

Dielectric properties of silicone rubber based composites with nano-sized fillers

Ajay James Thomas* and Nandini Gupta*

Room Temperature Vulcanized Silicone Rubber (RTVSR) is one of the forms of silicone rubber used in outdoor insulating systems, usually in order to improve the pollution performance of ceramic insulation. RTV silicones are used to withstand mechanical and thermal stress. Introduction of nano-sized particles inside RTVSR alters the dielectric properties of the polymer. In this work, we study the change in dielectric properties of silicone rubber on introduction of nanofillers. The dielectric properties investigated include complex permittivity and space charge accumulation. The nanofillers incorporated are alumina and barium titanate at 1% concentration (by volume). The nanofillers are mixed mechanically and by ultrasonication to obtain well dispersed nanocomposites.

It is seen from the dielectric spectroscopy measurements that the alumina nanocomposite shows a higher complex permittivity compared to the barium titanate nanocomposite. Space charge measurements are carried out by the Pulsed Acoustic Method (PEA). The results indicate that the incorporation of nanoalumina leads to more space charge accumulation. On the other hand, the depletion of charge is faster with the incorporation of nanofillers.

Keywords: RTVSR, nano composite, complex permittivity, space charge accumulation

1.0 INTRODUCTION

Research in nanodielectrics has thrown up some interesting possibilities in the design of dielectric materials. Nanoparticles are incorporated in the bulk matrix of an insulating polymer, and they have sizes comparable to the intermolecular distances in the bulk material and hence tend to alter the properties of the material. Earlier work in the author's laboratory indicated that the addition of nanofillers in silicone rubber improves its resistance to tracking considerably [1]. In this work, the effect of incorporation of nanofillers on complex permittivity and space charge accumulation are investigated. Complex permittivity provides a measure of the energy storage and losses in the dielectric. Dielectric Spectroscopy experiments are performed to study the changes in complex permittivity of

silicone rubber and its nanocomposites. Further, the application of electric field to a dielectric, results in the accumulation of space charge within the volume leading to local field intensification and degradation of the material. The nature of space charge evolution inside silicon rubber and its nanocomposites is investigated by using the Pulsed Electro-Acoustic Method.

2.0 MATERIALS

The bulk polymer used in the experiments is RTV8112 from Momentive Performance Materials Solutions Inc., USA. RTV8112 is a white viscous liquid with faint odor. The flash point is around 298 °C. The compound has a density of 1.18 g/cm³ and a relative density of 1.2. It is insoluble in water but soluble in toluene or xylene. The hardener used is RTV9858 which

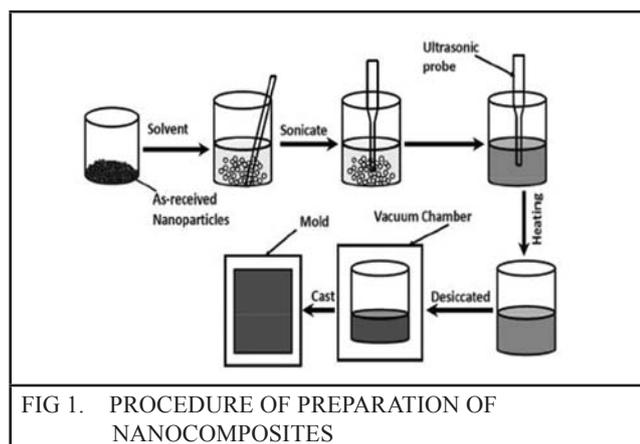
*Department of Electrical Engineering, Indian Institute of Technology, Kanpur-208016, India, email-ajay@iitk.ac.in

*Department of Electrical Engineering, Indian Institute of Technology, Kanpur-208016, India, email-ngupta@iitk.ac.in

is a white viscous liquid with faint odor. The boiling point and flash point are 285 °C and 93.5 °C respectively. The density and relative density are 1.61 g/cm³ and 1.61. It is insoluble in water and stable under normal conditions. The two nanofiller materials used are aluminum oxide and barium titanate. The alumina particles are on an average 50 nm in diameter with a density of 4 g/cm³. The barium titanate particles are on an average 100 nm in diameter with a density of 6.08 g/cm³. Both are in powder form. All particles are used as received or after heating. No surface processing is done. The solvents used are acetone and ethanol.

3.0 SAMPLE PREPARATION

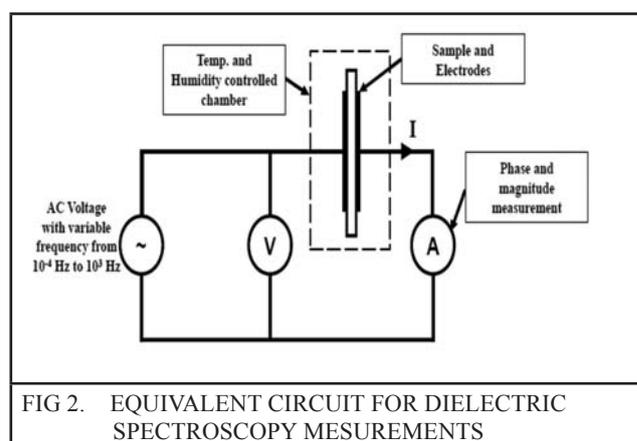
Pure silicone rubber samples are prepared by taking the required amount of RTVSR in the beaker and mixing with hardener in the ratio 10:1. The mixing is done slowly to ensure homogeneous mixing. The mixture is then degasified in vacuum for 15 minutes and finally poured into mould. The sample in the mould is allowed to cure for 24 hours at room temperature. Nanocomposites are prepared by taking required amount of nanoparticles in 100 ml of ethanol and mechanically stirred for 10 minutes. It is then ultrasonicated for 9 seconds on and 9 seconds off pulse mode. After ultrasonication, the ethanol-particle mixture is added to RTVSR and the final mixture is heated at 100 °C to remove the ethanol. Required amount of hardener is added to the mixture and degasification is done for 15 minutes in dessicator. The mixture is then poured to the mould and allowed to cure for 24 hours at room temperature. Samples are stored in a moisture-free, dust-free atmosphere.



4.0 EXPERIMENTS

4.1 Dielectric Spectroscopy

Dielectric spectroscopy experiments are carried out to measure complex permittivity. Figure 2 shows the equivalent circuit for dielectric spectroscopy measurements. An ac voltage of magnitude 1 V with variable frequency from 10⁻⁴ Hz to 10³ Hz is applied to the sample in a temperature and humidity controlled chamber. The voltmeter measures the voltage across the sample and the electrometer measures the phase and magnitude of the current through the specimen. From these measurements, the capacitive reactance and thus the complex permittivity of the sample is calculated. The real and imaginary permittivities are plotted separately for silicone rubber and its nanocomposites. All experiments are performed at room temperature.



4.2 Space Charge Measurements

The space charge measurements are performed by the Pulsed Electro Acoustic (PEA) method. The PEA is a non-destructive technique for measuring space charge accumulation under ac or dc fields. The sample is placed between the high voltage electrode and the earth electrode. High voltage is applied across the electrodes leading to the formation of space charge. A series of voltage pulses of very short duration is applied across the electrodes. This electric transient electric field interacts with the space charge and the surface charges creating an electrical force displacing the internal charges. This disturbance travels to the

piezoelectric transducer near the earth electrode in the form of pulsed acoustic pressure waves. The transducer converts this pressure signal to a voltage signal which is then calibrated and deconvoluted to obtain the charge density distribution in the dielectric. Figure 3 shows the schematic of a PEA system.

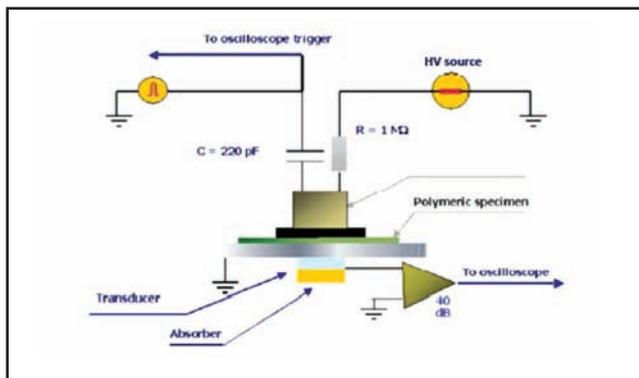


FIG 3. SCHEMATIC OF PEA SYSTEM

5.0 RESULTS AND DISCUSSIONS

5.1 Dielectric Spectroscopy

In this section we present the results and discussions of dielectric spectroscopy experiment performed on pure silicon rubber, silicon rubber alumina nanocomposite and silicon rubber barium titanate nanocomposite over a broad frequency range. This is expected to identify the principal polarization mechanisms in silicone rubber and how that is affected by incorporation of nanofillers. Figure 4(a) and 4(b) shows the real and imaginary characteristics for silicone rubber and its nanocomposites. The incorporation of nanoparticles has increased the real and imaginary permittivities in the entire frequency range. The alumina nanocomposite shows higher complex permittivity than pure silicone rubber and barium titanate nanocomposite.

Results in the past have shown a major polarization mechanism in it is dipolar polarization; this is expected since silicone rubber is polar in nature. Interfacial relaxation mechanisms have also been observed at lower frequencies [2]. This is corroborated in the present experiment from the imaginary characteristics of pure silicone rubber in figure 4(b).

The loss peak for pure silicone rubber appears between 0.01 Hz and 0.001 Hz. In case of silicone rubber alumina nanocomposite it appears around 0.001 Hz. The imaginary

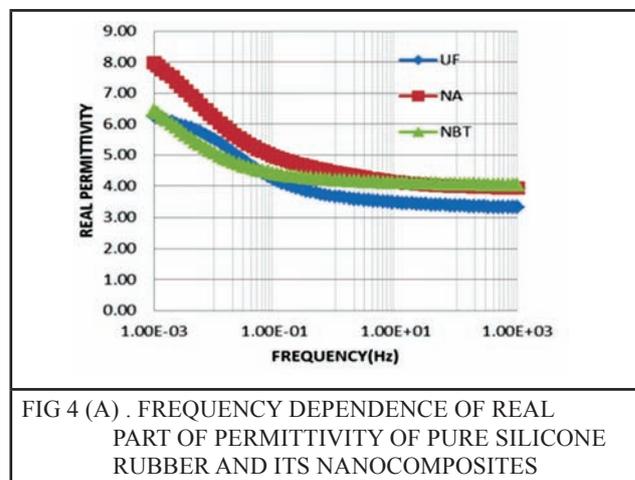


FIG 4 (A) . FREQUENCY DEPENDENCE OF REAL PART OF PERMITTIVITY OF PURE SILICONE RUBBER AND ITS NANOCOMPOSITES

characteristics of silicone rubber barium titanate nanocomposite shows a positive slope at 0.001 Hz; it is indicated that the loss peak would appear at frequencies less than 0.001 Hz. In this work, the loss peaks seems to be shifted to lower frequencies by the incorporation of nanofillers. This is corroborated by previous experimental findings [3-4].

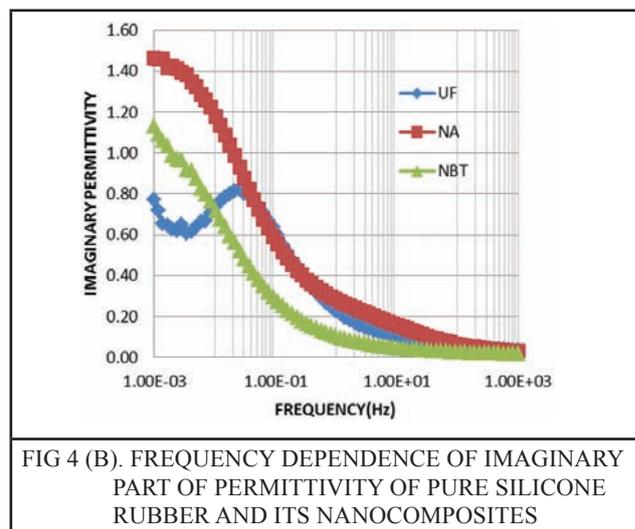


FIG 4 (B). FREQUENCY DEPENDENCE OF IMAGINARY PART OF PERMITTIVITY OF PURE SILICONE RUBBER AND ITS NANOCOMPOSITES

The real permittivity shows an overall increase when nanofillers are introduced. As pointed out earlier, the increase in the hydroxyl group density on the surface of nanoparticle increases the dielectric constant of the nanocomposites over the entire frequency range [5]. As nanoparticles are added into the matrix, a larger fraction of

atoms of nanoparticles get exposed at the surface increasing the moisture absorption and thereby hydroxyl groups at the surface [6].

UF- unfilled silicone rubber

NA- silicone rubber alumina nanocomposite

NBT- silicone rubber barium titanate nanocomposite

5.2 Space Charge Measurements

In this section we present the results and discussions of space charge measurements of pure silicone rubber and its nanocomposites. Figures 5, 6 and 7 shows the depolarization signals for pure silicone rubber, silicone rubber alumina nanocomposite and silicone rubber barium titanate nanocomposite at different instants of depolarization respectively.

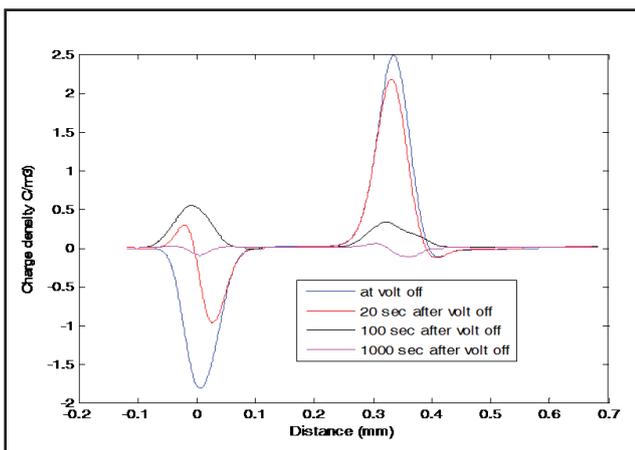


FIG 5. DEPOLARISATION SIGNALS OF PURE SILICONE RUBBER

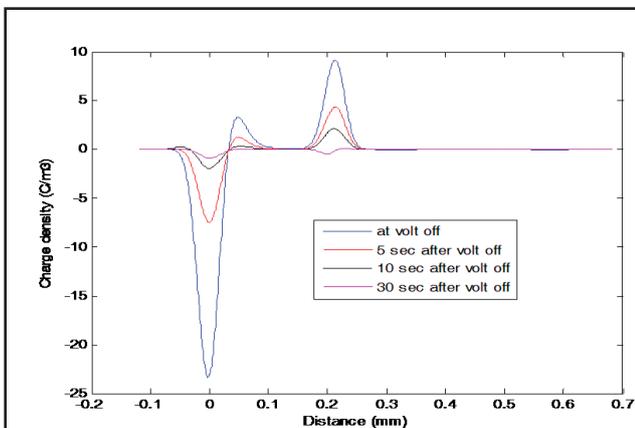


FIG 6. DEPOLARISATION SIGNALS OF SILICONE RUBBER ALUMINA NANOCOMPOSITE

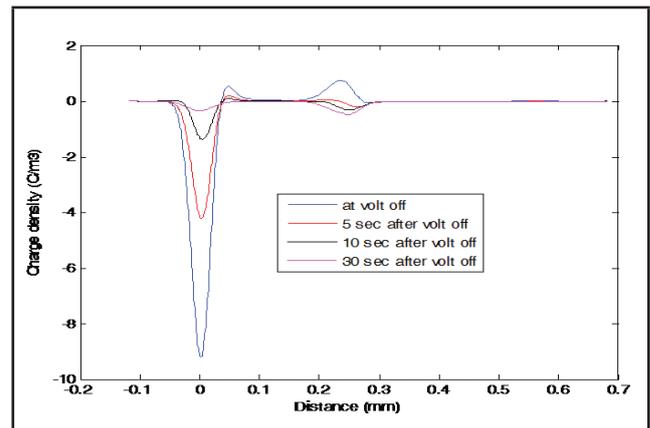


FIG 7. DEPOLARISATION SIGNALS OF SILICONE RUBBER BARIUM TITANATE NANOCOMPOSITE

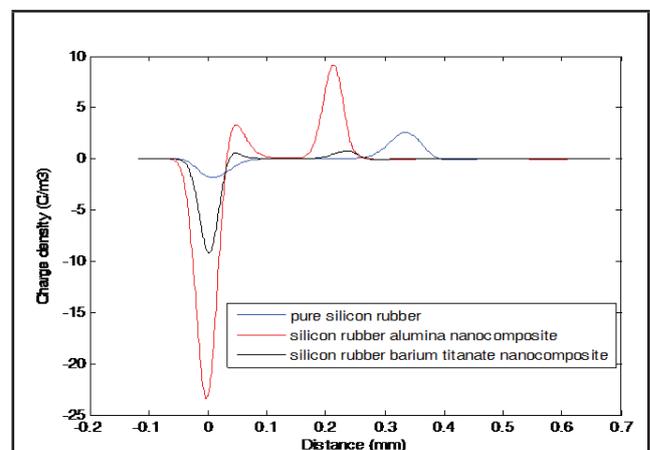


FIG 8. SPACE CHARGE SIGNALS OF PURE SILICONE RUBBER AND ITS NANOCOMPOSITES AT THE END OF POLARISATION

The depletion of space charge in pure silicone rubber takes around 1000 seconds. But for nanocomposites the depletion is much faster (within 30 seconds). Figure 8 shows the space charge signals at the end of polarization for pure silicone rubber and its nanocomposites. Space charge accumulation in alumina nanocomposite is found to be highest followed by barium titanate nanocomposite. In general the nanocomposites accumulate more charge inside the bulk compared to pure silicone rubber. All experiments were repeated on several specimens, and the deviation was found to be within 5%.

6.0 CONCLUSIONS

It can be concluded that the incorporation of nanofillers have increased the real permittivity

and losses. Alumina nanocomposite shows the highest real permittivity and losses. Earlier work however has indicated that resistance to surface tracking is higher with barium titanate rather alumina nanofillers [7]. It needs further experimentation to conclude if this is a material effect or a size effect.

In the case of space charge accumulation, the addition of nanofillers intensified the space charge accumulation but the depletion was faster in nanocomposites compared to pure silicone rubber.

REFERENCES

- [1] Rakesh Kumar, Nandini Gupta, "Tracking and surface degradation of barium titanate filled silicone rubber nanocomposites,"
- [2] IEEE Conference on Electrical Insulation and Dielectric Phenomena (CEIDP), pp. 495-498, 2015,
- [3] U. H. Nguyen I, A. Sylvestre I, P.Gonon I, S. Rowe, "Dielectric properties analysis of silicone Rubber" , 2004 International Conference on Solid Dielectrics", Toulouse, France, July 5-9, 2004
- [4] Chao Zhang and Gary C Stevens." The Dielectric Response of Polar and Non PolarNanodielectrics",IEEE Transactions on Dielectrics and Electrical Insulation Vol.15, No.2; April 2008
- [5] N.Fuse et al.:Effects of Nanofiller Loading on the Molecular Motion and Carrier Transport In Polyamide.IEEE Transactions on Dielectrics and Electrical Insulation Vol.16, No.2; April 2009
- [6] Xingyi Huang, Liyuan Xie, Ke Yang, Chao Wu, Pingkai Jiang," Role of Interface in Highly Filled Epoxy/BaTiO3 Nanocomposites. Part I-Correlation between Nanoparticle Surface Chemistry and Nanocomposite Dielectric Property", IEEE Transactions on Dielectrics and Electrical Insulation Vol. 21, No. 2, April 2014
- [7] Parimal Maity "Dielectric Behaviour of Epoxy Resin based Composites with Nanosized Alumina Fillers", PhD Thesis,Indian Institute of Technology Kanpur March, 2009
- [8] Rakesh Kumar, HK Chakerwari, Nandini Gupta, "Improvement of dielectric performance of Silicone rubber by incorporation of nano-sized fillers", IEEE International WIE Conference on Electrical and Computer Engineering (WIECON-ECE), pp. 147-150, 2015

