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MECHANICAL AND ELECTRICAL STUDIES OF NANOGRAPHITE REINFORCED SILICON ELASTOMERNANOCOMPOSITES

Janita Saji
E-Mail Id: janita.saji@rediffmail.com
Department of Sciences and Humanities
Christ University Faculty of Engineering, Bangalore, India

Abstract-Nanographite reinforced silicon elastomer nanocomposites were prepared. The morphology of nanocomposite samples have been studied from scanning electron microscope (SEM) photographs. It shows excellent distribution of nanographite in the silicon matrix. The effect of nanographite loadings (2, 4, 6, 8 parts per hundred parts of rubber) on mechanical properties like hardness, tensile strength, elongation at break, modulus (100%, 200%, 300%) have been studied. The study shows increase in tensile strength, hardness and modulus and decrease in elongation at break with nanographite loading. Dynamic mechanical properties of nanocomposites have been studied as a function of temperature (-100 to 100°C) at a constant frequency 1Hz and strain 1%. The effect of nanographite loadings on storage modulus, loss modulus, and loss tangent has also been studied. The non-linearity in tan delta, storage modulus and loss modulus was explained on the basis of nanographite elastomer interaction. The smooth cole-cole plots explain the non-linearity in the elastomer nanocomposites as well as good distribution of nanographite in the elastomer matrix. The effect of nanographite loading on cole-cole plots is explained on the basis of relaxation phenomenon. The phenomenon of percolation has been discussed, based on the measured changes in electric conductivity and dielectric permittivity. The percolation threshold occurred in the range of 6 phr of nanographite loading.

Keywords: elastomer, nanocomposite, modulus, dynamic mechanical, conductivity, percolation.

1. INTRODUCTION

For all practical applications (except adhesives), elastomers are reinforced with fillers to improve their performance by incorporating inorganic fillers. Traditionally this has been achieved by using materials such as carbon blacks, clays, talc and silica. But new generation fillers like carbon nanotubes, carbon silica dual phase filler, nanoclays and nanographite are continuously challenging the hegemony of these traditional fillers. The advantage of using nano scale fillers is their very high surface to volume ratio which leads to greater polymer-filler interactions thereby giving rise to significantly improved properties. Mechanical and dynamic-mechanical properties of composites reinforced with nano sized fillers show better properties than conventional micro-sized fillers. Decreasing size of the filler gives larger specific surface area thereby providing greater interactions between the polymer and filler leading to better properties. In recent years, there has been considerable research on nano particle based composites. Especially nanoclay reinforced plastics [1-3] and elastomers [4, 5] have been extensively studied. Graphite can provide additional advantages like excellent electrical and thermal conductivity. They also show good affinity for both organic compounds and polymers, therefore some monomers and polymers can be absorbed into the pores and galleries of graphite. Though there have been many studies on the effect of addition of nano-graphite in polymer matrix, it hasn't been reported in elastomer matrices. Fillers also strongly influence the visco-elastic properties of elastomer compounds. The dynamic properties of elastomers filled with structural fillers are influenced by factors like type of fillers, volume fraction of filler, processing conditions and strain history.

The objective of the present work is to investigate the mechanical and electrical properties of nanographite reinforced with silicon elastomer nanocomposites. The effects of filler and temperature on dynamic viscoelastic properties of the nanocomposites were studied.

2. EXPERIMENTAL

2.1. Materials

Silicon Elastomer (Mooney viscosity ML_{1+4} at 100°C =60) was procured from Bayer AG. Nano graphite of purity ≥ 95% were purchased from Nanoshel LLC, USA. Other chemicals like zinc oxide specific gravity of 5.4, stearic Acid and di cumyl peroxide used as curing agent were chemically pure grade procured from standard suppliers.

2.2. Compounding and Sample Preparation

The rubber was compounded with the ingredients according to the formulation of the mixes (Table-2.1).

Compounding was done in a brabender plastograph at 60rpm followed by laboratory size (325x150 mm) two roll mixing mill at a friction ratio of 1:1.25 according to ASTM D 3182 standards while carefully controlling the temperature, nip gap, time of mixing, and uniform cutting operation. The temperature range for mixing was 65–70°C. After mixing the elastomer compositions were molded in an electrically heated Moore hydraulic press at a pressure of 10MPa and at 160°C using moulding conditions determined by Monsanto Rheometer (R-100) according to ASTM D2084 and ASTM D5289 procedures.

Table-2.1 Formulation of Unfilled and Nano Graphite Reinforced Silicon Elastomer Nano-Composites

Ingredients	**SiR1**	**SiR2**	**SiR3**	**SiR4**	**SiR5**
Silicon Rubber	100	100	100	100	100
Nano Graphite (phr)	0	2	4	6	8
Each mix contains ZnO 5 phr, Stearic acid 2 phr, DCP 2 phr.					

2.3. TESTING

2.3.1. Scanning Electron Microscopy (SEM)

The morphology of the SiR-NG nanocomposites (Gold coated sample) was imaged using a scanning electron microscope.

2.3.2. MECHANICAL STUDIES

Hardness of Silicon elastomer nanocomposites was measured using a Shore-A Durometer as per ASTM D 676-59T. Mechanical properties of dumbbell specimen like modulus, tensile strength and elongation at break were measured using a Hounsfield H10KS Universal Testing Machine according to ASTM D 412 procedure. At least five specimens per sample were tested for each property and mean values are reported.

2.3.3. Dynamical Mechanical Analysis (DMA)

DMA is a technique that is employed in the structural analysis of visco-elastic materials, that is, materials displaying both elastic and dissipative components of deformation. DMA was carried out using TA instrument over a temperature range of -100 to 100 °C, at a frequency of 1 Hz and at 1% strain. Storage modulus (E'), loss modulus (E") and loss tangent (tanδ) curves were plotted versus temperature from measured data.

2.3.4. Dielectric Studies

Dielectric relaxation spectra of the nanocomposites were obtained by a Hioki 3530 *HiTester* LCR meter in the frequency range of 0.1 Hz to 10^6 Hz using aluminum foil as blocking electrodes. The dielectric characteristics have been observed as a function of frequency. Electrical conductivity (σ) has been evaluated from dielectric data in accordance with the relation:

$$\sigma_{AC} = \omega \varepsilon_0 \varepsilon' \tan \partial \qquad 2.1$$

Where ω is $2\pi f$ (f is frequency), ε_0 is permittivity of the vacuum and ε' dielectric constant or dielectric permittivity.

$$\varepsilon' = C_p / C_0 \qquad 2.2$$

Where C_p is the observed capacitance of the sample and C_0 is vacuum capacitance of the cell and is calculated using the expression $\varepsilon_0 A / d$ (where A is the area of sample and d , the thickness of the sample) and $\tan \partial$ is the dielectric loss tangent.

3. RESULTS AND DISCUSSION

3.1. Morphology

The photomicrographs of SiR/NG nanocomposites containing different wt% of nano graphite are shown in Fig. 3.1. It is evident from Fig. 1(a–d) that the nano graphites are randomly oriented throughout the silicon matrix and

interconnecting network is formed. In Fig. 1a, nano graphites are dispersed individually in the silicon matrix. The silicon/NG nanocomposites with 6wt% (Fig. 1c) and 8 wt% (Fig. 1d) nano graphite loading also display well dispersion, even though very small agglomerates are appeared due to increase in wt% of nano graphite in silicon matrix. which is clearly observed from the SEM photomicrographs of samples containing 8 phr filler loading. This observation appears to be in good agreement with the results of mechanical properties.

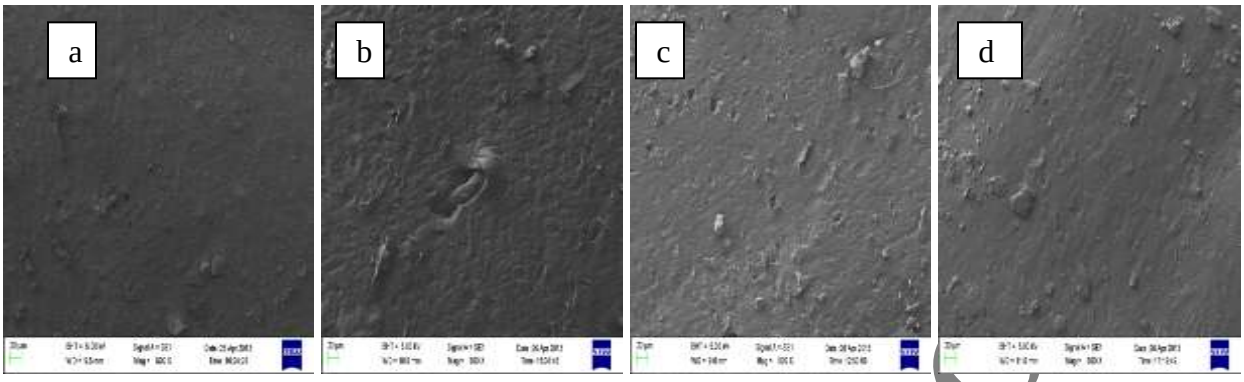

Fig. 3.1 SEM Photomicrographs of Razor Cut Surfaces of Silicon Elastomer Nano-Composites: Effect of Nano Graphite Loadings, (a) 2 PHR (b) 4PHR (c) 6 PHR (d) 8 PHR

To observe the dispersion and distribution of the nano graphite and their physical interaction with the silicon matrix, the SEM microphotographs of cryo-fractured surface morphology of the silicon/NG nanocomposites are shown in Fig. 3.2.

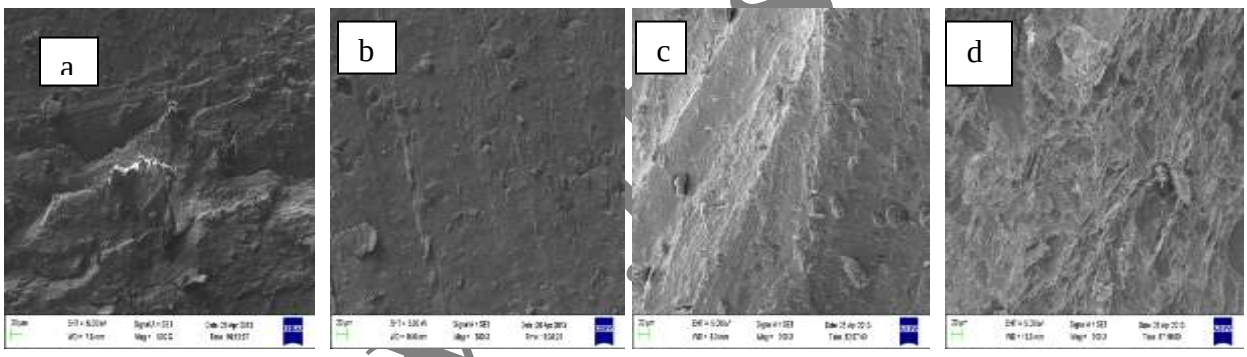

Fig. 3.2 Fractographs of Nano Graphite Loaded Filler Agglomerates Initiate Crack During Tensile Deformation. (a) 2 PHR (b) 4PHR (c) 6 PHR (d) 8 PHR

The bright dots and tread-like structures in the images are attributed to the nanographites. It is apparent from Fig. 2 that there are more bright dots and thread-like structures in the composites with higher NGs loading. The dispersion of NGs in the silicon matrix is clearly observed from SEM images with low magnification that NGs at lower loading (2 and 4 wt%) are homogeneously dispersed throughout the silicon matrix, but high agglomeration is seen at higher (8 wt%) loading. At higher nanographite loading agglomerates acted as foreign body and initiate cracks in the composites under stress and also reduce elongation at break. Although the size of agglomerates were big but sphere-like agglomerate has lessen their effect as stress concentrator in composites, so, did not show pronounced effect on tensile properties and enhance the modulus. However, high amount of nanographite nanoparticles dispersed in elastomer matrix significantly increases the hardness and decreases the impact strength of the composites. Micrographs 2(a) with 2 wt% and (b) with 4 wt% show an abundance of well-dispersed nanographite throughout the entire surface without any agglomerates. From two micrographs, it seems that induced van der Waals force between the polymer and NGs during sonication exceeds the existing forces. In micrograph 2c, high agglomeration of NGs is seen, which may be due to self-aggregation of NGs by van der Waals bonding and also due to lack of interfacial phase represents the adhesion between NGs and silicon.

3.2 Mechanical Properties

Mechanical properties like tensile strength, elongation at break, modulus and hardness of the nanocomposites are shown in Table-3.1.

Table-3.1 Mechanical Properties of Unfilled and Nano Graphite Filled Silicon Elastomer Nano-Composites

Mix No	Hardness (shore A)	Tenile Strength (Mpa)	Elongation at Break (%)	Modulus (100%) Mpa	Modulus (200%) Mpa	Modulus (300%) Mpa
SiR	41	5.5	530	1.03	1.86	2.72
SiR/NG1	42	5.647	506	1.12	1.98	2.92
SiR/NG2	44	5.864	476	1.24	2.16	3.2
SiR/NG3	45	6.15	457	1.34	2.28	3.5
SiR/NG4	47	6.24	442	1.42	2.4	3.7

It is observed that Tensile strength, modulus (100%, 200%, 300%) and hardness values increases with nano graphite loading, while elongation at break decreases which is also shown in Fig. 3.3, Fig. 3.4, Fig. 3.5 and Fig. 3.6 respectively.

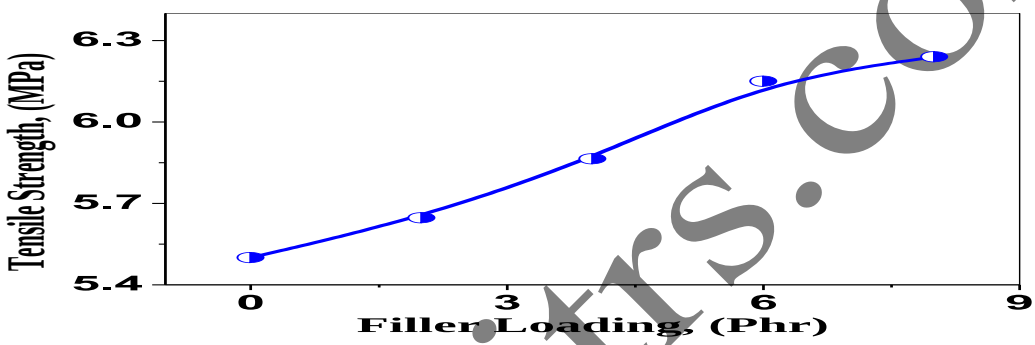

Fig. 3.3 Variation of Tensile Strength with Nano Graphite Loading in Silicon Elastomer Nano-Composites

The effect of incorporation of a nano graphite (be it a reinforcing filler or not) on the mechanical properties of elastomeric materials can be partially explained as follows: If particles of high elastic-modulus are dispersed through a low elastic-modulus matrix, it is obvious that the modulus of the mixture will be higher than that of the matrix, mainly because of the decrease in volume content of matrix substance. However, for a filler to be really reinforcing in nature, many other factors come into the picture, the most important being the force of adhesion of the matrix to the particle. If the adhesion between the filler and the polymer matrix is very low then no significant increase in modulus (except due to hydrodynamic effect) can be observed. But, if the particle-to-matrix adhesive force is large, then the modulus of the mixture is primarily determined by the magnitude of the polymer filler interactions. At 8 phr filler loading silicon elastomer gives the highest value of tensile strength, which means that at this loading polymer-filler interaction is maximum. Unlike conventional filler, nano graphite have very high surface area and this complex structure of the branched filler aggregates attributes to a strong surface polymer interaction. Incorporation of fillers is a major source of energy dissipation thereby increasing the tensile strength of nanocomposites.

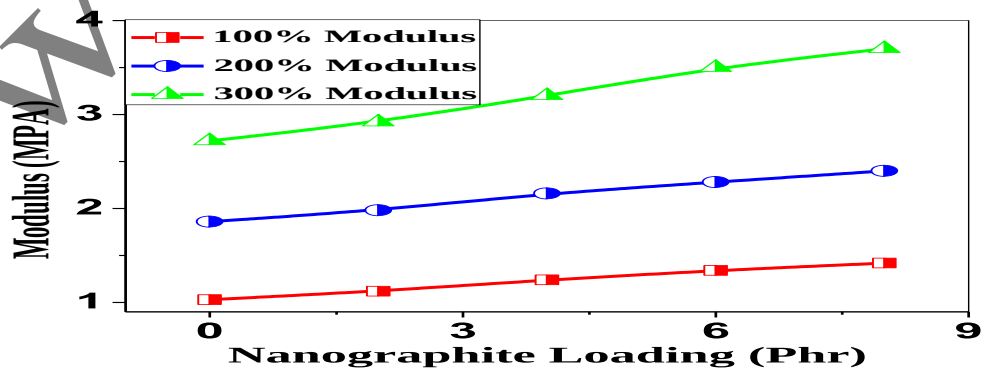

Fig. 3.4 Variation of Modulus (100%, 200%, 300%) With Nano Graphite Loading in Silicon Elastomer Nano-Composites

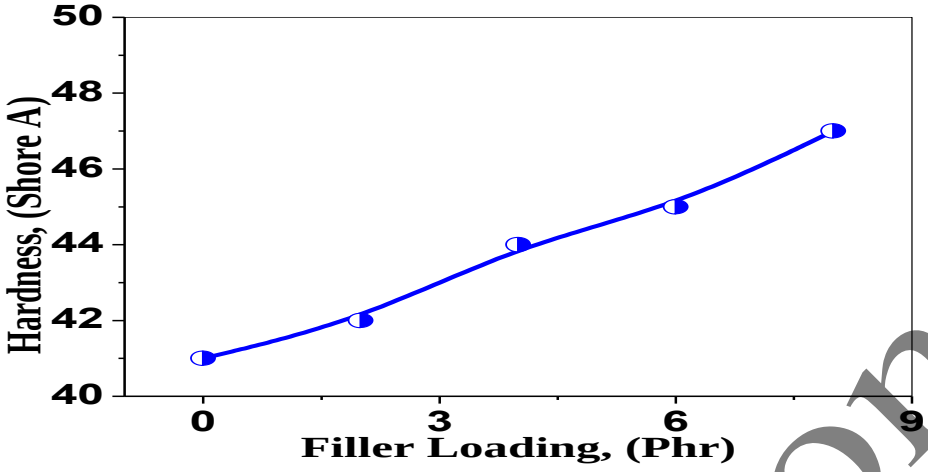

Fig. 3.5 Variation of Hardness With Nano Graphite Loading in Silicon Elastomer Nano-Composites

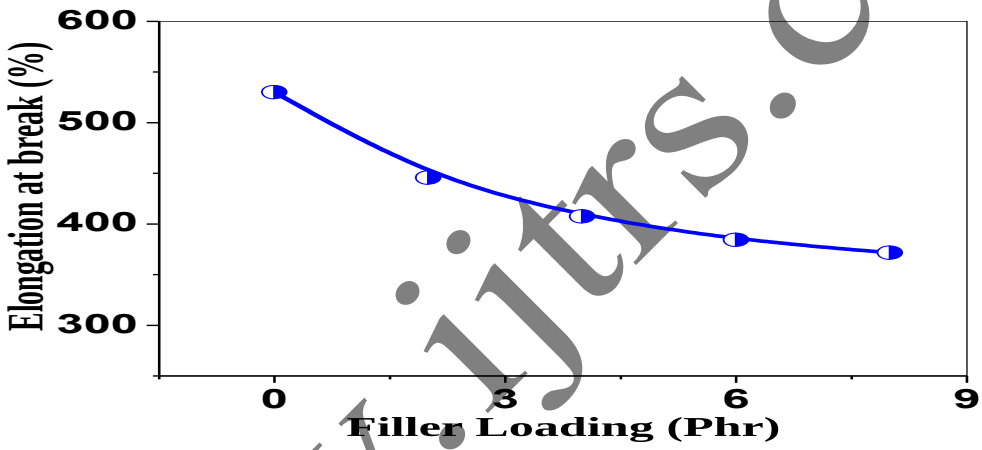

Fig. 3.6 Variation of Elongation at Break With Nano Graphite Loading in Silicon Elastomer Nano-Composites

On addition of filler like nano graphite, free space between the chains is filled up thus depriving the chains to straighten thereby reducing elongation. The higher the filler loading the more is the reinforcement and more crosslinks are formed during vulcanization, thereby trapping the free ends of polymer chains. As the degree of crosslinking increases, the hardness progressively increases. The more compact the networks, the shorter are the molecular segments between the crosslinks and hence the tighter is the network, which causes increase in hardness [6]. Very high value of hardness is due to highly reinforcing nano graphite having very high surface area.

4. DYNAMIC MECHANICAL ANALYSIS

4.1 Effect of Temperature on Loss Tangent

Dynamic mechanical analysis (DMA) measures the cyclic response of a material as a function of the temperature. Figure 7 shows the loss tangent spectra of silicon elastomer nanocomposites reinforced with increasing amounts of nano graphite as a function of temperature. From the figure it can be observed that the Increasing amount of filler in the nanocomposite has no significant effect on the temperature of location of $\tan\delta_{max}$ value of glass transition, but the magnitude of the peak decreases with increase in filler loading. All the samples show the glass transition in the narrow temperature range of -11^{0}C to -6^{0}C. This indicates that the addition of filler does not shift the glass transition temperature of the vulcanizates. This also indicates that no polymer chains are absorbed onto the filler surfaces, thereby causing no restriction to the mobility of polymer chains. But recent studies show that the dynamics of polymers near the immediate surface of the filler are very much different from that of the bulk.

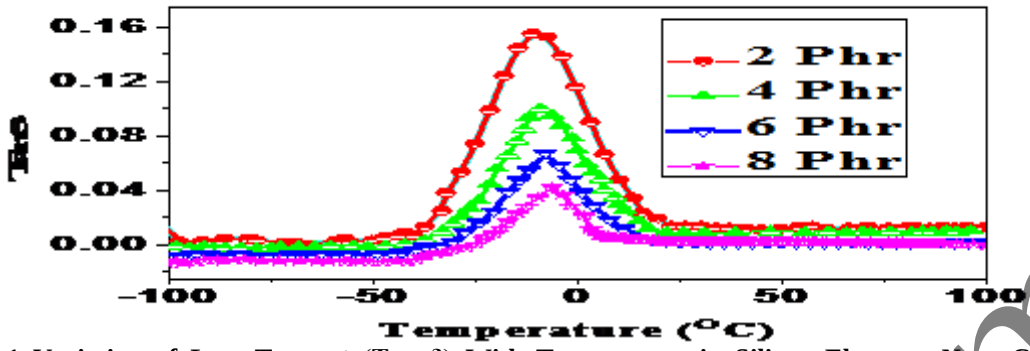

Fig. 4.1 Variation of Loss Tangent (Tan δ) With Temperature in Silicon Elstomer Nano-Composites at Increasing Nano Graphite Loadings

Increasing amount of filler in the nanocomposites has no significant effect on the temperature of location of $\tan\delta_{max}$ value of glass transition, but the magnitude of the peak decreases with increase in filler loading. Nano graphite is a new generation filler in rubber compounding, which has the advantage of providing improved dynamic mechanical properties It is observed that beyond glass-transition temperature, i.e., in the rubbery region, the curves of different nanocomposites crossover around a wider temperature interval. This may be attributed to the relaxation of the interfacial region of the polymer-filler in the composites.

4.2 Effect of Temperature on Storage and Loss Modulus

Fig. 4.2 and 4.3 shows the variation of storage modulus E' and loss modulus E'' for silicon elastomer nanocomposites with increasing filler loading over a wide range of temperature (-100 °C to 100°C). The applied dynamic strain amplitude and frequency test conditions were 1% strain and 1 Hz, respectively. Increasing filler loading results in increase of storage modulus E' at all experimental temperatures. It is theorized that at low strains the filler is the main contributor to the reinforcing of the polymeric system. The nanographite dispersed in the system do not show any form of "network" because of the absence of van der Waal forces between the polymer matrix and the filler. Thus, the reinforcing effect is purely hydrodynamic in nature. There is a continuous increase in the storage and loss modulus with increasing filler loading at all the test temperatures. This increase is only due to the hydrodynamic effect. Previous studies showed that there is not much influence of crosslinking on the dynamic mechanical properties such as E' and E''. But, the amount filler loading, the filler–filler interaction, and the filler–polymer interactions play a crucial role in the dynamic viscoelastic properties of a filled polymer system. If adhesion between filler and polymer matrix is very low then significant increase in modulus (except due to hydrodynamics effect) is observed. But, if the particle-to-matrix adhesive force is large, then the modulus of the mixture is primarily determined by the magnitude of the polymer filler interactions.

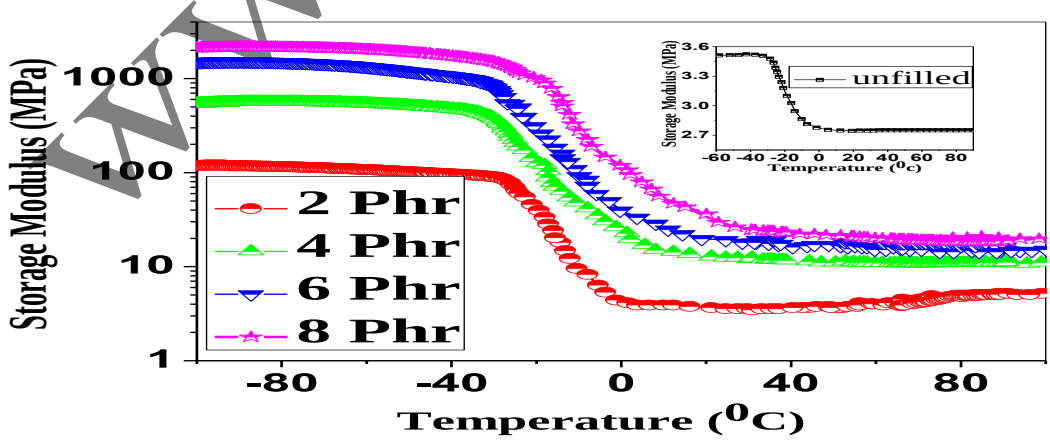

Fig. 4.2 Variation of Storage Modulus With Temperature in Silicon Elstomer Nano-Composites at Increasing Nano Graphite Loadings

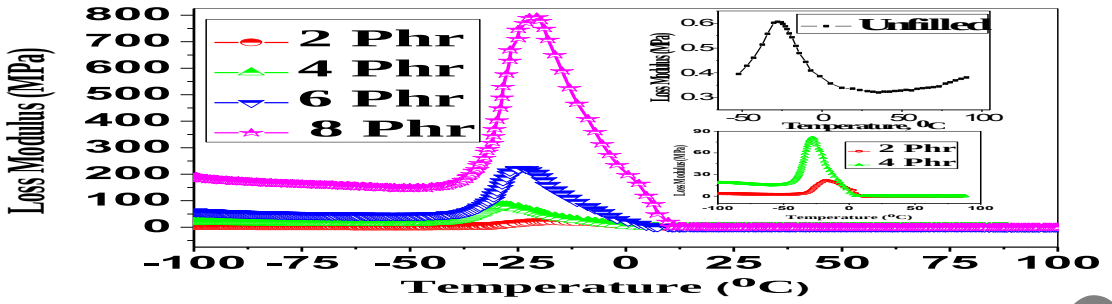

Fig. 4.3 Variation of Loss Modulus With Temperature in Silicon Elastomer Nano-Composites at Increasing Nano Graphite Loadings

Fig. 4.3 represents the temperature dependence of the loss modulus (E'') for various compositions. For all filled compositions, a distinct peak (α transition) is observed around -25^{0}C. The α peak is located in the transition region between glassy and rubbery state. The type and loading of filler do not affect the α peak temperature; however the intensity of α peak increases with increasing particle size of the filler. The cross over point has been observed in the region of -30 to -25 ^{0}C. In the rubbery region, E'' also shows the same trend as observed in storage modulus, that is E'' decreases with lower structure and lower particle size of nanographite. To explain the influence of filler on the loss modulus, Kraus [7] noted a close relationship between the decreases in storage modulus with agglomeration-deagglomeration of the primary aggregates which are formed during process of vulcanization. Whereas, Wang [8] postulated that for a filled compound, there is always a difference in the surface energy between filler and polymer, so that even for a system in which the filler is well or uniformly dispersed in the polymer matrix, the filler aggregates tend to agglomerate during vulcanization and storage of the compound forming a filler network. This effect termed as flocculation in a colloid systems like filler-elastomer was extensively reviewed by Bohm et. al. [9,10] Therefore, it can be said that particle size, filler-polymer interaction have profound influence on the loss modulus.

4.3 Effect of Temperature on Cole-Cole Plot

The relaxation dynamics can also be expressed in terms of cole-cole plots i.e. relationship between storage modulus (elastic property) and loss modulus (viscous property). Cole-cole plots for nano graphite reinforced silicon elastomer nanocomposites are shown in figure 10. Irrespective of nano graphite loading at all concentrations of the filler, the usual depressed semi-circular loop can be observed which clearly indicates the presence of reinforcing elements. The large change observed on the arc radius in the Cole–Cole plot indicates the relaxation dynamics and a significant alteration of chain conformation due to nano graphite interaction [11]. McLachlan et al. correlated the shape of Cole-Cole plots with the homogeneity of filler dispersion in the polymer matrix. Irrespective of filler concentrations, smooth arcs are observed with no humps indicating good dispersion of nano graphite in the polymer matrix [12].

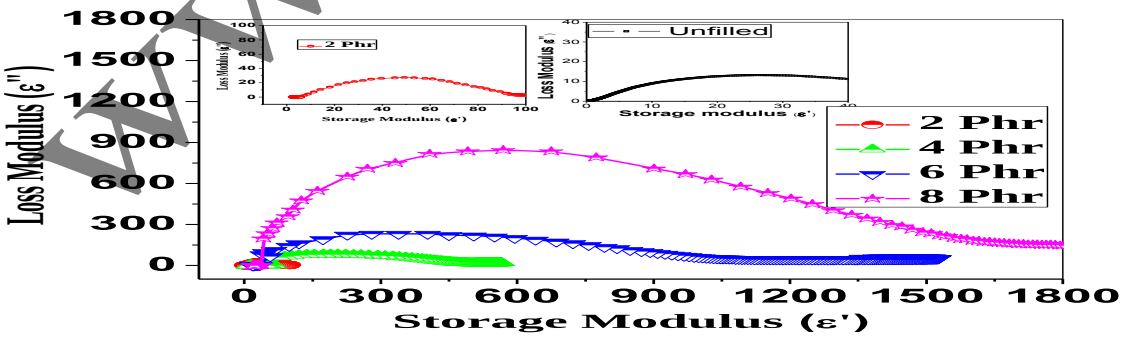

Fig. 4.4 Cole-Cole Plots of Silicon Elastomer Nano-Composites With Increasing Nano Graphite Loadings

5. ELECTRICAL PROPERTIES

5.1 Dielectric Permittivity

Fig. 5.1 represents the variation in dielectric permittivity as a function of frequency for 2, 4, 6 and 8 phr nano-graphite reinforced silicon elastomer nanocomposites. For the silicon nanocomposites, nano-graphite is conductive

and silicon is insulating, for which space charge accumulates at the interface of nanocomposites and charge is stored. From the figure it is found that the dielectric permittivity decreases with the frequency at all loadings and increases with the filler loadings. With the increase in nano-graphite loading, more electrons supplied by nano-graphite contribute the remarkable space charge polarization. Saji et al. [13] have reported a reduction in ε' with increase in frequency for MWCNT reinforced with silicon rubber nanocomposites. Renukappa et al. [14] have noticed a similar behavior in conductive carbon black filled SBR rubber.

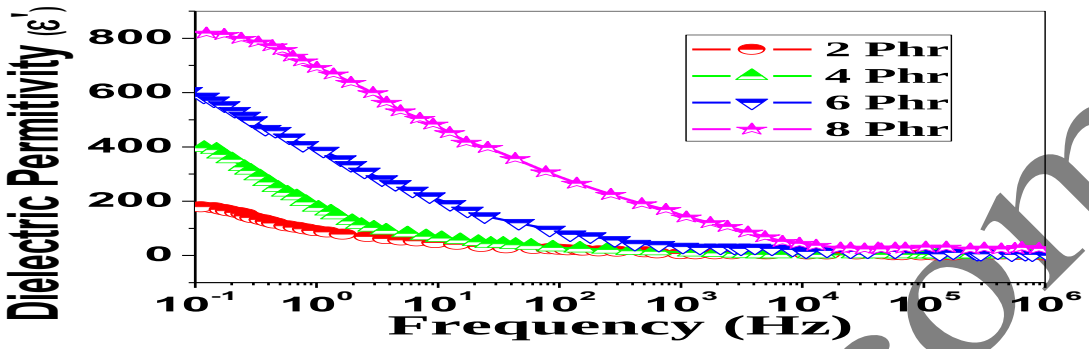

Fig. 5.1 Effect of Nano Graphite Loadings on Dielectric Permittivity (ε') of Silicon Elastomer Nano-Composites With Increasing Frequency

The decrease in dielectric constant with increase in frequency can be explained as follows. At low frequencies, the change in polarity of electric field is slow, so molecules get sufficient time to be polarized, by which polarization at low frequency is high. At high frequencies, the polarity of electric field changes much faster than orientation of dipoles, as a result the degree of polarization is less. When the polarization of a material cannot follow the change in external electric field, the relaxation happens so that the corresponding dielectric constant decreases quickly [15, 16].

5.2 Electrical Conductivity

Fig. 5.2 shows the variation of electrical conductivity with increasing nano-graphite concentrations in silicon elastomer nanocomposites at different value of frequency. At any given frequency, tremendous increase in conductivity with increasing nano-graphite concentration can be observed. Figure also shows the variation of conductivity with frequency for silicon elastomer nanocomposites at increasing nano-graphite concentrations. As observed from the figure, there are three different regions namely frequency independent region (Region 1), exponential growth with increasing frequency (Region 2) and finally a plateau region (Region 3). The shift from Region 1 to Region 2 depends on the nano-graphite concentration.

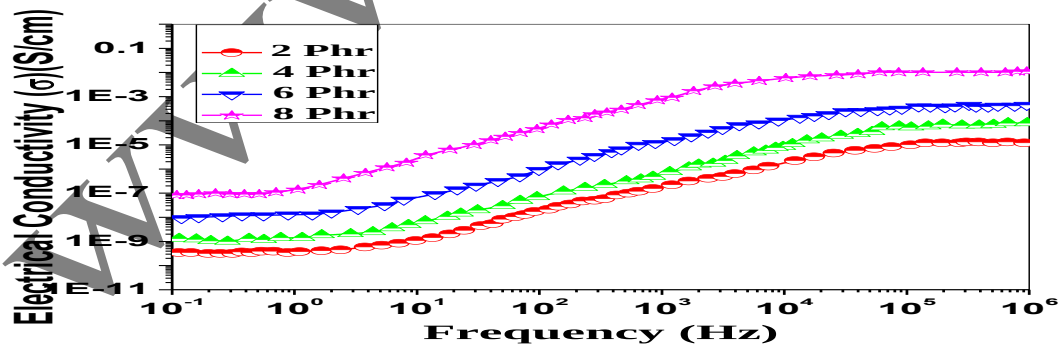

Fig. 5.2 Effect of Nano Graphite Loadings on Electrical Conductivity (σ) of Silicon Elastomer Nano Composites With Increasing Frequency

It is widely believed that electrical properties of reinforced polymers depend primarily on the way the filler particles are distributed through the polymer matrix also called as meso-structure. At low levels of filler loading, the conductivity of the composite is slightly higher than that of the base polymer, since the filler particles are isolated from each other by the insulating polymer matrix. When concentration of filler is low, the conductivity between the grains is expected to be primarily via hopping and tunneling mechanisms. In this mode of conduction, the electron

transport may be coupled strongly with the molecular and ionic processes in the polymer matrix. Usually, hopping transport between localized sites is the main reason for frequency dependence of conductivity in polymer composites. The dispersion of filler is heterogeneous, localized and disordered. This disorder results in a wide distribution of hopping rates, giving a strong dispersion of the ac conductivity [17].

5.3 Percolation

The variation of electrical conductivity and dielectric permitivity with nano-graphite loadings in silicon elastomer nanocomposites is shown in fig. 5.3 and 5.4 respectively.

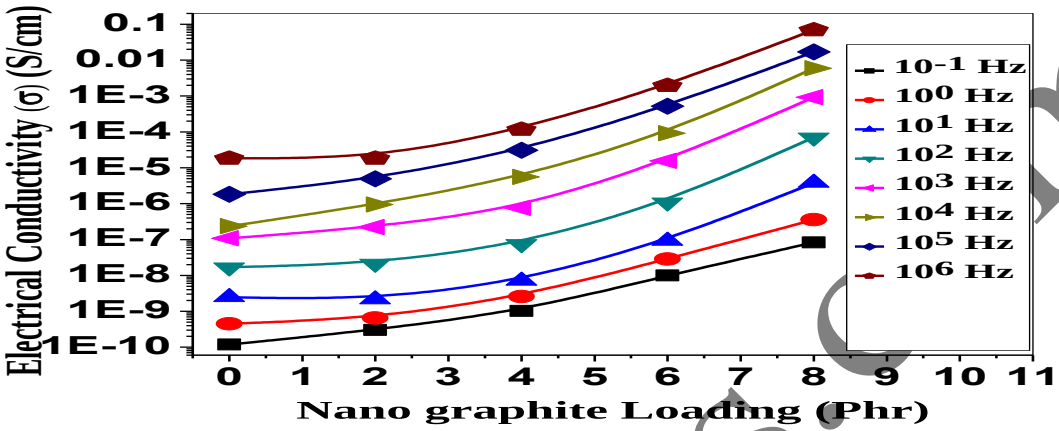

Fig. 5.3 Effect of Nano Graphite Loading on Electrical Conductivity of Silicon Elastomer Nano-Composites at Different Frequencies

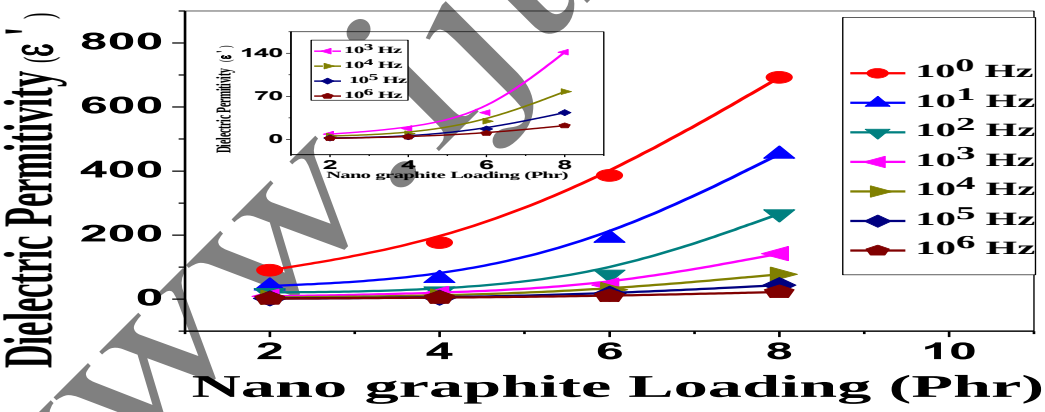

Fig. 5.4 Effect of Nano Graphite Loading on Dielectric Permittivity of Silicon Elastomer Nano-Composites at Different Frequencies

The electrical conductivity and dielectric permittivity of a composite is generally characterized by its dependence on volume fraction of filler. It is observed at all frequencies (10^{-1}, 10^{0}, 10^{1}, 10^{2}, 10^{3}, 10^{4}, 10^{5} and 10^{6} Hz) above 6 phr nano-graphite loading, there is an abrupt increase in the conductivity, implying the occurrence of a percolation limit. As the nano-graphite amount in the composites is increased, the filler particles begin to touch each other, and a continuous path is formed through the volume of the sample for the electrons to travel. The formation of this conductive network is based on the principle of percolation theory. Experimental results for composites consisting of an insulating matrix and conductive filler particles are often analyzed with the help of statistical percolation theory. Beyond a critical concentration of the filler, known as the percolation threshold, an increase in the composite conductivity of several orders of magnitude is witnessed.

CONCLUSIONS

We have demonstrated the preparation of silicone elastomer nanocomposites consisting of 2-8 wt% of nanographite. The nanocomposites were prepared through a mechanical mixing technique. SEM micrographs show excellent distribution of filler in the polymer matrix. Tensile strength and modulus increase with filler loading, whereas elongation at break gradually decreases. With filler loading hardness also gradually increase, due to increase in crosslink density. The dynamic mechanical analysis of the nanocomposites shows that the addition of the filler has no significant effect on the glass transition temperature, T_g. However on increasing filer loading the intensity of $\tan\delta$ curve decreases. This has been explained in terms of filler polymer interaction and interaggregate interaction. Broadening of the relaxation region was observed in the plots of storage and loss modulus with temperature. Electrical conductivity increases exponentially with frequency and filler loading. The percolation limit of the nano graphite in the composite has been studied by electrical conductivity and dielectric measurements. It has been observed that the percolation limit was occurring in the range of 6 phr nano graphite loading.

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