Impedance Behavior of Conducting Piezoresistive Polymer Composite

Brijesh Prasad, Vikas Rathi, Varij Panwar, Fateh Singh Gill, Pravin Patil

***Abstract:** Conducting polymers (CP) are used as piezoresistive sensors for the various application of monitoring human and structural health monitoring, flexible electronic devices, automobile, aerospace and for biomedical device applications. In the present work we have developed the polyvinylidene fluoride (PVDF) based CP using Carbon Nano Fiber (CNF). The samples were prepared using the solvent casting technique. The conductivity plays crucial role in improving the sensitivity of the sensors. The impedance behavior of the developed PVDF/CNF membranes was analysed using the E4900A impedance analyzer using the parallel plate probes. The impedance was measured as the function of frequency in the range of 20 Hz-1 MHz. The piezoresistive behavior of PVDF is an important parameter for sensing application. The objective of the present work was to study the impedance spectroscopy of the PVDF/CNF samples. The sample composition of PVDF/CNF (90/10) and PVDF/CNF (85/15) showed better results with lower resistance and higher conductivity.*

***Keywords:** Conducting, Impedance, Sensing, Piezoresistive*

I. INTRODUCTION

The interest about PVDF polymer based piezoresistive sensors have attracted the researchers and is studied due to the wide application range in the field of sensors, actuators, flexible devices, soft robotic muscles, stretchable conductors, automobiles, aerospace, wearable antenna etc. [1,2,3]. The polymers are not natively conducting but bade conductive by the infiltration of the conducting fillers such as carbon nano tubes (single wall and multiwall), carbon black, graphene, graphene oxide and CNF to development of CP [4,5,6]. Among all CNF has been of a great advantage due to long fiber length and highly conducting behavior. Long fiber length helped in reducing the leakage current and improved the conductivity of the polymer membrane [5]. PVDF based CP show highly appreciated physical and chemical properties of high mechanical strength, resistance to corrosion and toxic acids, light weight easy availability and low cost [6,7]. The conductive behavior of these CP is analysed using the instrument called impedance analyzer which measures the conductivity, impedance, resistance, capacitance and various other parameters on the application of potential difference across the thin membrane in varying frequency range.

In the present article we have focused on developing the PVDF/CNF thin membranes using the solvent casting technique by varying the concentration of the conducting fillers (CNF). The developed CP membranes were used for the sensing application and discussed in our previous work done [3,9]. Here we have concentrated on studying the electrical behavior of the PVDF/CNF polymer membranes using impedance. Impedance encompasses the concept of resistance to the Alternating Current (AC) circuit possessing both phase and magnitude. Impedance is very much helpful in analysing the AC electrical circuits used in the application of soft electronic devices for monitoring human health and structural health.

II. EXPERIMENTAL

A. Materials

PVDF was taken as the polymer material for matrix development and to make the polymer conductive CNF was added to form the PVDF/CNF membrane. Solvent casting technique was applied for the development of PVDF/CNF membrane.

B. Methodology

Firstly, the PVDF was added to DMF to form homogeneous solution with heating at temperature of 60^{o}C with regular mixing using the magnetic bead on the hot plate magnetic stirrer. After 4 hours continuous mixing the CNF was added to the homogeneous solution of PVDF. Further the solution was allowed to heat and mix for another 5 hours. After five hours the solution was poured on the glass petridish and allowed for solvent evaporation to obtain dry CP film. The dry film of PVDF/CNF was peeled off from the petridish and cut into the circular shape for the impedance characterization. Before characterisiatation the sample were coated both sides with silver paste to form conductive surface.

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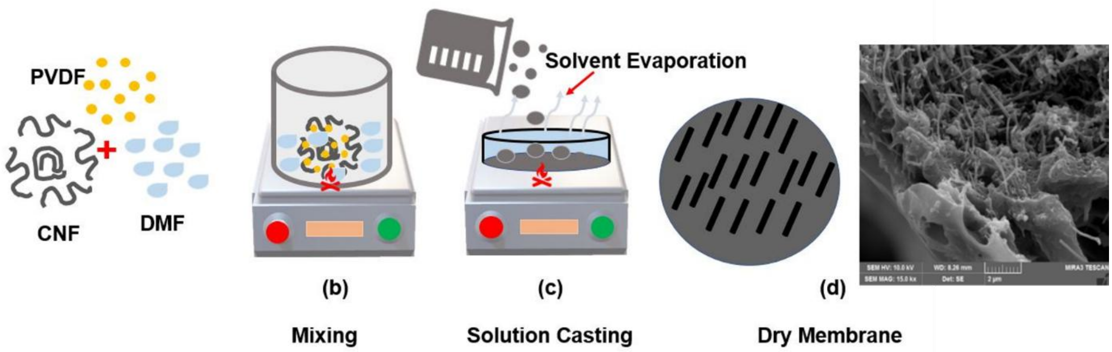

Figure1. Development of PVDF/CNF conducting polymer composite

III. CHARACTERISATATION

The microstructural morphology of the various PVDF/CNF membranes were analysed using FESEM (MIRA3b TESCAN, USA) machine. Before doing the characterization the samples were coated with the thin gold layer using sputtering technique to clearly obtain the reflectance patterns of the internal structure.

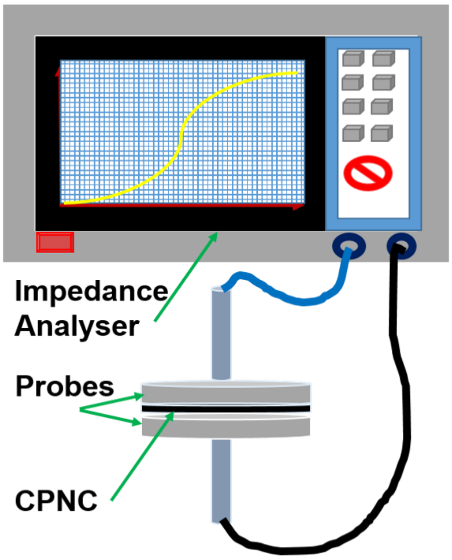

Figure2. Model of impedance analyser.

The AC electrical behavior of the PVDF/CNF membranes were analysed using the E4900A impedance analyser supplied by Keysight technologies. The impedance was measured by taking frequency as the function in the range of 20Hz – 1MHz at ambient temperature. The values of impedance were directly obtained from the impedance analyser in respect of varying frequency. Figure 2. shows the model of impedance analyser used for the AC analysis.

IV. RESULTS AND DISCUSSION

A. *Morphology analysis*

Figure 3. shows the FESEM images of the pure PVDF. It can clearly be observed that the PVDF membrane just plain without any fibers structure. While, when CNF was added in the PVDF matrix it helped in developing the conductive network for charge transportation to make the membrane conductive. The threads like structure shown in the Figure 2 (b) is clear indication of the presence of CNF.

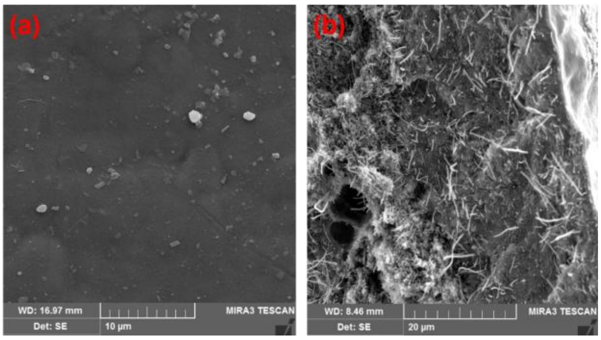

Figure 3. (a) Microstructure of pure PVDF and (b) shows the PVDF/CNF membrane

B. *Impedance analysis*

This section deals with the measurement of impedance using the two probe parallel plate impedance analyser. The system involves an impedance analyzer, test fixture and high power module. The arrangement of the system components is shown in Figure 2 open circuit potential was measured across the polymer membranes of PVDF/CNF. The AC conductivity of the carbon based composites is measured by measuring the impedance of the sample. Electrical impedance is considered as the total opposition produced by the circuit to the AC current. Impedance includes the resistance (R), inductance (X_L) and capacitance (X_C) of the material. In case of AC conductivity, the ohm's law includes total impedance of the circuit and is shown by equation 1 which shows the current (I) is directly proportional to the applied voltage and inversely proportional to the total impedance [8,9].

$$I = \frac{V}{Z} \qquad (1)$$

The impedance of the sample is measured by applying the potential and the current is measured through the sample. When the potential is applied in the sinusoidal form the response is obtained in the form of current signals. The obtained current signal is the sum of analysed sinusoidal function. The excitation potential signal is expressed in equation 2 with respect to time function.

$$V_t = V_o \sin(\omega t) \qquad (2)$$

Here, V_t is defined as the potential at particular time (t), V_o defines the amplitude and ($\omega = 2 \pi f$) as radial frequency. Whereas, the current I_t as response signal at time (t) is shifts by phase angle (ϕ) with amplitude I_o and the expression for the response signal is given by equation 3.

$$I_t = I_o \sin(\omega t + \phi) \qquad (3)$$

Combining the equitation 3 and 4 the expression for ohms' law for calculating the impedance of the system.

$$Z = \frac{V_t}{I_t} = \frac{V_O \sin(\omega t)}{I_O \sin(\omega t + \phi)} \qquad (4)$$

The impedance of the CP for AC conductivity is measured at different frequencies. It provides the real and imaginary part of the of the impedance as function of frequency. The AC analysis is very much helpful in measuring the resistance of higher magnitude materials [10].

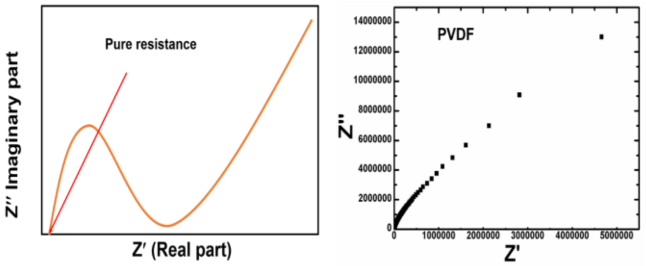

Figure4. shows the real and imaginary parts of the impedance and the plot for pure PVDF.

Figure 4 represents the Nyquist plots for the pure PVDF samples and Figure 5. shows Nyquist plots for the varying concentration of PVDF and CNF. Impedance as the relationship of real (Z′) and imaginary (Z′′) curve as the impedance components. The real resistance is obtained from the Nyquist plot by the intercept of the curve made with the X axis which defines the real part of the impedance [11,12].

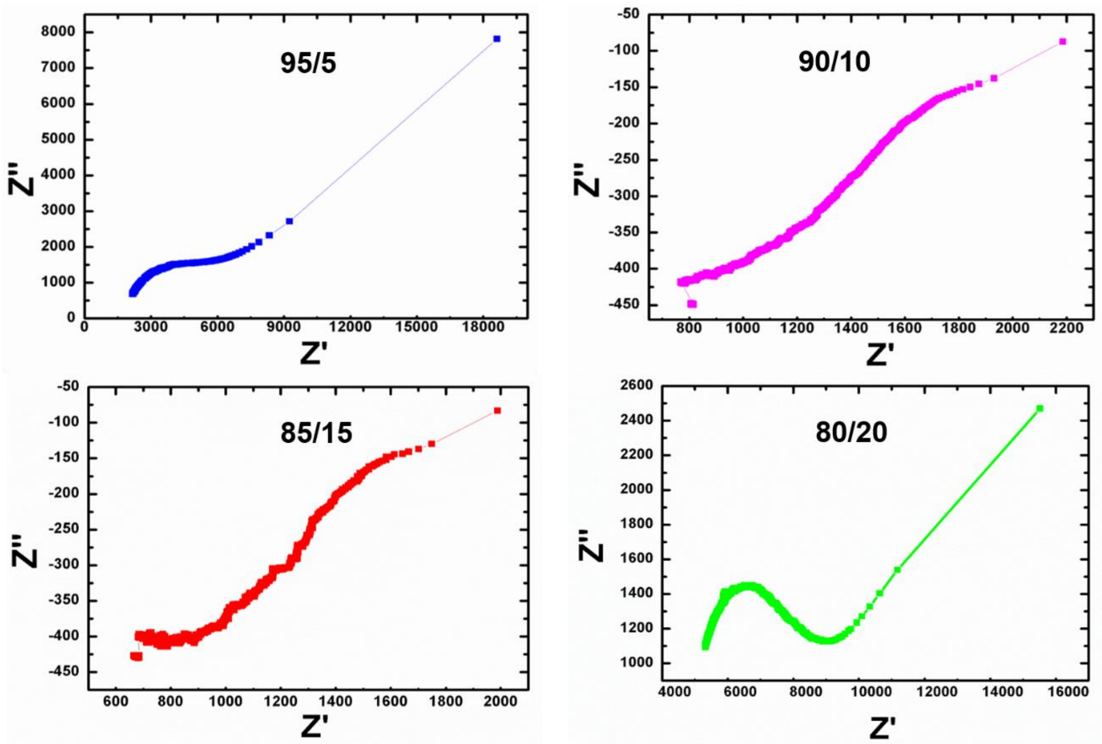

Figure 5. shows Nyquist plots different compositions of PVDF and PVDF/CNF

Figure 5 the samples having varying concentrations of the PVDF and CNF. PVDF/CNF (95/5) has the lowest percentage of CNF and has high resistance of ~2540 Ω While in sample PVDF/CNF (90/10) due to the higher filler content the conduction has improved due to which there is decrease in the value of resistance ~658 Ω. Which proves that the CNF developed the conducting network for easy transportation of the charges through the membrane. Further the values obtained for resistance in ohms for the various compositions are shown in Table1.

Table1. Values of resistance obtained from impedance analyzer

Sample Name/Composition	Resistance (Ω)
PVDF (100/0)	25417.70
PVDF/CNF (95/5)	2022.40
PVDF/CNF (90/10)	762.45
PVDF/CNF (85/15)	658.32
PVDF/CNF (80/20)	5247.21

This occurred due to the higher percentage of CNF which lead to the bulging and made clusters of the CNF with agglomeration which in result effected the conductive channels with the increase in the leakage current. Therefore, it can be concluded from here that the sample PVDF/CNF (90/10) and PVDF/CNF (85/15) showed better results.

The reduced resistance is very much helpful for the development of piezoresistive sensors as the small change in the resistance can be picked with the occurrence of change in the surrounding.

V. CONCLUSION

PVDF based polymer composite is attracting the world because if their good physical and chemical properties. Developed CP of PVDF/CNF has put the polymers one step forward for the piezoresistive sensing application. As the addition of CNF in optimized condition can help in achieving low resistance. The PVDF/CNF samples were prepared using the solvent casting technique and analysed using the FESEM for surface morphology and the impedance was characterized using the E4900A impedance analyzer. It can be concluded from the above study that the sample PVDF/CNF (90/10) and PVDF/CNF (85/15) showed better results. It was also observed that addition of the filler content above 2 wt% level leads to agglomeration of the filler contents and disturbs the conductive network and effects the conductivity and performance of the CP membranes. These membranes can be applied for the strain sensing and for actuator application for flexible devices.

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