piezoresistor. The measurement results demonstrate the feasibility of the presented flexible piezoresistive nanocomposite rubber elastomers for force detections of tactile sense applications. Measurements also demonstrate that the proposed device chips can serve as bendable, foldable, and wearable applications, even further as touch sensing in the robotic and prosthetic fields.

CONCLUSION

This work presents flexible tactile applications using piezoresistive nanocomposite rubber elastomers, which are black (C) powders (26nm) filled polymethylsiloxane (PDMS 10:1, Sylgard 184) using solvent-wetting methods with n-Hexane to produce well-dispersed conductive polymeric nanocomposites. This device chip has the following characteristics and advantages: (i) nanocomposites directly homogeneously deposited on stretchable polymer substrates by multi-casting and multi-stacking processes, (ii) short processing time for rapid prototyping, (iii) available for large-area and low-cost fabrication, and to demonstrate its functions and tactile sense applications by the force-induced strain sensing of normal loads.

For the prototype demonstration, the measured resistivity (ρ) of 30 wt% solvent-wetting C-PDMS nanocomposites was ~1.88 (Ω -m). Measurements in electrical resistance changes of C-PDMS piezoresistors (R₁~R₄) versus normal loads were respectively ~4.3%, ~2.92%, ~4.36%, and ~4.45% (Δ R/R₀/mN) while the flexible sensor attached to a flat surface. Further measurements of electrical resistance changes of R₂ and R₄ (under normal loads) respectively become ~0.98% and ~0.92% (Δ R/R₀/mN) while the flexible sensor attached to a curved surface (r = 2.25 cm). In conclusion, the proposed piezoresistive nanocomposite rubber elastomers can function as flexible MEMS sensors, and has the potential to realize the touch sensing for robotic and prosthetic applications.

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REFERENCES

- [1] J.-H. Kong, N.-S. Jang, S.-H. Kim, and J.-M. Kim, "Simple and Rapid Micropatterning of Conductive Carbon Composites and Its Application to Elastic Strain Sensors", *CARBON*, vol. 77, pp. 199-207, 2014.
- [2] K. Noda, K. Hoshino, K. Matsumoto, and I. Shimoyama, "A Shear Stress Sensor for Tactile Sensing with The Piezoresistive Cantilever Standing in Elastic Material", Sens. Actuators A, Phys., vol. 127, pp. 295-301, 2006.
- [3] H. Takahashi, A. Nakai, N. Thanh-Vinh, K. Matsumoto, and I. Shimoyama, "A Triaxial Tactile Sensor without Crosstalk Using Pairs of Piezoresistive Beams with Sidewall Doping", Sens. Actuators A, Phys., vol. 199, pp. 43-48, 2013.
- [4] R. S. Dahiya and M. Valle, *Robotic Tactile Sensing*, Springer Netherlands, 2013.
- [5] H.-B. Yao, J. Ge, C.-F. Eang, X. Wang, W. Hu, Z.-J. Zheng, Y. Ni, and S.-H. Yu, "A Flexible and Highly Pressure-Sensitive Graphene-Polyurethane Sponge Based on Fractured Microstructure Design", *Adv. Mater.*, vol. 25, pp. 6692-6698, 2013.
- [6] N. Lu, C. Lu, S. Yang, and J. Rogers, "Highly Sensitive Skin-Mountable Strain Gauges Based Entirely on Elastomers", Adv. Funct. Mater., vol. 22, pp. 4044-4050, 2012.
- [7] X.-Z. Niu, S.-L. Peng, L.-Y. Liu, W.-J. Wen, and P. Sheng, "Characterizing and Patterning of PDMS-based Conducting Composites", *Adv. Mater.*, vol. 19, pp. 2682-2686, 2007.
- [8] J.-J. Wang, M.-Y. Lin, H.-Y. Liang, R. Chen, and W. Fang, "Piezoresistive Nanocomposite Rubber Elastomer for Stretchable MEMS Sensor", *Proc. IEEE MEMS'16*, Shanghai, Jan. 24-28, 2016, pp. 550-553.
- [9] M. Zhang, J. Wu, L. Wang, K. Xiao, and W. Wen, "A Simple Method for Fabricating Multi-layer PDMS Structures for 3D Microfluidic Chips", *Lab on a Chip*, vol. 10, pp. 1199-1203, 2010.
- [10] M. Hussain, Y.-H. Choa, and K. Niihara, "Fabrication Process and Electrical Behavior of Novel Pressure-sensitive Composites", *Composites: Part A*, vol. 32, pp. 1689-1696, 2001.
- [11] A. Teichler, J. Perelaer, and U. S. Schubert, "Inkjet Printing of Organic Electronics - Comparison of Deposition Techniques and State-of-the-art Developments", J. Mater. Chem. C, vol. 1, pp. 1910-1925, 2013.
- [12] C. A. Goss, D. H. Charych, and M. Majda, "Application of (3-mercaptopropyl)trimethoxysilane as a Molecular Adhesive in the Fabrication of Vapor-deposited Gold Electrodes on Glass Substrates,", Anal. Chem., vol. 63, pp 85-88, 1991.

CONTACT

*W. Fang, tel: +886-3-5742923; fang@pme.nthu.edu.tw