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Review of Silicone Rubber Nanocomposites for Outdoor Insulation

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Polymer nanocomposites have attracted wide interests in high voltage insulation due to their excellent electrical, thermal and mechanical properties. Among the new insulating materials extensively used as high-voltage outdoor insulation, silicone rubber (SIR) has received the most attention. Indeed, SIRs are gaining popularity as an effective counter-measure to handle the high voltage insulation problems. In recent times nano sized fillers (Nano fillers) have sought great deal of research scholars' attention and has brought revolution in the polymer industry due to its better interaction with polymer. A relatively less quantity of nanofillers in comparison to micro filler is reported to achieve the required properties in SIR. In this review, the effectiveness of the most common nanofillers of SIR for dielectric applications along with various dispersion methods is presented. But numerous experimental investigations on nanocomposites have indicated a significant decrease in electrical, thermal, mechanical and surface properties, due to the agglomeration of nanofillers. Agglomeration can be reduced by using modified nanofillers & optimum quantity of nanofillers, high shear mixing, or by efficient filler dispersion methods but it is almost impossible to entirely eliminate this effect with the presently available technologies.

Keywords: *Silicone rubber, Nanocomposites, Outdoor insulation, Filler dispersion methods*

1.0 INTRODUCTION

In the recent past polymer nanocomposites (PNC) have gained great deal of importance and attention. Nano-composites are composed of nanometer sized fillers (popularly referred as nanofillers) which are homogeneously distributed within the polymer matrix. It is well known that pure silicone rubber does not exhibit effective resistance to tracking and erosion. So, some properties of silicone rubber need to be improved to extend its service life. The fillers & additives are added to the polymer to enhance specific properties.

There is still scope of improvement in properties of commercially available silicone rubber [1,2]. The property modification of silicone rubber brought about by addition of micro and nanofillers has found to be effective [3] for outdoor insulation applications. This article presents the most common nanofillers for dielectric applications. A need for comprehensive review of nanofiller used in the area of outdoor insulator is felt necessary as frequent failures are being reported. Literature survey indicates some filler as potential candidate for outdoor insulator application. However, there is scope to fully characterize and to optimize their property to establish for commercial application especially in the absence of standard and standardization of materials.

All over the world, composite insulators have been accepted by utilities and equipment manufacturers along with its challenges in the field, ever since they got introduced in the 1960s. But for this approach, there would not have been any scope for alternate material to come into existence which is proven in field. Among their advantages, they offer lighter weight resulting in economic design of towers

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or alternatively enabling voltage up gradation of existing systems without changing the tower design dimensions. The other attractive features that SIR exhibits are less breakage, improved seismic performance and effective flexibility in design than ceramic insulators. The light weight of the composite insulators also obviates the need to employ heavy cranes for their handling and installation, rendering them cost effective in terms of labour and maintenance.

Composite insulator design generally comprises of: i) a fibre glass rod or hollow core offering mechanical strength, ii) external weather-sheds made from either Silicone Rubber (SIR), Ethylene Propylene Diene Monomer (EPDM), Ethylene Propylene Rubber (EPR) or any composite and iii) metal fittings for connection. Silicone rubber showed better performance than most organic polymers such as EPDM for outdoor insulator application [4]. Therefore, the present work focuses only on silicone rubber materials for insulator applications. The hydrocarbon methyl groups of SIR are hydrophobic. Additionally, they exhibit the ability to restore its hydrophobicity even after formation of pollution layer on its surface, which can suppress the development of leakage currents, dry-band arcing and flashover [5,6,7]. Literature survey indicates that, comprehensive studies have been carried out to apprehend the dynamic behavior of recovery and transfer of hydrophobicity [8,9] however efforts are ongoing in addressing the problems of agglomeration of nanoparticles because of their high surface energy, non-homogeneous dispersion of nanofillers and problems associated with interface [10].

2.0 POLYMER NANOCOMPOSITES

Polymer Nanocomposites (PNC) are the special class of composite material, having nano-scale dispersion, typically 1-100 nanometers of the filler phase in a polymer matrix. PNC was discovered at Toyota Central Research and Development Laboratory, Inc. in 1985 [11]. As in the case of conventional composites, the properties of nanocomposites are determined not only by the bulk properties of each of the components but also by complex interactions between different phases and interphases between them. In polymer nanocomposites, the interaction level between the polymer and filler is much higher when compared with conventional fillers at the same Volume fraction due to very high specific areas and aspect ratios of fillers involved. The interfacial region is responsible for 'interaction' between the matrix and filler and is conventionally ascribed property different from the bulk matrix because of its proximity to the surface of the filler [12]. A simplified diagram illustrating the constituents of Polymer nanocomposites is as shown in Figure 1. Nano technology is now recognized as one of the most promising fields of research in 21st century. The concept of creating both structural and functional multiphase nanocomposites with improved performance is currently under development in a wide variety of metallic, ceramic, and polymeric matrices, although the emphasis to date has been on polymeric systems. Similarly, the filler particles can be organic or inorganic with a wide range of material compositions and structure.

In recent years organic-inorganic hybrid nanocomposites with inorganic nanoscale building blocks have attracted great interest of academic and industrial scientists, since they combine the advantage of inorganic materials (rigidity, hardness, thermal stability, abrasion resistance, gas-barrier property, electrical conductivity, hydrophobicity, flame retardancy etc.) with the organic polymers (flexibility, ductility, process ability and low density). Polymer nanocomposites represent a radical alternative to conventional filled polymers or polymer blends-a staple of the modern plastic industry.

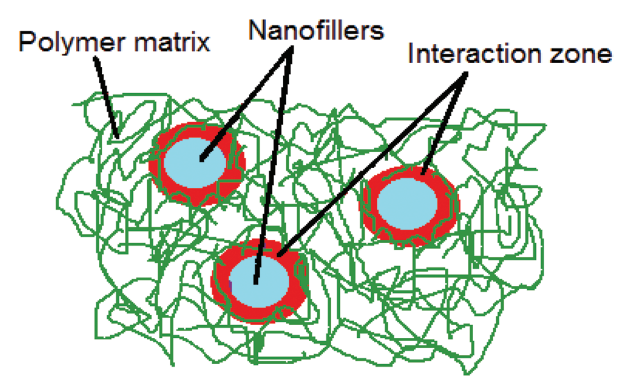

FIG. 1 MECHANISM OF POLYMER/NANO-FILLER INTERACTION

The value of PNC technology is neither solely based on the enhancement of mechanical properties of the virgin polymer nor the direct replacement of current filler or blend technology. Rather its importance stems from providing value-added properties not present in the neat polymer, without sacrificing the polymer inherent processability, low density and mechanical properties.

3.0 SILICONE RUBBER

The chemical building block of silicone rubber is shown in the Figure 2. It consists of inorganic silicon-oxygen (SiO) back bone and two organic side chains attached to the silicone atom. The side chain in most silicone rubbers used for HV insulators is methyl (CH_3) group, hence a polymer with this type is known as poly-dimethyl-siloxane (PDMS). The side chains can be of other organic groups, such as phenyl or vinyl.

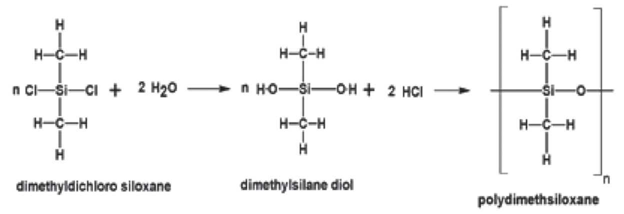

FIG. 2 STRUCTURE OF SILICONE RUBBER

Silicones are synthesized material from silicon dioxide (sand) and coal. The coal is used to reduce the silicon dioxide to form silicon. In a next step the silicon is reacted with methyl chloride to form a mix of different chlorosilanes. After purification and separation of the different chlorosilanes by a careful distillation, cyclic and/ or linear organosiloxanes are produced by the reaction of the chlorosilanes with water (hydrolysis). A further reaction step leads to the polymeric gums which are basic products for the formulation of silicone rubber compounds. Polymer (gum) is available in a wide range of molecular weight; from very low (water like) to very high viscosity. A typical silicone rubber contains polymer (gum), fillers to reinforce the silicone polymer material, processing additives and crosslinking agent to enable a proper curing to the elastomer. With these, a large variety of different products result varying in viscosity, hardness, mechanical and electrical properties. All these product variations inside the silicone rubber family require tailor-made processing methods.

Pure silicone rubber gives poor tracking and erosion resistance. In order to extend service life and improve service effect, some properties of silicone rubber need to be improved. The fillers are added to the polymer to improve specific properties and also to reduce costs. With increasing filler content tracking performance will be higher but this could hinder its hydrophobic property [13]. The properties of SIR nano-composite depend on filler concentration, filler morphology, such as particle size and structure, the degree of dispersion and orientation in the matrix, and also the degree of adhesion with the polymer chains. Both inorganic and organic fillers have been employed in improving certain electrical, thermal and mechanical properties of basic material-PDMS to achieve enhanced service life. The fillers are used in their oxide forms and the most common inorganic oxide materials found in the literature are Alumina trihydrate (ATH- (Al_2O_3 $3(H_2O)$)), Silica (SiO_2), Titanium oxide (TiO_2), Zinc oxide (ZnO), Magnesium oxide (MgO), Calcium carbonate ($CaCO_3$) and Barium titanate ($BaTiO_3$).

4.0 NANO-FILLERS

Filler (in a plastic) is a relatively inert solid material added to a plastic to modify its strength, performance, working properties, or other quantities or to lower cost as per definition in IS 1885 standard. The nanofillers used in entire polymer nanotechnology can be classified into three main categories as per their shape:

- Spherical or 3-dimensional e.g., silica nanoparticles
- Platelet type or 2-dimensional sheet e.g., organo clays, synthetic mica, graphite etc.
- Rod or fiber type or 1-dimensional e.g., synthetic whiskers, carbon nanotubes, sepiolite etc.

4.1 Inorganic Oxide Materials

The use of silicone rubber compounds with inorganic fillers is widely studied. The most common inorganic nanofillers found in the literature are described below.

4.1.1 Alumina Trihydrate

Numerous studies have reported on the benefits of using alumina trihydrate (ATH) as fillers mainly on tracking and erosion resistance in silicone rubber. Typically, ATH is added to the silicone rubber as an anti-tracking and flame retardantagent. Venkatesulu et al. reported that less amount of nano ATH filler can minimize material erosion of SIR nanocomposites at dry band arcing sites by improving thermal property[14]. Vas et al. evaluated the performance of the insulators under positive and negative dc stresses. Nano sized Alumina fillers in SIR matrix improves the resistance to tracking and erosion. Results suggest that SIR composites perform better under negative dc than under positive dc voltages [15]. DC insulation characteristics of liquid silicone rubber (LSR) with nano ATH were studied by Hwang et al. Their study revealed that nano ATH/ LSR nanocomposites have various advantages such as short curing time and the ease of maintenance and better performance as a HVDC insulation material for cable joints. Figure 3 shows nano-filler uniformly dispersed in LSR which provides better electrical performance [16]

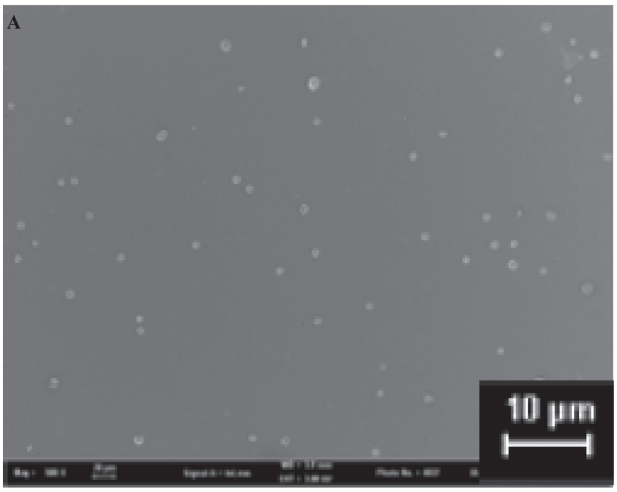

FIG. 3A 1 PHR

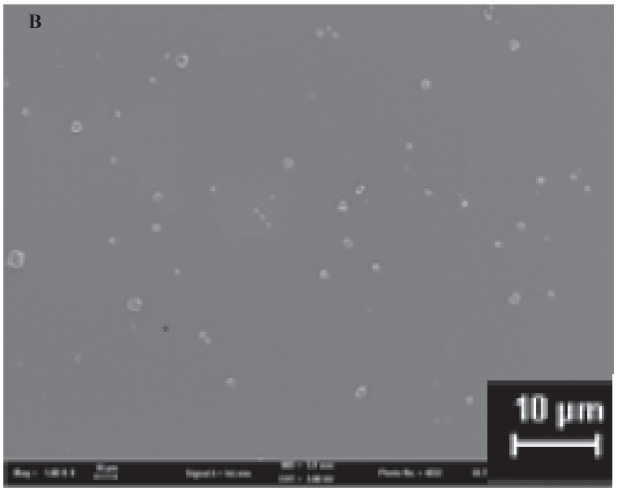

FIG. 3B 3 PHR NANO - ATH DISPERSED IN SIR [16]

4.1.2 Silica

The nanosilica filled SIR composite is a promising material which offers resistance to high voltage arcs. El-Hag et al. shown that only 10 wt% of nano silica provides remarkably better erosion resistance, using both ASTM D 2303 inclined-plane test and infrared laser technique [17].

The effect of nano-scale SiO_2 on the dc flashover voltage of silicone rubber nanocomposite in the presence of charged water droplet was studied by B.X.Du and Jie Li [18]. It is noticed that the surface charge density of SIR nanocomposite increased with very little concentration (1%) of nano-SiO_2. This increase of surface charge density adversely affects the electric filed resulting in lower DC flashover voltage. Venkatesulu and Thomas studied corona aging on SIR/SiO_2 nanocomposite and attributed that the nanocomposite performs much better than the unfilled SIR [19].

4.1.3 Titaniumoxide (TiO_2)

Silicone rubber is a polymer with low dielectric permittivity which needs to be improved for outdoor insulation applications. The addition of nano TiO_2 to SIR improves its dielectric properties viz. thermal stability, and photocatalytic effect [20]. Dang et al. confirmed that modified TiO_2 gives even better electrical insulation and mechanical properties than its unmodified counterpart [21,22]. The properties are also depended on particle size and filler concentration.

4.1.4 Calcium carbonate

Great improvement of the mechanical, rheological and flame retarding properties of PDMS reinforced by nano calcium carbonate($CaCO_3$) has been reported [23]. A study conducted by Mishra et al. reports that PDMS filled with nano-$CaCO_3$ when modified with the silicone di-cumyl peroxide system showed enhanced mechanical properties [24]. The tensile strength (8.5 kg/cm^2) and elongation at break (780%) showed greatest improvements at 10 wt% loading with treated nano-$CaCO_3$ in comparison to the untreated nano and commercial ($CaCO_3$ + silicone rubber) composites. The improvement in the properties is attributed to their good compatibility with the rubber chains and uniform dispersion.

4.1.5 Barium Titanate

Barium titanate ($BaTiO_3$) has been used as a filler for SIR to improve dielectric constant and high electrical breakdown strength. Dang et al. modified SIR with nano structured $BaTiO_3$ and carbon black and studied the dielectric property and elastic modulus of the nanocomposites. The paper accounts to how nano carbon black and $BaTiO_3$ gives improved dielectric property and elastic modulus of the nano carbon black/ $BaTiO_3$/ SIR nanocomposites by lowering the percolation threshold, dielectric permittivity and with a low dielectric loss [25]. Gurukarthik Babu etc. confirmed that $BaTiO_3$/ TiO_2/SIR nanocomposites gives better dielectric permittivity and dielectric loss than pure rubber over a frequency range 100Hz-10MHz. [26]

4.1.6 Organo-clay

The effect of organically modified montmorillonite (OMMT) nano-clay on improvement of dielectric properties, actuation stress and its relaxation response was studied by Garavi et al. OMMT modified room temperature vulcanized (RTV) nanocomposite showed that the actuation stress for a given electric field intensity is higher for composites than that for pristine SIR and the dielectric relaxation time of the actuation stress for nanocompositesis also is less than for the pristine rubber [27]. Kong et al. studied the influence of synthetic Fe-montmorillonite (Fe-MMT) and Natural montmorillonite (Na-MMT) clay filler on the thermal degradation in silicone rubber/clay nanocomposites. The thermal stability, gel fraction and mechanical property of the nanocomposites were improved by the incorporation of modified nano clay [28]. ZnO and MgO are also used as fillers in some cases.But the use of nano ZnO, nano MgO fillers for SIR is seldom explored.

5.0 PROBLEMS OF USING NANOFILLERS

Several investigations on silicone rubber composites indicated that the Mechanical, thermal and electrical properties of nanocomposites is much better than the composites filled with microfillers [29-32]. The prime impediment to achieve uniformity in nanocomposites is filler agglomeration. Small size nanocomposites and the strong forces between the filler particles result in agglomeration.

Lan et al. [33] studied the properties of nano-silica-x and nano-layered silicate added to room temperature vulcanizing (RTV) silicone rubber. It is reported that both nanocomposites exhibited better corona resistance than the virgin RTV material. In other work it is concluded that thermal conductivity has little impact on the improvement of the nanocomposite erosion resistance[34]. On the contrary, it is also reported that the thermal conductivity of composites increases with the increasing filler concentration [35-36].

Thus inconsistent observations are hindering development of the physical properties of nano filled silicone rubber. It is necessary to obtain clarity on properties of different nanofillers.

5.1 Filler Dispersion

Although many other parameters are affecting the properties of nano filled composites, the improper dispersion of nanofillers appeared to be the key factor in the improvement of composite insulators properties [37-42].

There are several mixing techniques to improve filler dispersion. Mainly they can be classified as physical and chemical methods.

5.1.1 Physical Dispersion Methods

In this method for dispersing fillers it involves conventional mechanical mixing and ultrasonic agitation. Shear force is used to activate the filler surface in mechanical mixing methods by changing the external crystal structure and physicochemical properties. Vibrations release trapped gases in the ultrasonic agitation preventing the void formation in the composites. The advantage of physical methods is that no extra additives are required.

Physical mixing cannot break when the filler size is in the nano-scale because of the interfiller forces dramatically increases. It is reported that conventional mechanical facilities often fail to break apart the nano-aggregations in polymer because of the intense interfiller interaction [43-44]. Space complexity is also reported for Single or twin screw extruders in physical dispersion methods [45].

5.1.2 Chemical Dispersion Methods

This method is basically dependent on extra additives for modification of filler surface. Electrostatic repulsive forces in between the fillers become stronger because of the electrified dispersants change the surface properties of the fillers effectively [46]. Only when the repulsive forces are higher than the attractive forces between the fillers, the filler agglomeration can be broken.

Surfactant lowers the surface energy of the nanofillers and the interfacial tension between the fillers and the matrix, facilitating the separation of the fillers and enhancing their dispersion during mixing. Triton™ X-100 (Triton) is used by Ramirez et al. for the physical surface modification of nanosilica and to improve the filler dispersion in the RTV silicone rubber matrix. The improvements in filler dispersion are confirmed by the SEM images however it is emphasized that there is an adverse effect to the chemical bonding of composites. So, the choice of effective surfactant is application dependent and optimized quantity of surfactant needs to be decided to reduce the side effects [47].

5.1.3 Electro-spinning Method

Due to the limitations of existing mixing methods, an alternative simple mixing method is adopted and is depicted in Figure 4.

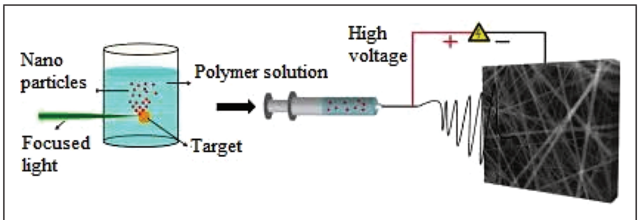

FIG. 4 ELECTROSPINNING PROCESS

In this process, the polymer solution is pushed through a needle at a constant speed by controlling a syringe pump and a high voltage DC source is used to electrify the polymer solution.

Shanshan Bian prepared SIR samples filled with nano fumed silica by conventional mixing and by electro-spinning method. Electro-spinning has significantly improved the thermal properties compared to those of conventional SIR composites because of improved filler dispersion [48].

6.0 CONCLUSION

In this review, nanofiller reinforced silicone rubber for outdoor application is presented. Various filler dispersion methods are also reviewed which have accounted for reduction in agglomeration, improved erosion resistance and reduced thermal degradation of SIR nanocomposites. The study reveals that there is scope for further improvement in SIR nanocomposites' processing method.

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