A facile approach to fabricate graphene based piezoresistive strain sensor on paper substrate

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Sensors, FETs and chemi resistors are few of the devices which show potential in the area of flexible electronics for health monitoring applications. In the present work, piezoresistive strain sensors based on graphite and graphene on cellulose paper substrate has been reported. Graphite sensor has been fabricated by rubbing pencil on paper and graphene sensor by directly coating graphene ink using paint brush. The resistance of the fabricated sensor increases with outwards bending and vice-versa, further the piezoresistive effect has also been evaluated by applying variable longitudinal stress. A comparative study of gauge factor (GF) depending upon different type of strains has been presented and it has been observed that the GF of graphene piezoresistive strain sensor decreases with increase in number of layers, the GF for graphene sensor is higher as compared to graphite sensor. Fabricated piezoresistive strain sensors may find applications as human body motion detection, gait analysis etc.

1 Introduction

Piezoresistive strain sensors or strain gauge are one of the most important devices in the application area of wearable sensor[1], human body motion detection[2], vibrations detection[3], gait analysis[4], strain sensors[5,6], etc. These devices work on the principle of piezoresistive effect in which stress/strain is converted into a resistive change by the sensing material that can be easily detected by electrical measurements. In the literature, numbers of piezoresistive strain sensors have been fabricated using variety of materials, such as metals, semiconductors and composites on various rigid and flexible substrates[7-10]. The conventional silicon-based force sensor uses a cantilever beam for providing stress/strain, but the limitation of these sensors is that they are rigid and fragile in nature which limits its application in flexible electronics applications. In order to fabricate flexible sensors, many attempts have been made using flexible and stretchable substrate materials[11-15]. In this regard, two allotropes of carbon, namely graphite and graphene have emerged as promising materials with piezoresistive effects. Graphite is a three dimensional hexagonal

structure with irregular edges. Easiest way to obtain graphite layer is by means of graphite pencils. The pencil consists of varying mixture of macroscopic graphite particle with clay[16]. The amount of clay decides the hardness and the conductivity of pencil traces. Graphene also exhibits piezoresistive effect along-with other superlative material properties like high electrical conductivity[17], superior mechanical flexibility and stretch ability[18], etc. It is a single atom thick layer of carbon atoms arranged in honeycomb lattice structure. It can be easily deposited on paper substrate which is advantageous from the point of view of portability, cost effectiveness, environment friendly and exhibits piezoresistive effect[19,20]. Although both graphite and graphene have drawn a lot of attention to the researchers for fabrication of piezoresistive sensors and FET[21] but due to its fascinating properties graphene based piezoresistive strain sensors have upper edge.

In the present work, a facile approach to efficiently fabricate the piezoresistive strain sensors based on graphite and graphene on cellulose paper substrate has been demonstrated. The comparative study of graphite and graphene based piezoresistive sensor with distinct stresses is presented. The GF of graphene sensor is found to be 20.47 which is the highest

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among previously reported large geometric sensors to the best of our knowledge.

2 Experimental

2.1 Device Fabrication

Cellulose paper was used as a substrate for the realization of flexible strain sensors. For fabricating graphite based piezoresistive sensor, pencil lead (10B) was exfoliated on cellulose paper and the graphite particles adhere to the paper fibres thus forming a resistive layer. Multiple graphite coatings were applied in order to make a conductive trace.

Copper contacts were used on both the ends to realize good electrical connection for resistance measurement purposes. The dimension of fabricated piezoresistive sensor was 70 mm×10 mm. Figure 1(a,b) shows the deposition method of graphite on cellulose paper and fabricated graphite based piezoresistive

strain sensor. A 10B-pencil, lead-diameter 2 mm was used. The area $(70 \times 10 \text{ mm}^2)$ to be coated was scaled and the 10 mm width was divided into 5 equal columns of 2 mm each. In order to cover the whole area, 5 traces of pencil along the length were made and whole of the process was repeated 5 times.

The fabrication of graphene based piezoresistive strain sensor started from preparation of graphene ink. Graphene was prepared by liquid phase exfoliation method from raw graphite[22]. Then the graphene layer was directly coated on paper by using paint brush. Several coatings of graphene layers (such as 2, 4 and 6) were applied. For the precise coating of graphene layer a rectangle of desired length/width (70 mm/ 10 mm) was marked on the paper substrate and the marked area was brush coated by graphene ink as shown in Fig. 2(a,b).
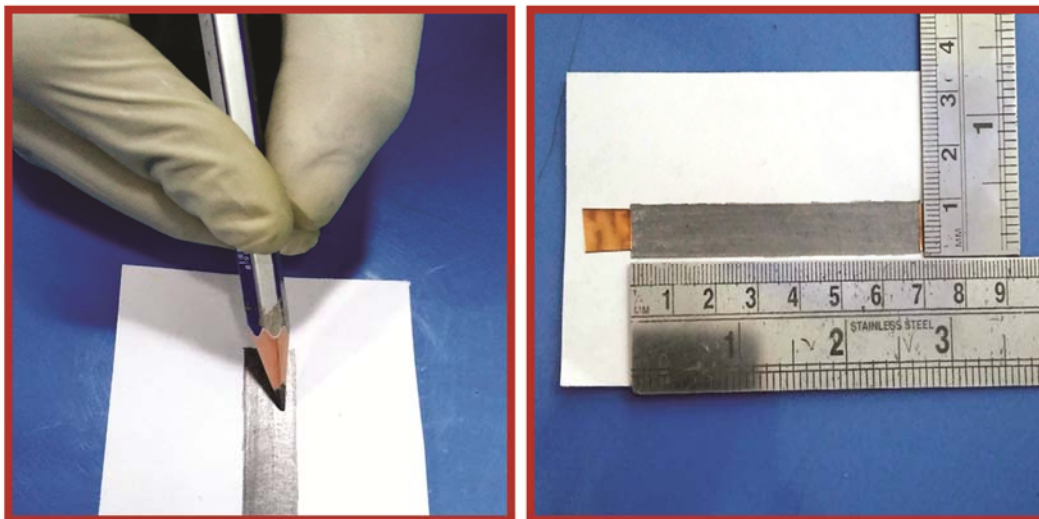

Fig. 1 – (a) Deposition of graphite pencil layer on paper substrate and (b) fabricated graphite based piezoresistive strain sensor.
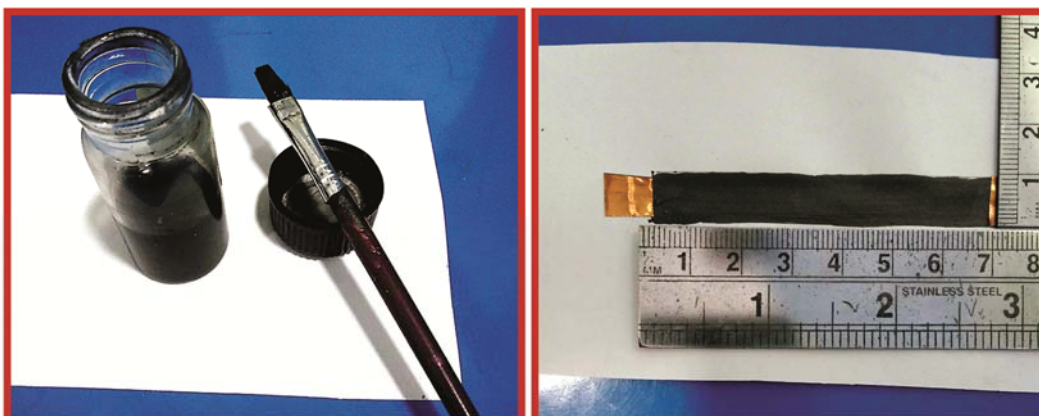

Fig. 2 – (a) Graphene ink and (b) schematic structure of graphene based piezoresistive strain sensor where graphene is used as sensing material.

2.2 Characterization of carbon traces deposited on paper

The surface morphology of the conductive traces prepared from graphite and graphene materials were analysed by field emission scanning electron microscope (FE-SEM). The images were recorded using Hitachi SU8000 FESEM (Hitachi High-Technologies Corporation, Japan) as shown in the Fig. 3. The recorded image of cellulose paper has shown in Fig. 3(a) which reveals its porous nature essential for proper adhesion of the layers to be deposited. The surface morphology of graphene and graphite coated paper is shown in Fig. 3(b,c), respectively. In order to analyze the continuity of the deposited layers the SEM images were recorded. Raman characterization (Renishaw Raman Spectrometer) of the graphite and graphene resistive layers has been done using 785 nm wavelengths as an excitation source. The Raman spectrum shows the characteristics of D and G bands for graphite and graphene layers (Fig. 3(d,e)). It is observed that the ratio of D/G bands in graphene is lower than graphite which indicates less defects and high quality graphene.

3 Results and Discussion

The fabricated sensors were subjected to the tensile stress by bending outward and inward directions. In graphite and graphene layers, during outward bending the interlayer spacing increases which results in cracks occurred between the layers and led to increase in the resistance of sensors[1]. On the contrary, when the sensor was subjected to compression by bending it inward, the overlapping of graphite and graphene layers results in decrease of resistance. Relative resistive change for the piezoresistive strain sensor was calculated using following relation:

$$\Delta R/R = (R_o - R)/R \qquad \text{... (1)}$$

Where, R_o is resistance in bending state and R represents resistance of the sensor in relaxed state. The resistance values were measured as a function of chord length, which is the arc length of the sensor under bending state. These measurements were performed using Keithley 2450 source meter. The resistance of sensors was measured by changing the chord length from 70 mm to 20 mm as shown in Fig. 4.

Figure 4(a,b) shows the tension and compression resistance with different chord length for graphite sensor while Fig. 4(c,d) exhibits the change in resistance of graphene sensor with tension and compression for different layers of coatings. It was observed that the resistance of the piezoresistive strain sensors depends on the bending directions. The
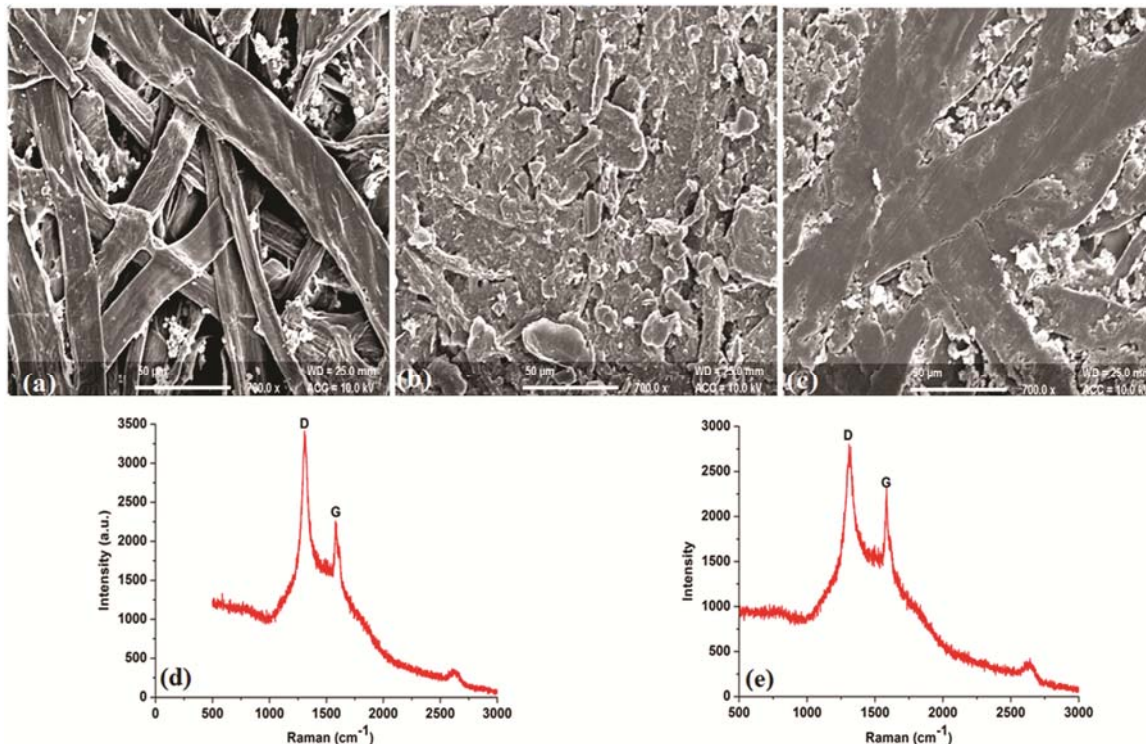

Fig. 3 – (a) SEM image of cellulose paper shows the cellulose fibres, (b) SEM image of graphene coated paper, (c) SEM image of graphite coated paper, (d) Raman spectrum of graphite and (e) Raman spectrum of graphene.
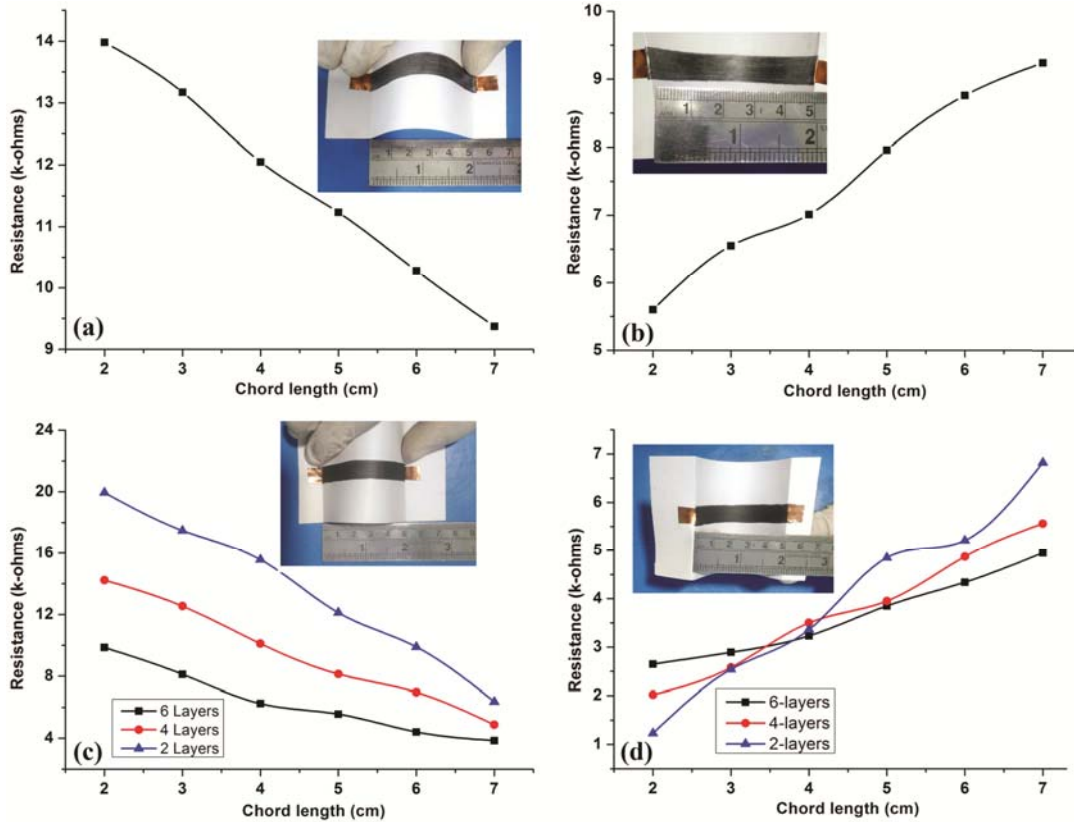
Fig. 4 – (a) Tension, (b) compression for graphite based piezoresistive strain sensor, (c) tension and (d) compression of graphene based piezoresistive strain sensor.

resistance of the sensor increases by bending it outward (tension) and decreases by bending it inward (compression) with bending limit as 38°. A relation between the change in resistance as a function of chord length can be derived from this observation.

The sensitivity of the piezoresistive strain sensor to mechanical deformation was analysed by calculating gauge factor (*GF*), which is defined as the ratio of change in resistance to strain[23]:

$$GF = (\Delta R/R)/\varepsilon \qquad \qquad ... (2)$$

Where, $\Delta R/R$ is the relative change in normalized resistance and ε is the mechanical strain.

The value of ε depends upon the type of stress applied to the sensor and can be computed as

For longitudinal stress applied to sensor[24,25]:

$$\varepsilon = \Delta L/L \qquad \qquad ... (3)$$

Where ΔL change in length is after mechanical stress applied to the sensor, *L* is the length of sensor.

For compression and tension applied to sensor[26]:

$$\varepsilon = h/2r \qquad \qquad ... (4)$$

Where *h* is thickness of the sensor and *r* is radius of the curvature. The value of *r* can vary depending upon
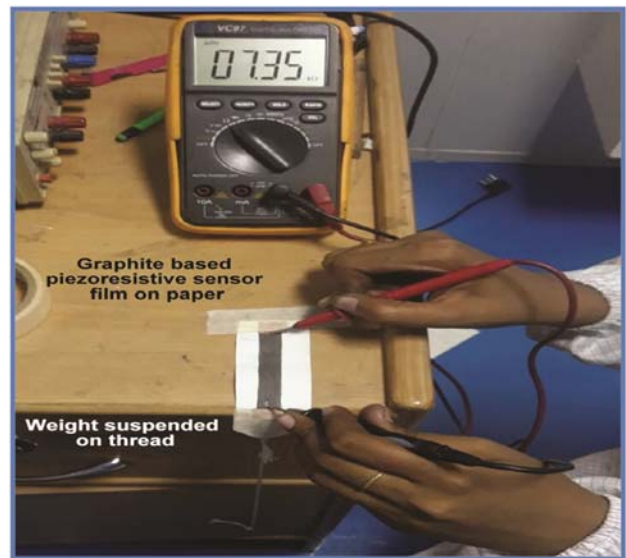

Fig. 5 – Piezoresistive sensor with applied longitudinal stress.

the arc and chord length of the sensor during tension and compression.

As shown in Fig. 5, longitudinal stress is applied to the sensors using different weights of 5 g, 10 g, 20 g and 50 g at one end keeping the other end fixed using
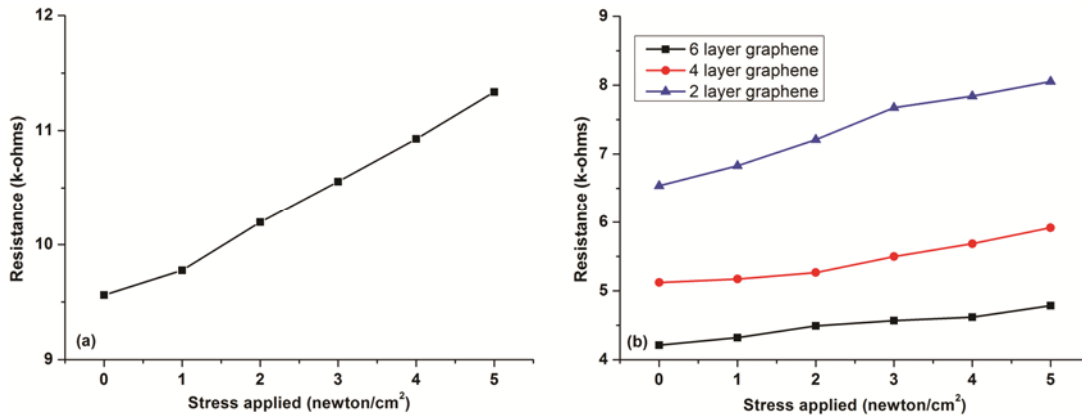
Fig. 6 – Resistance change in (a) graphite and (b) graphene based piezoresistive strain sensor with applied longitudinal stress.

Table 1 – Comparison of Gauge factor with earlier works.

Sensing material used		Gauge factor			Reference
Graphite		Strain 1		Strain 2 (longitudinal)	
		Tension	Compression		
		0.30	0.24	12.51	Present work
Graphene	2-layers of coating	1.16	0.44	20.47	
	4-layers of coating	0.93	0.31	18.77	
	6-layers of coating	0.69	0.25	16.74	
rGO		66.6 ± 5	--	--	[1]
Graphene		--	--	9.49	[20]
Graphene epoxy		--	--	11.4	[24]
Graphene		--	--	1.6	[25]
Carbon paper and PDMS elastomer		0.917	0.731	--	[27]

adhesive tape on the wooden table. The gauge factor for longitudinal stress was measured as 12.51 for graphite sensor and 20.47, 18.77 and 16.74 for graphene sensor with 2, 4 and 6 layers of coating, respectively, as shown in Fig. 6(a,b). It is observed that for both types of piezoresistive strain sensors, GF is higher under the tensile strain than the compressive strain. GF factor decreases with increase in number of layers due to distribution of applied stress more evenly among all the layers. A comparative study of present work with previously fabricated piezoresistive strain sensors for various types of strains is given in the Table 1.

4 Conclusions

In summary, we have demonstrated the fabrication and characterization of graphite and graphene piezoresistive strain sensors using facile approach on cellulose paper. The behaviour of piezoresistive sensors is investigated by measuring resistance values as a function of inward and outward chord length as well as with different types of weights applied that exhibits longitudinal strain. The resistance decreases with increase in outward chord length because of respective decrease in tension which results in reducing inter layer spacing, while increases with increase in inward chord length because of respective decrease in compression force which results in reducing overlapping of neighboured layers. During tensile and compressive strain the GF for graphite is 0.30 and 0.24 while for graphene sensors (2, 4 and 6 layers of coating) is 1.16, 0.93, 0.69 & 0.44, 0.31, 0.25, respectively. In case of longitudinal strain the GF for graphite is 12.51 and for graphene sensors (2, 4 and 6 layers of coating) is 20.74, 18.77 and 16.47, respectively. It has been observed that the GF of graphene piezoresistive strain sensor decreases with increase in number of layers. Also the GF for graphene sensor is higher as compared to graphite sensor. Fabricated piezoresistive strain sensors may find applications as human body motion detection, gait analysis etc. In future polymer based flexible substrates can be exploited for enhanced sensitivity and small sensor size.

Acknowledgement

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