Filler Dispersion & its Influence on Performance of Nanocomposite Materials

Arathi Mohan Sharma, Faisal Aldawsari, Chitral Angammana, Shesha Jayaram
Dept. of Electrical and Computer Engineering
University of Waterloo, Ontario, Canada
phone: (1) 226-978-3724
e-mail: amohansh@uwaterloo.ca

Abstract—**This paper explores the influence of alumina nanofiller (fumed aluminium oxide hydrophobized with an organosilane) on the properties of silicone rubber based nanocomposites. The composite samples made of 10wt% and 20wt% filler loadings were prepared using high shear and electrospinning techniques with the aim of achieving maximum dispersion of fillers in the silicone matrix. The effects of filler concentrations and mixing method on the mechanical and thermal properties were analyzed. The results of mechanical tests show improvement in strength with an increase in the loadings of nanofillers in the silicone matrix. Additionally, composites prepared using electrospinning method resulted in higher enhancements in mechanical strength and erosion resistance as compared to the conventionally prepared samples. Thermogravimetric Analysis (TGA) of the samples reveal better thermal stability with increasing filler loadings and electro spun composite samples showed improved stability than high shear composites.**

I. Introduction

It is well-known from earlier practices that good electrical properties, mechanical strength and erosion resistance are crucial for the long-term performance of materials used in insulation applications. This enhancement in material property can be achieved with the use of inorganic fillers such as silica, alumina trihydrate (ATH), titanium, alumina; and further enhancement can be achieved by adding nanofillers [1-3]. However, due to their high surface energy, the nanoparticles tend to agglomerate and form clusters, which ultimately leads to characteristics similar to micro particles. Therefore, it is extremely important to break these nano agglomerates and increase the polymer filler interface, in-order to achieve synergistic properties of both phases [4].

Achieving maximum dispersion of nanofillers in the polymer matrix is quite challenging. Over the past years, several processing methods have been implemented with the aim of improving the dispersion of nanofillers [5, 6]. Some of the compounding methods include high shear mixing (HS), ultrasonic agitation, chemical treatments with surfactants or coupling agents and electrospinning (ES) [6-8], all of which impart specific property enhancements to the bulk material. Among these methods, the electrospinning technique has proven to be the most effective in improving the dispersion of nanofillers in polymer matrix [8].

In this research, an attempt has been made to investigate the influence of different mixing techniques; conventional high shear mixing and electrospinning on the mechanical and thermal properties of alumina filled silicone rubber nanocomposites, with a greater focus on different loadings of electrospun composites. The electrospinning setup (presented earlier) was modified in such a way that sample mixtures were pushed through the column with an air pressure of 40 psi and let to fall onto a rotating cone plate controlled by an air motor operating under the same pressure [9]. The pressure in the column was adjusted according to the viscosity of the composite mixtures. As the material is ejected to the rotating cone, and as the melt polymer composite get expelled onto the collector, strong shear forces are developed. These forces break apart any agglomerates of nano particles and multiple cycles of this process enables good dispersion of the nano particles in the base matrix.

II. EXPERIMENTAL

A. Materials and Sample Preparation

Alumina AluC 805 (a fumed aluminum oxide, highly hydrophobized with an organosilane) received from Evonik, was the nanofiller used in this research work. RTV-615, a two part room temperature cured silicone rubber (SiR), manufactured by General Electric (10:1 ratio of Part A- Silicone rubber and Part B- curing agent) was used as the base polymer. One set of composite samples containing 10 wt% and 20 wt% nano filler was prepared using a conventional high shear mixer at 12000rpm and another set was prepared using the electrospinning technique to achieve maximum dispersion. The mixtures were degassed in a vacuum oven at 100kPa vacuum (~29 inHg) and were then cured in a hot press at 150℃ and 15 metric tons pressure for 15 minutes.

B. Testing Methods

B.1. Mechanical Tests

The measurements for tensile strength and elongation at break were done according to the standard ASTM D1708 using a Q series Mechanical Test machine. For each formulations, five specimens (thickness <3.2 mm) were tested at room temperature and the tensile strength was calculated using the equation, $\sigma=f/A_0$, where f and A_0 are the applied force and initial cross-sectional area of the sample, respectively.

B.2. Infrared Laser Tests

Laser based thermal tests were conducted to evaluate the erosion resistance of the composites [10]. The test specimens were irradiated with an infrared laser source (wavelength of 802 nm) set at 16A. In order to ensure that all samples absorbed uniform heat from the laser beam radiation, the samples were mixed with iron oxide (amount equivalent to 5 wt% of filler in each formulation) prior to curing. In this test, 6mm thick samples were placed at 5cm from the laser source. Temperature distribution profile was recorded using a FLIR-SC500 infrared camera.

B.3. Thermogravimetric Analysis (TGA)

Thermogravimetric analysis (TGA) test monitors the weight change with respect to increase in temperature and reflects the thermal stability of the composites. The tests were

performed using TA instrument TGA-Q500 and measurements were carried out in air atmosphere with temperature increasing from 200 to 800 ℃ and a ramp rate of 20 ℃/min.

III. RESULTS AND DISCUSSION

The high shear and electrospun samples of silicone rubber nanocomposites containing nano alumina were compared to explore the mechanical strength, elongation at break and erosion resistance of the materials. The thermal stability and bonding of the filler with the matrix was further investigated using TGA.

A. Mechanical Properties

The study on mechanical properties of the nanocomposites show a good correlation between the type of filler, filler concentration and the dispersion of fillers in silicone rubber matrix. Alumina based composites showed higher strength and elongation compared to pure silicone rubber samples. The tensile strength of high shear samples decreased with increasing loading of the filler at 20%, which can be due to the nanofiller agglomerations present at such high loadings. However, the elongation at break improved by a small margin of 8%. The alumina composites made of electrospinning had nearly the same tensile strength but exhibited significant improvement in percentage elongation from low to high loadings. From Table 1, it can be seen that 10 and 20% filled alumina composites prepared using ES method had higher strength and elongation compared to the high shear samples. The TGA results in Figure 1 support this observation.

TABLE 1: MECHANICAL EVALUATION OF ALUMINA FILLED SILICONE RUBBER NANOCOMPOSITES

Composite Sample	Tensile strength (MPa)	Elongation @ Break (%)
Pure SiR	3.5	126.8
HS 10%nAlu805	4.6	128.1
ES 10%nAlu805	5.6	179.6
HS 20%nAlu805	4.0	138.5
ES20%nAlu805	5.5	229.9

B. Laser Erosion Test

Table 2, shows the eroded mass for samples subjected to a constant energy laser source at 16A. The samples containing 10% nano alumina mixed using the electrospinning technique had ∼40% less eroded mass than its high shear counterpart. This shows that the samples have better erosion resistance and can withstand the heat better than HS composite samples. At higher loading of 20%, electrospun composites had ∼48% less weight loss than the high shear composites. These observations indicate improved dispersion of the filler in the polymer matrix achieved by mixing using the electrospinning technique. Additionally, the

eroded mass of HS and ES alumina composites increased with increasing concentration of the fillers from 10 to 20%. However, the increase is minimal in case of conventionally mixed samples from10 to 20% loadings. At high loadings of alumina, viscosity is very high that conventional mixing becomes difficult leading to more agglomerations and poor dispersion of filler in the rubber matrix.

TABLE 2: ERODED MASS OF SILICONE RUBBER COMPOSITES EXPOSED TO INFRARED LASER

Composite Sample	Eroded mass(mg)
HS 10% nAlu805	100.3
ES 10% nAlu805	61.05
HS 20% nAlu805	95
ES 20% n Alu805	49.1

C. Thermogravimetric Analysis (TGA)

It can be seen from TGA plots in Fig. 1, that nanocomposites have higher residual weight than pure silicone rubber as expected. With the increase in percentage loading of fillers from 10 to 20%, there seems to be a significant increase in the residual weights. Weight loss is less for higher loadings of alumina composites which is an indication of better thermal stability of the material at 20% loading. Furthermore, comparing electrospun and conventionally mixed alumina composites, there is 10% less weight loss for composites prepared using electrospinning. This is due to the better dispersion and bonding of the filler with silicone rubber, which enhances the thermal stability of the ES nanocomposites [11].

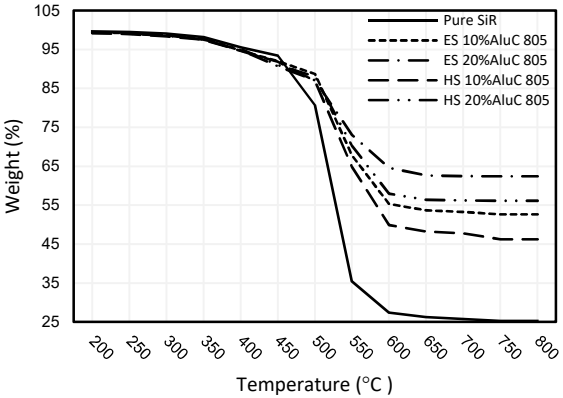

Fig. 1. Thermal Degradation of HS and ES 10% and 20% alumina SiR Nanocomposites

IV. SUMMARY

From this study of silicone rubber nanocomposites based on alumina nano fillers, it is evident that electrospun samples have better mechanical strength, thermal stability and erosion resistance than those samples prepared using high shear mixing. The higher the loading of fillers, higher is the thermal stability of the composites. Tensile strength of the nano composites did not vary much from 10 to 20% loading of alumina in high shear and electrospun samples, however, elongation at break improved significantly.

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