



# Dielectric properties of micro-composites based on acrylic coated conducting carbon particles and silicone elastomer

Ajit Shankar Singh, Vishal Das, Preeti Mishra & Arvind Kumar Pandey\*

Polymer Science Division, Defence Materials and Stores Research & Development Establishment (DMSRDE),  
Defence Research & Development Organisation (DRDO), G.T. Road, Kanpur, Uttar Pradesh 208 013, India

Received: 25 August 2020

The present embodiment provides an insight on the fabrication and characterization of polymeric dielectric composites for energy storage applications required in mobile electronic devices, stationary power systems, hybrid electric vehicles and pulse power generation. In present study, conducting carbon particles of appropriate dielectric properties have been selectively functionalized with acrylic resin using micro-ballooning method to form core shell conducting particles in order to tailor its conductivity and dielectric properties (dielectric loss in particular). The core shell conducting particles were then mixed with silicone resin in varied proportions (10, 20, 25 and 30 % wt/wt) to form flexible dielectric micro-composites which exemplify concomitant increase in dielectric constant without impairing dielectric loss owing to selective coating of carbon particles via micro-ballooning method. Formulation containing 25wt% core shell conducting particles showed most linear variation in dielectric constant and dielectric loss factor ( $\tan \delta$ ) with respect to frequency due to feeble contribution of interfacial polarization.

**Keywords:** Dielectric, Composites, Conducting carbon, Coating, Polarization

## 1 Introduction

Materials with high electrical energy storage capabilities are known to play a pivotal role in the development of mobile electronic devices, stationary power systems, hybrid electric vehicles, and pulse power applications. In present times, there is an ongoing demand for flexible polymeric capacitors that can accumulate a large amount of electrical energy and deliver it nearly instantaneously when triggered. This form of “pulse power” is predominantly required for a variety of military and commercial applications<sup>1,2</sup>. Over the period of time, these applications have amplified their dependency on the use of these high dielectric materials and presently demand materials with even higher energy storage capabilities and power densities. Dielectric materials have been known to store electrical energy in the form of charge separation when the electron distributions around constituent atoms or molecules are polarized by an external electric field<sup>1</sup>. The maximum energy density that can be stored by a dielectric material in the form of a closed capacitor configuration can be obtained through equation 1 as:

$$W_{\max} = \frac{1}{2} \epsilon_0 \epsilon_r E_{\text{break}}^2 \quad \dots (1)$$

where,  $W_{\max}$  is the maximum energy storage density (in  $\text{Jm}^{-3}$ ),  $\epsilon_0$  is the permittivity of free space ( $8.85 \times 10^{-12} \text{ Fm}^{-1}$ ),  $\epsilon_r$  is the relative permittivity or dielectric constant of the material and  $E_{\text{break}}$  is the breakdown field strength. Thus, in order to attain the maximum possible value of energy storage density, the material should have both high dielectric constant and high breakdown field strength at the same time. However, in most practical applications, the strength of electric field employed across the dielectric material is less than the breakdown field. On the other hand, the permittivity of an insulating material is itself not a constant quantity but depends on the frequency ( $\nu$ ) of the applied electric field and can be expressed as the complex permittivity represented through equation 2.

$$\epsilon^*(\omega) = \epsilon'(\omega) + j\epsilon''(\omega) \quad \dots (2)$$

where,  $\omega$  is the angular frequency and  $\omega = 2\pi\nu$ ,  $\epsilon'(\omega)$  is the relative permittivity or dielectric constant of the material and the imaginary part,  $\epsilon''(\omega)$  is related to the dielectric loss. The relative dielectric permittivity or the dielectric constant is hence a frequency dependent quantity as well. The imaginary part  $\epsilon''(\omega)$  shows possible dissipative effects arising due to charge migration (i.e., conduction) or conversion into thermal energy (e.g., molecular vibration)<sup>1</sup>. The dissipative

\*Corresponding author (E-mail: akpandey@dmsrde.drdo.in)

behaviour is also characterized by means of the dielectric loss tangent ( $\tan \delta$ ) or dielectric loss factor which can be defined through equation 3.

$$\tan \delta(\omega) = \frac{\varepsilon''(\omega)}{\varepsilon'(\omega)} \quad \dots (3)$$

The occurrence of a dissipative process of any nature in the frequency region of interest is unwanted as it will lead to energy losses. Thus, to positively enhance the electrical energy storage capability of a dielectric material, the effective loss factor should be kept as low as possible. As a matter of fact, the dissipative losses not only waste part of the input electrical energy, but also deteriorate the insulation properties of the materials<sup>3</sup>. Thus high dielectric losses can bring down the effective breakdown field strength substantially.

Since last three decades, polymers have been tailored to suit the demanding areas of energy storage owing to their outstanding formability into intricate shapes. Although polymers possess high breakdown field strength in the range  $10^6$ – $10^8$  Vm<sup>-1</sup> and low losses<sup>3</sup> but their dielectric constant only ranges from 1–12, the highest being that of fluorinated polymers<sup>4</sup>. Thus, the contemporary challenge is to substantially increase the dielectric constant of polymers while retaining their excellent mechanical and processing properties. Consequently in recent times, methodologies to fabricate random composites of polymers, field structured composites and new polymers have been studied in detail to improve the dielectric properties of polymer based high dielectric materials<sup>3</sup>. Of these, the most common solution is the addition of fillers in polymeric matrices to make composites. The fillers which are used to make these composites include metals<sup>5-8</sup>, ceramics<sup>9-14</sup>, carbon based materials<sup>15-18</sup> and organic fillers, such as semi-conductive oligomers<sup>19, 20</sup> and conducting polymers<sup>21-23</sup>. In general, the advantages of the high dielectric ceramic filler based polymer composites include predictable dielectric properties, relatively low dielectric loss and easy fabrication<sup>3</sup>. However, there are other factors which restrict the use of the ceramic-polymer composites which are primarily related to the deterioration of mechanical and processing properties due to the high filler concentration of rigid ceramic particles in the flexible polymer matrix. A methodology to counter this drawback is the replacement of ceramic particles

with conductive particles to fabricate percolative polymer composites which can demonstrate drastic increase in dielectric constant in the vicinity of the percolation threshold of the conductive particles<sup>5-8, 15-18</sup>. However, the drastic increase in dielectric constant is always seen to be accompanied by considerable increase in electrical conductivity and dielectric loss due to the 'insulator-conductor' transition taking place at the percolation threshold. This type of transition is also reported to result into tremendous sensitivity of the dielectric constant to the concentration of the conductive fillers<sup>3</sup>. Hence even a small deviation from the percolation threshold could result in substantial drop of the dielectric constant, thus making the parameters of the preparation process quite difficult to control.

To overcome the negative aspects of ceramic as well as conductive fillers mentioned above, fillers with core-shell type of structure have been dispersed into polymers in recent times whereby the dielectric properties of the filler/polymer composites were reported to be improved upon tailoring the thickness and characteristics of the core-shell fillers<sup>24,25</sup>. Shen and co-workers have reported extremely stable high dielectric permittivity together with low dielectric loss of the polymer dielectrics through incorporation of core-shell fillers<sup>25</sup>. They have used silver particles coated with organic dielectric shells which not only act as inter-particle barricade to prevent silver particles from forming direct conductive pathway but also produce admirable compatibility between the fillers and the polymer matrix thus resulting in fair dispersion of fillers in the polymer matrix. Core-shell particles with conducting cores and insulating shells are thus ideal candidates to be used as fillers in polymer based percolative composites. The core-shell particles not only provide a barrier layer between the conductive fillers but they can also tune the dielectric properties of the resultant composites. This has led to the possibility of tailoring the dielectric properties of the polymer based composites through the incorporation of core-shell particles where the conducting filler particles are selectively coated with another polymer which has very low dielectric loss. Present study is thus an account of work done in this novel direction to evolve soft polymeric dielectric composite using innovative technique of conducting filler modification for enhanced dielectric performance (dielectric constant, dielectric loss and dielectric loss factor).

## 2 Experimental

### 2.1 Materials used

Acrylic resin (PMMA, polymethyl methacrylate powder), xylene, dichloromethane (DCM) and hexane were purchased from Sigma Aldrich and were used as received without further purification. Conducting carbon (Cabot VULCAN XC-605) was obtained from Cabot, USA and the silicone resin (Sylgard 184) was purchased from the Dow Chemical company.

### 2.2 Characterization techniques

Infrared spectra were recorded on a Perkin-Elmer FT-IR RXI spectrophotometer. Dielectric constant and dielectric loss factor ( $\tan \delta$ ) were evaluated using  $33 \pm 0.1$  mm diameter circular disc samples of thickness  $1.0 \pm 0.1$  mm obtained from the cast films of micro-composites on a DS6000 dielectric thermal analyzer from Lacerta Technology (U.K.) in the frequency range of 1-100 kHz. Thin coats of conducting silver paste were applied on both sides of the circular discs prior to the conduct of dielectric analyses. Morphology of the neat conducting carbon particles, the coated conducting carbon particles and the prepared formulations were studied through Carl Zeiss EVO50 scanning electron microscope. The samples containing coated carbon particles were cryo-fractured in liquid  $N_2$  followed by etching with a solution of 30% DCM in hexane in an ultrasonic bath in order to preferentially remove the coated carbon particles for carrying out SEM analysis.

### 2.3 Preparation of surface modified core shell conducting carbon particles

#### 2.3.1 Preparation of carbon particles

Carbon particles purchased were in the form of small particles of size less than 1 mm which were then crushed in a grinder for 5 min with a rotation speed of  $500 \text{ min}^{-1}$ , followed by ball milling at a set speed of 900 rpm using 1 mm sieve balls and then finally sieving was done using a sieve shaker to get particles size ranging from 5-20  $\mu\text{m}$  which could then be utilized for further coating as well as direct mixing with silicone elastomer.

#### 2.3.2 Coating of acrylic resin on conducting carbon particles through micro-ballooning method

5 gm of acrylic resin powder (PMMA) was dissolved in rectified xylene (75 mL) in a flask to form acrylic solution. A suspension of conducting carbon particles (15gm) in 750 mL of hexane was sonicated for 30 min at  $40^\circ\text{C}$ . This suspension was then allowed to stir vigorously under sonication using

overhead stirrer and acrylic solution was added drop-wise in a period of 1 hr, followed by stirring for additional 2 hours. The solution was then allowed to settle and the particles were then filtered and washed using vacuum suction to obtain core-shell type selectively coated conducting particles in the form of micro-balloons. These coated conducting carbon particles were then used for further mixing with silicone elastomer to form micro-composites.

#### 2.3.3 Preparation of conducting carbon particles filled silicone elastomer films

Crushed and sieved neat conducting carbon particles were mixed with silicone elastomer along with curing agent in a dual planetary mixture for 10 min to ensure proper mixing and degassing as per formulation provided in Table 1 and were labelled 10UCMC, 20UCMC and 30UCMC for micro-composites containing 10, 20 and 30 wt% uncoated conducting carbon filler respectively. The resultant composite solution was then poured in a flat mould which was closed to cast film of thickness  $1.0 \pm 0.1$  mm. The film got cured and solidified after 24 h and then it was further subjected to heat treatment for 1h in an air circulating oven at  $60^\circ\text{C}$  to ensure complete curing. The film was then cut into circular disc specimens of diameter  $33 \pm 0.1$  mm for dielectric analysis. Similar process was used to cast the films of silicone elastomer containing coated conducting carbon particles as per formulation provided in Table 1 which were labelled as 10 CMC, 20 CMC, 25 CMC and 30 CMC for micro-composites containing 10, 20, 25

Table 1 — Formulation of varied uncoated and coated carbon particles based micro-composites.

S. No.	Micro-composite batch (sample code)	Carbon particles (weight in gm)	Silicone resin with 10% curing agent (weight in gm)
1	Silicone resin + 10wt% uncoated carbon particles (10UCMC)	2	18
2	Silicone resin + 20wt% uncoated carbon particles (20UCMC)	4	16
3	Silicone resin + 30wt% uncoated carbon particles (30UCMC)	6	14
4	Silicone resin + 10wt% coated carbon particles (10CMC)	2	18
5	Silicone resin + 20wt% coated carbon particles (20CMC)	4	16
6	Silicone resin + 25wt% coated carbon particles (25CMC)	5	15
7	Silicone resin + 30wt% coated carbon particles (30CMC)	6	14

and 30 wt% coated conducting carbon filler respectively.

### 3 Results and Discussion

#### 3.1 Characterization of acrylic resin coated conducting carbon particles

The coated conducting carbon particles were characterized using FT-IR and SEM. The FT-IR analysis of the coated and uncoated samples of conducting carbon particles as well as neat acrylic resin showed clear distinction in transmission peak pattern. The coated carbon particles showed peak at  $1738\text{ cm}^{-1}$  corresponding to carbonyl peak. Peak at  $1371\text{ cm}^{-1}$  corresponds to aliphatic C-C stretching band. The peak at  $1218\text{ cm}^{-1}$  corresponds to etheral – C-O stretching. These peaks were absent in uncoated carbon as illustrated in Fig. 1.

The SEM photomicrographs of the uncoated and coated particle are illustrated in Fig. 2(a) and 2(b). These SEM photomicrographs demonstrated that uncoated as well as coated core-shell type particles were of size ranging from 5-20  $\mu\text{m}$ . However, the core-shell type of coated particles illustrates almost spherical shape instead of irregular structure of uncoated particles and a far better size distribution as well.

#### 3.2 Dielectric analysis

Sylgard 184 which is a vinyl terminated polydimethyl siloxane (PDMS), was used in this work as the matrix material based on the concept to fabricate foldable flexible dielectrics. The curing of the resin proceeds through a hydrosilylation reaction assisted by the organo-Pt catalyst (illustrated in

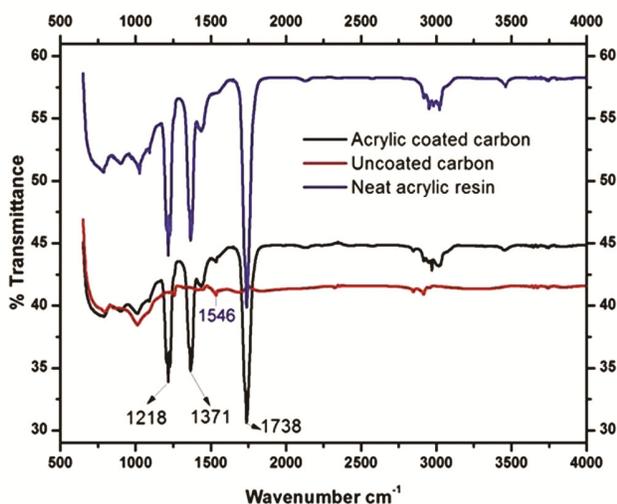


Fig. 1 — FTIR analysis of uncoated and acrylic coated carbon particles.

Fig. 3). The resin was selected on basis that there is almost negligible shrinkage upon curing with no side products. The resin itself demonstrated a low dielectric constant of 3.23 with a dielectric loss factor of 0.013 at 1 kHz.

Cabot VULCAN XC-605 conducting carbon filler was chosen as the filler in this study, as it has medium conductivity with excellent dispersion due to surface functionalities. The conductivity of particles is attributed to the surface graphitization<sup>26</sup>, resulting in both high dielectric constant as well as dielectric loss due to formation of conducting channel. The surface conjugated structure and hopping of charges illustrated through Fig. 4 is thus primarily responsible for the conductivity and origin of dielectric properties under applied electrical field.

The dielectric analyses of the samples were carried out at 30 °C by keeping the aluminium foil below and above the conducting silver paste coated polymer micro-composite discs of size  $33 \pm 0.1$  mm diameter and  $1 \pm 0.1$  mm thickness for uniform contact with the circular electrodes of diameter  $33 \pm 0.05$  mm. Figure 5 demonstrates a semi-log plot of dielectric constant values at varied applied frequencies of the electric field with respect to filler content. It can be seen from

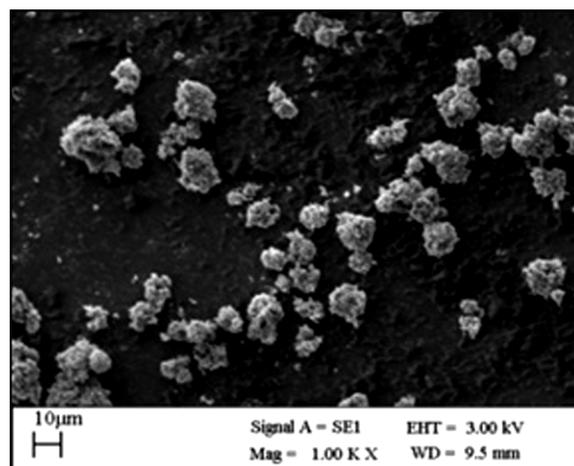


Fig. 2 — SEM images of (a) uncoated and (b) acrylic coated carbon particles.

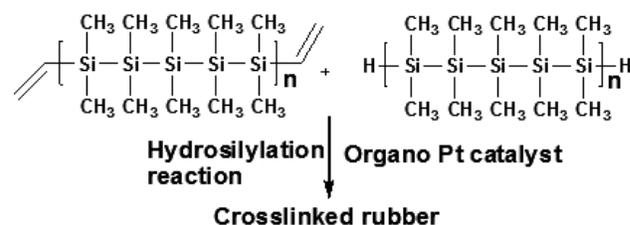


Fig. 3 — Organo-Pt catalyzed curing reaction of the silicone resin.

Fig. 5 that there is a substantial enhancement in dielectric constant (of the order of 280 times) in case of micro-composite containing 30 wt% uncoated carbon particles (30UCMC) when compared to neat silicone film. Subsequently all the other micro-composites show a modest increase in dielectric constant with respect to increasing carbon content. One more interesting feature that was observed was that micro-composites containing coated carbon particles (CMC) showed a much linear enhancement in dielectric constant with increase in filler content whereas in case of micro-composites containing uncoated carbon particles (UCMC), the enhancement in dielectric constant was by and large exponential.

Figure 6 denotes the effect of frequency of the applied electric field on the dielectric constant values of each of the micro-composites when compared to neat silicone resin. Many researchers have reported strong frequency dependence of dielectric permittivity in conducting fillers/polymer composites<sup>3</sup>. It can be easily construed from Fig. 6 that there was an exponential reduction in the dielectric constant values of the micro-composites containing uncoated carbon particles when the frequency was increased by each order of magnitude. However the micro-composites containing coated carbon particles, in this case, also show a much linear response to a change in frequency from 1 kHz to 100 kHz. This phenomenon could be attributed to the loss of interfacial polarization with the rise in the frequency of the applied electric field where dipoles get less time to orient themselves. Much more predominant effect of interfacial polarization was therefore observed at low frequency. Interfacial polarization<sup>27</sup> generally occurs in electrically heterogeneous materials (system containing phases of different specific conductivities) such as composites. This effect is commonly known as the Maxwell-Wagner-Sillars (MWS) effect<sup>28</sup>. Thus, at significantly low frequencies, the frequency dependent behaviour of dielectric constant is attributed to MWS effect which is largely associated with the confinement of free charges between insulator/conductor interfaces. The interfacial polarization thus causes an enhancement in dielectric constant due to motion of trapped virtual charges at the interface of components of a multiphase material of dissimilar conductivities. At higher frequencies this interfacial polarisation was not a predominant phenomenon so the dielectric constant decreased drastically. In case of coated carbon based micro-composites, the almost stable behaviour of the

dielectric constant could be attributed to low concentration of free charges between the interface of coated particles and polymer matrix due to an additional insulating layer of acrylic resin between the two, thus resulting in poor interfacial polarization

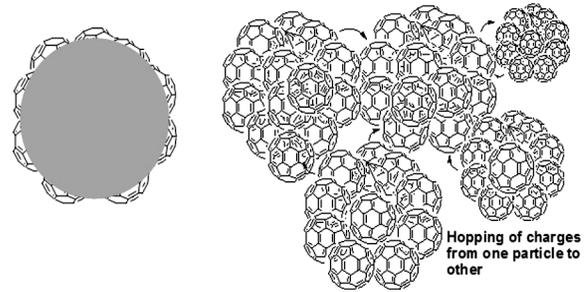


Fig. 4 — Structure of conducting carbon filler showing surface quasi-graphitic structure and hopping of charges via conducting conjugated pathway offered by sp<sup>2</sup> hybridized carbon skeleton.

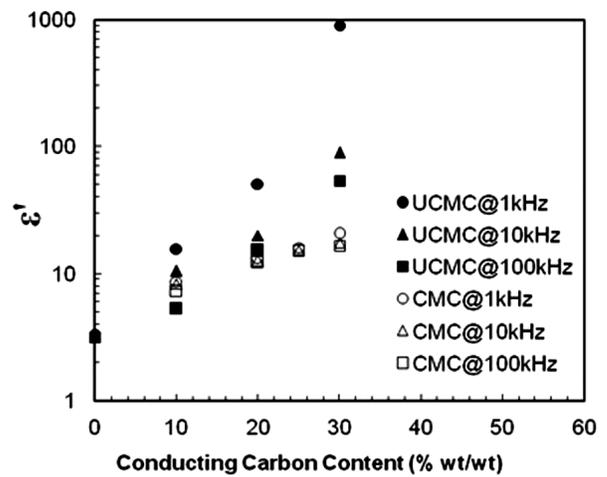


Fig. 5 — Dielectric constant of the micro-composites as the function of conducting carbon content measured at varied frequencies of the applied electric field.

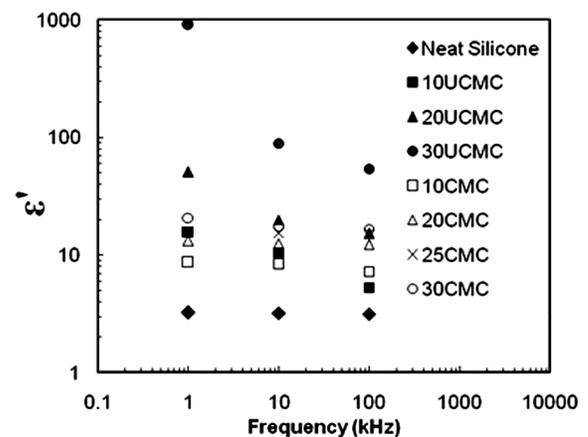


Fig. 6 — Dielectric constant of the micro-composites as the function of frequency of the applied electric field.

which is predominant phenomenon in composites having wide difference in conductivity of filler and matrix at low frequencies. Thus there was very feeble change in the dielectric constant values of the micro-composites containing coated carbon particles with an increase in frequency of the applied electric field. Also the dielectric constant in case of 25 CMC shows the most stable values at different frequencies which could be again attributed to the feeble contribution of the interfacial polarization because of optimal carbon content and presence of additional insulation layer of the acrylic resin coating on carbon particles.

Dielectric loss factor ( $\tan \delta$ ) gives an indicative figure of the energy consumption during charge storage, as it is directly proportional to the dielectric loss in the materials as illustrated in equation 3. Materials with a high dielectric loss exhibit disadvantages of high heat dissipation and low energy storage efficiency. Figure 7 illustrates the role of filler content on the dielectric loss factor ( $\tan \delta$ ) of the micro-composites. Here also, the micro-composites based on uncoated carbon particles show a much severe increase in  $\tan \delta$  values with respect to the filler content. In case of the micro-composites containing coated carbon particles there was hardly any significant change in the dielectric loss factor with respect to the increasing filler content. Most of the micro-composites containing coated carbon particles (CMC) show dielectric loss factors which are almost two orders of magnitude less than the corresponding micro-composites containing uncoated carbon particles (UCMC). Figure 8 shows the effect of frequency of the applied electric field on the dielectric loss factor of the micro-composites. As revealed in Fig. 6, in this case also, the micro-composites containing uncoated carbon particles show a substantial reduction in dielectric loss factor with increasing frequency whereas the micro-composites containing coated carbon particles show insignificant change in  $\tan \delta$  values with a change in frequency.

There are presumably two key sources of dielectric loss in these micro-composites. The first one is interfacial polarization loss caused by the enhancement of the frequency of the applied electric field which the charge carriers accumulated at the interface are not able to sustain it, especially in low frequency range. The second source is the conduction loss that arises as the result of leakage current induced by defects, pores, and closely contacted conducting carbon particles in the micro-composites. As

illustrated in Fig. 7, it can be observed that the dielectric loss factor of the 30 UCMC micro-composite is the maximum among all the micro-composites studied. For the 30 UCMC micro-composite with a very high filler loading, conduction loss was the main source of the dielectric loss. The uncoated carbon particles contact each other forming several conductive pathways in the micro-composites, so the dielectric loss increased with the addition of conductive carbon particles. In contrast, there is no appreciable change in the dielectric loss factor of micro-composites containing coated carbon particles with an increasing filler loading. The reason can be mainly attributed to the insulating core-shell structure of the acrylic coated carbon particles. The conductive carbon particle core was tightly wrapped by the insulating acrylic shell, which prevents direct contact between conducting carbon particles and restrains the

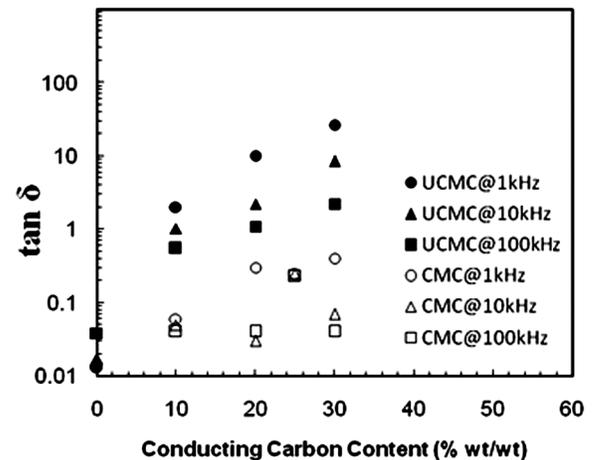


Fig. 7 — Dielectric loss factor ( $\tan \delta$ ) of the micro-composites as the function of conducting carbon content measured at varied frequencies of the applied electric field.

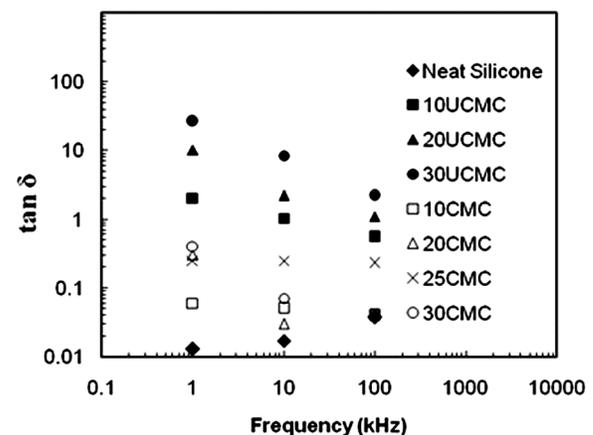


Fig. 8 — Dielectric loss factor ( $\tan \delta$ ) of the micro-composites as the function of frequency of the applied electric field.

mobility of free charge carriers. Similar results were also reported by Wang<sup>29</sup> and co-workers where they have explored a feasible strategy for improving the dielectric properties (enhanced dielectric constant with low dielectric loss) of carbon black/epoxy composites by utilizing core-shell particles. In this case also the dielectric loss factor in case of 25 CMC shows the most stable values at different frequencies which could again be accredited to the reasons already discussed in preceding paragraphs.

### 3.3 Morphological analysis

SEM images of uncoated particles showed highly irregular and coarse-spherical structure which upon coating formed globular structure through the employed micro-ballooning methodology (Fig. 2). Consequently, the SEM images of 10 UCMC and 10 CMC were illustrated in Fig. 9(a) and 9(b) respectively. The SEM image corresponding to 10 UCMC shows carbon particles which were quite far from each other and hence do not form a percolative structure as a consequence of which there was hardly any overwhelming improvement in the dielectric constant as compared to 30 UCMC. The SEM image corresponding to 10 CMC (Fig. 9(b)) also portrays some coated particles which were adhering to the silicone matrix while some particles were etched away. However, in the same manner as in the case of 10 UCMC, the coated carbon particles in 10 CMC were also far off from each other thus contributing much less in raising the dielectric constant of the micro-composite. A common phenomenon observed in both these micro-composites is that an increasing loading of fillers resulted in an increased dielectric constant, i.e. ca. 5 times at 1 kHz and ca. two times at 100 kHz in case of 10 UCMC when compared to base silicone resin. In the same manner the dielectric

constant of 10 CMC was increased by ca. 3 times at 1 kHz and ca. 2.5 times at 100 kHz when compared to the base silicone resin. Thus it could be assumed that the primarily source for the enhancement of dielectric constant at these filler loadings was through interfacial polarization arising out of MWS effect. However, the dielectric loss factor of 10 CMC was observed to be approximately two orders of magnitude less than that of 10 UCMC. This is primarily because of the added insulation layer interface of acrylic resin which was coated on the conducting carbon particles.

The SEM micrographs of 30 UCMC and 30 CMC have been illustrated in Figs. 10(a) and 10(b) and portray two extremely different morphological features. The SEM image of 30 UCMC showed a distinct percolative structure where the uncoated conducting carbon particles form several conducting pathways, resulting into substantial increase in the dielectric constant in the order of ca. 280 times at 1 kHz and ca. 18 times at 100 kHz when compared to base silicone resin. Usually, when the filler content of the uncoated carbon particles in the micro-composites was close to the percolation threshold, the uncoated carbon particles start to be in contact with each other to form a conductive network. In this network, lots of micro-capacitors consisting of uncoated carbon particles as electrodes and silicone resin as the dielectric are in parallel or in series in the micro-composites. Therefore, UCMC micro-composites with a high number of micro-capacitors have a greater capacity for charge storage, thus leading to a higher dielectric constant. On the other hand, the SEM image of 30 CMC showed a distinct droplet type of morphology where the droplets in the form of holes were formed due to the etching of the core-shell type

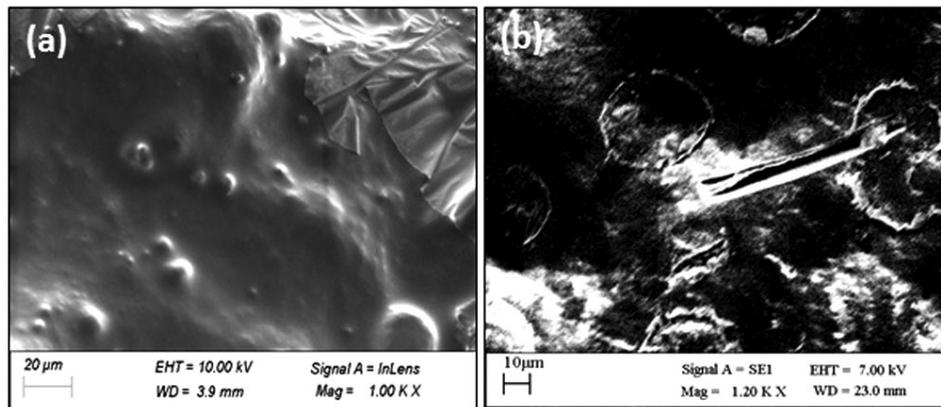


Fig. 9 — SEM images of micro-composites based on (a) uncoated and (b) acrylic coated carbon particles showing dispersion of fillers.

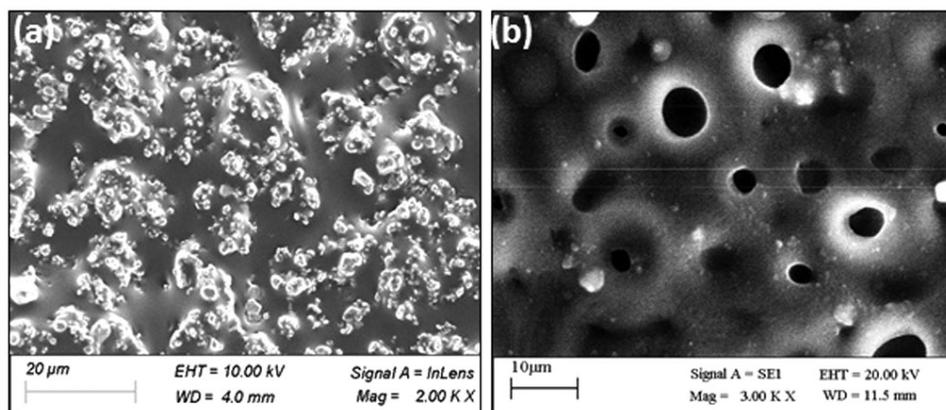


Fig. 10 — SEM images of micro-composites based on (a) uncoated and (b) acrylic coated carbon particles showing percolative and droplet type morphological features respectively.

of acrylic coated carbon particles. In this case, the key source of enhancement of dielectric constant was estimated to be from the interfacial polarization as seen in the case of 10UCMC and 10CMC resulting in the form of gradual improvement in the dielectric constant in the order of ca. 6 times at 1 kHz and ca. 5 times at 100 kHz when compared to neat silicone resin.

However, the percolative feature witnessed in case of 30UCMC (Fig. 10(a)), also led to a drastic increase in the dielectric loss factor of 30UCMC by more than three orders of magnitude at 1 kHz and by almost two orders of magnitude at 100 kHz in comparison to neat silicone resin. Contrary to this, the droplet type of morphological feature witnessed in case of 30CMC which had core-shell type of particles embedded in silicone matrix led to a stable dielectric loss factor where there was no change in the dielectric loss factor at 100 kHz when compared to that of the base silicone resin.

This is one of the proven ways to decrease the unwanted dielectric losses by using core-shell type of particles<sup>25, 29, 30</sup> where the conducting core was often coated with an insulating material having low dielectric loss factor. To cite an example, Shen and co-workers<sup>25</sup> studied percolative nano composites using core-shell structured particles as fillers where conductive Ag cores were coated with organic carbonaceous shells (denoted as Ag@C) which were mixed with an epoxy resin matrix. The organic carbonaceous shell was found to play a very significant role on the frequency dependence of the permittivity of the Ag@C/epoxy composite. In the similar way, Dang and co-workers have also reported that, the Ag@TiO<sub>2</sub> core-shell particles with thin TiO<sub>2</sub>

layer resulted in a weak frequency dependence of dielectric permittivity and dielectric loss in the Ag@TiO<sub>2</sub>/PVDF composites<sup>30</sup>. These studies strongly support that the frequency dependence of the dielectric properties in conducting fillers/polymer composites can be substantially minimized by changing the composition of composites and employing the core-shell structure particles with insulating shells.

#### 4 Conclusions

The goal of this research work was to evolve flexible dielectrics based on easily available and cost effective micron sized conducting carbon particles in the quest for obtaining composites with large electrical energy storage capabilities. In this study, the high dielectric loss shown by the conducting filler based micro-composites was overcome by incorporating a core-shell type of filler in the form of insulation layer coating on the conducting carbon filler. This coating methodology employed in the present study in the form of micro-ballooning has two key consequences. Firstly, favourable polymer-filler interactions promote better dispersion of the filler in the polymer matrix which leads to higher effective energy storage capabilities of the micro-composites. Secondly, the insulating layer coating on the conducting carbon particles gives rise to a set of dielectric properties which are stable at varied frequencies of the applied electric field, further leading to a more effective energy storage capability of the micro-composites. Thus, with the optimization of filler content, size of the filler and the insulation layer material, tailored flexible dielectric materials can be fabricated which can have a vast canvas of

applications ranging from mobile devices to pulsed power supply systems.

### Acknowledgements

The authors are thankful to the Director, DMSRDE, Kanpur for extending his support to carry out this work. The authors would also like to thank Dr. D.N. Tripathi, Scientist 'G' & Directorate Head, DPPMCC and other members of the Polymer Science Division of DMSRDE, Kanpur for their guidance and help rendered throughout this work. Additionally the authors are also grateful to Dr. Kavita Agarwal and Mr. Rakesh Kumar for their support in the SEM analysis.

### References

- Barber P, Balasubramanian S, Anguchamy Y, Gong S, Wibowo A, Gao H, Ploehn HJ & ZurLoye HC, *Mater*, 2 (2009) 1697.
- Osaka T & Datta M, *Energy Storage Systems for Electronics, Gordon and Breach: Amsterdam*, 2001.
- Dang ZM, Yuan JK, Zha JW, Zhou T, Li ST & Hu GH, *Prog Mat Sci*, 57 (2012) 660.
- Nalwa H. Handbook of Low and High Dielectric Constant Materials and Their Applications, Academic Press: London, 1999.
- Pothukuchi S, Li Y & Wong CP, *J Appl Polym Sci*, 93 (2004) 1531.
- Qi L, Lee BI, Chen S, Samuels WD & Exarhos GJ, *Adv Mater*, 17 (2005) 1777.
- Dang ZM, Peng B, Xie D, Yao SH, Jiang MJ & Bai J, *Appl Phys Lett*, 92 (2008) 112910.
- Li YJ, Xu M, Feng JQ & Dang ZM, *Appl Phys Lett*, 89 (2006) 072902.
- Ang C, Yu Z, Guo R & Bhalla AS, *J Appl Phys*, 93 (2003) 3475.
- Bai Y, Cheng ZY, Bharti V, Xu HS & Zhang QM, *Appl Phys Lett*, 76 (2000) 3804.
- Dang ZM, Lin YQ, Xu HP, Shi CY, Li ST & Bai J, *Adv Funct Mater*, 18 (2008) 1509.
- Rao Y, Ogitani S, Kohl P & Wong CP, *J Appl Polym Sci*, 83 (2002) 1084.
- Kakimoto MA, Takahashi A, Tsurumi TA, Hao J, Li L, Kikuchi R & Yamada S, *Mat Sci Eng B*, 132 (2006) 74.
- Cho SD, Lee JY, Hyun JG & Paik KW, *Mater Sci Eng B*, 110 (2004) 233.
- He F, Lau S, Chan HL & Fan J, *Adv Mater*, 21 (2009) 710.
- Dang ZM, Wang L, Yin Y, Zhang Q & Lei QQ, *Adv Mater*, 19 (2007) 852.
- Dang ZM & Nan CW, *J Appl Phys*, 93 (2003) 5543.
- Jiang MJ, Dang ZM & Xu HP, *Appl Phys Lett*, 90 (2007) 042914.
- Wang JW, Shen QD, Yang CZ & Zhang QM, *Macromolecules*, 37 (2004) 2294.
- Huang C & Zhang QM, *Adv Mater*, 17 (2005) 1153.
- Huang C, Zhang QM & Su J, *Appl Phys Lett*, 82 (2003) 3502.
- Li JY, Huang C & Zhang QM, *Appl Phys Lett*, 84 (2004) 3124.
- Yuan JK, Dang ZM & Bai J, *Phys Status Solidi: Rap Res Lett*, 2 (2008) 233.
- Wei T, Jin CQ, Zhang W & Liu JM, *Appl Phys Lett*, 91 (2007) 222907.
- Shen Y, Lin YH & Nan CW, *Adv Funct Mater*, 17 (2007) 2405.
- Pantea D, Darmstadt H, Kaliaguine S & Roy C, *Appl Surf Sci*, 217 (2003) 181.
- Liu GZ, Wang C, Wang CC, Qiu J, He M, Xing J & Yang GZ, *Appl Phys Lett*, 92 (2008) 122903.
- Yu Z & Ang C, *J Appl Phys*, 91 (2002) 794.
- Wang X, Li Z, Chen Z, Zeng L & Sun L, *Adv Ind Eng Polym Res*, 1 (2018) 111.
- Dang ZM, You SS, Zha JW, Song HT & Li ST, *Phys Status Solidi A*, 207 (2010) 739.