Silicone composites cured under a high electric field : an electro-mechanical experimental study

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Abstract

Recently, dielectric elastomers (DEs) become enviable materials for the applications of electro-mechanical transducers. However, requirements of enhanced properties like high dielectric constants, low elastic moduli, high stretchability, and minimum viscous and dielectric losses are major challenges. The development of elastomeric composites to improve dielectric properties is essential for wide applications, particularly in the field of DE-based generators and actuators. To enhance dielectric properties, particle-filled DE can be prepared in the presence of an electric field during the curing process which leads to aligned particles in a regular fashion. Previous studies suggest that such an alignment enhances electrical properties of the resulted composite as compared to the one prepared under a normal condition, i.e., curing without an electric filed. In contrast to earlier studies, in this work, we develop a holistic experimental characterization approach to assesses how particle alignments enhance mechanical, electrical and electro-mechanical properties over the randomly particle filled composites. For that, a commercially available silicone polymer (Ecoflex) is used and is filled with high dielectric constant barium titanate fillers. In order to substantiate electro-mechanical performance enhancements, mechanical, electrical, electro-mechanical, and morphology experiments are conducted on both types of silicone composites. Dielectric composites prepared under an electric field during curing process outperform in almost all aspects of electro-mechanical tests. This comprehensive experimental characterization technique provides a framework that will guide future dielectric composite preparations and electrical, mechanical, and electro-mechanical experiments especially for the oriented dipole fillers prepared under an electric field.

Keywords: Electro-active polymers; Dielectric elastomer; silicone composites; barium titanate; electro-mechanical properties; electric field; polymer curing.

1. Introduction

Dielectric elastomers (DEs) belong to a family of electro-active polymers (EAPs). They have compliant electrodes on their top and bottom surfaces which respond to electrical stimulus by changing their size and shapes [1, 2, 3, 4]. EAPs have gained tremendous importance in the research community with a wide range of applications for their low mass, simple structure, low cost, robustness, large and complex actuation properties, and high energy density [5]. Large and complex actuations as a result of electro-mechanical transducing of DEs are key factors in many engineering applications such as in soft robotics, sensors, energy

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harvesters and artificial muscles [6, 7, 8, 9, 10]. At present, acrylic polymers such as VHB and silicone polymers are most commonly used dielectric elastomers due to their favorable behavior under the application of an electric field [11, 12]. A significant research efforts have been directed in recent years to characterize mechanical, electrical, and electro-mechanical properties of commercially available elastomers as well as customised new polymers and polymer composites as DEs [13, 14, 15]. Therein, a wide range of time-dependent viscoelastic characterization experiments such as single-step and multiple-step relaxation tests, loading-unloading cyclic tests of an acrylic elastomer were conducted to get complete understandings of their mechanical behavior. These studies reveal that acrylic elastomers have several disadvantages over silicones such as high viscoelasticity, high loss of tension, adhesiveness and high operating voltage [16], large variation in the stiffness modulus with a change in operating temperature, low thermal stability [2, 17, 18]. Although silicone rubbers have many advantages over acrylic elastomers however, they have low dielectric constants [19], low voltage induced strains [20, 21], and low glass transition temperatures [22]. So, there is a growing need for the fabrication of new materials which have high electromechanical properties with enhanced dielectric permittivities. Many research works have been carried out by which voltage induced strain can be enhanced by adding nano-fillers in the polymer matrix [2, 12, 18, 23, 24, 25, 26]. Polymer blends are being fabricated which can be also used as dielectric elastomers. Wang et al. [27] fabricated dielectric polymer capacitors by using a series of polymers sandwiched together which are mainly acrylic elastomers and ferroelectric particles diffuse into the polymer matrix as filler materials. Ellingford et al. [28] used grafting techniques to prepare polymer blends of styrene-butadiene-styrene block copolymer (SBS) with poly (vinylidene fluoride) (PVDF) and carbon nanotubes (CNTs). They reported enhanced dielectric and mechanical properties. But when the percentage of CNT increases near and beyond the percolation threshold limit, an agglomeration of nano fillers takes place. Poudel et al. [29] fabricated dielectric nanocomposites by mixing poly(styrene-ethylene/butylene-styrene) (SEBS), SEBS-grafted-maleic anhydride (SEBS-g-MA) and barium titanate as filler particles. They achieved a high dielectric constant elastomer with a low dielectric loss but the modulus of elasticity increased after a certain limit of filler addition.

Carbon nanotubes (CNTs) are widely being used to fabricate new composites of dielectric elastomers for desirable dielectric and mechanical properties for transducer applications, see Ellingford et al. [30] and Madsen et al. [31] for exhaustive reviews on the material perspectives of dielectric elastomers. Geng et al. [32] prepared multi-wall carbon nanotubes filled silicone rubber with the help of solvent evaporation method. They found enhanced dielectric properties of the silicone composites well below the percolation threshold limit. Saji et al. [33] tested dielectric relaxation behavior of in-house fabricated silicone composites made of multi-wall carbon nanotubes with increasing percentage of fillers. Zeng et al. [34] designed an electrothermal bimetallic actuators by using silicone rubber filled with multi-walled carbon nanotubes which can be driven even at a low voltage. Tan et al. [35] prepared multi-wall carbon nanotubes/fluorosilicone rubber composites by a direct mechanical mixing method and found enhanced dielectric properties. The main shortcoming as reported by all the researchers is that the dispersion of carbon nanotubes near the percolation limit hinders the dielectric and mechanical property of the composites.

A sizeable amount of research works have also been carried out by mixing various fillers such as barium titanate, titanium dioxide, carbon black, strontium titanate, zirconate titanate, lead zirconate titanate, and lead lanthanum with different types of polymers [29, 36, 37, 38, 39, 40, 41, 42, 43, 44, 45, 46, 47, 48, 49, 50]. However, most of these fillers have poor compatibility, non-uniform distribution in polymer matrices and also show high dielectric losses with an increase in filler content as reported by Jiang et al. [41]. These unfavourable circumstances hinder applications of filled polymers in actuators and generators. In order to overcome above shortcomings, fillers are surface treated by using commercial surfactants [39, 42]. This method improves the mixing and compatibility of filler particles in the polymer matrix. However, it results in high leakage current, dielectric losses, and agglomeration when filler content increases near the threshold limit [51, 52].

Several works have been reported that ferroelectric materials can be polarized and aligned when they are brought under an electric field during the curing process [44, 53, 54]. Electrophoretic assembly of fillers in a polymer matrix by using an electric field was successfully carried out by Tomer et al. [55] and Kashani et al. [40]. In one of the study [40], titanium dioxide was used as a filler in silicone rubber matrix and in the other study [55], barium titanate was used as the filler. Both works reported that the sample prepared under an electric field enhanced the dielectric properties. Note that except few dielectric permittivity tests, Tomer et al. [55] did neither mechanical nor electro-mechanical experiments. Despite the fact that Kashani et al. [40] did some tensile mechanical tests to show how the elastic modulus changes with the increase of randomly-distributed filler, a complete mechanical and electro-mechanical characterizations of oriented and randomly-filled composites at large strains through a rigorous study with loading-unloading cyclic tests, single and multi-step relaxation tests have not been carried out yet. Moreover, the enhancement of electro-mechanical properties has not been demonstrated with the help of any actuator or generator prototype. Particularly, how much electro-mechanical performances improved in the case of oriented fillers in contrast to randomly filled composites are yet to be demonstrated. Therefore, the aim of this contribution is to propose a holistic approach for characterizing aligned particle filled dielectric elastomers starting from composite preparations to actuation enhancements tests. Note that material modelling and numerical simulation of electro-active composites is an active field of research, see [56, 57, 58].

In this work, we develop silicone composites using barium titanate (BT) nano particle as fillers and a commercially available polymer, Ecoflex, as the matrix. To prepare oriented and randomly-filled composites with different percentage of fillers, Ecoflex silicone is being cured with and without an electric field during the curing process. At first a series of viscoelastic tests were carried out of both composites, randomly-filled and oriented-filled polymers. The oriented silicone composites were compared with those of the randomly dispersed filler composites of same filler volume percentage to show how orientations affect viscoelastic properties. Afterwards, dielectric properties, e.g., dielectric constants are measured with various percentage of fillers of both composites. After studying mechanical, electrical, and morphology properties of both composites, an optimized filler content, that is free from any agglomeration, is determined for each type. Dielectric constant and dielectric losses of the composites are measured in a large frequency range at room temperature (25°C) and results are compared between the silicone composites prepared by the two distinct methods. In this work, we further compare the actuation performances of both types of silicone composites by making a circular actuator. Thus, we aim to prepare silicone composites which have high dielectric constants and a low operating driving voltage for a wide range of applications.

2. Experimental study

2.1. Material preparations

Ecoflex is a commercially available silicone rubber that is a trademark product of Smooth-On, an USA-based company. It is a platinum catalyzed silicone rubber which is curable at room temperature. The polymer consists of two parts; A and B. Both parts are equally mixed by weight or by volume and its properties are given in Table 1 [59]. The first part A contains polydimethylsiloxane having platinum as a catalyst and part

B contains a curing agent. However, the distributor has not disclosed the actual chemical compositions of Ecoflex. Barium titanate (BT), a high dielectric constant filler having particle size less than 100 nanometer (nm) with a cubic crystalline phase, was purchased from Sigma-Aldrich. The preparation of BT filler particle was carried out by drying it for 24 hours in a vacuum oven at 100°C. Figure 1 shows Scanning Electron Microscopic (SEM) images of BT particles having a size less than 100 nm.

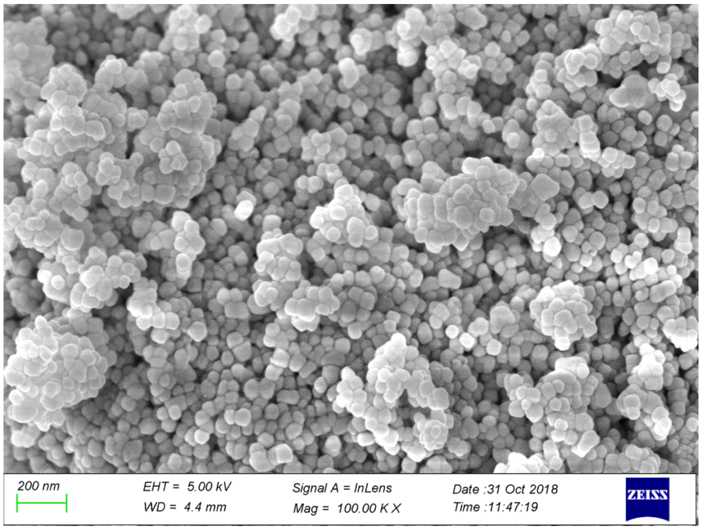

Figure 1: SEM image of barium titanate nano particles

Table 1: Properties of Ecoflex 00-30 [59]

Commercial name	mixed viscosity	shore hardness (A)	tensile strength	100% modulus	elongation at break
Ecoflex 00-30	3 Pa.s	00-30	1.37 MPa	0.069 MPa	900%

2.2. Preparation of silicone composites

For mechanical and electro-mechanical characterizations, samples were prepared by taking different weight fractions of fillers in silicone (Ecoflex) matrix. During material preparations, the solution of silicone rubber and filler particles were mechanically stirred for 10 minutes to ensure a homogenous mixing. While mixing, some air bubbles got entrapped which were removed by keeping the mixture in a vacuum oven for 15 minutes. After the degassing, the mixture was poured in a mold and kept it at room temperature for 4 hours. For experimental purposes, two different curing conditions were selected. The first type of curing was performed in the absence of an electric field which resulted in random oriented fillers in the matrix while the second type of composite was prepared in the presence of an electric field which yielded aligned filler dipoles.

In the first case, the mold was kept as it was for 4 hours at room temperature for the curing of Ecoflex-BT composite. For the second case, a high voltage DC source (Ionics Power Solution Limited, India) was used during the curing process. The mixture of silicone rubber and BT filler was spread on an aluminium plate

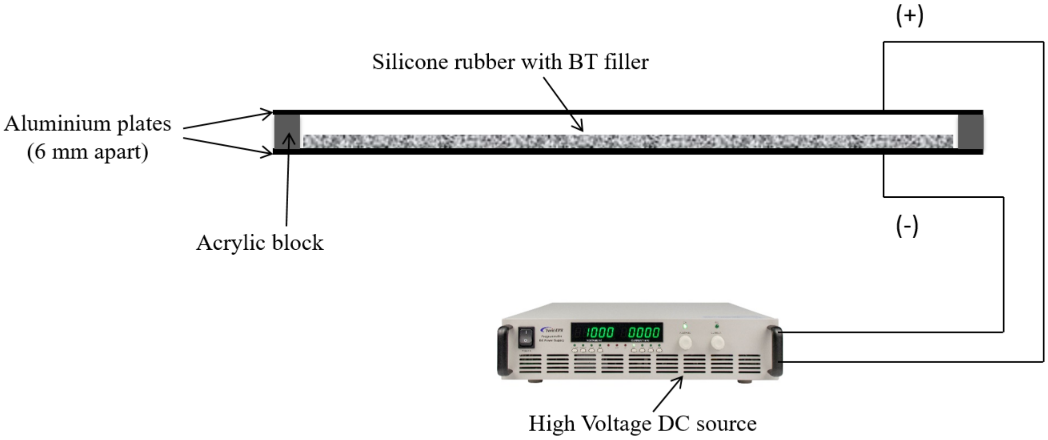

Figure 2: Schematic diagram of Ecoflex-BT curing process in presence of an electric field

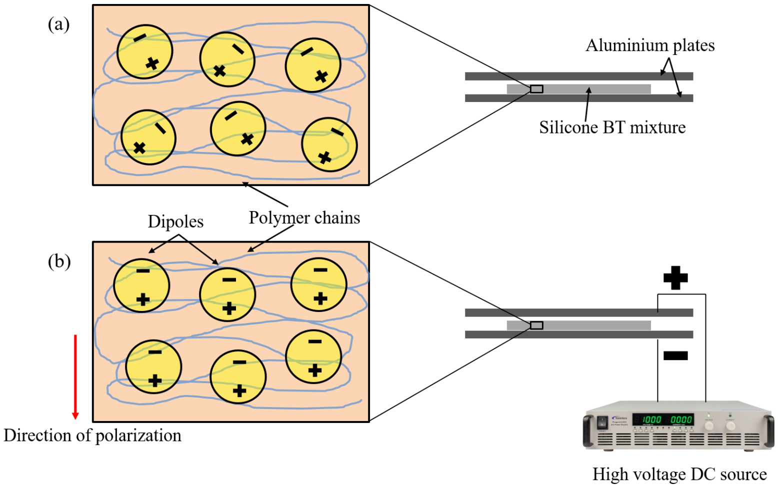

Figure 3: Arrangements of dipoles in polymer matrix during the curing process, (a) randomly distributed filler dipoles in polymer matrix, (b) aligned filler dipoles in polymer matrix due to the application of an electric field

having cavity of 1 mm depth and another plate was kept just above the polymer sheet at a distance of 6 mm. Then both the plates were connected with two terminals of a high voltage DC source which created an electric field of 1.67 kV/mm as schematically shown in figure 2. After curing the silicone films for 4 hours under an electric voltage, the electrical supply was disconnected and the film was peeled off gently from the mold. Figure 3 shows arrangements of filler particles (dipoles) for both the cases schematically. Figure 3(a) shows that filler dipoles are randomly oriented in the first case, i.e., curing in the absence of an electric field whereas, figure 3(b) shows alignments of filler dipoles thanks to an applied electric field in the second case. 5%, 10%, and 15% weight percents of BT particles were selected to prepare samples both for randomly distributed and oriented composites. Moreover, we tried to make 20% BT-filled composite which was successful for aligned fillers but unsuccessfully for randomly distributed particles as the agglomeration of BT starts at 10% filler content for the latter case. Note that as per manufacturer's (Smooth-On, USA) specification, the density of Ecoflex is 1.065 g/cm^3, while the density of barium titanate (BT) particle is 6.02 g/cm^3. If 5% BT filler is added to the Ecoflex matrix, then the composite density becomes 1.108 g/cm^3. In a similar way, densities of 10%, 15%, and 20% can be calculated as 1.151 g/cm^3, 1.193 g/cm^3, and 1.234 g/cm^3, respectively.

2.3. Mechanical and dielectric characterizations and actuation experiments

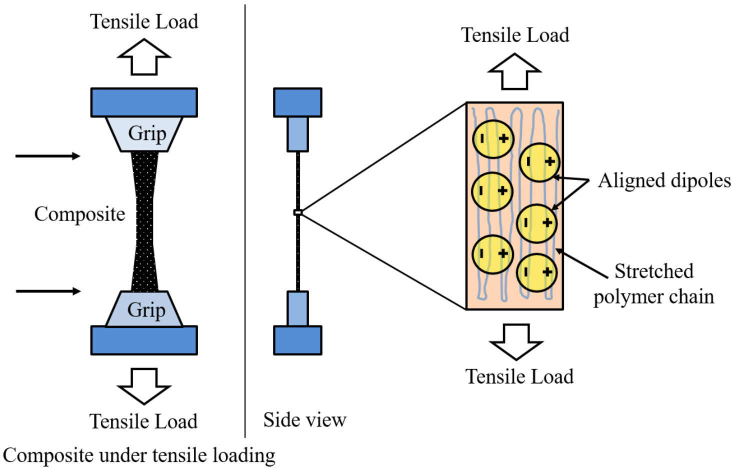

Figure 4: A schematic diagram shows how the aligned composites are taken to prepared samples for various mechanical tests

For the mechanical characterization, Ecoflex-BT composite samples were tested on a universal testing machine under a uniaxial mode of deformation. For this purpose, Zwick/Roell Z010 tensile testing machine was used which has a load cell capacity of 2.5 kN. Sample dimensions were of ASTM D412 standard obtained by using die punching device. Three samples tested for each case. A schematic diagram is presented in Figure 4 that shows how the aligned composites are taken to prepared samples for various mechanical tests. All tests were carried out at a cross head speed of 50 mm/min up to the sample failure. Data were acquired and analysed using TestXpert II, a built-in software of Zwick Roell. To characterize viscoelastic behavior of polymeric materials, loading and unloading cyclic experiments are widely used [60, 61]. For the composite materials under investigation, we have conducted loading tests up to a maximum strain of 200% with a repetition of three samples for each test. This extent of deformation is well enough for dielectric elastomers as they are being used in soft robotics and in energy harvesting mechanisms. To analyze equilibrium stress-strain behaviour, relaxation tests were carried out at holding strain of 200% and a relaxation time of 2400 seconds was chosen. SEM images of the fractured surfaces of both the samples, i.e., randomly distributed fillers and oriented dipole filler samples, were observed by using GeminiSEM 500 (Gemini technology, Germany) to determine optimum BT filler content.

To measure dielectric properties, tests were carried out by a Novocontrol spectroscopy at room temperature using one volt (1V) signal with a frequency range of 1Hz to 1MHz. The obtained thin composite films of silicone composites were fixed in a circular frame without pre-stretching. Then carbon conductive grease (MG chemicals) was used as an electrode on both sides of the film. 20 mm diameter gold plated copper electrodes were used on both sides of the films to carry out dielectric measurements with a repetition of three each. The results of dielectric constant and dielectric losses were analyzed on the 'windata' software from Novocontrol company. The electro-mechanical test setup comprises of a high voltage DC power source, a high resolution camera, and a sample holder. In order to measure actuation-induced strains, a circular actuator made of the composites without any pre-stretch was made. With Ecoflex-BT composite, thin layers of composite of 300 μm thickness were prepared and fixed in a circular frame of inner diameter of 50 mm without pre-stretching. Afterwards, a carbon grease was used as an electrode to create a circular actuator of 10 mm diameter. The prepared circular actuators were connected to a high voltage DC source with a ramp voltage of 100 to 150 V/s and the actuation was recorded using a high resolution camera (1080p, Logitech) as conducted by several other research groups [38, 41, 62, 63]. The recorded video was converted into images by using a video to image software, 'DVD video soft' with a frame rate of 2 per second. The converted images were analyzed by using an image processing software, 'Imagej'. The initial active area was measured in pixels and consequently the actuated active areas were measured further. Afterwards, the actuation strains A_s were calculated by using the following equation,

$$A_s = \frac{(A_2 - A_1)}{A_1} \tag{1}$$

where, A_2 is an actuated active area and A_1 is the initial area.

3. Results and discussions

3.1. Mechanical properties of Ecoflex-BT composites

Stress-strain curves for randomly distributed fillers in the polymer matrix are presented in figure 5(a). As expected, the mechanical stress increases as the filler content increases from 0% to 15%. One of the reasons of such an increment is due to the addition of high modulus inorganic BT filler particles to the silicone matrix. We can see from Table 2 that with the increase of the filler content to 15%, the modulus of elasticity increases due to the reinforcement of fillers into Ecoflex-BT matrix. On further increase of filler contents, an agglomeration of particles takes place which is also observed and explained by many other researchers for different fillers, depending upon the combination of filler and polymer matrix types, see [38, 39, 40]. Therefore, the filler addition results in a stiffening of the composites and the elongation at break decreases. In order to minimize such effects, the curing of filled silicone composite was carried out under an electric field as shown in figure 3 where the alignments of dipoles take place due to the applied field. For comparison purposes, stress-strain curves are presented in figures 5(a) and 5(b) for randomly distributed (R) and oriented dipole (O) composites, respectively. The elastic modulus of the oriented dipole silicone is higher for the same filler content as compared to that with a randomly distributed one as illustrated in Table 2. However, for both 5% R and 5% O filler contents, there is not much change in the elastic modulus and it is observed that the values of elastic modulus are approximately the same. As the amount of filler content is less, the distribution of particles in the polymer matrix is less and uneven so there is not much effect of alignment of BT filler particles on the elastic modulus of the composite. Further loading in filler content increases reinforcement which results in an increment in the elastic modulus which is more in aligned fillers as the polymer chains are constraint to slide. For both 10% and 15% filler contents, the elongation at break in the case of oriented dipole composite is higher than those of randomly distributed one as shown in figure 6. Furthermore, in the case of oriented composite, the loading capacity of BT-filled matrix increases due to an alignment of filler dipoles in the electric field direction. Thanks to the effect of dipoles, the agglomeration of filler particles also decreases that results in an increase of the elongation at break of the material as shown in figures 6 and 7.

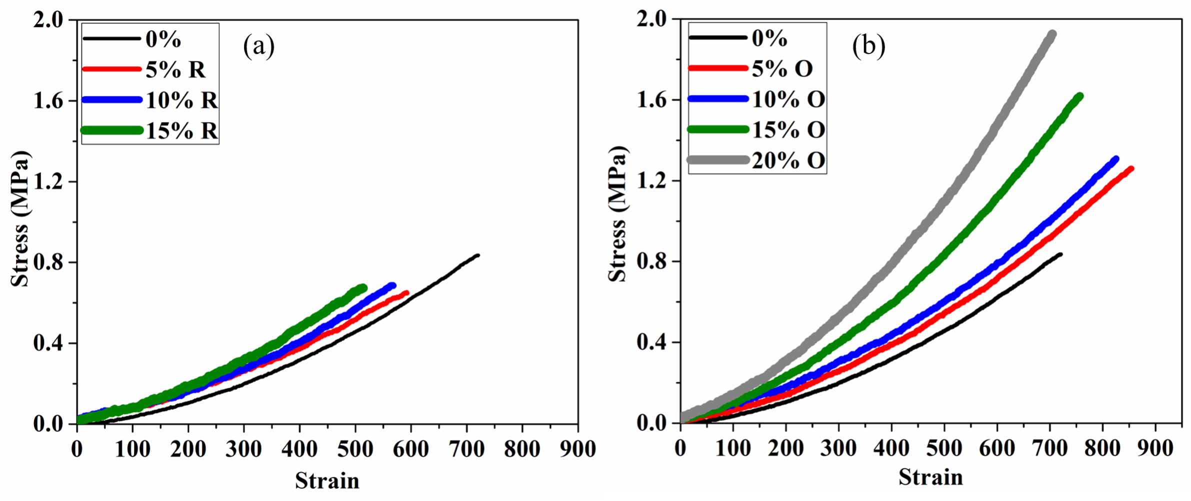

Figure 5: Stress-strain curves: (a) for randomly distributed silicone composites with filler contents of 0%, 5%, 10%, and 15% by weight; (b) for oriented filler dipoles in silicone matrix with contents of 5%, 10%, 15%, and 20% by weight

Table 2: Effects of filler addition to the modulus of silicone composites in both processes of curing

Barium titanate (wt%)		5%	10%	15%	20%
Random	Elastic modulus at 100% strain (MPa)	0.078±0.0010	0.082±0.0060	0.084±0.0053	-
	Elastic modulus at 200% strain (MPa)	0.161±0.009	0.166±0.017	0.187±0.015	-
Oriented	Elastic modulus at 100% strain (MPa)	0.065±0.0090	0.094±0.0014	0.104±0.0010	0.141±0.0014
	Elastic modulus at 200% strain (MPa)	0.141±0.019	0.178±0.013	0.235±0.012	0.305±0.016

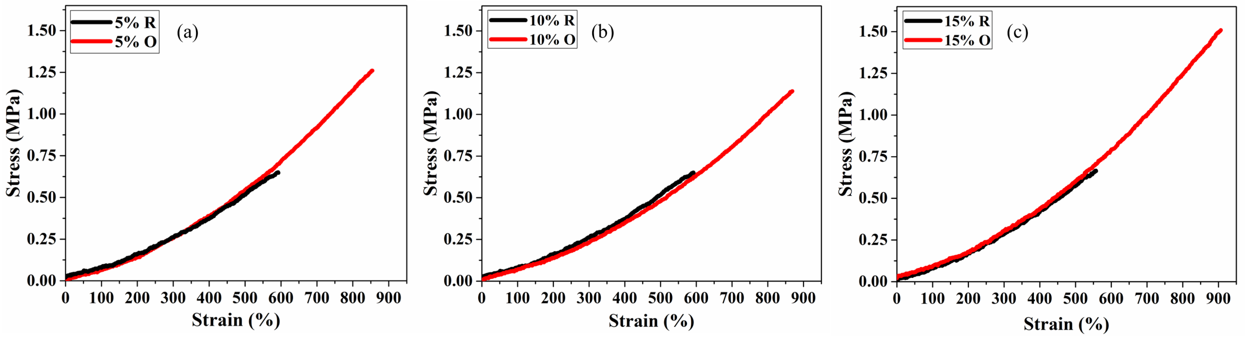

Figure 6: Comparison between randomly dispersed filler and oriented one with filler contents of (a) 5%, (b) 10% and (c) 15%

3.2. Loading-unloading cyclic experiments

Dielectric elastomers undergo cyclic deformations in most of their applications such as in actuators and generators. Hence, it is important to investigate hysteresis phenomenon of the manufactured materials through loading and unloading cyclic tests. For that, a fixed low strain rate of $0.021s^{-1}$ was chosen in

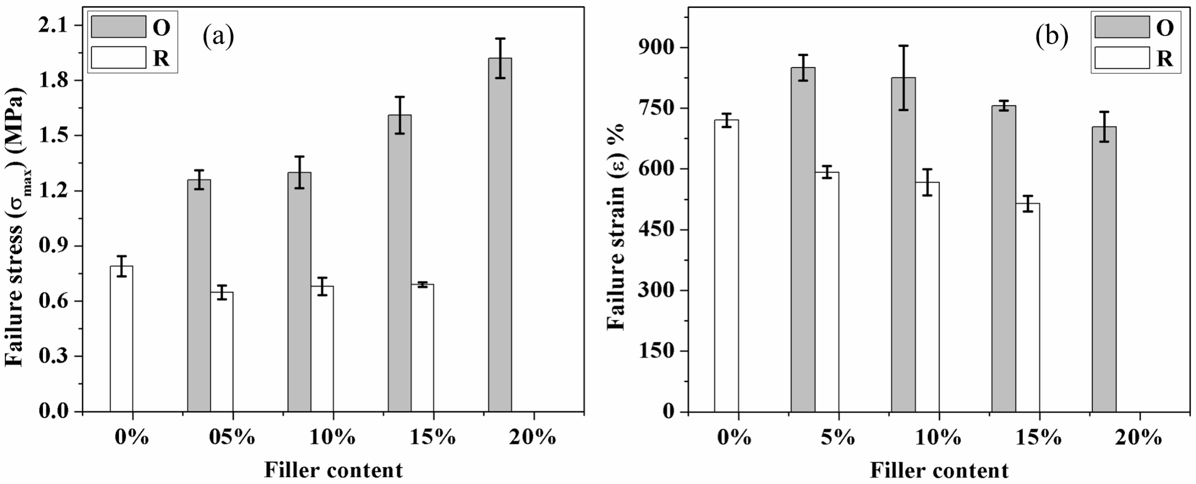

Figure 7: (a) Failure stress and (b) failure strain for silicone composites filled with randomly distributed and oriented fillers

this study as most of the devices made of dielectric elastomers are operated within the low strain rates, see [13, 14, 64, 65]. The hysteresis loss of a cycle is represented by the enclosed area of the loading and unloading curves. It can be observed that as the filler contents increase, the dissipative loss also increases. However, it is relatively less in the case of oriented composite. This would be possible because pure silicone polymer chains are free to slide on each other but as the fillers are incorporated, the particles present between the chains of the polymer matrix make them constraint. As a result, composites dissipate more energy due to frictions between the filler particles and the polymer chains. In the case of oriented filler composites, particles are arranged in a preferred direction that leads to a less dissipation of energy thanks to less frictions as illustrated in Table 3. For comparison, results of pure silicone elastomer, i.e., 0% filler content is also added. The dissipative loss (in percentage) increases as the filler content increases but it is comparatively less in the oriented filler composites as shown in Table 3.

Table 3: Energy loss due to dissipative behaviour characterized by loading and unloading curves

Sample (wt%)	Stress (kPa) at 200 % strain	Loss (%)
0%	200 ±38.36	36.62±13.31
5% Random	315±25.07	39.80±7.07
10% Random	325±28.56	39.09±10.42
15% Random	410±36.66	44.88±9.40
5% Oriented	420±18.93	38.44±4.69
10% Oriented	425±12.20	39.34±2.62
15% Oriented	440±15.06	41.70±1.07
20% Oriented	520±10.41	39.52±1.82

3.3. Stress relaxation experiments

When stress is applied, networks of polymer chains will deform and the deformed chains further will get relaxed slowly until an equilibrium state of stress is achieved for a sample holding at a constant strain. Stress relaxation, which a polymer realizes through a decrease of the mechanical stress at a constant strain, is an important phenomenon to understand the viscoelastic properties of polymers. The relaxation phenomenon of Ecoflex-BT (Ecoflex-barium titanate) composite was obtained experimentally at a strain of 200% by

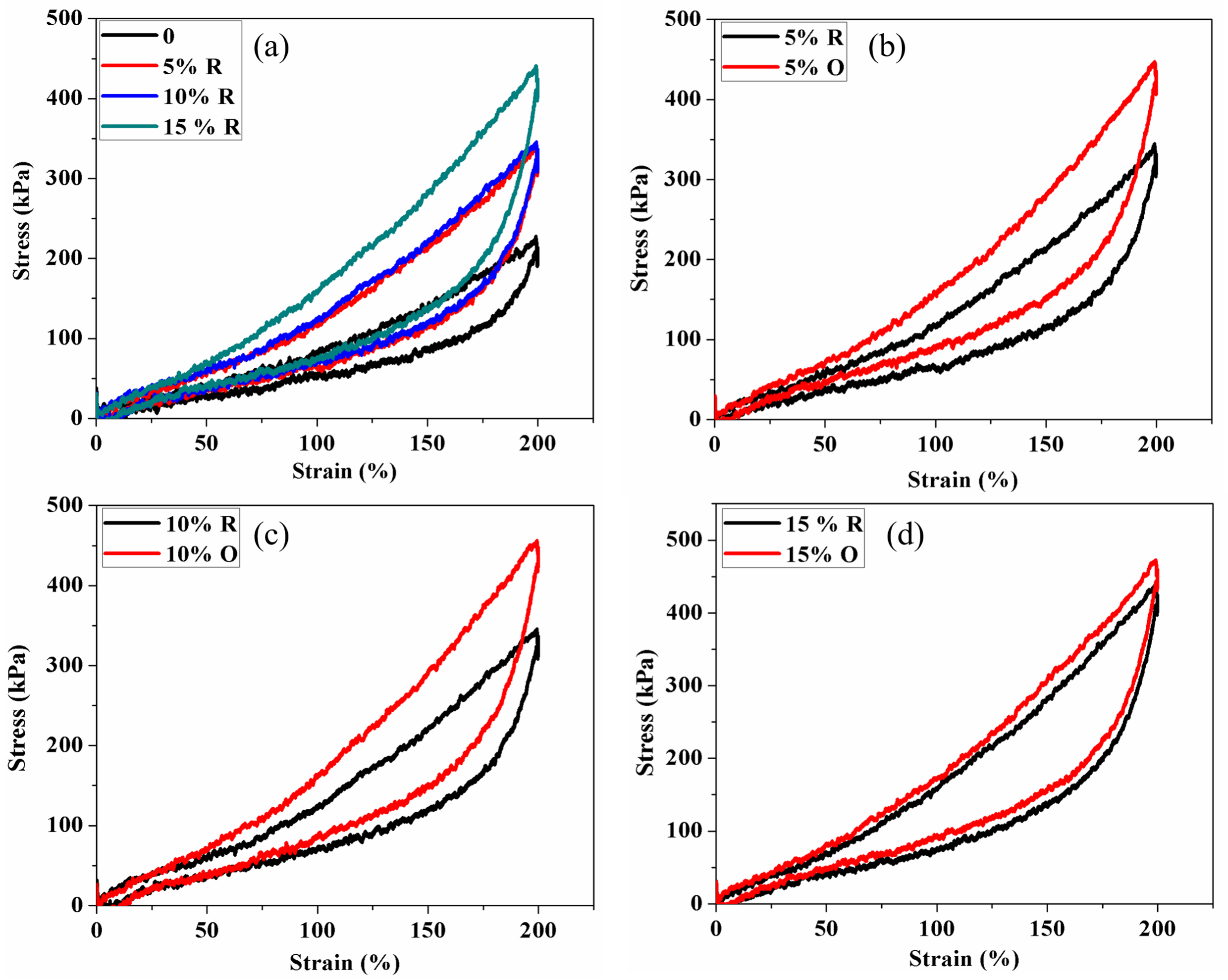

Figure 8: Loading and unloading hysteresis cycle of silicone composites at different percentage of fillers (a) only randomly distributed fillers, (b) 5%, (c) 10%, and (d) 15%, randomly oriented vs oriented filler composites

holding the samples for half an hour to attain the equilibrium condition. Figure 9 shows how the stress relaxation varies with the filler contents in the composites. In the absence of fillers there is a sudden drop in the stress within few seconds but as the holding time increases the stress decreases gradually with time. When the percentage of fillers increases, the stress takes more time to relax. This is due to the reason that the so-called free chains, which are mainly responsible for the viscous over-stress, will take a longer time to relax in the presence of fillers. From table 4, we can see that there is a fluctuation of the stress relaxation in the case of composites with fillers and no proper trend can be observed with a change in the percentage of their contents. In the case of oriented fillers, the BT dipoles are in an arranged way. Hence, oriented composites take even more time to go to a relaxed stress. From table 4, we can see that in the case of 15% oriented samples the stress relaxation is less as compared to others.

3.4. Scanning electron microscopy (SEM) of rapture surfaces

The amount of filler content has a strong influence on different mechanical properties for both types of Ecoflex-BT composites as described in earlier sections. However, the maximum amount of filler can be

Table 4: Stress in the beginning and after 2400 seconds of holding time for silicone composites of different filler contents

Sample wt %	Stress (kPa) at t= 0 sec	Stress (kPa) at t= 2400 sec	Stress Relaxation %
0%	121	32	74
5% Random	155	80	48
5% Oriented	180	95	47
10% Random	175	72	60
10% Oriented	190	92	51
15% Random	170	82	52
15% Oriented	210	120	42
20% Oriented	240	103	57

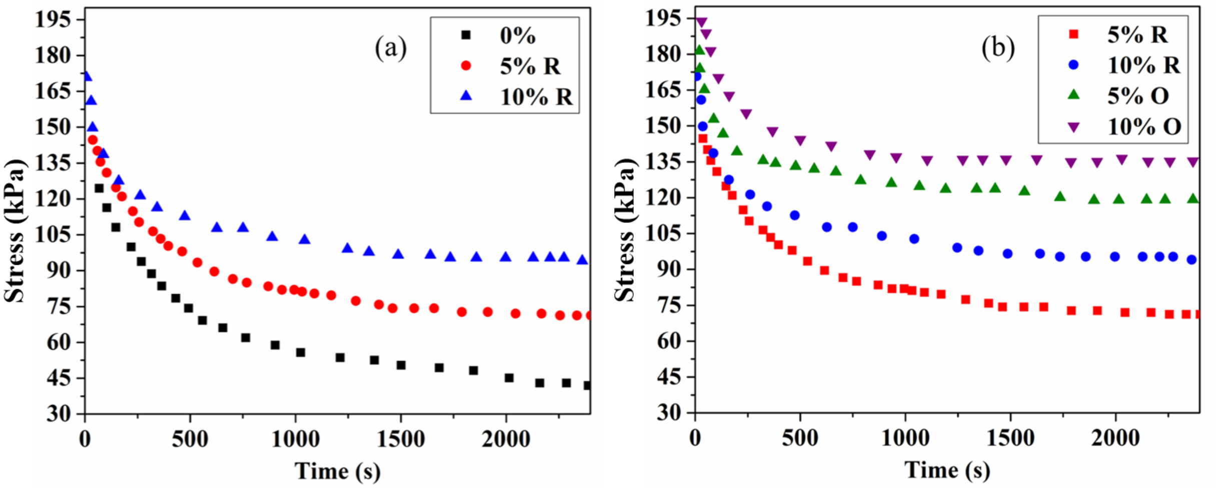

Figure 9: Stress relaxation curves of silicone composites at different percentage of filler contents (a) randomly filled Ecoflex-BT composites, (b) comparison of oriented Ecoflex-BT composites and randomly filled Ecoflex-BT composites

added is limited by the agglomeration of filler particles. The size and shape of particles also affect the agglomeration of them as explained in the literature [66, 67]. SEM images of rupture surfaces of different samples have been taken to visualize the agglomeration and alignment of filler particles for Ecoflex-BT composites cured in the absence of an electric field and in the presence of an electric field. SEM images of the cross section of the rupture surfaces of randomly distributed filled silicone composites are shown in figures 10 (a)-(d). We can see in these figures that there is no specific arrangement of the fillers in the matrix. As the filler content increases from 10% to 15%, agglomerations of particles can be seen clearly in figure 10(d), where BT particles in the polymer matrix make clusters. Figures 11 (a)-(c) illustrate the SEM images of the rupture surfaces of composites with oriented fillers. In these figures, we can see a circular contour type pattern due to an alignment of filler particles in a regular fashion. Thicker contour lines can be observed as the filler content increases. Further, the agglomeration of particles is diminished due to an orientation of dipoles of filler particles even though the filler content increases up to 20%.

3.5. *Dielectric properties of Ecoflex-BT composite*

Cubic crystalline $BaTiO_3$ (BT) particle at high temperature (above Curie temperature at 120°) is in cubic perovskite phase and paraelectric in nature [68, 69]. However, it transforms to ferroelectric structure with a tetragonal phase below the Curie temperature. As the operating temperature here is room temperature (25°),

Table 5: Summary and comparison of elastic modulus, failure stretch and dielectric constant

Silicone rubber	Filler	Filler content	Elastic modulus (MPa)		Failure stretch (%)		Dielectric constant (at 1000 Hz)	
Curing method			Random	Oriented	Random	Oriented	Random	Oriented
Sylgard 186 [40]	TiO_2	0 vol%	5.4	-	127	-	2.65	-
		2 vol%	3.1	-	150	-	3.50	3.9
		5 vol%	3.8	-	165	-	3.85	4.2
		8 vol%	6.1	-	145	-	4.10	4.8
		11 vol%	4.6	-	138	-	4.85	5.5
In house prepared [39]	BT	0 pph	0.095	-	2198	-	3.01	-
		5 pph	0.233	-	1196	-	3.90	-
Methylvinyl Silicone [38] (110-2)	BT	0 pph	0.250	-	478	-	3.2	-
		10 pph	0.275	-	440	-	3.4	-
		20 pph	0.290	-	435	-	3.6	-
		30 pph	0.305	-	400	-	3.9	-
Ecoflex 00-30 (This work)	BT	0 wt%	0.043±0.071	-	720±31	-	2.85±0.036	-
		5 wt%	0.078±0.094	0.065±0.063	592±25	850±26	3.00±0.039	3.20±0.025
		10 wt%	0.082±0.052	0.094±0.019	567±29	825±19	3.35±0.029	3.50±0.032
		15 wt%	0.084±0.049	0.104±0.086	514±28	756±22	3.65±0.021	4.15±0.033
		20 wt%	-	0.141±0.038	-	704±28	-	4.35±0.028

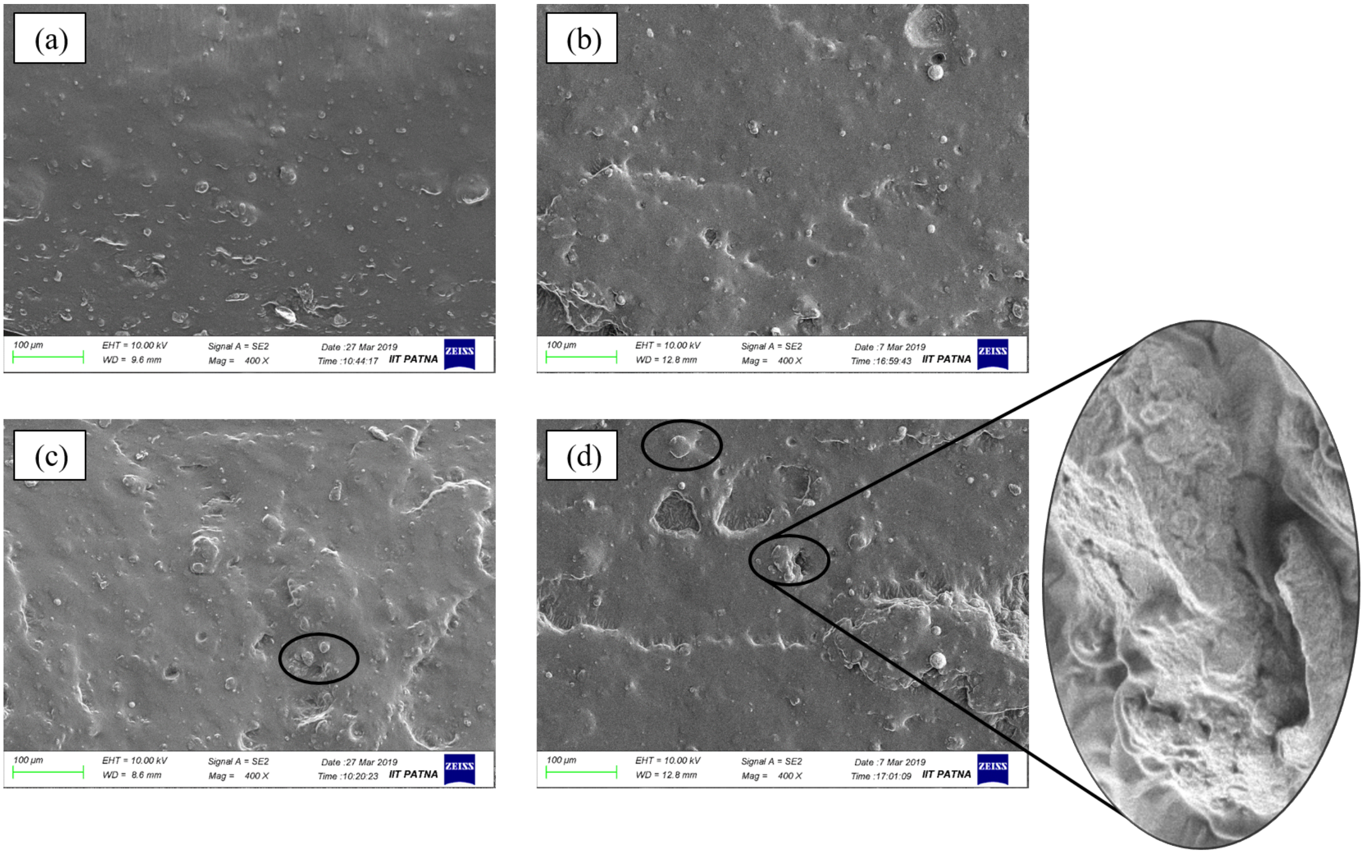

Figure 10: SEM images of fracture surface of Ecoflex composites filled with randomly distributed BT particles: (a) 0%, (b) 5%, (c) 10%, and (d) 15%. (agglomerations of filler particles can be seen with circles) and 400 magnification of agglomeration of particles

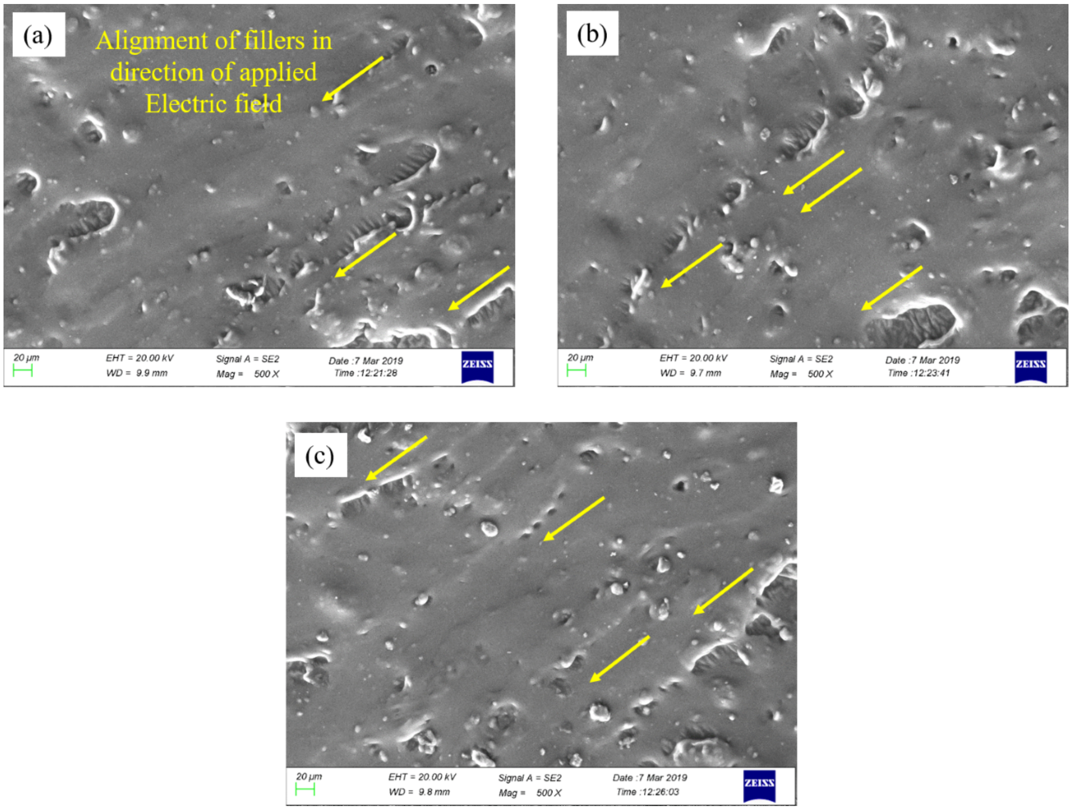

Figure 11: Morphology of fractured surfaces (a) 5% O, (b) 10% O, and (c) 15% O, yellow lines indicates alignment of fillers in the direction of applied electric filed

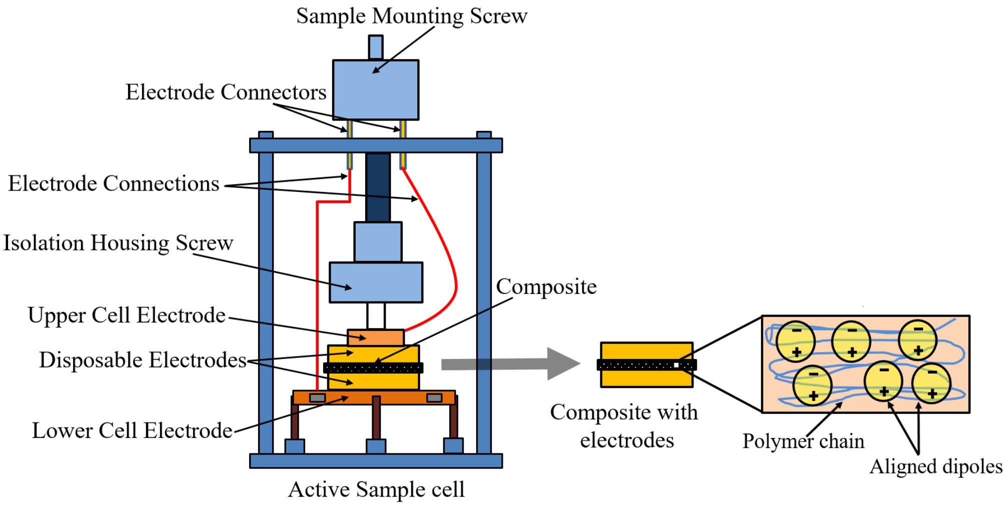

Figure 12: A schematic diagram that shows a sample preparation in a dielectric test

the BT particle is ferroelectric in nature. Hence, it has permanent dipole moment even in the absence of an electric field. It can be observed that ferroelectric barium titanate exhibits high dielectric constant [70]. When the ferroelectric ceramic particles of high dielectric constant are filled in polymer matrix, increase of dielectric constant of ferroelectric ceramic-polymer composite can be observed as explained by dielectric mixture models such as Maxwell-Garnett model, Lichtenecker model, Jayasundere and Smith model, etc [71, 72]. In the similar line the increase of dielectric constant of BT filled Ecoflex with increase of BT filler content can be explained. During the time of curing, the BT particles can move in the liquid silicone matrix and aligned in the direction of the applied electric field. Polymer chains after curing restrict the aligned dipoles to go back to their original positions. The differences in filler arrangements of the composites cured in the absence and presence of an electric field can be observed from the SEM images which were also explained by Kashani et al. [40] and Tomer et al. [55]. BT in the polymer matrix will also result in interfacial polarization which further increases the dielectric constant of Ecoflex-BT composites [70]. Figures 13(a) and 13(b) illustrate measured dielectric constants of composites with randomly distributed and oriented fillers, respectively. For a schematic diagram for the sample preparation in a dielectric test, see Figure 12. From figure 13 it is observed that the increasing percentage of filler content enhances the dielectric constant at all frequency ranges. On the other hand, the dielectric loss increases more with increasing filler concentration for the frequency range of 1 Hz to 10 kHz as compared to that of frequency range of 10 kHz to 1 MHz. This is because dipoles are unable to polarize with the rapid alteration of the electric field at a higher frequency. From figure 14, we can say that the frequency dependent dielectric losses are more in the randomly distributed silicone composite compared to oriented one. At a high frequency, the polarization of dipoles in the interface is less. Hence, there is a reduced loss at a high frequency, see Lim et al. [44]. In the case of oriented silicone composites, the dielectric behavior will be further enhanced as compared to the randomly distributed composites as shown in figure 13.

Note that Ecoflex-BT composites prepared during the curing process under an electric field help fillers to align in the direction of the applied field. Hence, there will be more electric fluxes which result in the enhancement of dielectric constants. Dipoles are aligned in the thickness direction that results in less translational and rotational motion of them which leads to a less dissipative polarization, i.e., interfacial polarization. Figures 15 (a) and (b) compare the dielectric constants and dielectric losses of both the silicone composites at 1 Hz frequency, respectively. We can see that the dielectric constants and dielectric losses improve significantly in the case of oriented filler composite as compared to the randomly one. To be specific, oriented composites with filler contents between 10 to 15% have high dielectric constant with minimum dielectric losses. Dielectric constants, elastic moduli, and failure stretches (%) of silicone composites prepared in this work are summarized and compared with other research works available in the literature in Table 5. Sylgard 186 used by Kashani et al. [40], on the one hand, has high dielectric constant but, on the other hand, has a high elastic modulus and a low failure stretch. The silicone composite prepared by Bele et al. [39] has a low elastic modulus and a high failure stretch but it has comparatively a low dielectric constant. Methylvinyl silicone has reasonably a high dielectric constant but it has a high elastic modulus and a low failure stretch [38]. The silicone composites prepared in this work has comparatively less stiffer with a high failure stretch and a high dielectric constant as compared to others. Although, it is frequently mentioned in the literature that an applied electric field will result in an anisotropic composite with a preferred direction. However, previous study by Kashani et al. [40], Tomer et al [55] and in the current study, we observed weak anisotropy which is in clear contrast with the magneto-active composites, where particle alignments are pronounced under an applied magnetic field during the curing process [73, 74]. This type of anisotropy observed in electro-active composites is frequently mentioned as a dispersion anisotropy/weak anisotropy/imperfect anisotropy. Very recently, Hosssain and Steinmann [4] developed an electro-elastic model for the dispersion type anisotropic composites. Table 6 shows percentage increase of dielectric constant values due to particle alignment under applied electric field in the present work and earlier works. It can be observed that the increase of dielectric constant is due to filler orientation is less and the maximum percentage increase in all these works lies in the range of 15-18%. Here the oriented composite direction is taken as 1-3 parallel composite (parallel to applied field direction) direction as mentioned in Tomer et al [55]. The value of dielectric constant of 0-3 random filled composite is almost equal to that of 1-3 perpendicular composite (perpendicular to applied electric field) direction [55]. Another reason for weak anisotropy of the filled elastomer composites can be the spherical shapes of the particles used in these works. The anisotropy of dielectric properties in the aligned filler in parallel direction of applied electric field can be increased significantly by increasing the aspect ratio of the filler as shown by Wang et al. [75] through finite element models. However, experimental verifications of the simulated results are not yet reported.

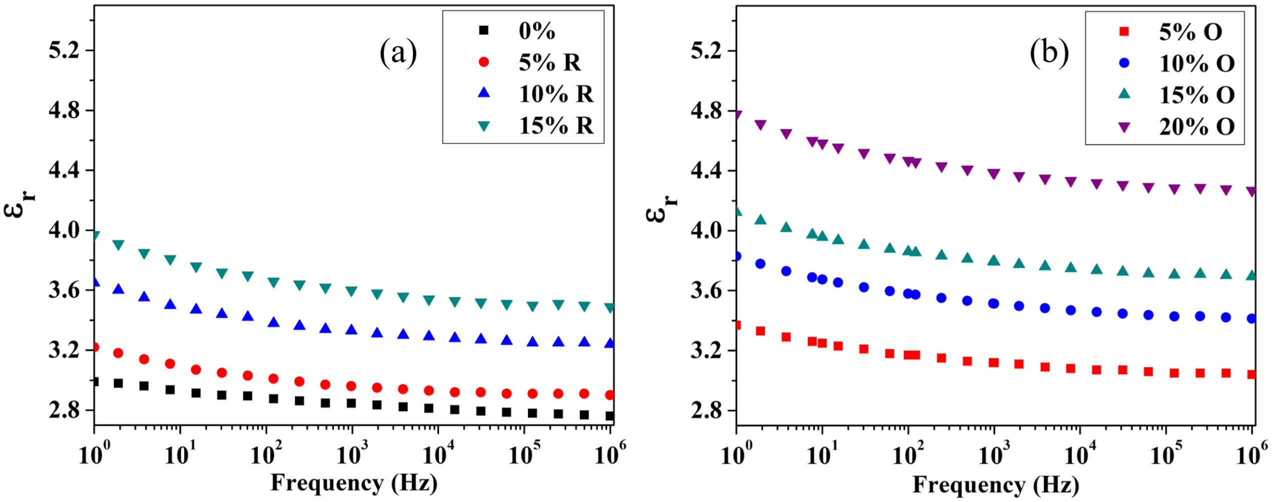

Figure 13: Dielectric constant of silicone composites filled with BT in, (a) randomly distributed, (b) oriented dipoles of fillers in silicone polymer matrix

3.6. Actuation strain measurements

In the previous sections, it has been demonstrated that Ecoflex composite with oriented BT filler has superior electro-mechanical properties. Now we want to conduct actuation experiments to test electro-mechanical performances of the manufactured Ecoflex-BT composites. For these, a circular-type actuator is prepared. These tests are conducted at different electric fields till the dielectric breakdown happens. As expected, the actuation strain of the active zone (electrode coated area) increases under the application of an electric field, see figure 16. The actuation strain is defined as the ratio of change in actuated area to the initial coated area which is calculated by measuring the area in terms of pixel using 'Imagej' image processing software. Figure 16 also shows that the initial coated area of 6132 pixel at 0 V increases to 7239 pixel area when the applied voltage reaches to 7 kV for the circular actuator made of a 15% oriented filler Ecoflex-BT composite. The calculated actuation strain in this case is 18%. Figure 17 shows that with an increase in the filler content, the actuation strain increases because it is directly proportional to the dielectric constant of materials. It was also observed that for the case of filled elastomer, actuation started at a lower voltage than that of unfilled Ecoflex samples. As the filler content increases from 10 to 15%, the stiffness of the silicone composites

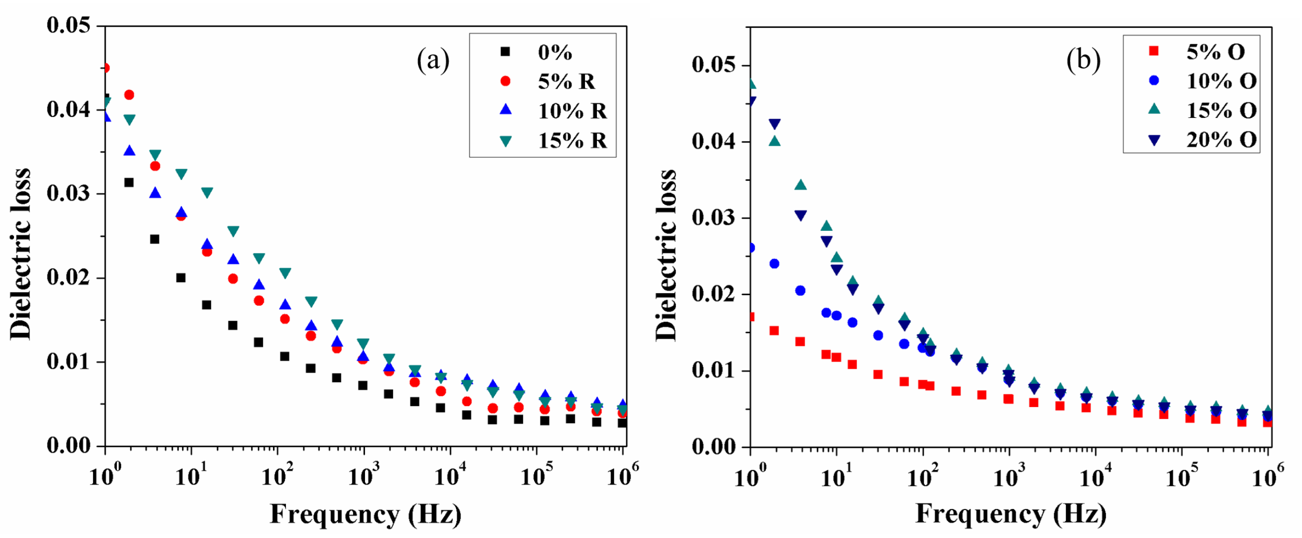

Figure 14: Dielectric loss of silicone composites with change in frequency from 1 Hz to 1MHz of: (a) randomly dispersed particles, (b) oriented filler composites

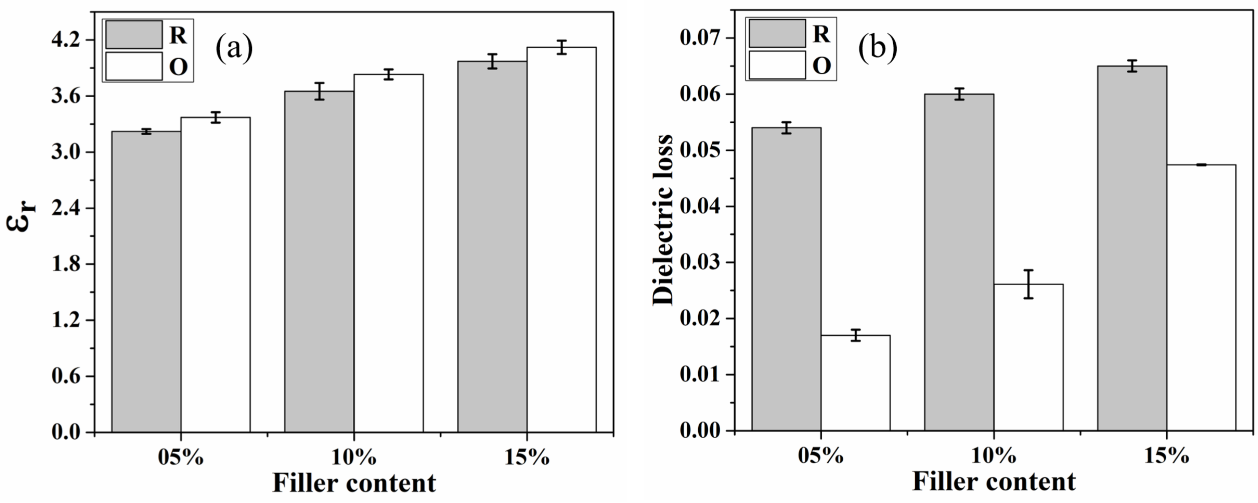

Figure 15: (a) Dielectric constant, (b) dielectric loss of silicone composites filled with randomly oriented (in the absence of an electric field) and oriented dipoles (in the presence of an electric field) at 1 Hz frequency.

increased that yielded a decrease in the rate of actuation strains. Furthermore, in the case of randomly filled composites, agglomeration of particles for 15% filler content occurs. This is also another reason for the decrease of rate of actuation strain with 15% filler content than that of 10% filler content. Overall, in the case of samples with oriented fillers, the actuation strain is more as compared to randomly distributed filler samples and the actuation strain is maximum in the case of Ecoflex composite with a 15% filler content. The actuation area at a particular electric field is summarized and compared with other works available in the literature in Table 7. In addition, we present a summary and comparison of dielectric breakdown strengths of various composites enhanced with BT particles in Table 8. It is vivid from the table 8 that the

Table 6: Percentage change in dielectric constant of different filled elastomer composites with oriented fillers in present work and earlier works

Silicone rubber	Filler	Filler content	% change in dielectric constant as result of filler orientation
Sylgard 186 [40]	TiO_2	2 vol%	9.70
		5 vol%	10.70
		8 vol%	10.90
		11 vol%	15.10
Sylgard 184 [55]	BT	5.0 vol%	9.33
		7.5 vol%	10.69
		10.0 vol%	11.70
		12.5 vol%	13.33
		15.0 vol%	17.67
Ecoflex 00-30 (This work)	BT	5 wt%	8.91
		10 wt%	10.47
		15 wt%	15.30

Table 7: Summary and comparison of some important parameters available in the literature : Different silicone matrices filled with BT particles

Silicone rubber	Filler	Filler content	Electric field (MV/m)	Actuated strain (planar area %)		Actuation improved %	
Curing method				Random	Oriented	Random	Oriented
Methylvinyl Silicone [38] (100-2)	BT	0 pph	30	3.8	-	-	-
		10 pph	30	4.9	-	-	-
		20 pph	30	5.3	-	-	-
		30 pph	30	4.7	-	-	-
Ecoflex 00-30 (This work)	BT	0 wt%	27.5	6.0±0.572	-	-	-
		5 wt%	27.5	7.5±0.657	7.8±0.572	25.0±0.88	30.0±2.61
		10 wt%	27.5	10.0±0.525	11.5±0.530	48.0±12.17	91.7±8.65
		15 wt%	27.5	5.5±0.326	18.0±0.524	-8.3∓3.05	200.0±18.13
		20 wt%	27.5	-	12.0±0.513	-	100.0±9.60

addition of BT filler to Ecoflex does not reduce the breakdown field significantly in compared to PDMS-BT composites. When we compare actuation results available in the literature for BT-filled elastomers, it was clear that the actuation area was more in the case of oriented filler silicone composites at a particular electric field as compared to the randomly distributed samples, see for example the works of Yang et al. [38]. In general, dielectric elastomers actuate at high voltages, i.e., in terms of several kilovolt (kV). Compared to other elastomers (VHB/acrylics, polyurethanes etc.) silicone elastomers actuate at high voltages because of their higher stiffness and very low dielectric permittivity. For instance, Opris et al. [76] observed actuation after 10 MV/m electric field. Another issue is that we did not prestretch the samples. Pre-stretching, on the one hand, enhances actuation. On the other hand, it reduces service life of a dielectric elastomer. Hence, our aim here is to observe actuation behaviour of Ecoflex-BT composites with any sort of pre-stretching.

4. Conclusion

In this contribution, our aim was to present a complete experimental characterization of two different types of Ecoflex-barium titanate composites to demonstrate which one is better in terms of electrical, mechanical, and electro-mechanical performances; one prepared by random fillers and another prepared by oriented fillers that are being prepared under an electric during the curing process. In order to quantify the influences

Table 8: Summary and comparison of dielectric breakdown strengths

Silicone rubber	BT Filler content	Dielectric breakdown strength (MV/m)	
		Random	Oriented
PDMS [77]	0 wt%	102.28	-
	5 wt%	5.76	-
	5 wt%	21.78	-
In house prepared [39]	0 pph	-	-
	1 pph	55.55	-
	2 pph	44.76	-
	5 pph	13.21	-
Ecoflex 00-30 (This work)	0 wt%	45.39±0.49	-
	5 wt%	38.79±0.62	40.62±0.32
	10 wt%	35.66±0.76	38.81±0.51
	15 wt%	31.48±0.59	34.57±0.72
	20 wt%	27.38±0.81	28.87±0.78

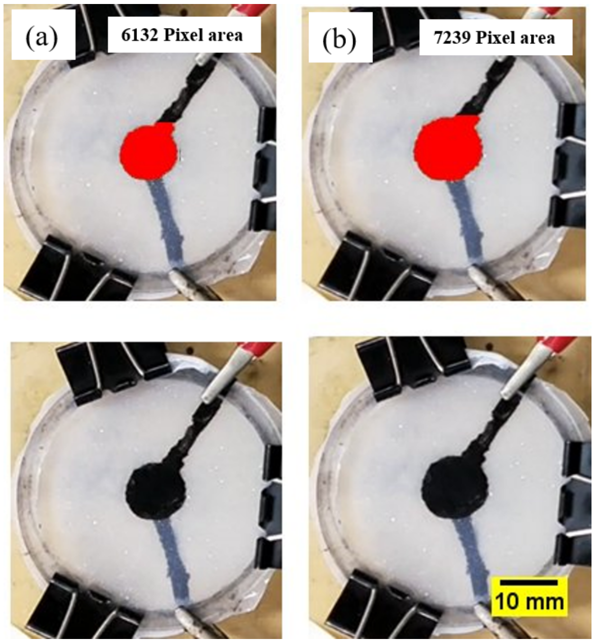

Figure 16: Oriented filler composite actuator sample with 15% filler content (unstretchered) for actuation experiments, (a) voltage off, (b) voltage on (7 kV)

of an electric field-induced curing, we have conducted a wide range of experiments, e.g., mechanical, dielectric, and electro-mechanical tests. With the help of various tests, we conclude that silicone composites cured in the presence of an electric field demonstrate better mechanical, dielectric, and electro-mechanical properties as compared to composite cured in the absence of an electric field. Furthermore, aligned filler particles due to an applied electric field during the curing process hinder agglomeration of particles near the percolation threshold. For example, the silicone composites filled with 15% filler content and cured in the presence of an electric field had a higher dielectric constant as compared to one cured in the absence of an electric field. In comparison to randomly filled composites, the elongation at break for aligned particles composite increases which is a desirable property for dielectric elastomer applications. When we prepare

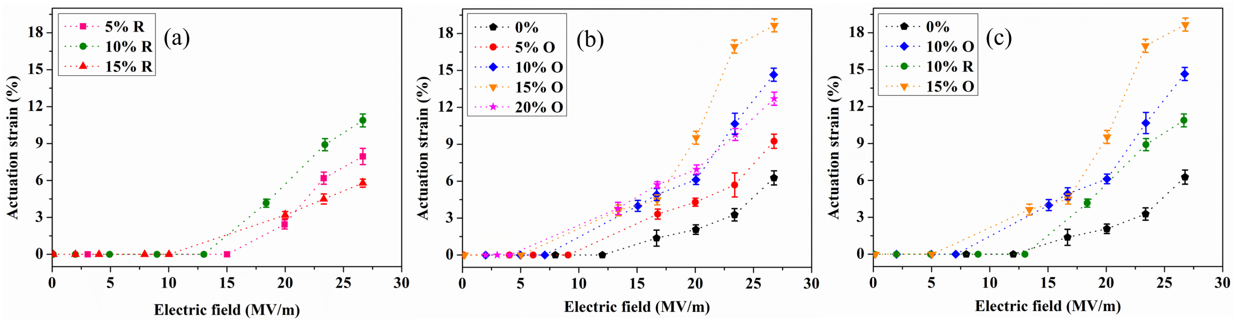

Figure 17: Actuation strain Vs applied electric field (a) random samples, (b) oriented samples, (c) maximum actuation and comparison between random and oriented samples

a circular actuator, it is observed that the actuation strain area is improved from 5.5% to 18% at a 15% filler loading for randomly filled and oriented dipole filled composites, respectively. These results indicate that silicone composites prepared by an electric field during the curing process have significant improved of various important properties. Note that barium titanate filler does not greatly improve dielectric permittivity of prepared composites. Recent literature [30, 31] demonstrate that carbon nanotube-based (CNTs) fillers massively improve dielectric permittivities. Therefore, experimental procedures established in this manuscript will be applied to carbon nanotube filled composites where materials will be cured under an electric field in a forthcoming contribution. This comprehensive experimental characterization technique provides a framework that will guide future dielectric composite preparations and electrical, mechanical, and electro-mechanical experiments especially for the oriented dipole fillers prepared under an electric field.

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Data Availability

The raw/processed data required to reproduce these findings cannot be shared at this time as the data also forms part of an ongoing study.

References

[1] A. O'Halloran, F. O'malley, P. McHugh, A review on dielectric elastomer actuators, technology, applications, and challenges, J. Appl. Phys. 104 (2008) 071101.

[2] Z. Suo, Theory of dielectric elastomers, Acta Mech. Solida Sin. 23 (2010) 549-78.

[3] M. Mehnert, M Hossain, P Steinmann, Numerical modelling of thermo-electro-viscoelasticity with field dependent material parameters, International Journal of Non-Linear Mechanics 106 (2018) 13-24.

[4] M Hossain, P Steinmann, Modelling electro-active polymers with a dispersion-type anisotropy, Smart Materials and Structures 27-20 (2018) 25010.

[5] R. Zhang, X. Huang, T. Li, P. Iravani, P. Keogh, Novel Arrangements for High Performance and Durable Dielectric Elastomer Actuation, Actuators 5 (2016) 20.

[6] S. C. B. Mannsfeld, B. C. K. Tee, R. M. Stoltenberg, C. V. H. H. Chen, S. Barman, B. V. O. Muir, A. N. Sokolov, C. Reese, Z. Bao Z, Highly sensitive flexible pressure sensors with microstructured rubber dielectric layers, Nat. Mater. 9 (2010) 859-864.

[7] A. T. Mathew, S. J. A. Koh, Maximum energy output of a two-phased self-priming dielectric elastomer generator, IEEE/ASME International Conference on Advanced Intelligent Mechatronics (July 2018) 1366-1371.

[8] R. I. Haque, P. A. Farine, D. Briand, Soft triboelectric generators by use of cost-effective elastomers and simple casting process, Sensors and Actuators A Phys 271 (2018) 88-95.

[9] F. Arena, L. Daniele, V. Fiamma, M. Fontana, G. Malara, G. Moretti, A. Romolo, G. P. Rosati Papini, A. Scial, R. Vertechy, Field Experiments on Dielectric Elastomer Generators Integrated on a U-OWC Wave Energy Converter. Ocean Renew. Energy, 37th ASME International Conference on Ocean, Offshore and Arctic Engineering. American Society of Mechanical Engineers Digital Collection (2018).

[10] P. Zanini, G. Rizzello, S. Seelecke, M. Homer, J. Rossiter, Self-sensing for robust automatic charge management of dielectric elastomer generators, Electroact. Polym. Actuators Devices 10594 52 (2018) International Society for Optics and Photonics.

[11] J. Huang, T. Lu, J. Zhu, R. D. Clarke, Z. Suo, Large, uni-directional actuation in dielectric elastomers achieved by fiber stiffening, Appl. Phys. Lett. 100 (2012) 211901.

[12] L. Duan, G. L. Wang, Y. Y. Zhang, Y. N. Zhang, Y. Y. Wei, Z. F. Wang, M. Zhang, High dielectric and actuated properties of silicone dielectric elastomers filled with magnesium-doped calcium copper titanate particles, Polym. Compos. 39 (2018) 691-697.

[13] M. Hossain, D. K. Vu, P. Steinmann, A comprehensive characterization of the electro- mechan-ically coupled properties of VHB 4910 polymer, Archive of Applied Mechanics 85(4) (2014) 523-537.

[14] Z. S. Liao, M. Hossain, X. Yao, M. Mehnert, P. Steinmann, On thermo-viscoelastic experimental characterisation and numerical modelling of VHB polymer, International Journal of Non-Linear Mechanics, 118 (2020) 103263.

[15] R. K. Sahu K. Patra, Rate-dependent mechanical behavior of VHB 4910 elastomer, Mech. Adv. Mater. Struct. 23 (2016) 170-179.

[16] Z. Suo, Mechanics of stretchable electronics and soft machines, MRS Bull. 37 (2012) 218-225.

[17] T. Vu-Cong, C. Jean-Mistral, A. Sylvestre, Impact of the nature of the compliant electrodes on the dielectric constant of acrylic and silicone electroactive polymers, Smart Mater. Struct. 21 (2012) 105036.

[18] J. Sheng, H. Chen, L. Liu, J. Zhang, Y. Wang, S. Jia, Dynamic electromechanical performance of viscoelastic dielectric elastomers, J. Appl. Phys 114 (2013) 134101.

[19] H. Liu, L. Zhang, D. Yang, Y. Yu, L. Yao, M. Tian M, Mechanical, Dielectric, and Actuated Strain of Silicone Elastomer Filled with Various Types of titanium oxides, Soft Materials 11 (2013) 363-370.

[20] A. Trols, A. Kogler, R. Baumgartner, R. Kaltseis, C. Keplinger, R. Schwodiauer, I. Graz, S. Bauer, Stretch dependence of the electrical breakdown strength and dielectric constant of dielectric elastomers, Smart Mater. Struct. 22 (2013) 104012.

[21] P. R. Dvornic, Silicon-containing polymers: the science and technology of their synthesis and applications, Thermal properties of polysiloxanes Netherland (Kluwer Academic Publishers) (2000) 185-212.

[22] R. Shankar, T. K. Ghosh, R. J. Spontak, Dielectric elastomers as next-generation polymeric actuators, Soft Matter 3 (2007) 1116-29.

[23] R. J. Nalbach, Design and characterization of polymer nanocomposites for engineering applications, Master's dissertation (Rowan University) (2018).

[24] S. Zhu, J. Guo, J. Zhang, Enhancement of mechanical strength associated with interfacial tension between barium titanate and acrylonitrile/butadiene rubber with different acrylonitrile contents by surface modification, J. Appl. Polym. Sci. 135 (2018) 2-11.

[25] C. Guo, M. Fuji, Effect of silicone coupling agent on dielectric properties of barium titanate/silicone elastomer composites, Adv. Powder Technol. 27 (2016) 1162-1172.

[26] J. Su, J. Zhang, Improvement of mechanical and dielectrical properties of ethylene propylene diene monomer (EPDM)/barium titanate) by layered mica and graphite flakes, Composites Part B: Eng. 112 (2017) 148-157.

[27] Y. Wang, J. Chen, Y. Li, Y. Niu, Q. Wang, H. Wang, Multilayered hierarchical polymer composites for high energy density capacitors, J. Mater. Chem. A 7 (2019) 2965-80.

[28] C. Ellingford, C. Wan, L. Figiel, T. Mcnally, Mechanical and dielectric properties of MWCNT filled chemically modified SBS/PVDF blends, Composites Communications 8 (2018) 58-64.

[29] A. Poudel, P. Walsh, J. Kennedy, K. Thomas, J.G. Lyons J G, A. Coffey, Thermal, mechanical, dielectric, and morphological study of dielectric filler-based thermoplastic nanocomposites for electromechanical applications, J. Thermoplast. Compos. Mater. 32 (2018) 178-204.

[30] C. Ellingford, C. Bowen, T. MnNally, C. Wan, Intrinsically Tuning the Electromechanical Properties of Elastomeric Dielectrics: A Chemistry Perspective, Macromolecular Rapid Communications 39 (18) (2018) 1800340.

[31] F. B. Madsen, A. E. Daugaard, S. Hvilsted, A. L. Skov, The Current State of Silicone-Based Dielectric Elastomer Transducers, Macromolecular Rapid Communications 37(5) (2016) 378-413.

[32] S. Geng, P. Wang, T. Ding, Impedance characteristics and electrical modelling of multi-walled carbon nanotube/silicone rubber composites, Composites Science and Technology 72 (2011) 36-40.

[33] J. Saji, A. Khare, S. P. Mahapatra, Impedance analysis, dielectric relaxation, and electrical conductivity of multi-walled carbon nanotube-reinforced silicon elastomer nanocomposites, Journal of Elastomers and Plastics 47 (2013) 394-415.

[34] Z. Zeng, H. Jin, L. Zhang, H. Zhang, Z. Chen Z, F. Gao, Z. Zhang, Low-voltage and high-performance electrothermal actuator based on multi-walled carbon nanotube/polymer composites, Carbon 84 (2019) 327-334.

[35] C. Tan, J. Gao, Y. Li, L. Qing, X. Zhang, Y. Yang, Development of a processing method for carbon nanotubes modified fluorosilicone rubber with enhanced electrical, dielectric and mechanical properties, Polym. Technol. Mater. 00 (2019) 1-12.

[36] K. C. B. Naidu, V. N. Reddy, T. S. Sarmash, D. Kothandan, T. Subbarao, N. S. Kumar, Structural , morphological , electrical , impedance and ferroelectric properties of $BaO-ZnO-TiO_2$ ternary system, J. Aust. Ceram. Soc. 11 (2018).

[37] A. Kumar, D. Ahmad, K. Patra, Barium titanate particle filled silicone elastomer composite: Preparation and evaluation of morphology and mechanical behaviour, IOP Conf. Series: Journal of Physics: Conf. Series 1240 (2019) 012049.

[38] D. Yang, S. HuanG, M. Ruan, S. Li, Y. Wu, W. Guo, L. Zhang, Improved electromechanical properties of silicone dielectric elastomer composites by tuning molecular flexibility, Compos. Sci. Technol. 155 (2018) 160-168.

[39] A. Bele, M. Cazacu, G. Stiubianu, S. Vlad, M. Ignat, Polydimethylsiloxane-barium titanate composites: Preparation and evaluation of the morphology, moisture, thermal, mechanical and dielectric behavior, Composites Part B: Eng. 68 (2015) 237-245.

[40] M. Razzaghi Kashani, S. Javadi, N. Gharavi, Dielectric properties of silicone rubber-titanium dioxide composites prepared by dielectrophoretic assembly of filler particles, Smart Mater. Struct. 19 (2010) 035019.

[41] L. Jiang, A. Betts, D. Kennedy, S. Jerrams, Improving the electromechanical performance of dielectric elastomers using silicone rubber and dopamine coated barium titanate, Mater. Des. 85 (2015) 733-742.

[42] M. Iijima, N. Sato, I. Wuled Lenggoro, H. Kamiya, Surface modification of $BaTiO_3$ particles by silane coupling agents in different solvents and their effect on dielectric properties of $BaTiO_3$/epoxy composites, Colloids Surfaces A Physicochem. Eng. Asp. 352 (2009) 88-93.

[43] A. Goshkoderia, S. Rudykh, Electromechanical macroscopic instabilities in soft dielectric elastomer composites with periodic microstructures, European Journal of Mechanics -A/Solids 65 (2017) 243-256.

[44] K. G. Lim, K. H. Chew, Tuning of polarization, internal electric field, and hysteresis loop behaviours in $BaTiO_3/Ba_xSr_{1-x}TiO_3$ superlattices, J. Appl. Phys. 123 (2018) 104101-10.

[45] F. Madidi, G. Momen, M. Farzaneh, Dielectric Properties of TiO_2/Silicone Rubber Micro- and Nanocomposites, Adv. Mater. Sci. Eng. 7 (2018).

[46] Y. Yao, G. Q. Lu, D. Boroyevich, K. D. T. Ngo, Effect of Al_2O_3 fibers on the high-temperature stability of silicone elastomer, Polymer 55 (2014) 4232-4240.

[47] H. Lu, F. Liang, J. Gou, Nanopaper enabled shape-memory nanocomposite with vertically aligned nickel nanostrand: controlled synthesis and electrical actuation, Soft Matter, 7 (2011) 7416-7423.

[48] M. Monti, M. Natali, L. Torre, J. M. Kenny, The alignment of single walled carbon nanotubes in an epoxy resin by applying a DC electric field, Carbon, 50 (2012) 2453-2464.

[49] R. Su, J. K. Tseng, M. S. Lu, M. Lin, Q. Fu, L. Zhu, Ferroelectric behavior in the high temperature paraelectric phase in a poly(vinylidene fluoride-co-trifluoroethylene) random copolymer, Polymer, 53 (2012) 728-739.

[50] P. Gupta, M. Rajput, N. Singla, V. Kumar, D. Lahiri, Electric field and current assisted alignment of CNT inside polymer matrix and its effects on electrical and mechanical properties, Polymer, 89 (2016) 119-127.

[51] E. Cholleti, J. Stringer, M. Assadian, V. Battmann, C. Bowen, K. Aw, Highly Stretchable Capacitive Sensor with Printed Carbon Black Electrodes on Barium Titanate Elastomer Composite, Sensors 19 (2019) 42-12.

[52] C. O. Blattmann, S. E. Pratsinis, Nanoparticle Filler Content and Shape in Polymer Nanocomposites, KONA powder and particle Journal 36 (2019) 1-30.

[53] S. Kim, G. P. Khanal, H. W. Nam, M. Kim, I. Fujii, S. Ueno, C. Moriyoshi, Y. Kuroiwa, S. Wada, In-situ electric field induced lattice strain response observation in BiFeO3-BaTiO3 lead-free piezoelectric ceramics J. Ceram. Soc. Japan 126 (2018) 316-320.

[54] R. Ui, F. Levassort, M. Lematre, L. P. Tran-Huu-Hue, M. Pham-Thi, Internal Electric Field in Co-Doped $BaTiO_3$ With $Co^{2+/3+}$, Nb^{5+}, Li^{+}, and F^{-}: Impact on Functional Properties and Charge Compensation With Niobium and Fluorine Ions, IEEE Trans. Ultrason. Ferroelectr. Freq. Control 66 (2019) 154-162.

[55] V. Tomer, C. A. Randall, G. Polizos, J. Kostelnick, E. Manias, High-and low-field dielectric characteristics of dielectrophoretically aligned ceramic/polymer nanocomposites, Journal of Applied Physics 103 3 (2008) 034115.

[56] M. A. Keip, P. Steinmann, J. Schroeder, Two-scale computational homogenization of electro-elasticity at finite strains, Computer Methods in Applied Mechanics and Engineering, 278 (2014) 62-79.

[57] R. Bustamante, L. Dorfmann, R. W. Ogden, On electric body forces and Maxwell stresses in nonlinearly electroelastic solids, International Journal of Engineering Science, 47(11-12) (2009) 1131-1141.

[58] R. Mao, B. Wu, E. Carrera, W. Chen, Electrostatically tunable small-amplitude free vibrations of pressurized electro-active spherical balloons, International Journal of Non-Linear Mechanics, 117 (2019) 103237.

[59] Smooth-On Ecoflex™Series https://www.smooth-on.com/productline/ecoflex/(Accessed on 23 March 2019).

[60] Z. S. Liao, M. Hossain, X. Yao, R. Navaratne, G. Chagnon. A comprehensive thermo-viscoelastic experimental investigation of Ecoflex polymer, Polymer Testing, 86 (2020) 106478.

[61] Z. S. Liao, M. Hossain, X. Yao, Ecoflex polymer of different Shore hardnesses: Experimental investigations and constitutive modelling, Mechanics of Materials, 144 (2020) 103336.

[62] D. Yang, F. Ge, M. Tian, N. Ning, L. Zhang, C. Zhao, K. Ito, T. Nishi, H. Wang, Y. Luane, Dielectric elastomer actuator with excellent electromechanical performance using slide-ring materials/barium titanate composites, J. Mater. Chem. A 3 18 (2015) 9468-9479.

[63] L. Jiang, A. Betts, D. Kennedy, S. Jerrams, The fabrication of dielectric elastomers from silicone rubber and barium titanate: employing equi-biaxial pre-stretch to achieve large deformations, J. Mater. Sci. 50 24 (2015) 7930-7938.

[64] P. S. Larsen, G. Kofod, M. H. Shridhar, M. Benslimane, P. Gravesen, Performance of dielectric elastomer actuators and materials, In Smart Structures and Materials Electroactive polymer actuators and devices (EAPAD) Vol. 4695 pp (2002) 158-166 International Society for Optics and Photonics. Larsen P S, Kofod G, Shridhar M H, Benslimane M and Gravesen P 2002 July.

[65] E. Biddiss E, T. Chau, Dielectric elastomers as actuators for upper limb prosthetics: Challenges and opportunities, Medical engineering and physics 30 4 (2008) 403-418.

[66] Z. Yang, H. Peng, W. Wang, T. Liu, Crystallization behavior of poly(ϵ-caprolactone)/layered double hydroxide nanocomposites, Journal of applied polymer science 116 5 (2010) 2658-2667.

[67] M. A. Ashraf, W. Peng, Y. Zare, K. Y. Rhee, Effects of size and aggregation/agglomeration of nanoparticles on the interfacial/interphase properties and tensile strength of polymer nanocomposites, Nanoscale research letters 13 1 (2018) 214.

[68] S. Saha, T. P. Sinha, A. Mookerjee, Electronic structure, chemical bonding, and optical proper-ties of paraelectric $BaTiO_3$, Physical Review B 62 13 (2000) 8828.

[69] C. Ang, A. S. Bhalla, L. E. Cross, Dielectric behavior of paraelectric $KTaO_3, CaTiO_3$, and $(Ln_{1/2}Na_{1/2})$ TiO_3 under a dc electric field, Physical review b 64 18 (2001) 184104.

[70] G.H.Haertling, Ferroelectric ceramics: history and technology, Journal of the American Ceramic Society, 82 4 (1999) 797-818.

[71] B. Luo, X. Wang, Q. Zhao, L. Li, Synthesis, characterization and dielectric properties of surface functionalized ferroelectric ceramic/epoxy resin composites with high dielectric permittivity. Composites Science and Technology 112 (2015) 1-7.

[72] L. K. Namitha, M. T. Sebastian, High permittivity ceramics loaded silicone elastomer composites for flexible electronics applications. Ceramics International 43 3 (2017) 2994-3003.

[73] E. Coquelle, G. Bossis, D. Szabo, F. Giulieri, Micromechanical analysis of an elastomer filled with particles organized in chain-like structure. J Mater Sci 41 (2006) 5941-5953.

[74] K. Danas, S. V. Kankanala, N. Triantafyllidis, Experiments and modeling of iron-particle-filled magnetorheological elastomers, J Mech Phys Solids 60 1 (2012) 120-138.

[75] Z. Wang, J. K. Nelson, H. Hillborg, S. Zhao, L. S. Schadler, Dielectric constant and breakdown strength of polymer composites with high aspect ratio fillers studied by finite element models. Composites science and technology, 76 (2013) 29-36.

[76] D. M. Opris, M. Molberg, C. Walder, Y. S. Ko, B. Fischer, F. A. Nueesch, New Silicone Composites for Dielectric Elastomer Actuator Applications In Competition with Acrylic Foil, Advanced Functional Materials, 21 (2011) 3531-3539.

[77] A. Bele, M. Cazacu, G. Stiubianu, S. Vlad, Silicone-barium titanate composites with increased electromechanical sensitivity. The effects of the filler morphology, RSC Advances, 4 (2014) 58522-58529.