Comparison of Piezo Resistive Property of Graphene based Polymer Films and Carbon Nanotube based Polymer Films to Optimize the Conductivity

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Abstract

**Aim:** The piezo resistive property of Graphene PVDF films and Carbon nanotube PVDF films is analyzed and the possibility of replacing solid state resistor (10 ohm) in electronic circuits is explored. **Materials and Methods:** Embedded hardware interface and Wheatstone bridge circuit is used to analyze the electrical conductivity of Graphene PVDF films (n=10) and Carbon nanotube PVDF films (n=10) of length 2.6 cm and width 1.1 cm. **Results:** Graphene PVDF films have significantly higher Conductivity (0.082 s/m) (p<0.05) than Carbon nanotube PVDF films (0.0108 s/m). **Conclusion:** Within the limits of this study Graphene PVDF films offer best Conductivity and can be used as a replacement for solid state resistors.

**Key-words:** Graphene, Carbon Nanotubes (CNT), PVDF (Polyvinylidene Fluoride), Nanocomposites, Nanotechnology, Novel Material, Wheatstone Bridge.

1. Introduction

Electrical Conductivity of novel materials like graphene and carbon nanotube-based polymer films have been explored (Georgakilas 2014) by replacing the solid state resistor with graphene and carbon nanotube based-polymer films in the electronic circuits (Georgakilas 2014; Jariwala et al. 2013). These films are low cost and light in weight. Graphene is the thinnest and strongest material
which conducts heat better than other materials and can be applied in electronic devices, circuits, sensors, batteries, membranes etc., Carbon nanotubes vary by length, purity, functionality and their applications can be found in energy storages, thermal materials and ceramics (Tripathy and Sahoo 2017).

Several research articles were published on graphene and carbon nanotube polymer nanocomposites in different journals for the past five years. 45 research articles were published in IEEE explore and 9809 research articles were published in Science direct. In recent times Christian Chandra Darmawan et. al, (Darmawan et al. 2017) explored the fabrication and characterization of Graphene-CNT hybrid material and improved heat dissipation in thermal measurement was observed. Wei Yan et. al, (Yan et al. 2017) explored the heterostructures of carbon nanotube and graphene. Ramos et. al, (Ramos et al. 2016) has investigated the performance and variability of electrical contacts in nanocarbon interconnects. Avash Badakhsh et. al, (Badakhsh et al. 2019) has done research on improving the electrical, mechanical and thermal properties of composites by using a network of length controlled-carbon nanotubes and graphene nanoplatelets. Effects of different lengths of carbon nanotubes were also studied.

Previously our team has a rich experience in working on various research projects across multiple disciplines (Sathish and Karthick 2020; Varghese, Ramesh, and Veeralayi 2019; S. R. Samuel, Acharya, and Rao 2020; Venu, Raju, and Subramani 2019; M. S. Samuel et al. 2019; Venu, Subramani, and Raju 2019; Mehta et al. 2019; Sharma et al. 2019; Malli Sureshbabu et al. 2019; Krishnaswamy et al. 2020; Muthukrishnan et al. 2020; Gheena and Ezhilarasan 2019; Vignesh et al. 2019; Ke et al. 2019; Vijayakumar Jain et al. 2019; Jose, Ajitha, and Subbaiyan 2020). Now the growing trend in this area motivated us to pursue this project.

The motivation behind doing this research is there is no better replacement for solid state resistance in electronic circuits (Cesano et al. 2020), (Navarro-Pardo, Martinez-Hernandez, and Velasco-Santos 2016), (Camilli and Passacantando, n.d.). Deepak et. al, (Deepak, Cherian, and Jenkins 2021) has done research on exploring the electronic applications of graphene based polymer films. The main aim of this research is to compare the conductivity of graphene and carbon nanotube based PVDF films and to explore the possibility of using it as replacement for solid state resistors in electronic circuits.
2. Materials and Methods

In this research work the materials are classified into two groups, one group refers to graphene based PVDF film and the other group refers to carbon nanotube based PVDF film. The pre-test analysis was done using clinicalcalc.com by keeping g-power at 80%, threshold at 0.05% (Tamburrano et al. 2013). Sample size of each group is 10 and the total sample is 20 having pre-test power analysis of 80%. Functionalized pure graphene, Multiwalled Carbon nanotubes (MWCNT), PVDF and solvents like DMF and acetone have been purchased commercially.

In sample preparation for group 1, graphene PVDF films were prepared by chemically mixing, sonicating and drying in an oven for 10 hours at a temperature of 65°C using standard solvent casting technique (Deepak et al. 2015).

In sample preparation for group 2, graphene powders were replaced with Carbon nanotubes and the same process is repeated in order to prepare MWCNT-PVDF films (Deepak, Cherian, and Jenkins 2021).

Embedded hardware is modified by replacing solid state resistor with Graphene PVDF film based resistor and later with Carbon nanotube PVDF film based resistor. Voltage regulator produces fixed output voltage which is connected to A/D converter and ATMEL Microcontroller through one of the ports, it is fetched and given to other ports such as RS-232 and 2x16 LCD (Liquid Crystal Display). RS-232 is a serial port which communicates and transmits the data from hardware to computer. Voltage(V) and resistance(ohms) are displayed in LCD as shown in Fig. 1.

The Wheatstone bridge is a circuit which is used to measure the unknown resistance. The Circuit consists of four different resistors R1, R2, R3, R4 of resistance 270Ω, 47Ω, 33Ω, 10Ω respectively. The circuit is later modified by replacing the solid-state resistor (R4) with graphene PVDF film based resistor and Carbon nanotube PVDF film based resistor. Voltage deflection was measured.

The Resistance of Graphene PVDF film and Carbon nanotube PVDF film is determined by applying pressure to the film ranging from P1 to P10 where P1 being the lowest pressure applied and P10 being highest pressure applied in pascals. Pressure is given as input, voltage and resistance are displayed as output and corresponding values are noted down. Conductivity can be calculated from resistance.

Statistical softwares used for plotting the graphs are ORIGINPRO V8.0 and SPSS. Independent variables are length, width, thickness and pressure applied to the film. Dependent
variable is resistance, resistivity and conductivity of the film. The analysis has been done on comparison of electrical conductivity of graphene and Carbon nanotube based PVDF films using Independent sample T test.

3. Results

As the pressure is applied to graphene PVDF and Carbon nanotube PVDF film the corresponding change in resistance and voltage are measured (Fig 1). Corresponding resistivity, conductivity of Graphene PVDF film are calculated and tabulated (Table 1). As the pressure increases from P1(low) to P10(high) resistance appears to decrease and conductivity appears to increase since resistance is inversely proportional to conductivity. Similarly, resistivity and conductivity of carbon nanotube PVDF film are tabulated (Table 2). As the pressure increases from P1(low) to P10(high) resistance appears to increase and conductivity appears to decrease for Carbon nanotube PVDF film. Resistance appears to decrease for graphene PVDF film and it appears to increase for CNT PVDF film. This phenomenon can be due to the inverse piezo resistive effect which is observed in graphene PVDF film. Voltage deflection of Wheatstone bridge with solid state resistor (Fig 2a) and graphene PVDF film (Fig 2b) are tabulated (Table 3). Resistance of graphene PVDF films appears to be gradually decreasing when the pressure applied is from P1 (low) to P10 (high) (Fig 3a). This phenomenon is due to the piezo resistance property of the film. Conductivity of graphene PVDF film appears to be gradually increasing (Fig 3b) since conductivity is inversely proportional to piezo resistance. Resistance of CNT PVDF film appears to be gradually increasing (Fig 4a) and hence conductivity of CNT PVDF film appears to be gradually decreasing (Fig 4b). T-test comparison of conductivity of graphene PVDF film and carbon nanotube PVDF film is tabulated (Table 4) which shows there is a statistically significant difference in conductivity of graphene PVDF film and carbon nanotube PVDF film. Conductivity of carbon nanotube PVDF film has the highest mean (0.012) over graphene PVDF film(0.037). The mean, standard deviation and significant difference of resistivity and conductivity of graphene PVDF film and carbon nanotube PVDF film is tabulated (Table 5) which shows there is a significance difference between the two groups since p<0.05 (Independent Sample T Test). Bar graph is comparing the mean (+/- 1 SD) conductivity of graphene PVDF films and carbon nanotube PVDF films (Fig 5) and there is a significance difference in resistance and conductivity of graphene and MWCNT based composite films p<0.05 (Independent sample T test).
Table 1- Voltage, Resistance, Resistivity and Conductivity of Graphene PVDF Films when Pressure from P1 (low) to P10 (high) is Applied to Films in Pascals

| Applied Pressure (Pascals) | Voltage (V) | Resistance (ohm) | Resistivity (ohm-meter) | Conductivity (S/m) |
|---------------------------|-------------|------------------|------------------------|-------------------|
| P1                        | 0.86        | 76               | 83.6                   | 0.011             |
| P2                        | 1.11        | 69               | 75.9                   | 0.013             |
| P3                        | 1.72        | 58               | 63.8                   | 0.015             |
| P4                        | 2.37        | 45               | 49.5                   | 0.020             |
| P5                        | 3.56        | 25               | 27.5                   | 0.036             |
| P6                        | 3.77        | 23               | 25.3                   | 0.039             |
| P7                        | 3.82        | 21               | 23.1                   | 0.043             |
| P8                        | 3.84        | 21               | 23.1                   | 0.043             |
| P9                        | 4.29        | 12               | 13.2                   | 0.075             |
| P10                       | 4.35        | 11               | 12.1                   | 0.082             |

Fig. 1- Embedded Hardware Showing Voltage and Resistance Reading when Pressure is Applied to Graphene PVDF Film

Table 2- Voltage, Resistance, Resistivity and Conductivity of Carbon Nanotube PVDF Films when Pressure from P1 (Low) to P10 (High) is applied in Pascals

| Applied Pressure (Pascals) | Voltage (V) | Resistance (ohm) | Resistivity (ohm-meter) | Conductivity (S/m) |
|---------------------------|-------------|------------------|------------------------|-------------------|
| P1                        | 1.86        | 56               | 61.6                   | 0.016             |
| P2                        | 1.37        | 65               | 71.5                   | 0.013             |
| P3                        | 1.29        | 66               | 72.6                   | 0.013             |
| P4                        | 1.33        | 68               | 70.4                   | 0.014             |
| P5                        | 0.96        | 72               | 79.2                   | 0.012             |
| P6                        | 0.94        | 73               | 80.3                   | 0.012             |
| P7                        | 0.92        | 74               | 81.4                   | 0.012             |
| P8                        | 0.54        | 80               | 88.0                   | 0.011             |
| P9                        | 0.37        | 83               | 91.3                   | 0.010             |
| P10                       | 0.33        | 84               | 92.4                   | 0.010             |
Table 3- Voltage Deflection of Graphene and CNT PVDF Films by Replacing Solid State Resistor (R4) with GRAPHENE and CNT Film based Resistors in Wheatstone Bridge Circuit

| Wheatstone bridge | Resistor (R1) | Resistor (R2) | Resistor (R3) | Resistor (R4) | Voltage deflection |
|-------------------|--------------|--------------|--------------|--------------|-------------------|
| Solid state resistor | 270Ω | 47Ω | 33Ω | 10Ω | 1.0V |
| Graphene PVDF film | 270Ω | 47Ω | 33Ω | 5.74Ω | 2.0V |
| CNT PVDF film | 270Ω | 47Ω | 33Ω | 5.74Ω | 2.0V |

Table 4- T-test Comparison of Conductivity of Graphene PVDF Film and Carbon Nanotube PVDF Film. There is a Statistically Significant difference in Conductivity of Graphene PVDF Film and Carbon Nanotube PVDF Film. Conductivity of Carbon Nanotube PVDF Film has the Highest Mean (0.012) over Graphene PVDF Film (0.037)

| GROUP | N | Mean | Std. Deviation | Std. Error Mean |
|-------|---|------|----------------|-----------------|
| VOLTAGE | | | | |
| Graphene PVDF | 10 | 2.9690 | 1.33119 | .42096 |
| MWCNT PVDF | 10 | .9910 | .48736 | .15412 |
| RESISTANCE | | | | |
| Graphene PVDF | 10 | 36.1000 | 24.02522 | 7.59744 |
| MWCNT PVDF | 10 | 72.1000 | 8.76166 | 2.77068 |
| RESISTIVITY | | | | |
| Graphene PVDF | 10 | 39.710 | 26.4277 | 8.3572 |
| MWCNT PVDF | 10 | 78.870 | 9.9616 | 3.1501 |
| CONDUCTIVITY | | | | |
| Graphene PVDF | 10 | .037970 | .0246872 | .0078068 |
| MWCNT PVDF | 10 | .012827 | .0017024 | .0005383 |

Table 5- Shows the Mean, Standard Deviation and Significance difference of Resistivity and Conductivity of Graphene PVDF Film and Carbon Nanotube PVDF Film. There is a Significance difference between the Two Groups since p<0.05 (Independent Sample T Test)

| GROUP | N | Mean | Std. Deviation | Std. Error Mean | Mean Difference | Std Error Difference | 95% Confidence Interval of the Difference |
|-------|---|------|----------------|-----------------|----------------|---------------------|----------------------------------------|
| Voltage | | | | | | | |
| Equal variances assumed | | | | | | |
| Equal variances not assumed | 17.747 | 4.412 | 18 | .000 | 1.97800 | .44828 | 1.03619 | 2.91981 |
| EQUAL VARIANCES | | | | | | |
| Equal variances assumed | | | | | | |
| Equal variances not assumed | 15.468 | 4.452 | 18 | .000 | -36.00000 | 8.08689 | -52.98992 | -19.01008 |
| RESISTIVITY | | | | | | |
| Equal variances assumed | | | | | | |
| Equal variances not assumed | 14.682 | 4.385 | 18 | .000 | -39.1600 | 8.9312 | -57.9237 | -20.3963 |
| Conductivity | | | | | | |
| Equal variances assumed | | | | | | |
| Equal variances not assumed | 13.207 | 3.213 | 18 | .000 | -0.0251430 | .0078253 | .0087026 | .0415834 |
Fig. 2a- Wheatstone Bridge with Solid State Resistor. Voltage Deflection Measured is 1 Volt

Fig. 2b- Wheatstone Bridge Circuit with Graphene PVDF Film. Voltage Deflection Measured is 2 Volts

Fig. 3a- Pressure vs Resistance of Graphene PVDF Films. Resistance Decreases as the Pressure is Applied from P1 (low) to P10 (high) in Pascals
Fig. 3b- Pressure vs Conductivity of Graphene PVDF Films. Conductivity Increases as the Pressure is Applied from P1 (Low) to P10 (High) in Pascals

Fig. 4a- Pressure vs Resistance of CNT PVDF Film. Resistance Increases as the Pressure is Applied from P1 (Low) to P10 (High) in Pascals

Fig. 4b- Pressure vs Conductivity of CNT PVDF Film. Conductivity Decreases as Pressure is Applied from P1 (Low) to P10 (High) in Pascals
4. Discussion

Electrical Conductivity of carbon nanotube based polymer films and graphene based polymer films are explored to optimize the conductivity and to use it as replacement instead of solid state resistor. Some of the past studies (Deepak et al. 2015) reported the possibility of using graphene based polymer film in electronic circuits. (Rafiee 2011) has presented an approach for preparation of graphene polymer composites by exfoliating the graphite. (Patole et al. 2012) has presented the analysis in the preparation of graphene and carbon nanotube by water based in situ microemulsion polymerization. All the results are in concurrence with the present findings.

Factors which can affect the conductivity of both graphene and CNT based polymer films are length of the film, width of the film, weight percentage of graphene in graphene PVDF film and carbon nanotube in CNT PVDF film, homogeneity and electrical noise in circuit (Namasivayam and Shapter 2017).

Modification of length and width of the film can affect the conductivity of the film. So precautions were taken to keep both length and width of the film constant during the research work. Weight percentage (6wt%) of graphene in Graphene PVDF film and CNT in CNT PVDF film were also kept constant during sample preparation. So the weight percentage of graphene will have the same effect on all the samples and hence conductivity will not get affected. Environmental factors
such as electric noise can affect circuit during its operation. During sample preparation special measures were taken to keep homogeneity of the film constant.

Our institution is passionate about high quality evidence based research and has excelled in various fields (Vijayashree Priyadharsini 2019; Ezhilarasan, Apoorva, and Ashok Vardhan 2019; Ramesh et al. 2018; Mathew et al. 2020; Sridharan et al. 2019; Pc, Marimuthu, and Devadoss 2018; Ramadurai et al. 2019). We hope this study adds to this rich legacy.

Limitations such as sample preparation delay, Inability to precisely measure the applied pressure, presence of impurities in the thin film and external disturbance like noise should be taken into consideration.

Carbon nanotubes are electrically conductive (Lekawa-Raus et al. 2014) and they have the potential to be a cost effective substitute for metal wires. (Qu et al. 2020), (Chae and Lee 2014), (Ghallab and Badawy 2006), (Yang et al. 2011). Graphene PVDF films with higher stiffness, durability, and electrical conductivity have a wide range of applications as lightweight materials, they can be used in aircraft, windmill blades, sports equipment (Ho and Wei 2013) and as flexible conductive materials for electronic devices (Kim et al. 2012). Graphene can also be used in biomedical applications like drug delivery, brain penetration improvement (Maiti et al. 2014).

5. Conclusion

Graphene based PVDF film has better conductivity when compared to carbon nanotube based PVDF film. As the pressure is applied to graphene PVDF film there is a significant decrease in resistance and increase in conductivity, whereas in carbon nanotube PVDF film, there is a significant increase in resistance and decrease in conductivity. Possibility of using graphene PVDF and CNT PVDF films as a solid state resistor in electronics is achieved by successfully implementing in wheatstone bridge circuit and also can be applied in other electronic circuits.

Declarations

Conflict of Interests

No conflict of interest in this manuscript.
Author Contribution

Author P. Manoj Kumar was involved in data collection, data analysis, manuscript writing. Author Dr. A. Deepak was involved in conceptualization, guidance and critical review of manuscript.

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