Enhancement of Transduction Performance of a Dielectric Elastomer Actuator Based on Acrylonitrile Butadiene Rubber

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Abstract: The synthetic elastomer based on acrylonitrile butadiene rubber (NBR) has been recently developed as a kind of dielectric elastomer actuators (DEAs). Its advantages are that its properties can be modified according to the requirements of applications. In this paper, we report a comprehensive study on the effects of plasticizers including diisodecyl phthalate (DIDP), dioctyladipate (DOA), and dioctyl phthalate (DOP) as well as a high dielectric additive, which is, barium titanate ($BaTiO_3$) with respect to the overall performance of the synthetic elastomer. We investigate how the significant parameters representing the actuator performance can be changed according to the composition of the additives and how the actuation performances can be improved. In addition, we address how the environmental conditions such as exposure to the light and the air have influence on the properties of the synthetic elastomer, and a method for extending the lifetime of DEA is also addressed.

Keywords: transduction, dielectric elastomer actuator, acrylonitrile butadiene rubber.

Introduction

Electro-active polymers (EAPs) are smart materials with unique properties, changing their shape or size in response to external stimuli. Dielectric elastomers (DEs) have recently attracted particular interest due to their dominant properties such as large deformation, large force, high efficiency, light weight, low cost and simple manufacture.[1-3] In fact, there are a number of commercial materials that can be used for dielectric elastomers, such as polyurethane, acrylic elastomer, and silicone rubber.[4-7] Due to the outstanding properties of dielectric elastomer materials, they are now applied to a wide variety of fields such as actuators and sensors for mechatronics and robotics systems, artificial muscles, biomimetics, and medical and haptic devices.[8] However, according to the requirements of applications, the properties of commercial materials sometimes do not satisfy these requirements, so modifying and adapting these properties is necessary.

Up to now, there have been many studies on the electrical and mechanical properties of dielectric elastomer materials,[11-13] as well as how to improve their properties to meet the requirements of applications.[14,15] Among the previous methods, using additives has been reported as an effective way in numerous studies.[16-19] In this paper, we discuss how

to improve the overall performance of dielectric elastomer actuators (DEAs) based on synthetic rubber material, by using plasticizers such as diisodecyl phthalate (DIDP), dioctyl adipate (DOA), dioctyl phthalate (DOP), and a dielectric additive, barium titanate ($BaTiO_3$).[9,10] Through analyzing the results of electrical and mechanical properties of synthetic rubber materials, a suitable plasticizer and optimized fraction are chosen to produce the best performance of DEAs.

The operational principle of a DEA is based on the compressive affect of the electrostatic force.[7] As shown in Figure 1, when a voltage is applied on dielectric elastomer film coated with two compliant electrodes on both sides, the film will be compressed in the thickness direction and expanded in the lateral.

The effective pressure induced by the electrostatic force, called the Maxwell stress, is calculated as follows:

$$p = \varepsilon_0 \varepsilon E^2 = \varepsilon_0 \varepsilon \left(\frac{V}{d}\right)^2 \tag{1}$$

where ε_0 and ε are the permittivity of free space (8.85×10^{-12} F/m) and relative permittivity or dielectric constant of the elastomer, respectively. V is the applied voltage, d is the thickness of the film, and E is the electric field acting upon it.

To evaluate the actuation performance of the DEA, the strain of the film in the thickness direction is obtained by:
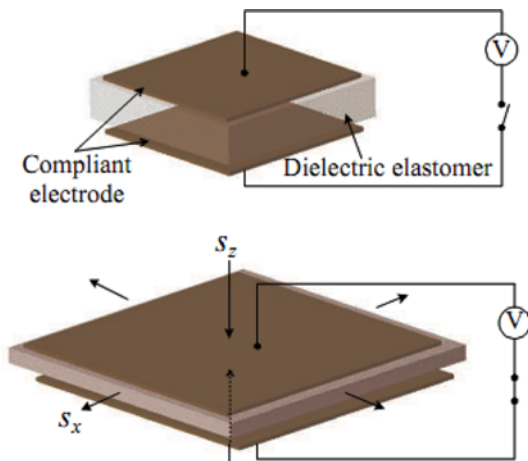
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Figure 1. The operational principle of dielectric elastomer material.

$$\delta_z = -\frac{p}{Y} = -\frac{\varepsilon_0 \varepsilon E^2}{Y} = -\left(\frac{\varepsilon_0 \varepsilon}{Y}\right)\left(\frac{V}{d}\right)^2 \qquad (2)$$

where Y is the Young's modulus of the dielectric elastomer material. From eq. (2), to improve the performance of the actuator, we need to improve three properties of the material including the relative permittivity, dielectric breakdown strength, and Young's modulus. In the same electrical field, the DEA will produce large deformation if its relative permittivity is higher or its Young's modulus is smaller. However, in the case that the elastomers have the same dielectric constant and Young's modulus, the higher breakdown strength will produce more deformation for the DEA. By optimizing these properties of dielectric elastomer material, we can produce desirable performance of the actuators that satisfies the requirements of applications.

As mentioned in previous work,[9] the viscosity of the elastomer reduces the stability and reliability of the DEA and should be minimized, in order to prevent hysteresis and relaxation. Dielectric loss is also an important parameter to clarify the possible dissipative effect of dielectric elastomer material. It reflects the loss of energy that goes into heating the material in an alternative electric field. Higher dielectric loss corresponds to more dissipated energy. As a result, the material will be hot and easily broken, so the loss factor should be minimized to improve the performance of the DEA.

Experimental

Additives for Synthetic Elastomer. As reported in previous work,[8] acrylonitrile butadiene rubber (NBR) can be considered as a promising candidate material for dielectric elastomers because its characteristics of high relative permittivity, small elastic modulus and low viscosity. Moreover, the mechanical and electrical properties of NBR can be changed by adding acrylonitrile, vulcanization agents, and modifying the conditions of vulcanization as well as the types and contents of additives.[20] Using additives is the most significant method to improve the properties of NBR material without adjusting the other factors.

In general, plasticizer and high dielectric filler are often used as additives to change the viscosity, modulus and dielectric constant of materials. The commercial plasticizers used are classified into three types: phthalates, adipates, and trimellitates. DOP is a common plasticizer accounting for greater than 50% of all the plasticizers used.[10]

In the present work, we use DOP, DIDP and DOA as plasticizers and evaluate their effects. $BaTiO_3$ with a relative permittivity of more than 1000[22] is selected as the high dielectric filler to improve the dielectric constant of the synthetic rubber. Furthermore, the effects of this inorganic powder on elastic modulus and breakdown strength of the material are also studied.

Material Preparation and Processing. The fabrication process to produce DEA based on NBR material consists of several steps. In the first, off-the-shelf NBR (LG Chem Co.) is dissolved in a solvent (Toluene) and mixed with vulcanizing agents and plasticizers using a stirrer. Then, $BaTiO_3$ powder is dispersed in the same solvent using a sonicator. The solution is mixed with the NBR compound and stirring is continued to achieve uniformity. Next, impurities or undissolved lumps in the solution are filtered out. Air bubbles in the material are removed by using a centrifugal bubble eliminator and the solution is dispensed to make a film using the robotic system shown in Figure 2.[21]

As the electrode material for measuring the performance of actuators, a mixture of graphite and carbon black particles dispersed in Ethanol solvent is used. The solution of the electrode material is then sprayed on the synthetic rubber film using an air spraying system integrated into the robotic system. The desired pattern of the electrode layers is easily obtained using a mask. The minimum thicknesses of the dispensed film and the sprayed electrode layer are 20 and 5 μm, respectively.
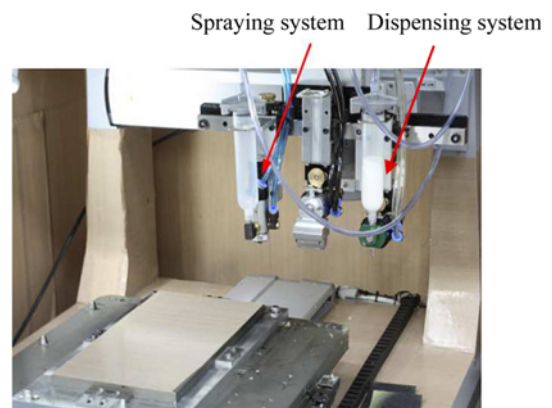

Figure 2. The automatic dispensing and spraying system.

The dispensed elastomer film is then kept in a 60 ºC oven for 48 h of aging to dry the solvent off. This process is very much important because the residual solvent inside the elastomer has a negative effect on the breakdown strength of actuators. To accelerate the evaporation of the solvent, heating the elastomer is suggested during dispensing. Finally, the synthetic elastomer is produced after curing in a 170 ºC oven.

Experimental Setup. Three kinds of experiments were conducted. First, different synthetic elastomers were prepared with different plasticizers including DOP, DIDP, and DOA, with different contents ranging from 0 to 100 phr (proportion by weight with respect to 100 parts of NBR used). The fractions between NBR and plasticizers in these materials are listed in the Table I.

The samples were used for testing the stress-strain properties and breakdown voltage. Based on these comparisons, one plasticizer was chosen for the next experiments.

After a suitable plasticizer was chosen, 0, 10, 20, 30, and 40 phr amounts of $BaTiO_3$ (Sigma-Aldrich Co.) were added to the synthetic rubber material. To verify the effect of $BaTiO_3$ on NBR, the performance of the synthetic rubber was evaluated with respect to its Young's modulus, stress relaxation, relative permittivity, electrical breakdown strength and electromechanical performance.

The Young's modulus and stress relaxation tests were measured with a material testing machine (Lloyd Co.). All specimens in the test have the same dimensions of 100-mm length, 15-mm width and 0.2-mm thickness. In Young's modulus tests, the specimens were extended to 100% strain with a constant rate of 50 mm/min, and tensile stress was automatically calculated by the machine. In the stress relaxation test, after the specimens were elongated to a predicted load, their positions were steadily maintained for 60 s. The relative permittivity of each specimen was measured by a dielectric spectrometer (DES100 from SEICO INST.) with a frequency range of 10 Hz to 10 kHz. To measure the electrical breakdown strength, we applied an electrical voltage
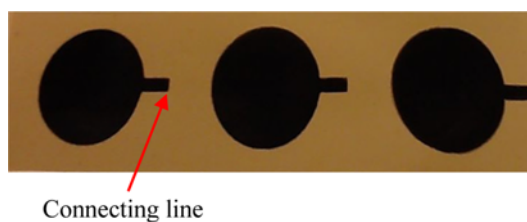

Figure 3. The specimens for the electrical breakdown strength measurement.

to a synthetic NBR film with a 200-µm thickness. Both sides of the film were covered by two compliant electrodes made by spraying the aforementioned electrode solution. Figure 3 shows the specimens for the test. The 1-Hz sinusoidal voltage is increased by ΔV=100 V every 10 s until the film reaches its breakdown strength voltage. The electric field strength is computed by the quotient between breakdown strength voltage and the thickness of the film.

The experimental setup to measure the electromechanical performance of the synthetic rubber material is shown in Figure 4.[10] We can indirectly calculate the thickness strain according to relation with the radial strain of the film. As shown in Figure 4, the synthetic elastomer specimen with 200-µm thickness is clamped between two annular rigid plastic frames with an inner diameter of 50 mm and a width of 5 mm. Both sides of the specimen have two circular electrode layers that have the same diameter as the frames. These electrode layers were made by brushing carbon black (Ketjenblack EC-600JD) powders on the films. A tiny mass is placed on the specimen at the center of the frame and we use a laser sensor (LK-G152 Keyence) to measure the displacement of the sample when it is activated. From that, the radial expansion of the film can be calculated as follows:

$$\Delta R = \sqrt{d^2 + R^2} - R \tag{3}$$

where ΔR, d, and R are the radial expansion, the displacement of the mass, and the inner radius of the frame, respectively. Hence, the radial strain is calculated by:
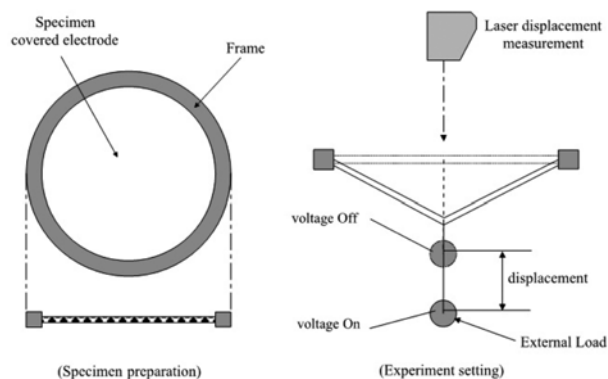
Table I. Different Samples of Synthetic Elastomers with Different Types and Fractions of Plasticizers (Unit: phr)

Samples	NBR	Plasticizer		
		DOA	DIDP	DOP
1	100	100		
2	100		100	
3	100			100
4	100	50	50	
5	100	50		
6	100		50	
7	100			50
8	100	25	25	

Figure 4. Schematic of the experimental setup to measure the thickness strain.
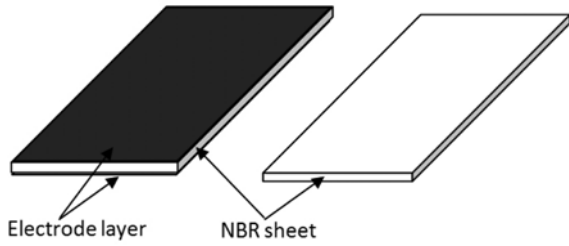
Figure 5. The samples for testing the effect of different aging conditions.

$$\delta_r(\%) = \frac{\Delta R}{R} \times 100 \qquad (4)$$

The actuation performance of different synthetic rubbers is verified based on this radial strain. The applied voltage in this experiment is the same one described in the electrical breakdown strength testing.

Another experiment was done to evaluate environmental effects such as oxygen and light. This experiment includes the two following steps:

· Step 1: we prepared two samples as shown in Figure 5. One is a synthetic elastomer sheet, 45 mm wide, 90 mm long and 0.2 mm thick. It was covered on both sides by two electrode layers made by spraying electrode material. The other sample is in the same way, but both its sides are bear without the electrode layers.

· Step 2: These samples were kept at room temperature with four different conditions: air and strong light, air and no-light, strong light in a vacuum, and no-light in the vacuum. After ten days, the surface condition and stress-strain properties of the samples were checked.

Results and Discussion

Effects of Plasticizers. The following figures describe the effect of different plasticizers on the stress-strain properties and electrical breakdown strength of the synthetic NBR.

As shown in Figure 6(a), the synthetic elastomer becomes softer when the amount of plasticizers increases. At the content of 100 phr of the DOP plasticizer, the sample reaches the largest strain (250%) and the tensile stress is the smallest one among the curves shown in the figure at the same percentage of strain. The stress is nearly half of the sample using 100 phr of DOA or DIDP. The electrical breakdown strength of these samples at 100 phr of the plasticizers is almost the same. Moreover, when a large amount (100 phr) of DOA or DIDP is used, a thin oil layer is formed on the surfaces of the samples after curing. This is due to the fact that the synthetic elastomer cannot keep such a high amount of plasticizers in the film. Therefore, since the main purpose of plasticizer is making the material softer, DOP is chosen as the plasticizer of the synthetic elastomer.

Effects of BaTiO₃. As mentioned previously, the relative
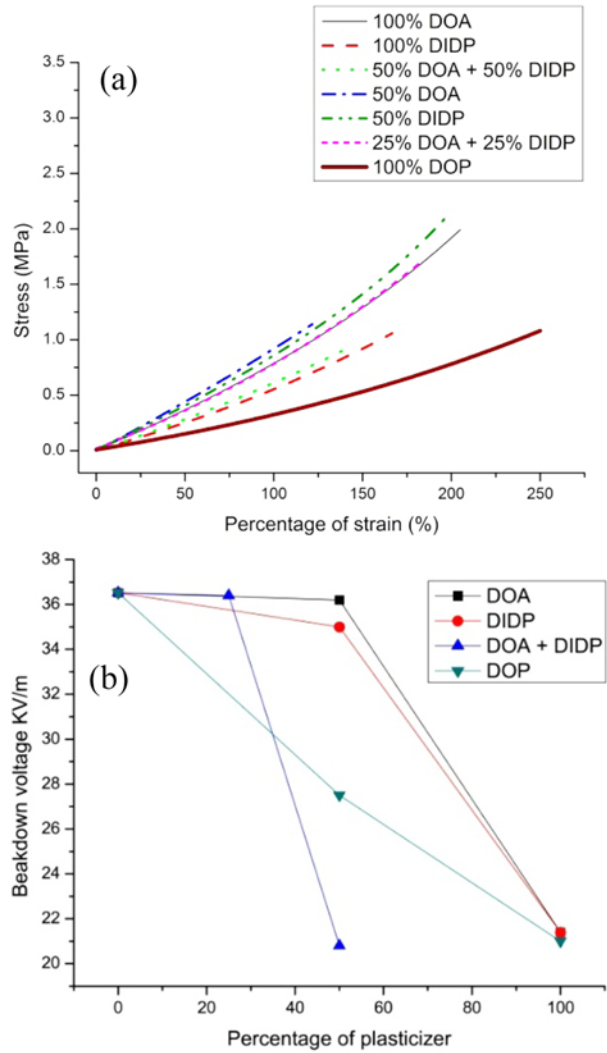

Figure 6. The stress-strain curves (a) and electrical breakdown strength (b) of synthetic NBR with different contents of plasticizers.

permittivity, the electrical breakdown strength and the Young's modulus are three important parameters to the overall actuation performance of the dielectric elastomer actuator. Consequently, these properties are investigated in the following subsections. The result of the actuation performance of the synthetic elastomer actuator will be addressed.

Relative Permittivity. According to the Experimental section, the effect of $BaTiO_3$ on the relative permittivity of the synthetic elastomer is the most expected one when it is added to the proposed material. Figure 7(b) shows the improvement of the relative permittivity when the content of $BaTiO_3$ increases from 0 to 40 phr. Greater content of $BaTiO_3$ resulted in higher relative permittivity of the material. From Figure 7(a), when the frequency increased, the relative permittivity of the proposed material decreased. This means that the actuation response of the synthetic elastomer actuators will be enhanced, but its deformation will be
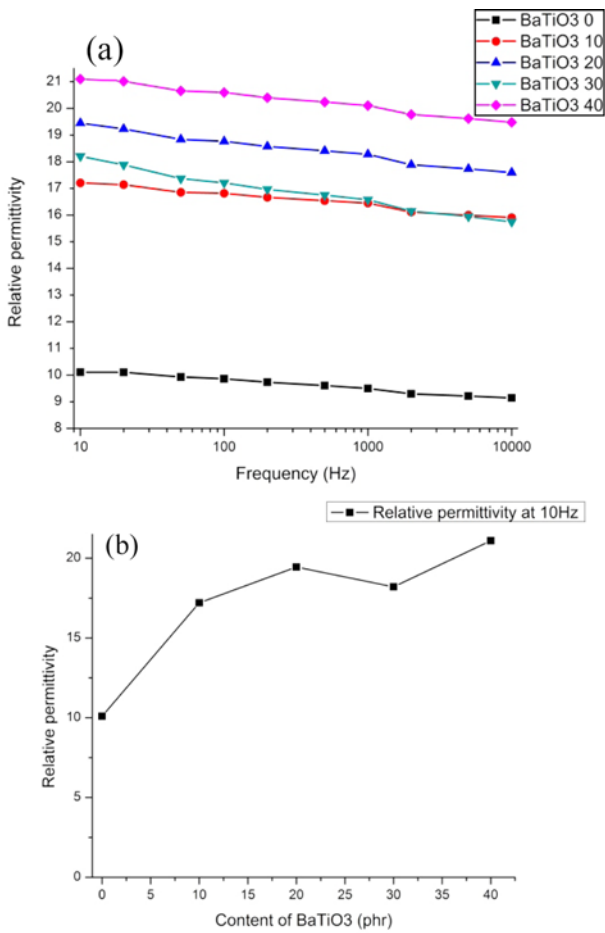
Figure 7. The relation between relative permittivity of synthetic NBR and frequency (a) and the effect of $BaTiO_3$ contents on the relative permittivity (b).

Figure 8. The stress-strain curves (a) and Young's modulus (b) of synthetic NBR with various contents of $BaTiO_3$.

reduced. According to our previous work,[10] the maximum relative permittivity of the synthetic elastomer is 11 at 40 phr of titanium dioxide (TiO_2) and 10-Hz frequency. However, this value is only half of that when using 40 phr of $BaTiO_3$ as show in Figure 7(b). This is because the relative permittivity of TiO_2 is much smaller than that of $BaTiO_3$.[22]

Young's Modulus and Stress Relaxation. The effects of $BaTiO_3$ on the Young's modulus and stress relaxation of the proposed material are presented in Figure 8. When the content of $BaTiO_3$ increases from 0 to 30 phr, the Young's modulus of the material also increases. However, exceeding 30 phr of $BaTiO_3$, the Young's modulus of the material decreases. The increase of Young's modulus corresponding to the content of $BaTiO_3$ from 0 to 30 phr can be explained by the $BaTiO_3$ particles restricting the movement of NBR chains and making the material stiffer. However, at over 30 phr of $BaTiO_3$, the particles seem to accumulate and agglomeration occurs. Therefore, the elastic modulus of the material will be reduced.

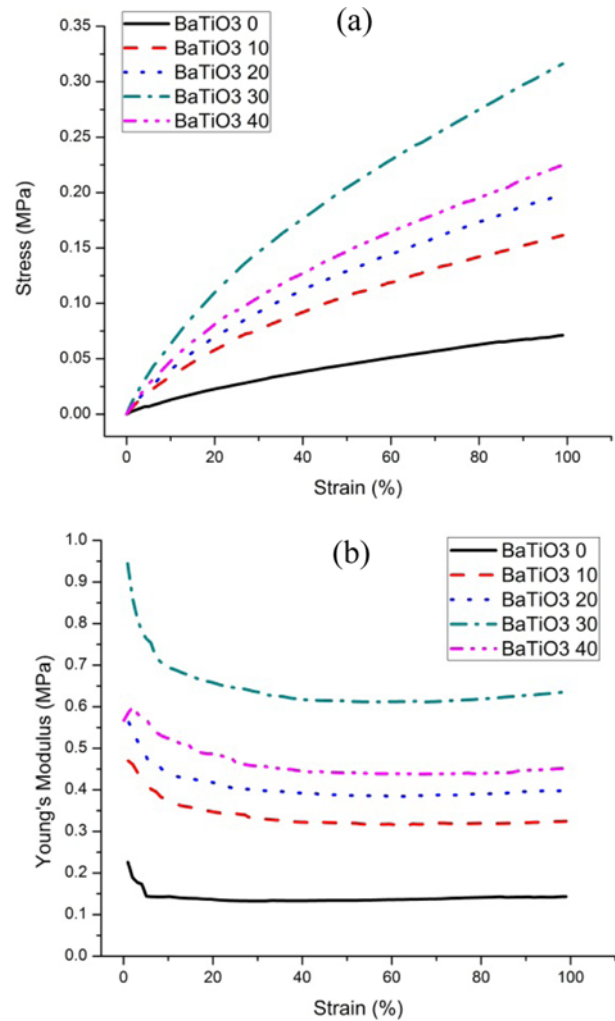In Figure 9, we can see that after reaching a predicted load

of around 0.21 N, all specimens suffer similar load after 80 s due to stress relaxation. Hence, the concentration of $BaTiO_3$ has no effect on the stress relaxation of the material.

Electrical Breakdown Strength. Figure 10 presents the electrical breakdown strength of the synthetic NBR materials based on various fractions of $BaTiO_3$. According to this result, the effect of $BaTiO_3$ on the breakdown field strength is complicated and unpredictable. In addition, the breakdown strength of the material reaches the highest value of 35.15 MV/m at 20 phr of $BaTiO_3$.

Actuation Performance. By implementing the experiments described in Figure 4, we can measure the radial strain of the synthetic NBR film, which reflects the actuation performance under the impact of the applied voltage. According to Figure 11, at the same applied electrical field, the synthetic elastomer using 20 phr of $BaTiO_3$ has higher radial strain than the others. Moreover, it also has the highest performance with 3.8% of radial strain at 14.22 MV/m.
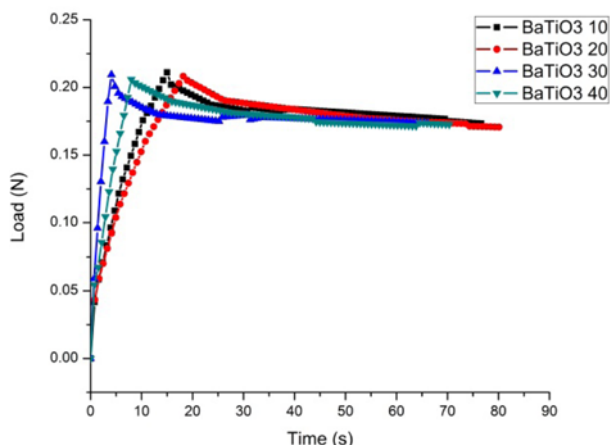
Figure 9. The stress relaxation of the materials with various contents of BaTiO₃.
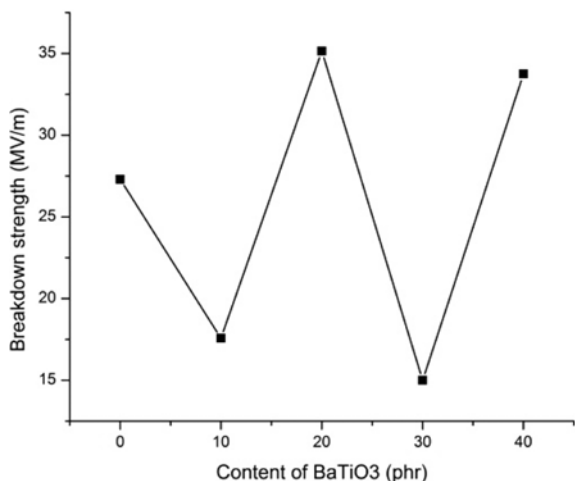

Figure 10. The effect of BaTiO₃ on the electrical breakdown strength.

Figure 11. The actuation performance of the materials based on different contents of BaTiO₃.
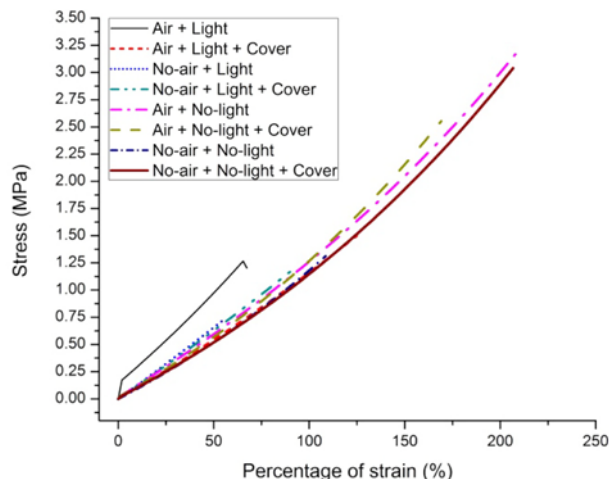
Figure 12. The stress-strain curves of NBR sheets after 10 days in different environments.

In Figures 10 and 11, it can be seen that the maximum electrical fields applied in the actuation performance test of the synthetic elastomers are smaller than that in the breakdown field strength test. Thus, the synthetic NBR films seem to be broken before reaching their breakdown field strength. While applying a certain voltage to both sides of the films, their thicknesses are reduced due to the Maxwell stress. Moreover, under the loading effect of the mass, the stress inside the film increases especially around the inner edge of the rigid frames. Therefore, due to the appearance of the stress concentration, the film cannot suffer the same voltage as in the breakdown field strength test. Consequently, it is suggested that in the experiments for testing the actuation performance of DEA, the thickness of the film should be thicker than that in the breakdown field strength test, and the mass should be selected properly so as to have little influence on the performance of the samples.

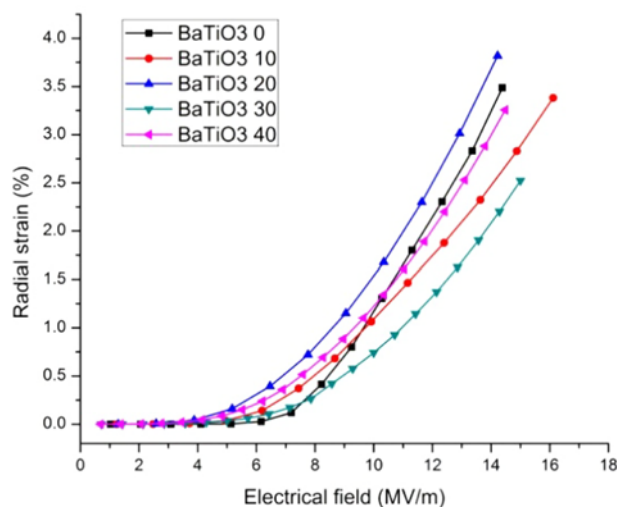**Effects of Environments.** After ten days of storage in different environments, the surface conditions and stress-strain properties of the samples were checked. Figure 12 presents the stress-strain curve of these samples. It can be seen in this figure that the surfaces of the exposed synthetic elastomer sheet kept in strong light and air conditions are very different from the other specimens. This sheet is stiffer and easier to crack as bending. The curve of this sample, which has the highest stress in the range of 0 to 65% strain, looks strange. In fact, the outer surfaces of the sample seem to be harder than the inner layers. At the beginning of the curve, it is quite difficult for deformation to occur. However, after some cracks form on the surface, deformation happens more easily.

As shown in Figure 12, the sample covered by electrode layers and kept in vacuum and no-light conditions (the bold curve in the table) can reach the largest strain of about

200%, with lower maximum stress than the exposed sample. According to this, we can realize two positive roles of the electrode layers. They act as antioxidant layers that protect the NBR sheet from the attack of light and oxygen in the air. In the second, it prevents DOP escaping and keeps it inside the NBR sheet. Hence, the synthetic NBR will maintain its softness and the lifetime of the DEA will be extended.

As a result of this analysis, a combination of the two following manners is proposed to protect the synthetic NBR material after curing. First, both surfaces of the NBR film should be covered by electrode layers. Second, these actuators need to be maintained in a dark and no-air environment.

Conclusions

The properties of DEA based on synthetic NBR were investigated with different contents of additives including plasticizers and a high dielectric filler. DOP, DOA, and DIDP plasticizers were proposed for synthetic NBR, and evaluated through experiments on the stress-strain properties and breakdown field strength. The DOP plasticizer was selected as the main plasticizer for the synthetic elastomer due to its advantage of reducing the stiffness of the NBR material more than the others. The high dielectric filler $BaTiO_3$ was used to improve the relative permittivity of the material. Different fractions of $BaTiO_3$ were added to the NBR to create different synthetic rubber materials. The electrical and mechanical properties of these materials were investigated, and their electromechanical responses were measured. The synthetic elastomer with 20 phr of $BaTiO_3$ produced the largest deformation, satisfying the main goal of our applications. The effects of environmental conditions were also discussed.

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