

## 10. SUMMARY AND SUGGESTIONS FOR FUTURE WORK

### 10.1 SUMMARY

Graphene-based materials are promising and potentially effective candidates for the reinforcement of PNCs owing to their impressive physical properties, making it possible to develop multifunctional nanocomposites. A lot of attention has been diverted towards the oxidized form of graphene i.e. Graphene oxide (GO). GO possess the similar properties to graphene as well as the special surface structure with the introduction of hydroxyl and carboxyl groups available for the synthesis of GO-based composites. GO has become a well-established reinforcement for PNCs and it has widest potential to achieve high-performance nanocomposites because it can be prepared on a large scale to meet the demand of nanofillers for composite preparation and can be chemically tailored to maximize its interaction with the polymer matrices. GO contains various oxygen-containing functional groups which promote the complete exfoliation in different solvents and uniform dispersion of GO with polymer matrices. The presence of these functional groups imparts GO with a good degree of matrix-reinforcement interactions leading to improved dispersion and stress transfer. Thus, motivated by its unique layered structure, desirable and superior physical properties, easy accessibility and graphene orientated application, GO was chosen as a primary filler for the fabrication of PNCs in the present study.

GO was first synthesized from natural graphite powder using modified Hummers method in presence of concentrated  $H_2SO_4$ ,  $NaNO_3$  and  $KMnO_4$ . The successful preparation of GO was confirmed using Raman spectroscopy, FTIR spectroscopy and XRD studies. The synthesized GO was integrated with various polymer matrices such as HPMC, PVA, WPPy, PVDF, PSSA and PEG to prepare various polymer composites using colloidal processing technique. In addition, various fillers such as ZnO,  $TiO_2$

and  $V_2O_5$  were used in combination with GO to form hybrid fillers and subsequently incorporated into these polymer matrices to form multiphase polymer composites. The physicochemical properties of all the prepared composites were investigated using various analytical techniques such as FTIR spectroscopy, Raman spectroscopy, UV-vis spectroscopy, XRD, TGA, POM and SEM. The dielectric properties of all the composites were investigated as a function of frequency and temperature using an impedance analyzer to ascertain their feasibility for energy storage applications.

HPMC/PVA/GO/ZnO nanocomposite films with good flexibility were prepared and characterized using various analytical techniques. The FTIR and XRD results indicate complete exfoliation of GO within the HPMC/PVA blend matrix leading to the formation of exfoliated nanocomposites. The SEM results infer the uniform distribution of GO and ZnO nanoparticles throughout the polymer matrix, indicating the strong interfacial interaction between the polymer blend matrix and nano-fillers. The enhancement in the mechanical properties of nanocomposites can be ascribed to the homogeneous dispersion of GO sheets and ZnO nanoparticles in the polymer matrix and strong interfacial interaction between all the components of nanocomposites. In addition, the nanocomposites showed high dielectric constant and low dielectric loss. The increase in dielectric constant of nanocomposites is due to increase in the interfacial polarization between GO/ZnO nano-fillers and the polymer matrix. A high dielectric constant (103.51) at 50 Hz and at 150°C with the low dielectric loss (3.14) at 50 Hz and at 150°C was obtained for nanocomposites with 2.5 wt % GO and 25 wt % ZnO nanoparticles loadings.

Flexible dielectric composites consisting of WPPy, PVA and GO were prepared at different GO loadings varying from 0-3 wt %. FTIR studies indicate the strong chemical interaction between GO and the polymer systems. SEM results confirm that GO was homogeneously dispersed within the polymer matrix. The composites exhibited a significant enhancement in the dielectric constant with low dielectric loss values as a function of GO

loading which resulted from the fine dispersion of GO within the polymer matrix. The dielectric constant increases from 27.93 (50 Hz, 150°C) for WPPy/PVA (50/50) blend to 155.18 (50 Hz, 150°C) for composites with 3 wt % GO loading. The dielectric loss increases from 2.01 (50 Hz, 150°C) for WPPy/PVA (50/50) blend to 4.71 (50 Hz, 150°C) for composites with 3 wt % GO loading.

Novel three-phase PNCs comprising of PVDF, TiO<sub>2</sub> nanoparticles and GO were prepared. The FTIR and XRD results infer good interaction between the constituents of nanocomposites. The microscopic studies infer homogeneous dispersion and distribution of TiO<sub>2</sub> nanoparticles and GO within the PVDF matrix. A notable improvement in the thermal stability of PVDF was observed by the addition of TiO<sub>2</sub> and GO as hybrid fillers. The dielectric performance of PVDF/TiO<sub>2</sub>/GO nanocomposite films was significantly improved as compared to PVDF/TiO<sub>2</sub> (90/10) nanocomposite film. The dielectric constant increases from 18.57 (50 Hz, 150°C) for PVDF/TiO<sub>2</sub> (90/10) nanocomposite film to 165.16 (50 Hz, 150°C) for PVDF/TiO<sub>2</sub>/GO nanocomposite film containing 7 wt % TiO<sub>2</sub> and 3 wt % GO loading. In addition, the dielectric loss also increases from 1.71 (50 Hz, 150°C) for PVDF/TiO<sub>2</sub> (90/10) nanocomposite film to 3.68 (50 Hz, 150°C) for PVDF/TiO<sub>2</sub>/GO nanocomposite film containing 7 wt % TiO<sub>2</sub> and 3 wt % GO loading.

GO reinforced novel polymer composites comprising of PSSA and PVA blend matrix have been developed. GO sheets were homogeneously dispersed in the polymer matrix and stable dispersions were obtained owing to the presence of the highly hydrophilic oxygen-containing functional groups of GO. The excellent reinforcement of composites was achieved due to the strong interfacial interaction between PSSA, PVA and GO via hydrogen bonding as confirmed by FTIR spectroscopy. The XRD results clearly indicate that GO was fully exfoliated into individual graphene sheets. The higher decomposition temperatures were observed for PSSA/PVA/GO

composites as compared to PSSA/PVA blend indicating the improvement in the thermal stability of composites. The improved thermal stability of composites can be attributed to the excellent dispersion of GO into the polymer matrix and the high aspect ratio of GO. The improved dielectric performance of PSSA/PVA/GO composites is attributed mainly to the homogeneous dispersion of GO in the polymer matrix and the strong interaction between PSSA, PVA, and GO. The PSSA/PVA/GO composites constitute a new type of graphene-based composites to be used as a high dielectric constant material in advanced electronic devices such as embedded capacitors.

Novel flexible dielectric composites consisting of PVA, PEG and GO with high dielectric constant and low dielectric loss have been developed. The structure and surface morphology of the PVA/PEG/GO composites were evaluated and the dielectric behavior of PVA/PEG/GO composites was studied in the wide range of frequencies from 50 Hz to 20 MHz and temperature in the range 40 to 150°C. The dielectric constant for PVA and PVA/PEG (50/50) blend film was found to be 10.71 (50 Hz, 150°C) and 31.22 (50 Hz, 150°C) respectively. The dielectric constant for PVA/PEG/GO composite with 3 wt % GO was found to be 644.39 (50 Hz, 150°C) which is 60 times greater than the dielectric constant of PVA and 20 times greater than that of PVA/PEG (50/50) blend film. The PVA/PEG/GO composites not only show a high dielectric constant but also show low dielectric loss which is highly attractive for practical applications.

A novel ultra-high dielectric constant composite comprising of PVA as a polymer matrix and  $V_2O_5$  and GO as fillers have been developed successfully. FTIR studies indicate the strong chemical interaction between GO and polymer matrix. SEM results confirm that GO was homogeneously dispersed within the polymer matrix. The dielectric results reveal that PVA/ $V_2O_5$ /GO composites exhibit ultra-high dielectric constant and low dielectric loss. The dielectric constant increases from 189.40 (50 Hz, 40°C)

for PVA/V<sub>2</sub>O<sub>5</sub> (95/05) composites to 5610.76 (50 Hz, 50°C) for PVA/V<sub>2</sub>O<sub>5</sub>/GO composites with 2.5 wt % GO loading. The dielectric loss also increases from 3.76 (50 Hz, 80°C) for PVA/V<sub>2</sub>O<sub>5</sub> (95/05) composites to 9.77 (50 Hz, 150°C) for PVA/V<sub>2</sub>O<sub>5</sub>/GO composites with 2.5 wt % GO loading. The dielectric properties of all the composites studied in this work at different filler loadings are summarized in Table 10.1.

**Table 10.1: Comparison of dielectric properties of all composites**

Sr. NO.	Composites	Dielectric Constant	Dielectric Loss	Filler Loadings
1	WPPy/PVA/GO	155.18, 50 Hz, 150°C	4.71, 50 Hz, 150°C	3 wt % GO
2	PSSA/PVA/GO	297.91, 50 Hz, 150°C	2.64, 50 KHz, 150°C	3 wt % GO
3	PVA/PEG/GO	644.39, 50 Hz, 150°C	5.58, 50 Hz, 150°C	3 wt % GO
4	HPMC/PVA/GO/ZnO	103.51, 50 Hz, 150°C	3.14, 50 Hz, 150°C	2.5 wt % GO, 25 wt % ZnO
5	PVDF/TiO <sub>2</sub> /GO	165.16, 50 Hz, 150°C	3.68, 50 Hz, 150°C	3 wt % GO, 7 wt % TiO <sub>2</sub>
6	PVA/V <sub>2</sub> O <sub>5</sub> /GO	5610.76, 50 Hz, 50°C	9.77, 50 Hz, 150°C	2.5 wt % GO, 2.5 wt % V <sub>2</sub> O <sub>5</sub>

It can be seen that all the composites showed high dielectric constant and low dielectric loss values. However, PVA/V<sub>2</sub>O<sub>5</sub>/GO composites showed maximum values of dielectric constant as compared to other composites studied. Thus, based on the comparison of dielectric constant values of all composites, it can be concluded that PVA/V<sub>2</sub>O<sub>5</sub>/GO composites seem to be a promising candidate for energy storage applications. These findings may aid researchers in the development of novel and flexible high dielectric constant composites featuring graphene or various other graphene-based nanofillers as a primary component for full exploitation of their excellent properties.

## **10.2 SUGGESTIONS FOR FUTURE WORK**

This thesis work was mainly focused on the development of high dielectric constant polymer composites based on GO and inorganic metal oxide nanoparticles. The material design, development and processing of high dielectric constant PNCs is challenging. Therefore, there are several topics for future work which can be extended to improve the performance of high dielectric constant PNCs and realizing their potential for energy storage applications.

- ❖ Attempts could be made in future to investigate the dielectric properties of PNCs comprising of core-shell structured nanofillers. The effect of shell layer thickness and its characteristics on the dielectric properties of different polymer matrices can be studied.
- ❖ The effective reinforcement of nanofillers in the polymer matrix plays a significant role in improving the dielectric properties of PNCs. Therefore, nanofillers of different particle size can be incorporated into the polymer matrix and the effect of particle size variation on the dielectric performance of PNCs can be investigated.

- ❖ The effect of PNCs film thickness on its dielectric properties can also be investigated as a function of frequency and temperature.
- ❖ Graphene nanoplatelets (GNPs) can be used as an excellent nanofiller to improve the dielectric properties of polymer matrix. GNPs can also be used as a co-filler in combination with suitable metal oxide nanoparticles to form hybrid fillers. High dielectric constant PNCs can be realized by filling these hybrid fillers into the polymer matrices.
- ❖ Furthermore, graphene can be functionalized using different ionic liquids (cationic or anionic) to achieve better solubility in organic solvents. The functionalized graphene can be incorporated into polymer matrices to improve their dielectric properties.