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Analysis of Relative Permittivity and Tan Delta Characteristics of Silicone Rubber Based Nano-composites

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***Abstract* - Owing to the continuous development towards the miniaturization of electronics, newer dielectric materials were sought which would enable to achieve high energy density for many applications. To achieve a compact and reliable design of electrical equipment for the present day requirements, there is an urgent need for better and smart insulating materials and in this respect, the reported enhancements in dielectric properties obtained for polymer nanocomposites seems to be very encouraging. Such change was often favourable for engineering purpose. In this case, we obtain a high dielectric permittivity and low dielectric loss. So, it can be used in many applications like electrical insulation for power apparatus, power cables, insulated wires and outdoor insulators too.**

***Keywords*- High voltage insulation Nanocomposites,FTIR,Relative permitivity and tan delta**

I. INTRODUCTION

Ceramics possessing very high dielectric permittivity are being used as voltage capacitors due to their high breakdown voltages. However, they are brittle, suffer from poor mechanical strength and hence cannot be exposed to high fields. Though polymers possess relatively low dielectric permittivity, they can withstand high fields, are flexible and easy to process.]By combining the advantages of both, one can fabricate new hybrid materials with high dielectric permittivity, and high breakdown voltages to achieve high volume efficiency and energy storage density for many applications. The development of nanocomposites represents a very attractive route to upgrade and diversify properties of old polymers without changing polymer composition and processing. In contrast to conventional filled polymers, nanocomposites are composed of nano-fillers which are homogeneously dispersed within the polymer matrix. Polymer nanocomposites are defined as polymers in which small amounts of nanometer size fillers are homogeneously dispersed by only several weight percentages. Addition of just a few weight percent of the nano-fillers has profound impact on the physical, chemical, mechanical and electrical properties of polymers. The insertion of nano-scale fillers may improve the electrical and dielectric properties of the host polymers and the properties can be tailored to a particular performance requirement. In all an agog, our project focuses at developing a novel polymer composite for dielectrics and electrical insulation. Recently these 'hi-tech' materials with excellent properties to meet the future demand of insulators for higher voltages with minimized cost and size have begun to attract research people in the field of dielectrics and electrical insulation [5].

II. SAMPLE PREPARATION

A. Sample selection

The samples are prepared by choosing silicone rubber as base matrix in which the nanofillers of alumina, Titanium di Oxide and barium titanate are selected to produce the Polymer Nano - composites which has good insulating and dielectric properties .

B. Synthesis of samples

The samples are prepared at M/S Acme Industries, Madurai. Initially Silicone rubber is in the form of latex that is the combination of lactic acid and water. Then Silicone rubber is passed through the two roll mill and nano fillers are added and rolled. The process is repeated for 10-15 times. Finally it is passed through the dye under high temperature of 200°c and compressed to get the desired shape and thus the samples are prepared. The following figure shows the diagram of two roll mill

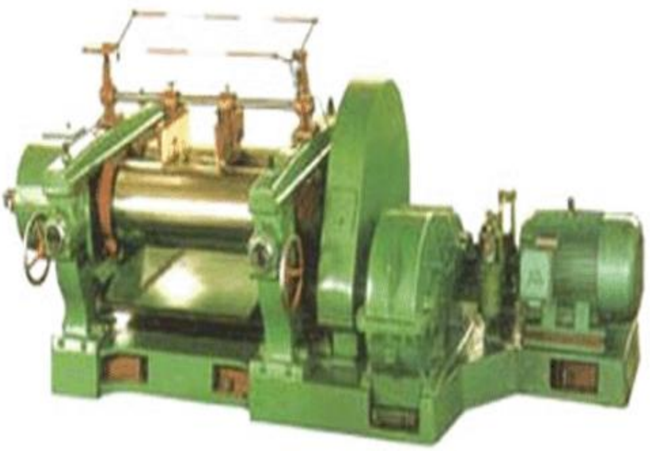

Figure 1. Two roll mill

Silicone rubber nanocomposites at various weight percentage of nano fillers like alumina, titanium oxide, barium titanate are prepared with the a diameter 90mm and thickness of 2mm/4mm/6mm/8mm are prepared [2].

Totally there are forty number of samples are prepared for our analysis. That is pure silicone rubber of Four numbers with the dimensions of 2mm, 4mm, 6mm and 8mm are prepared. In addition to that the nano fillers of Alumina ,Rutile(Titanium di Oxide) and Barium Titanante($BaTiO_3$) are mixed at 1%,2%,3% weight percentage with the pure Silicone rubber of twelve numbers each.

III. FOURIER TRANSFORM INFRARED SPECTROSCOPY

A beam of infrared light is produced and split into two separate beams. One is passed through the sample, the other passed through a reference which is often the substance the sample is dissolved in. The beams are both reflected back towards a detector, however first they pass through a splitter which quickly alternates which of the two beams enters the detector. The two signals are then compared and a printout is obtained.

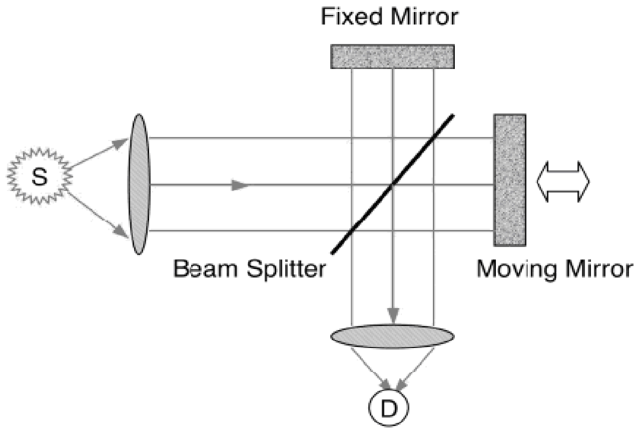

Figure 2. Fourier Transform Infrared Spectroscopy (FTIR)

Fourier Transform Infrared Spectroscopy (FTIR) is a powerful tool for identifying types of chemical bonds in a molecule by producing an infrared absorption spectrum that is like a molecular "fingerprint". FTIR is most useful for identifying chemicals that are either organic or inorganic. It can be utilized to quantitative some components of an unknown mixture. It can be applied to the analysis of solids, liquids and gases.

A. Characterization results of Fourier Transform Infrared (FTIR) Spectrography

The FTIR Spectrographs were taken and it is found that the transmittance is reduced by increasing the nano-filler content [7].

From the figure, it is clear that the densely arranged region at the last part of waveform is known as finger print region.it shows the transmittance value will be fluctuated at the beginning where as the transmittance value will be decreased at higher wave number.It is abvious that, the transmittance value will be decreased by varying the nanofillers of TiO_2 (2%), BaTiO3 (2%), when compared to pure rubber.

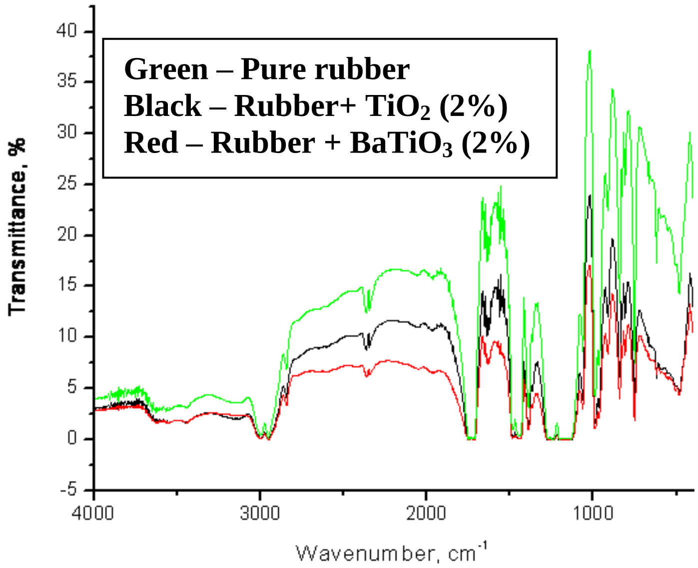

Figure 3. FTIR Spectra of synthetic rubber with Nano – composites

B. Measurement of AC impedance

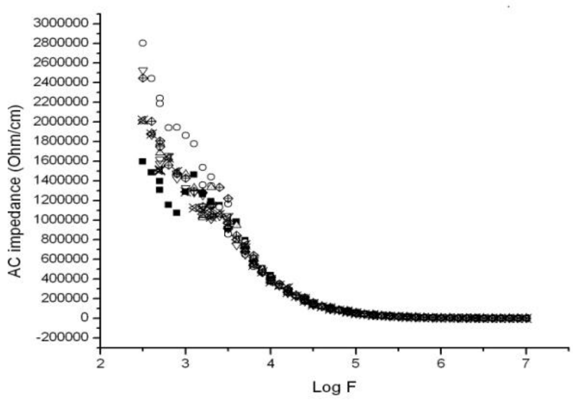

Figure 4. log f vs AC impedance

The measurement of AC impedance was carried out using a EC & G, USA- Potentiostat/ Galvanometer. The frequency dependent AC impedance behaviour of synthetic rubber – TiO_2 composites is shown in figure. The AC impedance of synthetic rubber is in the order of 1.6 x 10^5ohms, and the composites exhibited higher AC impedance response than that of the pure synthetic rubber. The impedance of synthetic rubber decreases gradually up to 100

KHz and decreases sharply beyond 100 Hz. All the composites exhibited a similar trend, but the impedance value is almost constant after 10 kHz. In this work, it is observed that the ac impedance increases with filler concentration for low frequency and at high frequency there is no significant change.

IV. RELATIVE PERMITTIVITY TEST

For the permittivity test the samples are tested at Central Electronics Engineers Research Institute (CEERI), Pilani. The samples are tested with **AGILENT 4284A** LCR meter.

The present work looks into two of the dielectric properties in rubber nanocomposites - permittivity and tan delta. Permittivity determines the charge storage capacity of a dielectric material as well as dictates the electric field distribution in a composite insulation system

To further understand the dielectric behavior of polymer nanocomposites, this experimental work reports the trends of dielectric permittivities and tan delta (loss tangent) of rubber nanocomposites with single nanofillers of $BaTiO_3$ and TiO_2 at low filler concentrations [3].

A. Effect of filler material

Nano-filler is a reinforced in polymer or primary particle compound of nanometer in range. The various Nano-fillers of Titanium di Oxide – TiO_2, Barium Titanante – $BaTiO_3$ are selected to enhance the relative permittivity [1].

The variations of dielectric permittivity for various nano-fillers at different filler concentrations characteristics will be determined by orientation polarization effects (where dipoles tend to orient themselves to an applied electric field).

Apart from this, there is a possibility for unreacted dipolar groups to exist which can influence the permittivity of the nanocomposite medium. These functional groups having definite dipole moments will contribute to the polarization mechanisms in the epoxy nanocomposites which in turn influences the permittivity of the system [3]. Using Lichteneker-Rother rule [equation (1)],

$$\text{Log } \varepsilon_c = x \text{ Log } \varepsilon_1 + y \text{ Log } \varepsilon_2 \quad .... (1)$$

Where, ε_c is the effective composite permittivity, ε_1 and ε_2 are the permittivity of filler and polymer and x, y are the concentrations of filler and polymer respectively.

The nanocomposite effective permittivity will be governed by the occurrence of two processes – minimum effect of nano-filler permittivity and strong polymer-nanoparticle interactions. This is probably the reason why the lowest effective permittivities in both $BaTiO_3$ and TiO_2 filled rubber nanocomposites are observed

The following table clearly shows the relative permittivity value is increased when nanofiller is added.

Table 1. Effect of nano fillers at 308K for 100 HZ

Materials	**Relative permittivity**
Pure rubber	7.5
1% TiO_2	8
2% TiO_2	14
1% $BaTiO_3$	16
2% $BaTiO_3$	23.5

B. Effect of filler concentration

The other interesting observation evident from the figures is that up to a certain nano-filler concentration, the permittivities of the rubber nanocomposites are less than that of the unfilled rubber in the entire frequency range [5].

An increase in the nano-filler concentration in rubber results in an increase in the number of nanoparticles and with the individual permittivities of each of these particles contributing to the permittivity of the composite, there is an increase in the effective permittivity[4].

The following table clearly shows how the relative permittivity value is increased when nanofiller is added at different weight percentages.

Table 2. Effect of nano fillers concentration at 328K for 100 HZ

Materials	**Relative permittivity**
Pure rubber	11
1% TiO_2	11
2% TiO_2	14
1% $BaTiO_3$	16
2% $BaTiO_3$	23

C. Effect of frequency

The permittivity of pure rubber will marginally decrease with increasing frequency in the present frequency range of 100 Hz - 1 MHz due to reduction in the polarizations caused by dipolar groups.Similarly, the permittivity in $BaTiO_3$ and TiO_2 also reduces with increasing frequency.

The nanocomposites with different fillers at different frequencies interms of logf. The graph is plotted with different relative permittivities at different temperatures.The permittivity values are decreased as the frequency increases.

The relative permittivity is determined for various nanocomposites along its nanofillers, when the frequency changes and to keep the temperature remains constant.For example, the percentage rise of relative permittivity for pure rubber to that of 1% $BaTiO_3$ at 100 Hz is 113.33% where as for pure rubber to that of 1% $BaTiO_3$ at 1000 Hz is 136.36% by keeping the temperature remains constant for 308K.

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Table 3. Log f Vs Relative permittivity at 308 K

Log f	Pure rubber	1% TiO_2	2% TiO_2	1% $BaTiO_3$	2% $BaTiO_3$
2	7.5	8	14	16	23.5
3	5	5	9	7.5	12.5
4	4.5	4.5	7	5.5	8
5	4	4	5.5	4.5	6
6	3.75	3.75	4	4	5.5

D. Effect of temperature

The graph is plotted with different relative permittivities at different temperatures, as the temperature values are increased the relative permittivity also increases.

For example, the percentage rise of relative permittivity for pure rubber to that of 1% $BaTiO_3$ at 308K is 113.33% where as for pure rubber to that of 1% $BaTiO_3$ at 328K is 136.36% by keeping the frequency remains constant for 100 Hz.

Table 4. Log f Vs Relative permittivity at 1000Hz
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Temperature	Pure rubber	1% TiO_2	2% TiO_2	1% $BaTiO_3$	2% $BaTiO_3$
308K	5	5	9	7.5	12.5
328K	6	6	10	7.5	12
348K	8	8.5	11	7.5	15
368K	10	10	11.5	8.5	15.5

V.DIELECTRIC LOSS TEST

For the dielectric loss test the samples are tested at Central Electronics Engineers Research Institute (CEERI), Pilani. The samples are tested with **AGILENT 4284A** LCR meter

Tan delta values indicate the dielectric losses possible in an insulating material. For any electrical insulation system, a low tan delta value is always desired in the dielectric material whereas the desired permittivity of the material must be high depending on our application [3].

A. Effect of filler material

Nano-filler is a reinforced in polymer or primary particle compound of nanometer in range. The various Nano-fillers of Titanium di Oxide – TiO_2, Barium Titanante – $BaTiO_3$ are selected to decrease the dielectric loss [1].

In polymers or their composites; loss tangent is a function of the electrical conductivity (which depends on the charge carrier mobility) and the applied excitation frequency.At lower frequency ranges, the incorporation of fillers does not introduce too much variation in the tan delta values with respect to the unfilled rubber value.

The following table clearly shows the dielectric loss value is decreased when nanofiller is added.

Table 5. Effect of nano fillers at 308K for 100 HZ

Materials	Dielectric Loss
Pure rubber	0.225
1% TiO_2	0.325
2% TiO_2	0.305
1% $BaTiO_3$	0.27
2% $BaTiO_3$	0.18

B. Effect of filler concentration

In polymers or their composites, loss tangent is a function of the electrical conductivity (which depends on the charge carrier mobility) and the applied excitation frequency. It can be seen from these graphs that in both the $BaTiO_3$ and TiO_2 filled rubber nanocomposites, there is a marginal (but continuous) decrease in tan delta values with increasing frequency for all filler concentrations.

As the frequency increases, the tan delta values in filled rubber for all filler concentrations are observed to reduce in comparison to the unfilled rubber tan delta.

The following table clearly shows how the dielectric loss value is decreased when nanofiller is added at different weight percentages.

Table 6. Effect of nano fillers concentration at 328K for 100 HZ

Materials	Dielectric Loss
Pure rubber	0.230
1% TiO_2	0.375
2% TiO_2	0.325
1% $BaTiO_3$	0.28
2% $BaTiO_3$	0.21

C. Effect of frequency

At high frequencies, the motion of charge carriers contributing to the conductivity primarily occurs along polymer chains. A barrier to the charge transport in polymers (causing reduction in electrical conductivity) can occur due to defects, inter-chain charge transport and transport through interfaces [5].

Probably, in nanocomposites, the presence of a large number of interfaces and polymer chain entanglements inhibit the motion of charges in the system, which in turn causes a reduction in the electrical conductivity (hence a lower tan delta value).

For example, the percentage rise of relative permittivity for pure rubber to that of 1% $BaTiO_3$ at 100 Hz is -20% where as for pure rubber to that of 1% $BaTiO_3$ at 1000 Hz is 40% by keeping the temperature remains constant for

308K.The following table shows how the dielectric losses are decreased as the frequency increased.

Table 7. Log f Vs Dielectric Loss at 308 **K**

Log f	Pure rubber	1% TiO_2	2% TiO_2	1% $BaTiO_3$	2% $BaTiO_3$
2	0.225	0.325	0.305	0.27	0.18
3	0.125	0.275	0.255	0.195	0.175
4	0.075	0.175	0.235	0.18	0.17
5	0.05	0.105	0.23	0.175	0.16
6	0.025	0.1	0.225	0.165	0.155

E. Effect of temperature

There are basically two different interacting processes which might influence tan delta behavior in nanocomposites. The first one is the number of charge carriers available for electrical conduction and the other is the number of interfaces and polymer chain entanglements in the bulk [6].

The dielectric loss values are decreased as the temperature increases for various nanocomposites along its nanofillers, when the temperature changes and to keep the frequency remains constant.

For example, the percentage rise of relative permittivity for pure rubber to that of 1% $BaTiO_3$ at 308K is -20% where as for pure rubber to that of 1% $BaTiO_3$ at 328K is -9% by keeping the frequency remains constant for 100 Hz.

Table 8. Log f Vs Dielectric Loss at 10 KHz

Temperature	Pure rubber	1% TiO_2	2% TiO_2	1% $BaTiO_3$	2% $BaTiO_3$
308K	0.075	0.175	0.235	0.18	0.17
328K	0.1	0.25	0.235	0.195	0.2
348K	0.125	0.275	0.25	0.215	0.22
368K	0.225	0.3	0.255	0.215	0.245

VI. CHARACTERIZATION RESULTS

The following graphs show how the dielectric permittivity values and tan delta values vary with respect to log f.

A. Dielectric permittivity

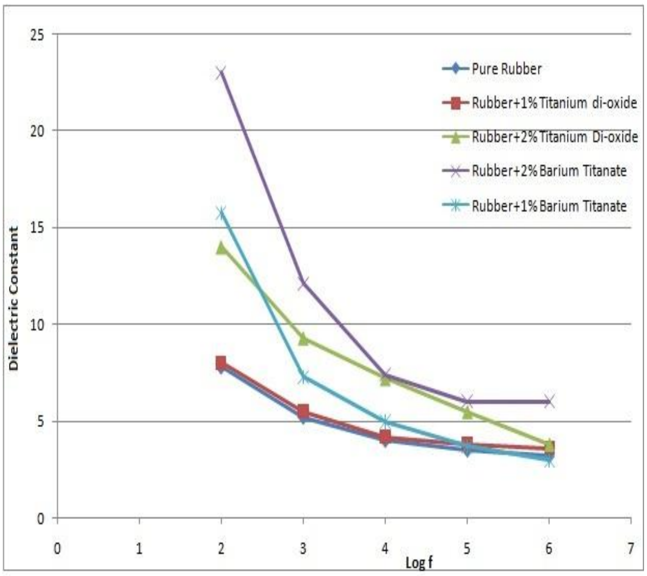

Figure 5. Dielectric Constant vs Log f

The relative permittivity for pure rubber, Titanium di Oxide, Barium Titanate is drawn against log f at the room temperature of 308k.The result shows the permittivity is increased when filler is added to the rubber when compared to the pure rubber.

C. Tan-Delta test

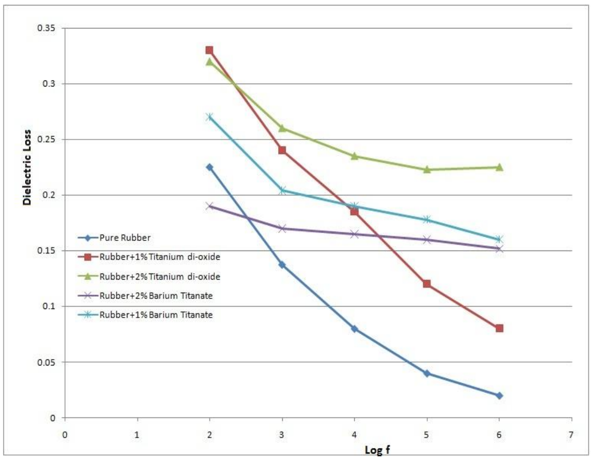

Figure 6. Dielectric Loss vs Log f

The dielectric loss for pure rubber, Titanium di Oxide, Barium Titanate is drawn against log f at the room temperature of 308k.The result shows the dielectric loss is decreased when filler is added to the rubber when compared to the pure rubber.

VII. CONCLUSION

Polymer nanocomposites could be advantageous over traditionally filled polymers in electrical and thermal properties as well as mechanical properties from the stand point of dielectrics and electrical insulation. This feature will technologically result in compact design of electrical equipments with high reliability and thereby in significant cost reduction for system integration and maintenance. In order to obtain excellent but low cost polymer nanocomposites, existing material processing technologies should be more advanced so as to match dielectrics and electrical insulation. Synthetic rubbers along with Titanium di Oxide, Barium Titanate nanocomposites are synthesized and their characterizations are studied. The characteristics of dielectric permittivity and dielectric loss in pure synthetic rubber along with Titanium di Oxide, Barium Titanate nanocomposites are found over a frequency range from 100Hz-10MHz. In summary, it is the rubber-filler interface which determines the behaviors of the effective permittivity and tan delta values in epoxy nanocomposites and a complete understanding of the interfacial phenomena is of utmost importance to fully understand not only these two dielectric parameters.The dielectric permittivity increases where as dielectric losses decreases with increasing filler concentration.

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