Investigation of Polymer Filler Interface Using Dielectric Spectroscopy

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***Abstract*— The increase in voltage level and electrical stress of the electrical equipment result in demands for electrical insulations that have high breakdown strength, high thermal conductivity, high electrical resistivity, and high mechanical performance. Use of dielectric polymer nanocomposites is a promising approach as nanocomposites have superior properties over traditional materials. Interaction between nanoparticles and base polymer (interfacial area) has a great effect on the properties of the composite. Therefore, dielectric spectroscopy and thermographic analysis (TGA) are used to study the effect of the interface on the electrical and thermal properties of nanocomposites. Incorporation of treated and untreated nano-alumina into RTV 615 silicone rubber with different weight percentages of nanofillers 5wt %, 7.5wt% 10wt%, and 20wt% has been investigated. Surface treatment of nanoparticles play a crucial role in determining dielectric and thermal properties of nanocomposites materials.**

I. INTRODUCTION

Polymer nanocomposites have received great attention in the last few years due to their significant improvements in electrical, thermal and mechanical properties over traditional polymer materials. These improvements are attributed to an interfacial region between nanofillers and base polymers [1]. However, depending on the strength of interface between nanofillers and base polymers, the improvements of the composites can be high or low. Therefore, there is a great need to investigate the effect of interface on the properties of nanocomposites. The interfacial interactions determine the status of the dispersion and the amount of the interfacial area [2]. Nanofiller agglomeration limit the interactions between base polymers and nanofillers. Therefore, in order to obtain strong interfacial interactions between polymer matrixes and nanofillers, good dispersion, uniform distribution, and surface treatments of nanofillers are required.

Nanocomposite fillers are distinguished from micro-composite fillers in their low loading level (up to 20 wt% for nanofiller, up to 50 wt% for micro-fillers) and their large specific surface area to volume ratio. The properties of nanocomposite are determined by shape, size, chemical nature, loading level, and dispersion of nanofillers, nature of polymer matrix and the interphase between base polymer and fillers [3]. In electrical insulations, many experimental works have shown promising result in terms of electrical breakdown strength, relative permittivity, space charge behavior, erosion resistance, tracking and electrical tree resistance, thermal conductivity and tensile strength [4-9].

This study is focused on the influence of interface between nanofillers and base polymers and their effect on electrical and thermal properties. Treated and untreated nano alumina were selected to investigate the effect of interface on the properties of the silicone rubber nanocomposite. Nanocomposites were prepared with different filler concentrations to study the impact of inorganic fillers on the properties. Dielectric spectroscopy was used to measure the relative permittivity and dissipation factor of the composite. The trends observed using dielectric spectroscopy are informative for composite materials especially at very low frequency where particle agglomeration and the Maxwell-Wanger effect start to influence the polarization [12]. Therefore, dielectric spectroscopy is a useful tool to study the improvements in the quality of the nanocomposite materials. Treated and untreated nano-alumina were investigated using frequency domain spectroscopy from 10^3 to 10^{-4} Hz. In addition, thermogravimetric analysis (TGA) was used to analyze thermal properties of the composites.

II. Experimental

A. Materials

The host polymer material used in this work was RTV 615 silicone rubber (SiR). It consists of a clear liquid silicone rubber (part A) and curing agent (part B). These two parts are clear and colorless liquid which can cure at room temperature. This kind of silicone rubber was chosen because of their low viscosity, low cost, low weight, containing no fillers or solvents in their chemical composition. In addition, curing rate of RTV 615 can be accelerated by heat for fast productions.

In this work, Aeroxide AluC 805 treated with organosilane with specific surface area 90±15 (m2/g,BET) and untreated Aeroxide AluC with specific surface area 100±15 (m2/g,BET) were used with different weight percentage; 5wt%, 7.5wt%, 10wt%, and 20wt% for investigations. Aluminum oxide (Al_2O_3) or alumina is used widely in dielectric polymer nanocomposites as fillers to improve both electrical and thermal properties.

B. Sample Preparations

The mixing ratio for silicone rubber and curing agent is 10:1 by weight, 10 parts of RTV615 (part A) to one-part RTV 615 (part B). To start with, nanofillers were added gradually to part A silicone rubber and then mixed at a speed of ~12,000 rpm using a high shear (HS) mixer (RossTM model HSM-100LSK) for approximately 20 minutes. During this process, high speed of the mixer and friction in the material cause the composite to heat, therefore the composite was kept in a cooling bath for about 5 minutes. After that part B was added to the composite and mixed for 3 minutes. The mixture was degassed in a vacuum oven at 28 inHg to remove any air bubbles. The resulting blend was poured into an aluminum mould and hot pressed at 1200 psi and 150 °C for 15 minutes to obtain cured slabs.

III. Dielectric spectroscopy measurements

An insulation diagnostic analyzer IDAX 300 from Megger™ was used to investigate the dielectric properties of the composites. Guarded electrode system was used to measure the dielectric properties of composite to minimize the effect of the fringe current during measurements. The relative permittivity and tan δ of the composites were measured over a frequency range from 10^3 Hz to 10^{-4} Hz to study the effects of interfacial polarization at 25 °C. The relative permittivity of composite samples was calculated based on the value of measured capacitance.

IV. Thermogravimetric analysis (TGA)

Thermogravimetric analysis (TGA) records the weight loss of a material as a function of temperature in a controlled atmosphere. TGA can provide useful information about thermal stability, composition and decomposition, static oxidation, volatiles and moisture content, filler content and life time estimation of materials [10]. In this work, the analysis was conducted from 200 to 800 °C at a heat rate of 20 °C/min in an air atmosphere using thermogravimetric analyzer (TGA-Q500) from TA instruments to analyze thermal stability and to confirm existence of surface treatments on the fillers.

V. Results and Discussion

Fig. 1 and Fig. 2 show the behavior of relative permittivity and tan δ (dissipation factor) as a function of frequency from 10^3 Hz to 10^{-4} Hz for treated (AluC 805) and untreated (AluC) nano-alumina filled SiR with different concentration as well as pure SiR at 25 °C. At high frequency (Fig.1), there is no major difference observed with increasing concentrations of nanofillers. However, relative permittivity increases proportionally with increasing filler concentrations of treated nano-alumina at very low frequency. For untreated nano-alumina, relative permittivity is higher than pure SiR at low frequency due to interfacial polarization effects. The relative permittivity of treated nano-alumina is similar to untreated nano-alumina at high frequency. On the other hand, untreated nano-alumina shows a significant increase in real permittivity at frequency below 0.1 Hz as compared to treated nano-alumina. In Fig. 2 higher loss tangent of nano-alumina composite was observed because of charge carriers. Since the addition of nanofillers increase the role played by the interphase, the loss tangent of 20 wt% composite is the highest compared to pure rubber and other compositions. The behavior of loss tangent of 5 wt% and 7.5 wt% treated nano-alumina composites is similar to that of pure SiR.

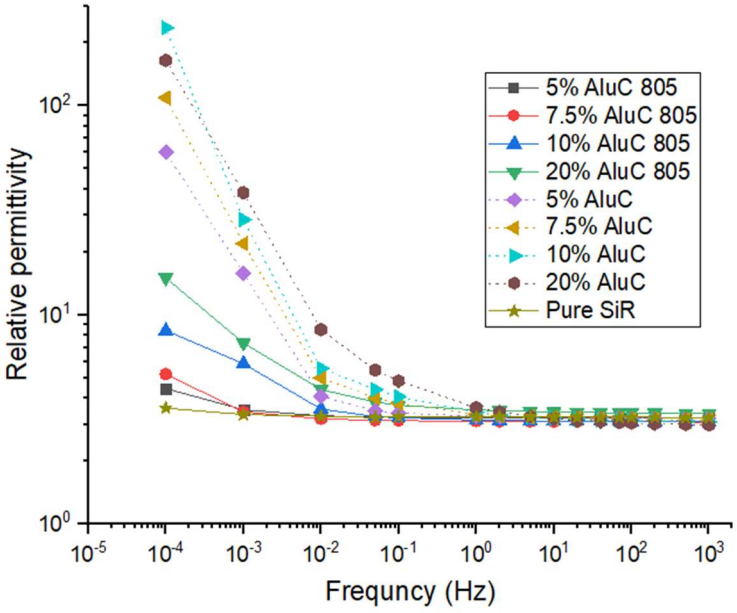

Fig. 1. Relative permittivity for treated and untreated nano-alumina filled SiR at different weight percentages and pure SiR as a function of frequency.

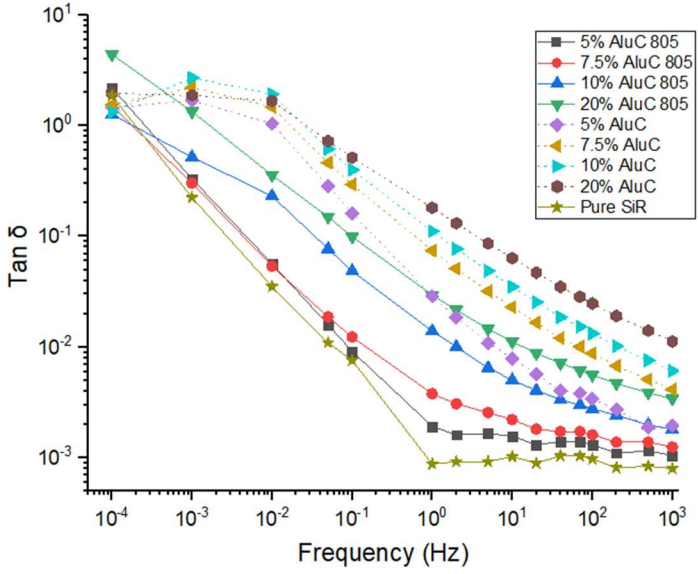

Fig. 2. Tan δ for treated and untreated nano-alumina filled SiR at different weight percentages and pure SiR as a function of frequency.

Treated nano-alumina has lower tan δ than untreated nano-alumina because the surface treatment of nanoparticles makes the bonding between treated filler and base polymer stronger. Good physical bonding between nanofillers and base polymer restrict the chain movements of the polymer [1]. The tan δ trends of untreated nano-alumina are different from those observed for a treated nano-alumina composites of SiR.

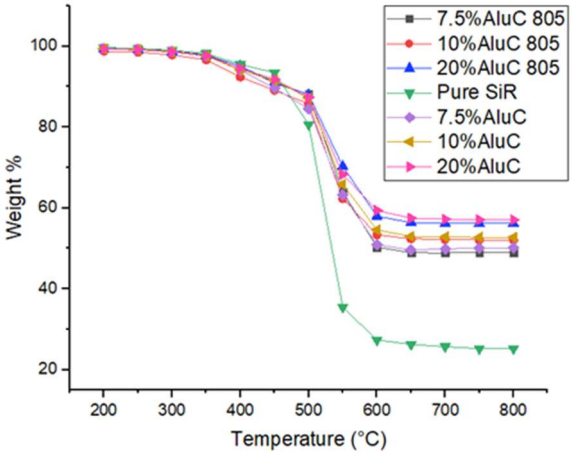

Fig. 3. TGA result of treated and untreated nano-alumina.

In Fig. 3, the result shows that treated and untreated nano-alumina composites have higher thermal stability than the pure polymer. Thermal stability increases with increasing addition of nanoparticles. The thermal stability of untreated nano-alumina composites is slightly better than the treated nano-alumina composites; but the difference is very small. This difference can be due to degradation of the silane group coating of surface treated AluC 805 particles at elevated temperatures [11].

VI. Conclusion

In this paper, treated and untreated nano-alumina filled silicone rubber (SiR) composites were studied to analyze the effect of surface treatment on the interaction between nano-alumina and SiR. Different weight percentages of nano-alumina Alu C and nano-alumina Alu C 805 were investigated. Dielectric spectroscopy is an effective method to get an in-sight into the filler polymer interaction. Observed lower relative permittivity and loss tangent obtained for treated alumina below 0.1 Hz can be due to restricted mobility of the polymer chains. Both treated and untreated nano-alumina improve thermal stability of SiR.

VII. ACKNOWLEDGEMENT

The financial support from the Natural Sciences and Engineering Research Council (NSERC) of Canada, and Prince Sattam bin Abdulaziz University, Saudi Arabia are acknowledged. The authors would like to thank Dr. Khadija Khanum for useful discussions.

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