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Silicone Rubber Dielectrics Modified by Inorganic Fillers for Outdoor High Voltage Insulation Applications

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ABSTRACT

The paper discusses the mechanisms by which inorganic fillers in silicone rubber dielectrics enhance the properties of thermal conductivity, relative permittivity, and electrical conductivity making them useful in outdoor high voltage insulation applications. The addition of alumina trihydrate or silica fillers to silicone elastomers, forming binary composites with enhanced thermal conductivity, is discussed in relation to filler type, particle size, shape, and concentration, and its use as a housing material for non-ceramic insulators to minimize material erosion at dry band arcing sites by lowering hot spot temperature. Also discussed is the enhanced relative permittivity of silicone dielectrics that is obtained through the addition of barium titanate powder which can be further increased with the addition of aluminium powder forming a tertiary composite, resulting in a significant grading of the surface electric field when applied as a housing material to high voltage bushings. Controlled electrical conductivity of silicone dielectrics is discussed through the use of antimony-doped tin oxide filler binary composites and when applied as a housing material to outdoor bushings, the pollution performance is greatly enhanced.

Index Terms - **Silicone composites, thermal conductivity, relative permittivity, electrical conductivity, outdoor insulation, inorganic fillers.**

1 INTRODUCTION

SILICONE elastomers of all types are known to be excellent dielectrics. They possess many desirable properties that make their application to electrical insulation successful. In outdoor high voltage applications, electrical insulation often experience stresses that can be withstood by only a few materials. These stresses are listed in Table 1. Of the stresses listed, silicone elastomers, when used as a housing material in outdoor high voltage insulation applications, are normally capable of withstanding all but the electrical stresses that give rise to partial discharge, dry band arcing, and corona discharge on the surface of the insulation, as all lead to the physical erosion of the material, and insulation failure. Of the discharges, dry band arcing is considered the most damaging.

The engineering problem of using silicone elastomers in outdoor high voltage applications is to reduce the surface electrical stress to a level whereby partial discharge, corona discharge or dry band arcing does not occur, and/or to modify the material to impart resistance to the effects of discharges which can be achieved through the addition of suitable inorganic fillers. Table 2 lists commonly employed inorganic fillers that are incorporated into dielectrics to achieve engineering materials for electrical insulation applications.

Fillers such as alumina trihydrate (properly aluminum hydroxide) and silica are commonly used to impart tracking and erosion resistance to silicone dielectrics and numerous studies have examined the effectiveness of filler type, particle size, and concentration, on material erosion in artificial tests such as the inclined plane test [1-10]. These fillers enhance the thermal conductivity of materials thereby removing heat from the area of dry band arcing. This is a means of producing a material that is resistant to the effects of arcing on its surface rather than to reduce the surface electrical stress.

Table 1. Stresses on outdoor insulation.

Classification	Stress
Environmental	UV, Moisture, Temperature, Chemical
Electrical	Dry band arcing, Partial discharge, Corona
Mechanical	Cyclic loads

Other fillers such as $BaTiO_3$ are incorporated into dielectric materials to increase the relative permittivity for applications involving stress relief in insulation systems and higher relative permittivity values are easily obtained through binary mixtures of ceramic and metal fillers. This is a method of reducing the electrical stress on the surface of insulation thereby minimizing the effects of arcing on the surface. Again, many studies have been reported on the increased relative permittivity that is obtained as a

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function of concentration and type of filler [11-17]; however, at this time there are no known commercial applications of these materials to the housings of bushings where a significant stress relief on the surface of the insulation can be obtained.

Table 2. Common fillers incorporated into dielectrics to achieve engineering materials for electrical insulation.

Filler	Property Modification	Engineering Insulation Improvement
$Al_2O_3.3H_2O$ SiO_2	Thermal Conductivity	Resistance to dry band arcing, partial discharge & Corona
$BaTiO_3$	Relative Permittivity	Electric field grading
$BaTiO_3$ + Al		
SiC		Contamination performance
Varistor (ZnO)	Electrical Conductivity	
Sb_2O_3 + SnO		Electric field grading

Conducting fillers are incorporated within dielectric materials yielding composite materials exhibiting electrical conductivity. As above, the electrical stress on the surface of insulation can be reduced to minimize the effects of arcing. In addition, heating of the surface provides some improvement to the outdoor contamination problem. Again, the literature contains many studies on the properties of such systems [18-21]. In these systems, the conducting particles are nearly in contact with each other and the conduction is limited to few conducting paths up to the percolation limit. However, above the percolation limit, the characteristic switches to a very conductive state and in most systems, the conductivity is much too great for insulation applications. However, the conductivity can be controlled by incorporating semiconductive fillers, whose conductivity can be controlled by doping. As for high permittivity materials, no commercial applications of this concept have been applied as yet.

The paper discusses the mechanisms in each of the three systems, provides some limits to designing materials, changes in some of the material properties, identifies some of the shortfalls in the related theories, and provides some interesting aspects in the practical application of each type of composite material.

2 THERMAL CONDUCTIVITY AND RESISTANCE TO DRY BAND ARCING

The performance and life of silicone rubber housings on outdoor insulators, surge arresters and bushings depends on a number of factors; namely, the design of the housing with respect to sheds, bonding of the housing, creepage distance, protected creepage distance, the surface electrical stress, the operating environment and the ability of the material to withstand surface discharge activity. While service experience provides the best guide to long term performance and life, this is obviously not practical and various accelerated tests are used. Most often two stages leading up to failure are considered; the first is the time period in which the surface chemistry changes and the material gradually loses hydrophobicity so as to permit wetting and the development of leakage current over the surface of the housing, and the second stage is the time period for which the material is able to withstand the discharge activity that follows. The ASTM D2303 inclined plane tracking and erosion test is one of many tests employed to evaluate the relative ability of materials to withstand surface discharges that occur during the second stage of aging.

Direct erosion of silicone compositions using the ASTM D2303 inclined plane test apparatus, but the method modified for a better measure of the eroded mass, has shown correlations to filler type, particle size and concentration [9]. One such result on RTV samples is shown in Figure 1 in which the effects of filler type, particle size and concentration on the eroded mass are evident. The corresponding correlation of the eroded mass to the thermal conductivity of the composites is shown in Figure 2 [9]. The eroded mass is clearly seen to be lower with increasing thermal conductivity and correlation between the eroded mass and thermal conductivity is clearly evident.

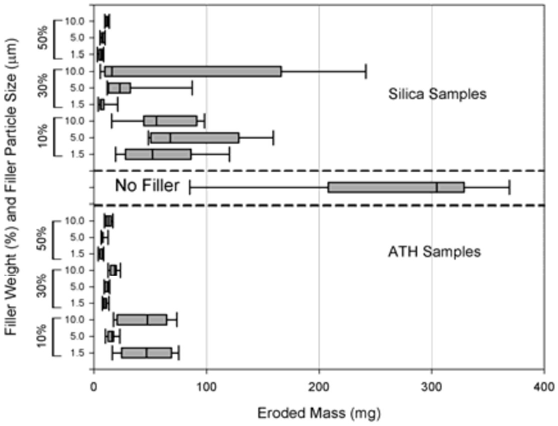

Figure 1. Mass eroded of RTV composite samples in the inclined plane test. Bars mark the 10 and 90 percentiles; extremities of the hatched bars are at the 25 and 75 percentiles, and the center line represents the statistical mean of the data [9]

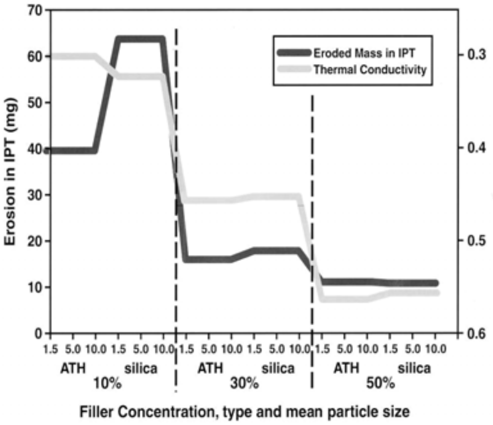

Figure 2. Correlation between mass eroded (statistical mean in Figure 1) of silicone composites in the incline plane test and thermal conductivity [9].

Table 3 lists the measured and calculated thermal conductivities for the RTV silicone samples in Figure 1 and for the same composites made using a hot pressed vulcanization (HPV) technique [9]. The effective thermal conductivities of the composites were obtained using a

Table 3. Thermal conductivity, measured and calculated, for RTV silicone composites as a function of filler type, particle size and concentration.

Filler	Wt %	Vol %	Particle Size µm	Thermal Conductivity W/m.K			
				Rule of Mixtures	Measured		Nielsen Equation
					RTV	HPV	
ATH	10	4.5	1.6	1.06	0.30	-	0.27
			5.0				
			12.0				
Silica		4.1	1.6	0.24	0.32	-	0.21
			4.1				
			10.5				
ATH	30	15.3	1.6	3.20	0.46	0.38	0.49
			5.0				
			12.0				
Silica		14.2	1.6	0.38	0.45	0.45	0.28
			4.1				
			10.5				
ATH	50	29.7	1.6	6.48	0.56	0.65	0.99
			5.0				
			12.0				
Silica		27.8	1.6	0.58	0.56	0.84	0.41
			4.1				
			10.5				

transient technique that has been previously described [7].

There are numerous models in the literature for estimating the thermal conductivity of composites which are of theoretical, semi-theoretical and empirical forms [23, 24]. Most models are applicable over a short range of filler volume fraction but Nielsen's formula has been shown to provide the best estimates over the widest volume fraction range [24].

The calculated thermal conductivities in Table 3 are based on two models, the first a very simple model corresponding to a weighted geometric mean of the thermal conductivities and volume fractions of the filler and the silicone matrix (1), and commonly referred to as the rule of mixtures. This model does not consider particle size and gives thermal conductivities that are much too high. The second model is based on Nielsen's equation that attempts to account for the shape and packing of the particles (equation (2)). For this model, the packing factor, Einstein coefficient, densities and thermal conductivity of the fillers are listed in Table 4.

$$k = k_m V_m + k_f V_f \text{...........}(1)$$

where k_m is the thermal conductivity of the silicone matrix, k_f is the thermal conductivity of the filler, V_m is the volume fraction of the silicone matrix, and V_f is the volume fraction of the filler. The thermal conductivity and density of the silicone polymer is 0.19 W/m.K and 0.93 g/cm^3, respectively.

$$k = k_m \left[\frac{1 + ABV_f}{1 - B\psi V_f} \right] \text{..............}(2)$$

$$A = k_E - 1$$

$$B = \frac{k_f / k_m - 1}{k_f / k_m + A}$$

$$\psi = 1 + \left(\frac{1 + P_f}{P_f^2} \right) V_f$$

where k, k_m, k_f and V_f are as in equation (1), k_E accounts for the shape of the filler particles, and P_f is the packing factor obtained from oil absorption data. As particle size gives rise to similar packing factors, the calculated thermal conductivity in Table 3 is independent on the size of the particles.

Table 4. Parameters in calculating thermal conductivity using Nielsen's equation.

Filler	Particle Size µm	Packing Factor P_f	Einstein Coefficient k_E	Thermal Conductivity k_f W/m.K	Density g/m^3
ATH	1.6	0.56	8	21.34	2.42
	5.0	0.56			
	12.0	0.60			
Silica	1.6	0.44	4.5	1.5	2.65
	4.1	0.54			
	10.5	0.61			

Figure 3 shows the thermal conductivity as a function of the percent volume fraction of filler for both RTV and HPV samples. The experimental values for the RTV show a linear relationship over the range of filler volume and the calculated thermal conductivities from Nielsen's formula gives reasonable estimates to the thermal conductivity over most of the range; however, departing significantly at higher filler volume fraction. The data points from HPV samples show scattered results when compared to both the RTV and Nielsen's formula.

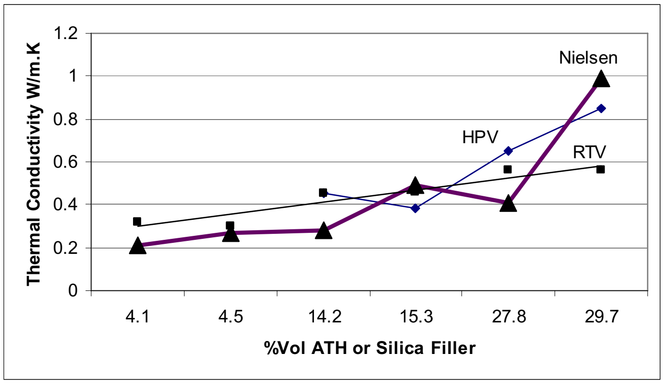

Figure 3. Thermal conductivity, measured and estimated by Nielsen's equation, as a function of volume fraction, in percent, of either ATH or Silica filler for RTV and HTV silicone composite samples.

Several of the HPV samples show higher thermal conductivity than RTV samples and from Nielsen's formula and also higher thermal conductivity with silica filler than with alumina tri-hydrate filler. Scanning electron microscope observations of HPV samples show improved bonding between the silicone polymer and the silica filler that develops through the moulding process when compared to the RTV process. The improved bonding can be expected to reduce the thermal boundary between the filler particles and the matrix thereby increasing the effective thermal conductivity of the composite. Silica, which is considered as an extending filler in silicone, forms bonds with the silicone polymer while alumina tri-hydrate filler does not exhibit this characteristic and considering that the thermal conductivity of the alumina tri-hydrate is an order of magnitude higher than silica filler, bonding appears to be an important aspect of heat transfer which is not taken into consideration by the various heat conductivity models in the literature. The lack of bonding in essence is equivalent to having a thin gas layer between the filler and the silicone polymer, thereby reducing the effective thermal conductivity of the particle.

3 RELATIVE PERMITTIVITY AND STRESS RELIEF

The use of high relative permittivity materials for stress relief in insulation systems is well known. For example, the concept is applied as a surface layer to coils in slots in rotating machines and as a sleeve in cable terminations. However this concept has not yet been applied to the housing material of bushings, surge arresters or cable terminations for stress reduction on the surface of these devices in which a significant reduction in stress relief can be obtained. As an example, the equipotentials at the grounded end of a silicone housed 230 kV bushing is shown in Figure 4; with relative permittivity of 2.7 in Figure 4a and 15 in Figure 4b. The effect of the higher relative permittivity housing in grading the electric stress on the surface of the bushing is quite clear which has a significant impact in preventing dry band arcing on the surface of the housing. In this example, which has not been optimized, the maximum electric stress is reduced by a factor of two as a result of increasing the relative permittivity from 2.7 to 15.

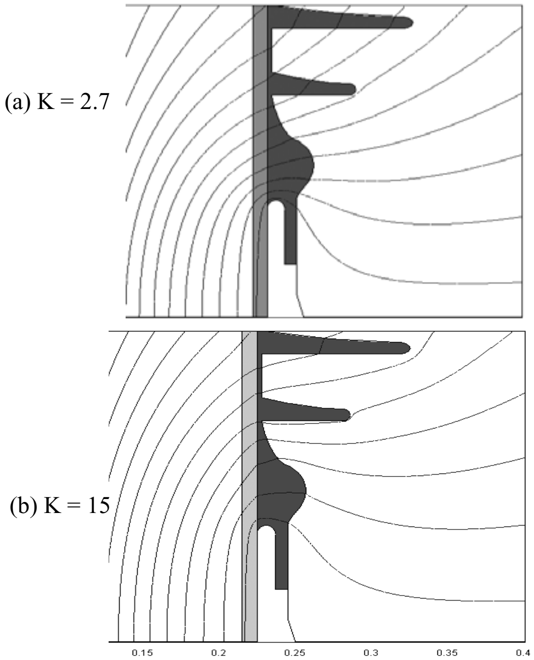

Figure 4. Equipotentials on a 230 kV bushing, silicone rubber housed, with relative permittivity of 2.7 in (a), and 15 in (b).

From Figure 5, the maximum tangential electric field for neat system with relative permittivity of 2.7 is about 0.65 kV/mm while, for the filled system with relative permittivity of 15 it is about 0.5 kV/mm. A greater reduction in stress relief can be realized with a higher relative permittivity material housing but a practical limit of filler concentration will limit the maximum stress relief.

There are numerous types of powders that are employed to enhance the relative permittivity of a composite and Table 5 identifies the classifications of powders that are in use for this purpose. Carbon based powders which are used in indoor stress relief applications cannot be considered for outdoor applications as heat from dry band arcing would most certainly release carbon forming tracks, leading to failure by tracking. Fillers such as $BaTiO_3$, TiO_2, and metal powders have been investigated for possible use in outdoor insulation [14,17] and extensive studies of the ferroelectric ceramic as a filler has been studied [15] and particle size, shape and concentration all play a role in the relative permittivity that can be attained in a composite [31]. In addition, ceramics and powders of $BaTiO_3$ can show a different dielectric behaviour, depending on particle size and packing as well as the electric stress, so their employment as a filler to enhance relative permittivity can vary considerably.

Figure 6 shows the measured relative permittivity of μm size $BaTiO_3$ in epoxy resin for volume fractions to 30 % [14]. The increase in relative permittivity is generally consistent with those reported in the literature for other composites [12, 25]. However, volume fractions greater than about 30% are difficult to mix and air entrapment in the resins is a problem in obtaining void free and homogeneous composites. Metal powder additions in resins can yield higher relative permittivity composites [26] but high loadings of metal powders in the housings of outdoor insulation limit their usefulness as insulation, so their volume additions must be kept quite low.

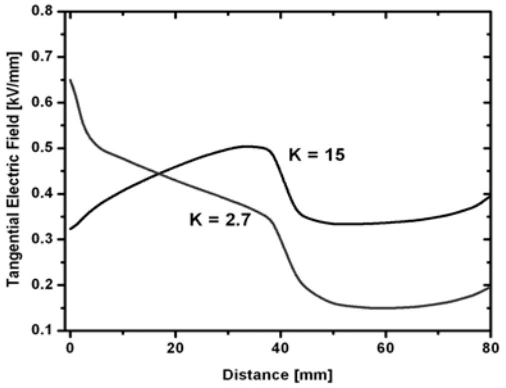

Figure 5. Comparison of the tangential electric stress on the surface of the bushing illustrated in Figure 4.

Table 5. Filler materials commonly used to increase relative permittivity.

Powder Classification	Type
Carbon	Blacks
Metals	Al, Cu, Ag
Ceramics	$BaTiO_3$, TiO_2, Al_2O_3, SiC
Metal Oxides	ZnO, Varistor (ZnO)

Models for multi-filled composites have been proposed and the literature contains numerous formulas for calculating the relative permittivity of the composites [27-29]. However, the main problem in all models is the numerical value for the relative permittivity of the individual filler particles. In aluminum composites, it has been suggested that the effective relative permittivity of the composite comes about from the polarization of each dispersed metal particle in the presence of an applied electric field. Each particle forms a dipole which cumulatively results in a polarization that simulates a true dielectric compound. The dipoles are formed when electrons tunnel from the aluminum particle into the aluminum oxide layer that surrounds the particle. A thin oxide layer results in more electrons tunnelling forming a stronger dipole moment and therefore the composite exhibits a higher effective relative permittivity [26]. A relative permittivity less than 50 has been reported for 40 nm Al_2O_3 coated 3 µm Al filler particles [26].

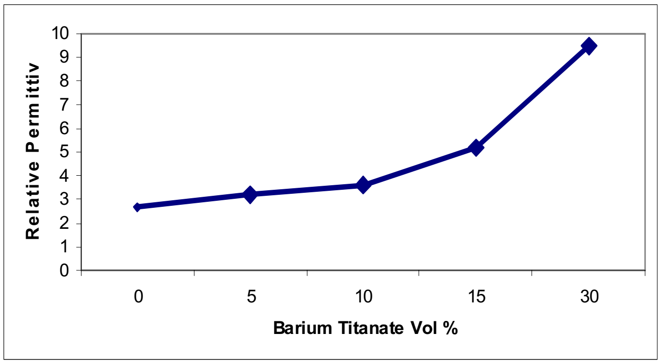

Figure 6. Relative permittivity of barium titanate in epoxy resin [14].

In Figure 7, the relative permittivity of the neat silicone material is 2.7, and 4.2 with 8.3 vol % of µm size aluminum powder, and 6 with 9.9 vol % µm size $BaTiO_3$ powder. It is also evident that the relative permittivity is almost linear with filler loading. A linear relationship between the relative permittivity and the volume fraction of $BaTiO_3$ over the range shown in Figure 7 has been reported in the literature [32]. Also, a linear relationship has been reported for aluminum filler [35]. So, if a parallel model is considered in which each filler component contributes to the overall effective relative permittivity, then the rule of mixtures can be considered to be a good approximation over the range of volume addition. In this case, the effective relative permittivity of the oxide coated aluminum powder is 21 and 36 for $BaTiO_3$ powder. The effective relative permittivity of the Al_2O_3 coated Al powder is in the range reported in the literature [26] but the effective permittivity for the $BaTiO_3$ powder is several orders of magnitude lower than that reported. However, most often the relative permittivity is estimated from various theoretical models or assumed to be the same as a crystal of barium titanate and actual measurements on the powder are not made [30]. In addition, the effective relative permittivity of the composites has been described by models that assume certain particle shapes and volume fractions. It is also known that ceramics and powders of $BaTiO_3$ show a different dielectric behaviour [33, 34, 36].

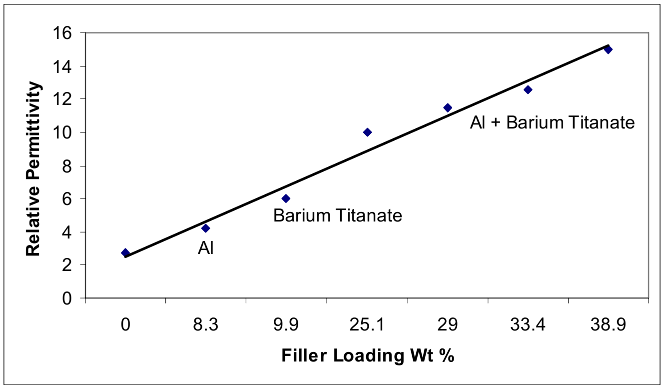

Figure 7. Relative permittivity of barium titanate and alumimum powder filled silicone composites [17].

In an experiment reported by Khastgir and Adachi [37], the relative permittivity of a suspension containing 0.137 volume fraction of $BaTiO_3$ powder in a polydimethylsiloxane (PDMS) resin and crosslinking agent mixture was measured while the resin crosslinked. Before crosslinking, the measured relative permittivity at low frequency was about 90 and as resin crosslinked, the relative permittivity decreased to about 6.4 for the crosslinked matrix. To explain the observations, the authors suggested that the $BaTiO_3$ particles in the liquid PDMS, which have strong dipole moments due to spontaneous polarization, align in an electric field giving rise to a high relative permittivity. However, due to Brownian motion of the $BaTiO_3$ particles there is no alignment of the dipoles and the rotational degree of freedom of the particles is frozen-in in the crosslinked matrix that gives rise to a lower relative permittivity.

Applying the rule of mixtures to the PDMS mixture, the effective relative permittivity of the $BaTiO_3$ is about 650 while in the liquid PDMS, which decreases to about 30 in the crosslinked matrix, while in a PDMS with relative permittivity of 2.84. The relative permittivity of a single crystal of $BaTiO_3$ is about 4000, 100 and 100 along the a, b and c axes, respectively, so depending on the alignment of the particles, the effective relative permittivity can indeed be very low in silicone composite materials.

4 ELECTRICAL CONDUCTIVITY AND POLLUTION PERFORMANCE

An improvement to the pollution performance of outdoor insulators is obtained by providing a resistive or partially conducting insulation material for the housing and a good example of this is the semiconductive glaze on porcelain insulators. The semiconductive glaze provides relief to pollution problems in three ways; namely, by controlling the electric field distribution, preventing moisture condensation by heating, and controlling dry band arcing across protective creepage. These characteristics are well known to porcelain and the concept was introduced some time ago to outdoor polymer insulators [38, 39]; however, no commercial applications to polymer insulation have been made.

Figure 8 shows the tangential component of the electric stress on the surface of the bushing illustrated in Figure 4, with a partially conductive silicone rubber housing having a conductivity of 10^{-6} S/m. Clearly a significant reduction in the maximum electric stress is evident when compared to the normal silicone rubber housing with relative permittivity of 2.7. The partially conductive silicone housing dominates the grading of the electric stress and the contribution due to relative permittivity is insignificant. A conductivity lower than 10^{-6} S/m is not as effective in grading the electric stress and a conductivity higher than this value is too conductive for this type of application.

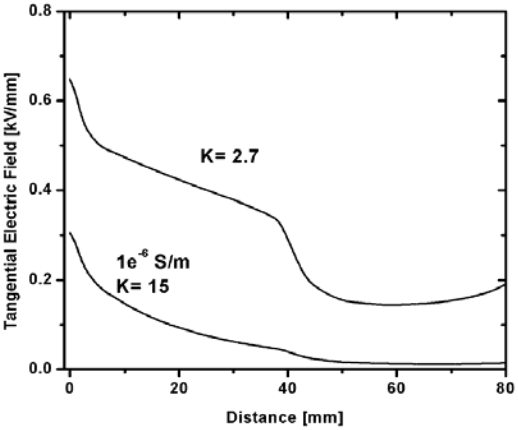

Figure 8. Tangential component of the surface electric stress on the bushing illustrated in Figure 4 with 10^{-6} S/m conductivity silicone rubber housing.

The main problem in using this concept is the dramatic change in the electrical conductivity that occurs within a very narrow range of volume addition of the conductive phase, making it difficult to obtain the required conductivity in production (Figure 9).

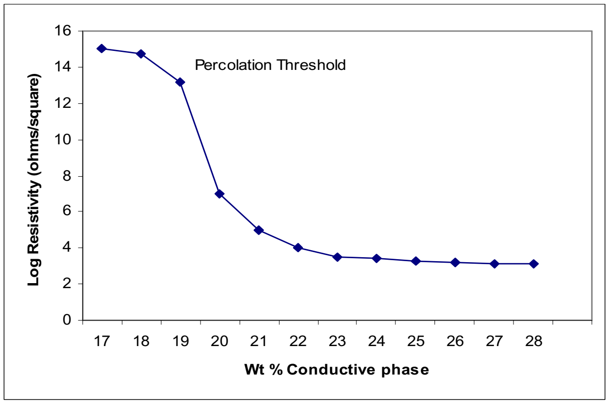

Figure 9. Illustration showing the change in electrical conductivity with volume addition of conductive phase in a composite illustrating the sensitivity of conductivity with dilution.

This sudden change in the relationship between electrical conductivity and filler concentration comes about due to the formation of continuous chains or network of the conducting phase that spans throughout the insulating matrix and is known as percolation. The percolation threshold will depend critically on many parameters such as the particle aspect ratio. However, normally the conductivity of a percolated composite is far greater than required for this application and a percolated conductive phase whose conductivity can be controlled is ideal. This can be attained using antimony-doped tin oxide. It has been suggested that the Sn^{4+} in SnO_2 can be partially replaced by Sb^{5+} forming an n-type semiconductor [41]. To further limit the conductivity, the conductive phase is applied as a film to the surface of titanium dioxide powder which forms an inert core. One such conductive powder is Zelec® [40].

Figure 10 shows resistivity as a function of volume addition of the conductive phase for a silicone composite made with Zelec® [21]. An asymptote in the resistivity is reached when the volume addition of Zelec® is about 24 w%. At this level, a reasonably uniform conductivity can be attained and the required resistance of the housing can be obtained through adjustment of the housing thickness. Figure 11 shows the temperature distribution, measured with an infrared camera, over a cylinder coated with the partially conductive silicone housing. The tube was prepared for a 69 kV experimental gas filled bushing, having the dimensions of 100 mm diameter and 650 mm in length [21].

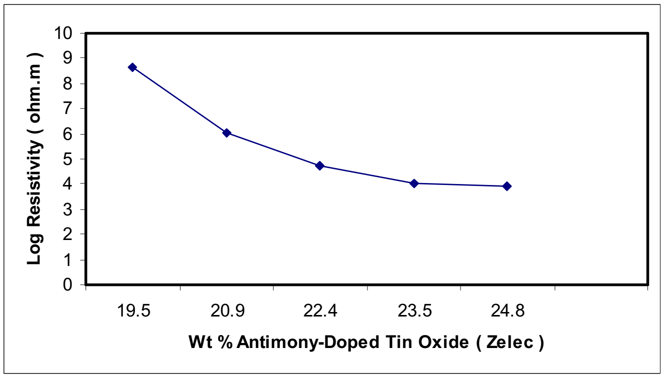

Figure 10. Resistivity as a function of volume dilution, in weight percent, of Zelec® in RTV silicone rubber [21].

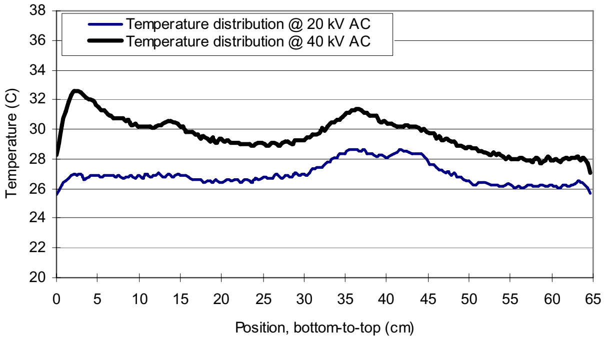

Figure 11. Temperature profile over a composite bushing tube, 100mm dia. and 650mm in length, with a partially conductive silicone housing [21].

5 CONCLUSION

Inorganic fillers added to silicone rubber dielectrics, enhance their properties to make them useful materials for outdoor high voltage insulation applications. Fillers such as barium titanate contribute to increasing the relative permittivity of silicone composites and when used as a housing material for outdoor insulators, the surface stress is significantly reduced while other fillers such as antimony-doped zinc oxide contribute to electrical conductivity achieving a similar result thereby preventing partial discharge, corona discharge or dry band arcing on the surface of the insulation. Other fillers such as silica and aluminum tri-hydrate improve the thermal conductivity of the composites and increase their resistance to erosion that results from the heat produced in dry band arcing.

Bonding of the filler particles in the polymer matrix, which is not taken into account by the various heat conductivity models, appears to be an important aspect of heat transfer and improvements to the predicted thermal conductivities should be possible if the thin gas layer between the filler and the polymer matrix is taken into account.

The relative permittivity of composites may be predicted with greater accuracy if the relative permittivity of the individual filler particles is known better. An oxide layer on metal particle filler and orientation of dipole moments of barium titanate filler contribute to modification of the relative permittivity of the individual particles thereby affecting the relative permittivity of the composite.

Controlled electrical conductivity of binary composites can be attained using antimony-doped tin oxide. A further improvement in the controlled conductivity should be possible by further controlling the doping of Sb^{5+} in SnO_2 making conductive silicone layers on insulator housings easier to manufacture.

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