Barium titanate particle filled silicone elastomer composite: Preparation and evaluation of morphology and mechanical behaviour

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Abstract: Barium titanate (BaTiO3) nano particles with different percentage by weight were added in silicone rubber as a filler to develop silicone elastomer composites. The morphology of the fractured surface of the prepared silicone elastomer composite film was investigated to see the distribution of BaTiO3 nano filler in the polymer matrix. The obtained results showed good distribution of BaTiO3 up to 10% by weight in silicon elastomer composite. Beyond this 10% weight percentage, agglomeration of BaTiO3 particles occurs. The mechanical characterizations of silicone elastomer composites with different weight % of BaTiO3 particles were performed through uniaxial tensile testing. Elastic modulus of composite increases when filler content increases up to 10%. However, further increase in the filler content results agglomeration of particles in the composite and decrease in elastic modulus of the silicone elastomer composite. The results show that the optimum value of filler content in the prepared composite is between 5% to 10%.

Key words: Nano composites, elastomer, modulus, agglomeration

1. Introduction
Silicone elastomer is one of most promising electroactive polymers which are being investigated for futuristic applications as sensors and actuators [1]. These elastomers are also being used in dielectric elastomer generators which are under investigation in generation of electrical energy from ocean current energy [2]. The presence of Si-O bond increases its high dielectric constant property as compared with non-polar organic polymers but due to presence of side methyl group Si-O dipoles doesn't approach each other's [3]. To overcome these shortcomings silicone elastomers are mixed with filler i.e. barium titanate and hence mechanical and dielectric properties are enhanced. On the basis of research data available three types of fillers groups are used: first is ceramic nano particles i.e, barium titanate, titanium oxide, lead magnesium niobate–lead titanate, strontium titanate and magnesium niobate [4-7], second is highly polarizable conjugated polymer, such as polyaniline, polythiophene and undoped poly (3- hexylthiophene) [5-6], and third one is the conductive particles, i.e. carbon black, carbon nanotube, copper–polyaniline [8]. Barium titanate is a ferroelectric material which show piezoelectric and photorefractive effects. It has high dielectric constant value which makes it use in high dielectric ceramic materials, capacitors, baseplate materials, microphones, transducers and communications in microwave range [9]. Barium titanate had been used by many researchers to prepare polymer composite matrix to enhance materials properties and electroactive elastomeric composites [10-11]. Barium titanate has been mixed with many polymer based materials such as polystyrenes, poly-ethylene–glycol-diacrylate, epoxides, etc [12-14]. The enhancement of
mechanical and electrical properties were studied and observed that dielectric constant increases with increase in filler content in the base materials [13-17].

In this paper barium titanate as a nano fillers were used as filler material and liquid silicone elastomer as polymer matrix. The curing of the prepared film was carried out at room temperature and morphology, mechanical properties were investigated. Adding filler increases the dielectric constant of elastomer materials but on the other hand elastic modulus also increases which makes it stiffer and harder [18]. The objective of this work is to see the effect of % of BaTiO₃ on the elastic modulus and fracture strain limit of the developed composite. This work also proposes the threshold limit of the filler materials which can be added to the BaTiO₃ filled silicone composite through the morphology and mechanical testing results.

2. Experimental

2.1. Materials

Eco-flex 0030 liquid silicone elastomer is a standard commercial blend used as a base material manufactured by Smooth-On, USA. This elastomer comes in two parts i.e. part A is the polyorganosiloxane and part B amorphous silica mixture [19]. Barium titanate (BaTiO₃) having particle size less than 100 nm, molecular weight 233.19 g/mol was purchased from Sigma Aldrich and was investigated by using SEM as shown in the figure 1.

![SEM image of barium titanate (BaTiO₃) particles](image)

**Figure 1.** SEM image of barium titanate (BaTiO₃) particles

2.2. Sample Preparation

Barium titanate powder was first crushed by using mortar and pestle to make homogenous size of the particles as it was obtained in agglomerated from. Then it was kept in a furnace for 1 hour to make it moisture free. The part A and part B of the platinum-catalyzed liquid silicone rubber were mixed in equal 1:1 ratio by volume or by weight and mechanically stirred for 5 minutes to make a homogeneous solution. After that 0%, 5%, 10% and 15% by weight of barium titanate powder were added and stirred for another 5 minutes ensuring the homogeneity of the mixture. The mixture was then kept in a vacuum for 2 minutes in order to escape the air bubbles trapped in it. Finally, the filled liquid silicone elastomer was spread in a rectangular acrylic mold (1 X 10 X 10 mm) and kept for 4 hours for curing at room temperature to get 1 mm thick filled silicone rubber films.
2.3 Characterization Techniques

2.3.1 Tensile stress-strain studies

Stress-strain properties of the prepared samples were carried out on a universal testing machine (UTM, Zwick-Roell Z010, Germany). The samples for the test were prepared by punching out from ASTM Die-C and tested as per ASTM D 412 method. The cross head speed was taken 50 mm/min and 10 kN load-cell was used. TestXpert II software of Zwick-Roell was used for the analysis of the results. The stress-strain diagrams at different % of BaTiO₃ fillers in silicone elastomer composite are shown figure 2. Three repetitions were carried out for each sample.

2.3.2 Scanning Electron Microscopy

The morphology of the fractured surface was obtained by using GeminiSEM 500 (Gemini technology Germany) to examine the barium titanate dispersion in silicone polymer matrix. The fractured tensile samples cross section was gold sputtered before capturing SEM images.

3. Results and discussion

To study the changes due to addition of barium titanate composites in silicone rubber, samples were experimentally tested under uniaxial loading condition. As we can see in the table 1 and figure 3 modulus decreases as filler content increases up to a certain value i.e. 5% further increasing the filler content increases modulus due to dispersion of the filler content between the gaps of the two chains. When filler content increases from 5% to 10% stress increases with strain in same trend due to reinforcement of filler in silicone rubber matrix. As it increases from 10% agglomeration of filler occurred as shown in the SEM images of the fractured surface in the figure 4, this pattern is seen in all the repetition of the test.

Table 1: Modulus during uniaxial tensile loading for different percentage of nano filler filled silicone elastomer composite

| Barium titanate (%) | 0% | 5% | 10% | 15% |
|---------------------|----|----|-----|-----|
| Modulus at 50% strain (MPa) | 0.0623 | 0.059 | 0.0463 | 0.0483 |
| Modulus at 100% strain (MPa) | 0.08 | 0.0953 | 0.0713 | 0.0837 |

Figure 4 (a), (b), (c) and (d) is the SEM images of the fractured surface of the filled silicone elastomer. We can see the rigged surface of filled rubber with the unfilled one and rigged surface increases as filler content increases. Agglomeration of particles can be clearly seen in the SEM images which increase as filler content increases results into weak interaction between agglomerated filler particles and silicone elastomer chain [20].
Figure 2. Mechanical properties of the filled silicone elastomer: (a) elongation at break, (b) modulus at 50% and 100% strain.

Figure 3. Stress–strain curves for silicone elastomer-barium titanate composites with random distribution of barium titanate nano fillers having filler content, (a) 0%, (b) 5%, (c) 10% and 15% in silicone rubber with repetition of three.
Figure 4. SEM micrographs of fractured surfaces of silicone elastomer barium titanate composites with (a) 0%, (b) 5%, (c) 10%, and 15% of nano filler. Blue circles show the agglomeration of the particles when filler content increases from 10% to 15%.

4. Conclusion
It was shown that strain at break decreases as filler content increases up to certain value. Further increase in filler content increases strain at break and again it decreases on increasing filler content due to agglomeration of the particles. Modulus increases as the filler content increases and decreases on further increase in the filler content. After reaching a threshold value, agglomeration of filler increases and hence modulus decreases. The optimum value of filler content from the results may be considered between 5% to 10% of silicone elastomer composite.
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