A PAPER-BASED DISPOSABLE STRAIN SENSOR BY DIRECT LASER PRINTING

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ABSTRACT
A paper-based, disposable strain sensor by means of a direct laser printing process with high sensitivity has been developed and demonstrated. Commercially-available printing papers are first soaked with the gelatin-Mo\textsuperscript{5+} ink and ablated by a CO\textsubscript{2} laser to convert molybdenum ions into conductive MoC and graphene nanocomposite flakes for disposable paper electronics. The strain sensor made of continuous MoC flakes has the capability of detecting and distinguishing both tensile and compressive strain. It is found that the stability of the sensor is preserved when there are defects in the device as well as in environments with heavy humidity for potential applications in wearable devices.

INTRODUCTION
Flexible and disposable strain sensors have attracted great attentions in recent years because of their potential applications in wearable systems, while graphene-based strain sensors have been a key focus area by different groups [1]–[4]. In these aforementioned research results, either tensile or compressive strain measurements with low gauge factors have been reported. Furthermore, some of the sensors use rather complex fabrication processes [5]. Here, we report a sensor with the capability of detecting and distinguishing both tensile and compressive strain using an easy fabrication method by direct laser writing.

Materials with the foam structure usually show good performances in strain sensing applications [6]–[8] because the foam structure can lead to stress concentration [9] or larger deformation [10] effects, compared to solid structures. Here, the direct laser writing method is used to make porous and conductive metal carbide-graphene composites on paper substrates for the strain sensing application, which adds to various prior demonstrations of the same scheme for applications in supercapacitors, energy harvesters and heavy metal detectors [11].

FABRICATION
The Gelatin-Mo\textsuperscript{5+} ink is prepared by adding MoCl\textsubscript{5} (2mol/L, Sigma-Aldrich, 95wt%) and Gelatin (60wt%, Sigma-Aldrich, porcine from skin, type A) in deionized water with continuously stirring under 65 °C until the whole system becomes homogeneous. Afterwards, the commercially available printing paper (Boise Aspen 30, thickness around 90 μm) is coated with the Gelatin-Mo\textsuperscript{5+} ink, both of which serve as precursors. Under the laser printing process (CO\textsubscript{2} laser by Tek Motion, wavelength of 10.6 μm), the mixture of paper and gelatin solution is transformed to the molybdenum carbide-graphene (MCG) composites, as shown in Figure 1(a). The laser power is controlled to only convert the top portion of the gelatin solution. Figure 1(b) shows the fabrication sequence from a white-color paper to the yellow-color paper after adding the gelatin solution and to the black-color after the laser writing process. On the other hand, the back side of the paper is still showing the white color as the laser power didn’t penetrate the paper. The sheet resistance of the composites is influenced by the laser power and the scan rate. Figures 1(c) and (d) show the experimental characterization results for sheet resistance. In general, the
resistivity decreases as the laser power decreases and increases as the scan rate increases as both higher laser power and slower scan rate can provide more energy into the precursors for better and more conversions. The optimal result shows that when the laser power is at 4 W and the scan rate is at 250nm/s, the composite sheet resistance has a relatively low value at 4 kΩ/□. This processing condition is adopted in the rest of this work.

The micro-morphology of MCG composites is characterized by the scanning electron microscope (SEM) with the top view (Figure 1(e)) and side view (Figure 1(f)). It is seen that MCG composites have porous coral-shape microstructures with pore sizes ranging from 10 to 50 μm. It is also observed that the thickness of the MCG composite and the paper substrate is 76.4 μm and 65.5 μm, respectively or about 27% of the paper (24.5 out of 90 μm in thickness) is converted to MCG composites, while the porous MCG expands 312% from the original thickness of 24.5 μm to the final thickness of 76.4 μm.

SENSING MECHANISM

MCG composites can be utilized for strain sensing application based on the cracked sensing mechanism [12]–[15]. This is examined by SEM images. Figures 2 (a) and (b) are the images of original MCG composites. Under a low tension force (Figures 2 (e) & (d)), small micro-cracks marked with red lines and circles, were formed on the surface driven by the external strain. These micro-cracks may break and block electrical current, leading to the increase in electrical resistance. Some other structures (the green circle) remain to be intact to conduct the electricity. However, a high external tension (Figures 2(e) & (f)) can generate more micro-cracks to break more structures (red lines and circles) to result in the increase of the electrical resistance.

On the other hand, under the external compressive strain, the foam structure is collapsed and MCG sheets overlap with each to increase the current paths and reduce the resistance. This is similar to the reported mechanism of some porous graphene/polymer composites [16]–[18].

RESULTS

SENSING PROPERTIES

We use the sensitivity and repeatability to evaluate the sensing performances of the MCG composite strain sensors. The gauge factors (GF) is used to quantify the sensitivity, which is defined as:

\[
GF = \frac{\delta(R/R_0)}{\delta\varepsilon}
\]

where \(\varepsilon\) is the strain; \(R\) is the resistance under a load; \(R_0\) is the original resistance. Therefore, GF can be characterized by plotting different resistance response values and their corresponding strain values in a graph to obtain the fitting slope.

We have used the curved state to test the strain sensor and the strain value is defined by the curvature and derived from the geometric relationship and formula:

\[
c = 2r \sin \left(\frac{\delta}{2}\right)
\]

\[
\varepsilon = \pm \frac{h}{2r}
\]

where \(l\) is the original length of samples; \(c\) is the chord length (which is controlled in experiments); \(r\) is the radius; \(h\) is the thickness of samples; “+” represents tension; and “-” represents compression. In our tests, \(l\) is 4 cm and \(c\) has the range from 3.0 to 3.9 cm with 0.1 cm as the interval. The strain state is determined by the bending direction.

![Figure 3: Sensing properties The sensitivity of the MCG composite sensor under (a) tension and (b) compression. The repeatability of the MCG composite sensor under 1000 cycles of: (c) tension and (d) compression tests.](image)

Figures 3(a) and (b) show the measured GFs of prototype sensors under different strain conditions, which are 73 under tension and 43 under compression. These are comparable or higher than those in reported works [1]–[4],[8]. Furthermore, it is observed that the resistance responses show good linearity within a wide range, which is an advantageous in practical applications [19].

Another important feature for the strain sensor is the stability. Here, the bending experiments were carried out
1000 cycles under both tensile and compressive states. For the tension test, the resistance response was observed to have a slight reduction at the beginning. After 200 seconds, it became relatively stable as shown in Figure 3(c) as micro-cracks were formed and expanded at the first 200s and the process became stabilized. Similar phenomena can be observed for the repeatability test under the compressive strain (Figure 3(d)).

APPLICATIONS

The application potential of the MCG composite sensors is further explored. Figures 4(a) and (b) are the two examples of the sensor used in wearable devices with good flexibility to enable good contacts with the human skin. In experiments, our sensors can be easily attached to the skin by using double-sided tapes for good convenience in real-life. For example, the sensor can be attached to human canthus to monitor the eye blinking signs. Figure 4(a) shows the resistance response of the sensor when detecting human wrist movements, where the sensor is attached to the backside of the wrist. The resistance changes differently against the movement direction, which can demonstrate that the sensing capability of the sensor to distinguish different strain types. Figure 4(b) shows a more complex strain condition, where the sensor is attached to the second knuckles on the back of the hand. The resistance changes of universal hand gestures for various numbers, from zero to five, were recorded. From these two demonstration examples, it is observed that the resistance change is opposite under the tensile and compressive strain conditions. Hence, it is clear that our sensor can distinguish either tensile or compressive strain directly, and is also functional for applications in rather complicated systems.

Meanwhile, the MCG composite sensor can detect small vibrations. For example, the sensor was attached at the microphone of one mobile phone (which has an app that can generate piano sound with different frequencies). The resistance responses were recorded when the mobile phone gave out different piano scales, like D4 and F4, at the same volume as shown in Figures 4(c) and (d), respectively. It is observed that the sensor has different response patterns with respect to different input sound signals. Hence, the MCG composite sensor is also able to distinguish small vibration signals.

DISPOSABILITY & ENVIRONMENT STABILITY

One possible feature of this paper-based sensor is the disposability. From the perspective of material and process, paper is not expensive and the direct-laser writing method is simple and repeatable to have low manufacturing cost. At the same time, as it is shown in Figure 5(a), this sensor can be burnt with only some ashes left (molybdenum carbide is non-flammable).

![Figure 5: Disposability and Environment Stability](image)

(a) The photograph of the sensor before and after being burnt, and its environmental stability tests – (b) line-cut and hole-cut; (c) stability in the wet water environment.

In addition, as a potential wearable device in real-life applications, some externally-induced defects may occur. Here, artificially introduced defects are produced such as a small line-cut and a hole-cut on the device. It is found that there were no obvious differences of the relative resistance changes (Figure 5(b)). This demonstrates that MCG sensors are capable of resisting potential external wear and tear during operations. Furthermore, since people may sweat and affect the operation of the sensors, the humidity test of the sensor is conducted by spilling some water on the sensor. The resistance of the sensor before and after water spilling was measured. It is found that the influence of water on the resistance is negligible with only 0.15% resistance changes (Figure 5(c)), which can be attributed to the hydrophobicity of graphene [20]. As such, we show the paper-based strain sensor has very good stability to resist changes in different environmental conditions and defects.

CONCLUSIONS

In conclusion, we have used the direct-write laser printing method to produce arbitrary patterns of MCG composites with form structures for strain sensing.
applications. Experimental results show the sensors have good sensitivity and stability and the ability to differentiate tensile and compressive strain states. These flexible sensors could find potential applications in wearable devices as well as the recognition of sound from a piano. The possible disposability and stability in humid environment are two other advantages of the MCG composites material for potential practical applications.

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