Chapter

Improvement of Elastomer Elongation and Output for Dielectric Elastomers

Seiki Chiba, Mikio Waki, Shijie Zhu, Tonghuan Qu and Kazuhiro Ohyama

Abstract

The need for light, high-strength, and artificial muscles is growing rapidly. A well-known type of artificial muscle meeting these requirements is the dielectric elastic (DE) type, which uses electrostatic force between electrodes. In hopes of utilizing, it practically for a variety of purposes, research and development is rapidly progressing all over the world as a technology for practical use. Much of the market demand is dominated by more output-focused applications such as DE power suits, DE motors, DE muscles for robots, and larger DE power systems. To meet these demands, the elasticity of the elastomer is very important. In this paper, we discussed what the important factors are for SS curves, viscoelasticity tests, etc. of the dielectric elastomer materials. Recent attempts have been also made to use new carbon foam materials such as SWCNTs and MWCNTs as electrodes for DEs. These electrodes bring the elastomers to a higher level of performance.

Keywords: Dielectric elastomer, Actuator, Sensor, Generator, Large deformation, High efficiency, Artificial Muscle

1. Introduction

The creation of artificial muscle has long been a scientific aspiration. It is well known that Wilhelm Conrad Röntgen, who discovered X-rays, conducted experiments using rubber strings as artificial muscles [1]. In the 1950s, artificial muscles using EPA (Electro Active Polymer) became mainstream. Since then, the need for light, high-strength, and artificial muscles has been growing rapidly.

EAP type artificial muscles which drive a polymer membrane by applying electrical stimulation, are actuators that realize movements similar to living muscles by electrical control. Because they move softly, they are also called soft actuators. **Figure 1** shows the following types are of EAPs: **1)** DEs (dielectric elastomers) which are driven by the generated coulomb force, made by sandwiching an elastomer between flexible electrodes. [2, 3], **2)** IPMCs (Ionic polymer-metal composites), which owe their power to the movement of ions and water molecules in the polymer film (combination of an electrolyte film and a thin metal electrode) [4], **3)** CPs (conductive polymer) which use a drive force moving ions by applying a voltage between the conductive polymers [5], **4)** ionic polymer gel Ion polymer gels, which utilize the movement of ions due to chemical changes (e.g., Ph changes)

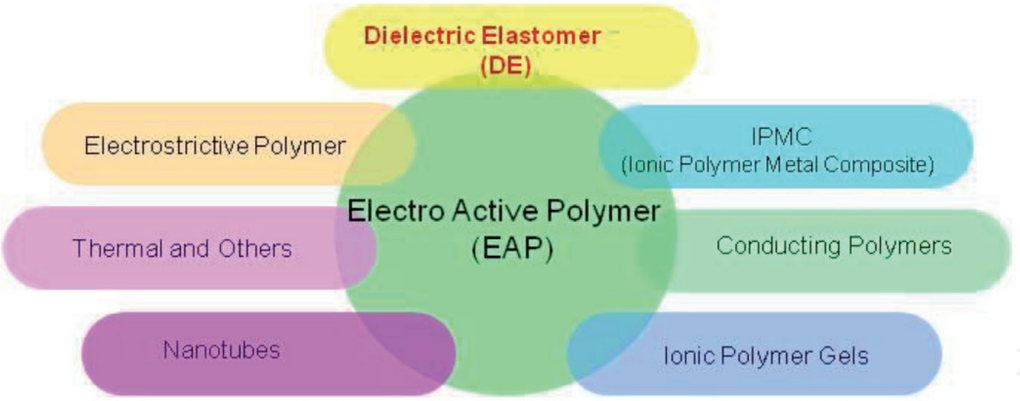

Figure 1.
Typical electroactive polymers (EAP).

within the gel [6], and **5)** CNT (carbon nanotube) actuators, which are ideally for nanomachines and do not require ion intercalation [7]. In addition, the piezopolymer utilizes a piezoelectric phenomenon [8], and some are driven by heat, air, light, etc. [9–13].

The most promising candidate from technologies above is the DE [2, 14]. In 1990, Chiba and Pelrine began research and development of dielectric elastomers for the first time in the world. [2], but now, research and development for their practical uses are rapidly progressing all over the world as a technology for practical use [2, 3, 14–34].

Most of the current market demand is for DE power suits, DE motors, DE muscles for robots, and systems that drive them in reverse to generate electricity efficiently. To meet these demands, the elasticity of the elastomer is extremely important. In this paper, we discussed important factors (including cross-linking agents and double bond breaks) through SS (strain stress) curves, viscoelasticity tests, etc. of DE materials. In addition, recent attempts have been made to use new carbon foam materials such as SWCNTs and MWCNTs as electrodes for DEs. These electrodes bring the above-improved elastomers to a higher performance. They will also be discussed in this paper.

2. Background of DEs

The structure of a DE is very simple and consists of a polymer film (elastomer), which is the main material, and two electrodes that sandwich it [2, 3]. When a potential difference is applied between the electrodes, the Coulomb force causes the polymer film to contract in the thickness direction and expand in the plane direction (see **Figure 2**).

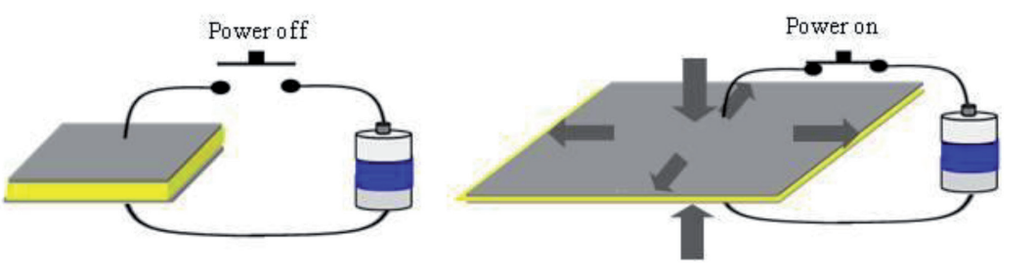

Figure 2.
DE artificial muscle actuator structure and operating principle: (a) The black sheet is the flexible and stretchable electrode, and (b) The yellow part is the elastomer.

At the material level, a DE actuator has a fast response speed (over 100 kHz), with a high strain rate (up to 680%) [15], high pressure (up to 8 MPa), and power density of 1 W/g [16]. A DE actuator having only 0.15 g of DE can lift the weight of 8 kg easily by 1 mm or more with the actuation speed of 88 msec, using Single-wall carbon nanotubes as electrodes [17]. Since the elongation and the output are in inverse proportion to each other, it is possible to suppress the output and increase the elongation. In addition, as mentioned above, power generation is possible by reversing the movement of the DE actuator. Its efficiency is excellent, at over 70% [18].

A mathematical model of the DE actuator can be described as follows: The strain (deformation) observed in the elastomer membrane is mainly caused by the interaction of electrostatic charges between the electrodes [19]. Opposite charges on the two electrodes attract each other, and the same charges repel each other. This phenomenon can be derived by using a simple electrostatic model to derive the effective pressure generated by the electrodes of the elastomer membrane as a function of the applied voltage [19]. Pressure ρ is

$$\rho = \varepsilon_r \varepsilon_o E^2 = \varepsilon_r \varepsilon_o \left(\mathrm{V}/\mathrm{t}\right)^2 \tag{1}$$

Here, ε_r and ε_o are the permittivity and the relative permittivity (dielectric constant) of the polymer in the free space, respectively, E is the electric field strength, V is the applied voltage, and t is the film thickness. The responsiveness of this polymer is similar to that of conventional electrostrictive polymers, and the pressure is proportional to the square of the electric field strength. For small strains with free boundary conditions, the actuator energy density, e_a, of the material can be written as

$$e_a = \mathrm{P}s_z = \mathrm{Y}s_z^{\,2} = \left(\varepsilon_r \varepsilon_o\right)^2 \left(\mathrm{V}/\mathrm{t}\right)^4 / \mathrm{Y} \tag{2}$$

where Y is the modulus of elasticity and s_z is the polymer thickness strain [14]. Conventionally, the elastic energy density $e_a = 1/2\ \mathrm{Y}s_z^{\,2}$ is often used (see **Table 1**).

Polymers investigated	**Presure (MPa)**	**Strain (%)**	**Young's modulus (MPa)**	**Breakdown Electric field (V/μm)**	**Dielectric constant (at 1 kHz)**	**Coupling efficiency, *k2* (%)**	**Elastic energy density (Jrcm^3)**
Fluoroelastomer 1	0.11	8	2.5	32	12.7	15	0.0046
Isoprene Natural Rubber 1	0.11	11	0.85	67	2.7	21	0.0059
Silicone 2	0.13	41	0.125	72	2.8	65	0.026
Fluorosilicone 2	0.39	28	0.5	80	6.9	48	0.055
Silicone 3	0.51	32	0.7	144	2.8	54	0.082
Polyurethane 1	1.6	60	17	160	7.0	21	0.087
Silicone 1	1.36	102	1.0	235	2.8	54	0.22
Acrylic 1	7.2	358	2.2	412	4.8	85	3.5

**Average engineering modulus at the maximum strain. Elastic energy density.*

Table 1.
The result of performance measurements of eight polymers (elastomers).

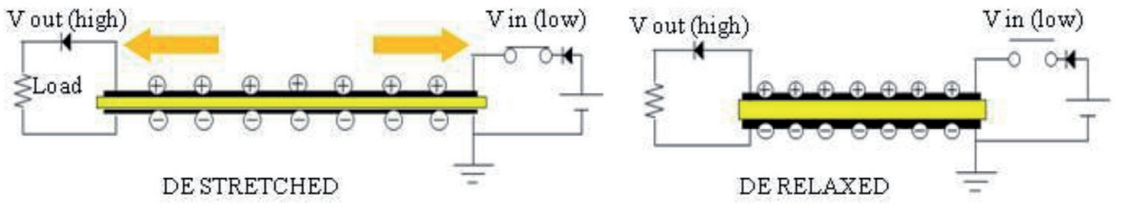

Figure 3.
Operating principle of DE power generation: (a) Thick lines are compliant electrodes, and (b) the yellow line between the thick lines is the dielectric elastomer.

As described above, when the movement of the dielectric elastomer actuator is reversed, the power generation mode is set. This field of power generation research has become more active around the world since [14, 15, 18, 19, 32–56] it was first tested with a DE generator on a buoy [34]. The principle is simple and utilizes the increase in electrostatic energy generated by changing the shape of the dielectric elastomer actuator with an external force (see **Figure 3**). That is, when some mechanical energy is applied to the dielectric elastomer to extend it, the thickness direction becomes thinner and the area expands (Increase in capacitance). At this time, electrostatic energy is generated on the polymer and stored as an electric charge. When the mechanical energy decreases, the elasticity of the dielectric itself increases the thickness in the thickness direction and reduces the area (Reduction of capacitance). At this time, the electric charge is pushed out toward the electrode. Such changes in charge increase the voltage difference, resulting in increased electrostatic energy [19]. The capacitance of the DE film "C" is given as follows:

$$C = \varepsilon_0 \varepsilon A / t = \varepsilon_0 \varepsilon b / t^2 \tag{3}$$

where ε0 is the dielectric permittivity of free space, ε is the dielectric constant of the polymer film, A is the active polymer area, and t and b are the thickness and the volume of the polymer. The second equality in Eq. (3) can be written because the volume of the elastomer is essentially constant, i.e., At = b = constant. The energy output of a DE generator per cycle of stretching and contraction is

$$E = 0.5 C_1 V_b^2 \left(C_1 / C_2 - 1\right) \tag{4}$$

where C1 and C2 are the total capacitances of the DE films in the stretched and contracted states, respectively, and V_b is the bias voltage.

3. Materials for DEs

The main parameters that improve the performance of the DE are the withstand voltage of the elastomer film, the dielectric constant (including the improvement of the dielectric constant due to additives), Young's modulus, the type of electrode used, use of a cross-linking agent, and the elastomer structure Improvements (such as the addition of monomers or cutting one of the double bonds).

Table 1 shows the measurement performance of some polymers [3, 20]. This table shows measurements of strain, electric field, modulus of elasticity, and permittivity. The pressure is calculated from Eq. (1) and the elastic energy density is estimated using the strain (measured value) and the pressure calculated from Eq. (2).

Note:

- Silicon 1 was made by mixing two types of silicon polymers.
- Acrylic 1 was modified by us after purchasing acrylic made in the United States.
- The polymers (elastomers) other than the above two types were made in the United States and were used as they were.

As shown in **Table 1**, the DE polymers (elastomers) that can obtain a value with a large strain has a large value of any one of elastic energy density, breakdown electric field, Young's modulus, or permittivity, or some of them are combined thereof. However, increasing these parameters will stiffen the elastomer and will not significantly deform the DE. In other words, the power obtained will not increase unless the elastomer is hardened and deformed (thickness) significantly.

A large deformation is important not only for actuators but also for power generation elements. That is, a large deformation produces more power (see Eq. (4)).

3.1 Elastomer properties obtained from SS curves/dynamic viscoelasticity tests

The SS curve and dynamic viscoelasticity were measured using Silicon 1 and Acrylic 1 [21]. The results are shown in **Figures 4** and **5**. First of all, we would like to point out that the research target is artificial muscles, and it is recommended to consider the tensile speed of the SS curve and the viscoelasticity test from the operating speed required for robots and power assist devices. In **Figure 4**, the SS curve was measured by changing the measurement speed in 4 steps, and the curve changed depending on the tensile speed. Similarly, in **Figure 5**, the curve of dynamic viscoelasticity changed depending on the measurement speed [14].

What is interesting here is that acrylic has higher viscoelasticity, so it depends more on tensile speed than silicon. This indicates that it is important to test it with the response required for the artificial muscle. In other words, until now, researchers have overlooked the importance of viscoelasticity. This meaning is easier to understand by looking at the results of the dynamic viscoelasticity test in **Figure 5**.

As the **Figure 5**, clearly shows, the acrylic is more affected by dynamic viscoelasticity than the silicon. As a result, when the driving voltage is increased and each elastomer is stretched, the silicon DE becomes harder, and the amount of stretching

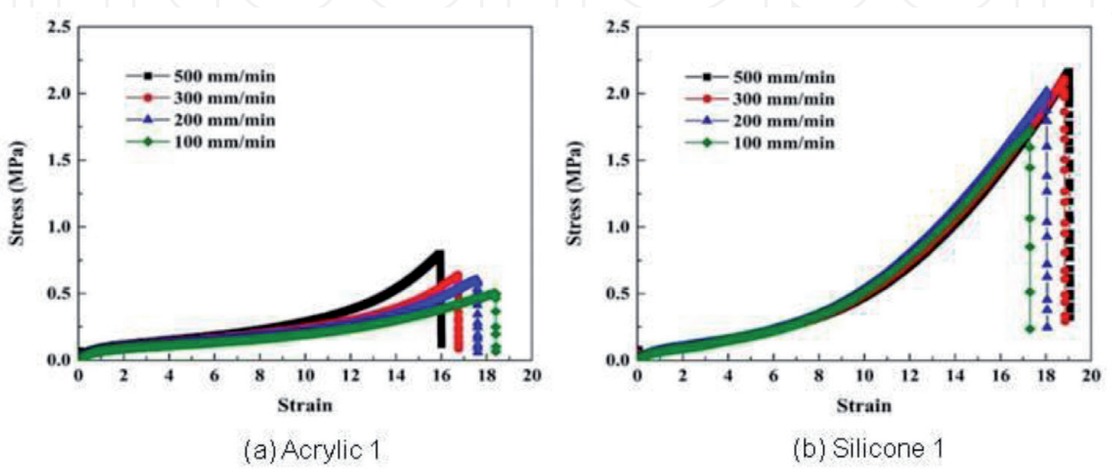

Figure 4.
Relationship of stress-strain for tensile tests: (a) Acrylic 1, (b) Silicone 1.

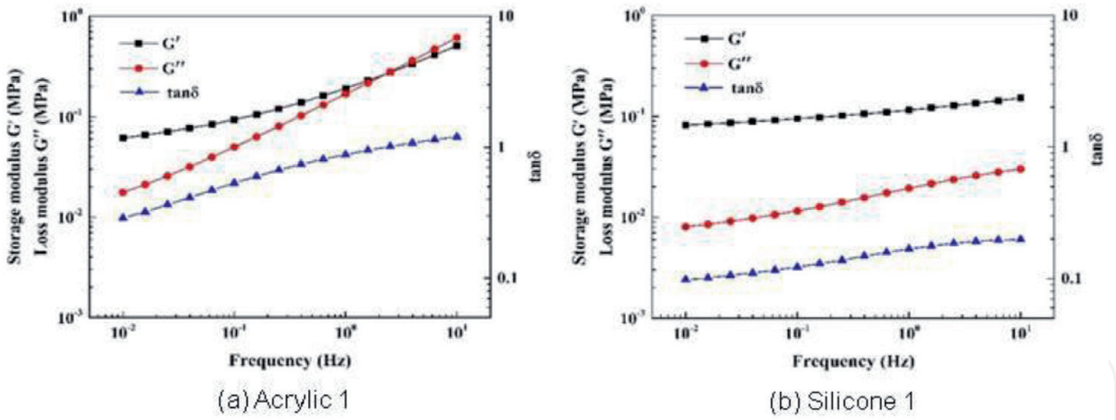

Figure 5.
The frequency dependence of G', G" and tan δ of (a) the acrylic 1 and (b) the silicone 1.

is smaller. Of course, it is also a fact that the difference in the dielectric constant and Young's modulus of both films is the cause (see Eqs. (1) and (2)). Nevertheless, the above behavior can be explained using the SS curves in **Figure 4**. The silicon curve stands up more. This does not mean that silicon has poor performance. It is a proposed that it is better to change the material depending on the application. Silicon has a faster drive speed and a higher rate than acrylic. Therefore, it is advisable to select the type of elastomer depending on where it is used, for example, for applications such as robots or power suits. Illustrating this point, in human muscles, there are slow muscles and fast muscles, each of which has an important mission. Moreover, Silicon can be used from a relatively high temperature to a considerably low temperature. Compared to acrylic, it could have a considerable advantage in devices that are used at higher or lower temperatures [21].

In terms of artificial muscles, e.g., for human uses, it seems that a flat hill-like shape with a gentle rise, like acrylic, is preferable.

3.2 Attempts to increase the dielectric constant of elastomers

As an attempt to increase the dielectric constant of the elastomer, there is a method of adding a monomer to change the structure or adding a substance having a very high dielectric constant such as Barium titanate. However, in general, these methodss make the elastomer harder and less stretchable. Examples of adding Barium titanate to our synthetic acrylic are shown below. Here, the acrylic we have synthesized is called a base acrylic.

As a method for dispersing barium titanate, a predetermined amount of barium titanate was added to the polymer-containing liquid and crushed with a homogenizer. As a result of particle size measurement by SEM, the median diameter was about 450 nm [57]. It was also confirmed by using SEM that the barium titanate was uniformly mixed.

Elastomer sheets (thickness: 400 μm) were prepared by a) adding 1 wt% of Barium titanate to the base acrylic, and b) adding a 2 wt% of Barium titanate to the acrylic. The SS curves of those elastomers were measured as shown **Figure 6**.
The acrylics, which are the base for the Barium titanate were slightly crosslinked. The permittivity of the acrylic was measured using the parallel plate capacitance method. The withstand voltage was measured using a general dielectric breakdown tester. The relationship between the withstand voltage and the capacitance of these films is shown in **Figure 7**.

From **Figures 6** and 7, as we initially expected, the withstand voltage and the amount of capacitance of the film containing a large amount of Barium titanate increased, and the film became harder and less stretchable by that amount. Circular

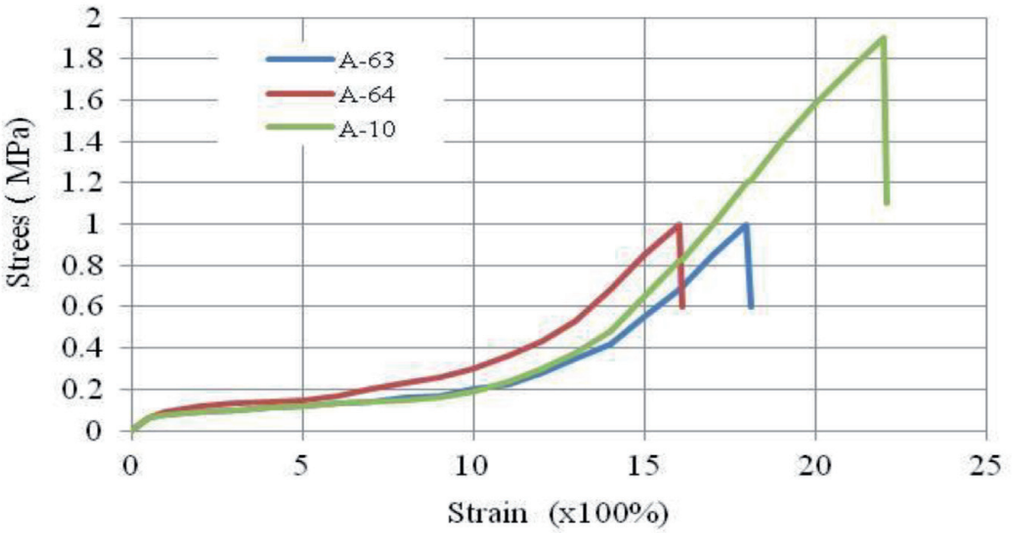

Figure 6.
The SS curve of the elastomer sheet with a small amount of a) 1wt% and b) 2wt% of Barium titanate added to the base acrylics.

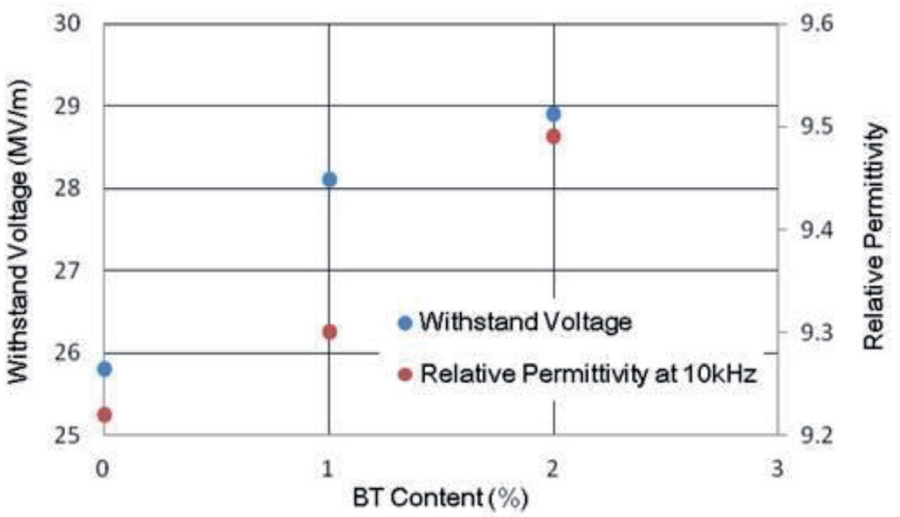

Figure 7.
Relationship between the breakdown of the electric field and the capacitances of these elastomer films.

actuators were produced using either the base acrylic film without adding the barium titanate, or the films containing 1 wt% or the barium titanate and 2 wt%, and the elongations of each were compared. As a result, the actuator using the base film showed the largest elongation. In fact, the film that was hardened by adding the barium titanate was superior in increasing the withstand voltage. From those results, it was found that even if a substance with a high dielectric constant was added, it did not give a significantly good effect. The dynamic viscoelasticity of the base acrylic +2% of Barium titanate is shown in **Figure 8** (**Figure 8** is in Section 3.3).

3.3 Adjustment of cross-linking agents/reduction of double bonds

Figure 8 shows the SS curve when the amount of cross-linking agent added to the above base acrylic is changed. Assuming that the amount of the cross-linking agent added to the base acrylic (blue) is 1, red, green, black, and orange are added at rations of 2:1, 1.5:1, 0.8:1, and 0.5:1, respectively. Not surprisingly, the less cross-linking agent we add, the better the elongation. Due to that reduced strength, circular actuators need to be manufactured and evaluated to determine how appropriate they are.

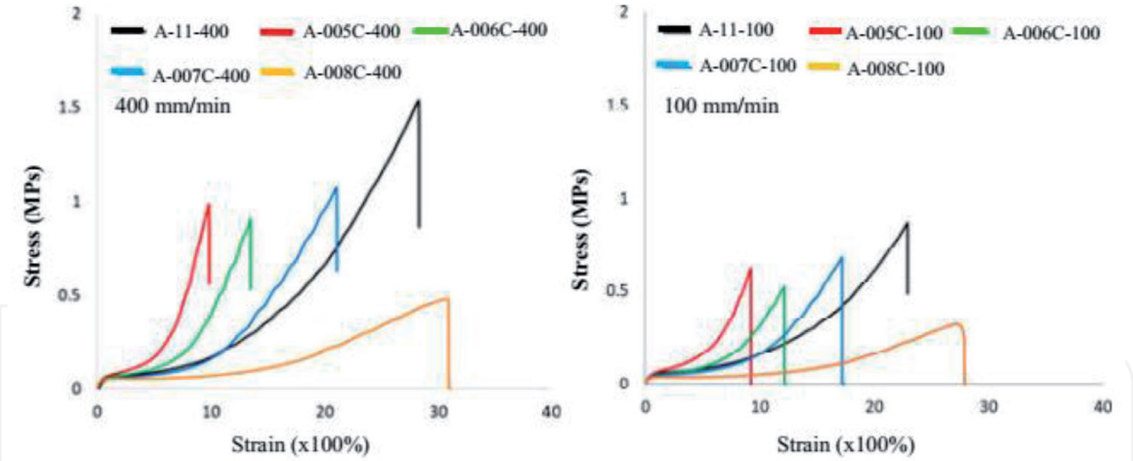

Figure 8.
The SS curve when the amount of cross-linking agent added to the above base acrylic changed. The case where the amount of the cross-linking agent added was changed and the case where the amount of the double bond was reduced by using HNBR (Hydrogenated acrylonitrile butadiene rubber).

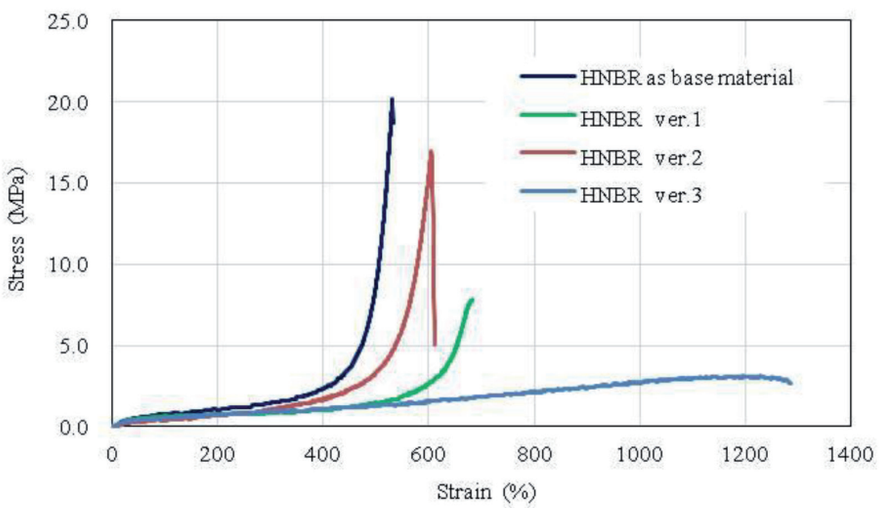

Figure 9.
The SS curve when the amount of cross-linking agent added to the above base acrylic changed: The case where the amount of the cross-linking agent added was changed and the case where the amount of the double bond was reduced by using HNBR (Hydrogenated acrylonitrile butadiene rubber).

In this experiment as well, the tensile speeds were set to 100 mm / min and 400 mm / min, but as mentioned above, such an evaluation is important for artificial muscles. That is, the test should be performed according to the actual running speed of the muscle. In this case, 100 mm / min / sec is clearly affected by viscoelasticity. In other words, even if the elongation increases, the stress does not increase so much (the inclination angle is gentle), and as a result, it could become easy to deform as the DE.

Next, attempts were made to not only change the amount of additive added, but also to reduce the amount of double bonds. **Table 2** shows the conditions of the case where the amount of the cross-linking agent added was changed and the case where the amount of the double bond was reduced by using HNBR (Hydrogenated acrylonitrile butadiene rubber). HNBR is a hard material with a dielectric constant of 15, but as shown in **Table 2**, when the ratio of double bonds is reduced, the slope of the SS curve becomes gentle (see **Figure 9**). HNBR Ver.3 has a dramatically reduced slope because the cross-linking agent has also been reduced. The capacitance was also 11.

	Crosslinker (%)	Double bond (%)
HNBR sheet as Base Material	8	10
HNBR ver.1	8	5
HNBR ver. 2	8	1
HNBR ver. 3	2	1

Table 2.
The case where the amount of the cross-linking agent added was changed and the case where the amount of the double bond was reduced by using HNBR (hydrogenated acrylonitrile butadiene rubber).

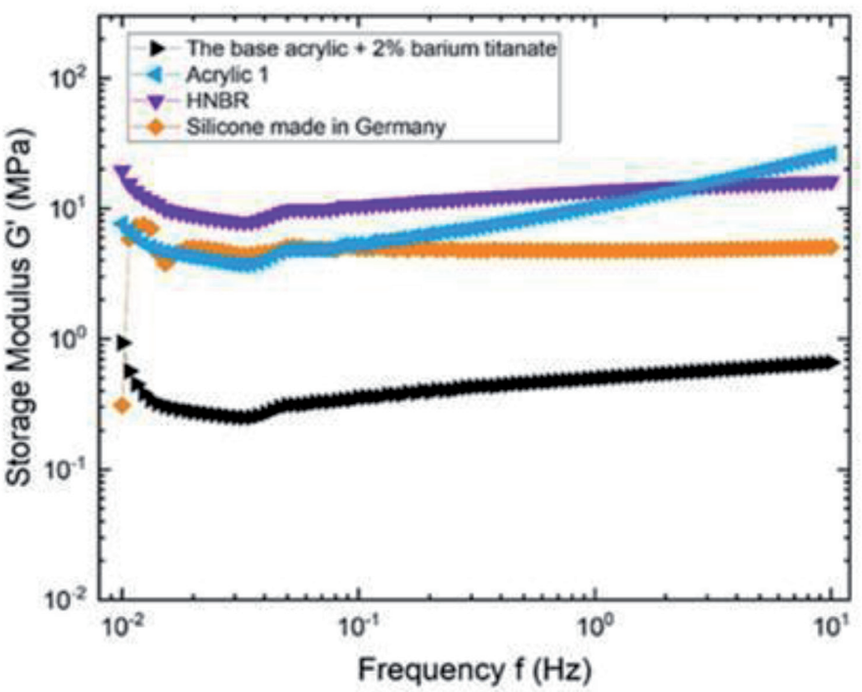

Figure 10.
The SS curves of HNBER (base material), HNBER ver.1, HNBR ver.2 and HNBER ver.3. Note: The silicon for this test was used the silicon German-made. This is because the silicon 1 was tested long time ago there is no remaining stock.

With these membranes, it is necessary to make a circular actuator and measure the elongation, but unfortunately it has not been done yet. Perhaps HNBR ver.3 is a little too soft and it could be difficult to make a DE. Or, because it is soft, the Coulomb force might be dispersed and it might not be possible to drive it well. Further studies are desired on the proportion of double bonds and the amount of cross-linking agent.

The dynamic viscoelasticity of HNBR was also measured (see **Figure 10**). Silicon and acrylic are also shown in this figure for comparison.

Figure 10 shows the frequency dependence of the storage modulus of five kinds of materials. From 0.032 Hz, it can be seen that the storage elastic modulus of the acrylic 1 (see **Table 1**) increases. It can be seen that the storage elastic modulus of HNBR ver.3 gradually increases, but the storage elastic modulus of the silicon (made in Germany) basically does not change as the frequency increases. Again, silicon could be a bit difficult to get the most out of as an artificial muscle. One of the reasons might be that the structure of silicon is generally a chain structure. Of course, silicon has excellent temperature characteristics and DE responsiveness, and can be driven efficiently. As for our recommendation, it is a good idea to use both fast (silicon) and slow (acrylic) muscles well, like human muscles. Since HNBR is rubber, it is resistant to humidity and can withstand temperature changes. In addition, the results of dynamic

viscoelastic research show that it is somewhat suitable for driving a DE. In particular, it seems that the amount of cross-linking agent added should be selected appropriately. We believe it is particularly suitable for ocean power generation. Since ocean power generation is exposed to a harsh natural environment, it is desirable to use a material that is tough and suitable for DEs. Another point that greatly contributes to power generation efficiency is that there are many changes in thickness (see Eq. (3)), and in that respect, acrylic is most suitable, but acrylic is not very suitable for harsh natural environments. We hope that moisture resistant acrylics will be developed. The film with 2% barium titanate added to the base acrylic is considerably harder than the other films as mentioned above, but the withstand voltage of this film is high (see **Figure 6**) and the elastic modulus is also increased. Therefore, if it could withstand a higher voltage, it might be used as a high-power DE in the future.

3.4 Pre-strain

Pre-strain will increase the performance of the DE. This is because when repeated tests were performed to know the SS curve of the elastomer sample, the film stretched and it could not return to its original length, so there was no choice but to stretch the film a little in advance and evaluate it [14, 20]. After that, if more pre-strain is applied, it will be advantageous because the strain is applied in advance compared to the case where it is not stretched, and the performance will be further improved [34]. In order to utilize the pre-distortion, it is advantageous to use a material having a gentle SS curve, such as acrylic (see **Figure 4**). As described above, as the degree of pulling increases, the film becomes harder and harder to stretch. However, since the curve of acrylic is gentle, it is harder to harden than silicon. In the dynamic viscoelasticity test, acrylic is more frequency dependent and has more storage modulus than silicon (see **Figures 5** and **8**). This means that even if the film becomes hard, it can function as a DE because the modulus increases. Again, this frequency dependence is also important for use as an artificial muscle.

3.5 Adopted CNTs as electrodes

Recently, attempts have been made to use new carbon foam materials such as SWCNTs and MWCNTs as electrodes for DEs. These electrodes bring the elastomers to a positively improved higher performance. **Table 3** shows how much weight can be lifted with a stroke of 5 mm due to the difference in electrodes. The elastomer used is acrylic 1, and its weight is 0.1 g [10]. Diaphragm actuators with a diameter of 8 cm were manufactured and those elongations were measured.

Since these electrodes are not optimized, it seems possible to lift heavier weights while having sufficient elongation in the very near future. In addition, these are single layers of DEs and are very light, so it is possible to have multiple layers of DEs, which is close enough to the range applicable to robots and power suits.

On the other hand, these electrodes are also promising as power generation elements. A power generation experiment was conducted using a drape type DE having a height of 120 mm and a diameter of 260 mm. The amount of power generation when the DE was pulled by about 60 mm is summarized in **Table 4** [15]. The drape weighs 4.6 g and uses acrylic 1. Carbon grease, Carbon black, MWCNTs (multi-walled carbon nanotubes), and SWCNTs (single-walled carbon nanotubes) were used as electrode materials.

Using MWCNTs or SWCNTs makes it possible to obtain more power, as shown in **Table 4**.

Electrode type	Weight that can be lifted with a stroke of 5 mm
Carbon grease	6.5 N
Carbon Black	10 N
Multi-walled carbon nanotubes (MWCNT)	16 N
Single-walled carbon nanotubes (SWCNT)	22 N

Table 3.
Types of electrodes and weight that can be lifted.

Type of electrode	Power obtained (mJ)
Carbon grease	179
carbon black*	274
multi-walled carbon nanotube	445
single-walled carbon nanotube**	630

**Carbon grease, Carbon black and MWCNT are manufactured in companies in United States*
***SWCNT (ZEONANO®-SG101) by Zeon Corp in Japan.*

Table 4.
Differences in power obtained when changing the electrode materials.

This is because the conductivities of MWCNTs and SWCNTs are much higher than that of carbon black or Carbon grease.

In this way, the highly conductive material significantly increases the elongation of the DE actuator, resulting in greater elongation and also increasing the amount of power generated by the DE element.

4. Conclusion

From the above experimental results and those discussions, the following was found:

- The elongation of the DE is greatly influenced by the elastomer material. If the material is too hard or too soft, it may not produce the desired result. DE performance needs to be adjusted according to product requirements. The same applies to the output of the DE.

- There are several methods for changing the properties of the elastomer, but two easy methods are to adjust the amount of the cross-linking agent added and/or the percentage of double bonds. Mixing additives with a high dielectric constant seems to be less effective.

- In order to know the properties of the polymer materials for the DE, it is desirable to measure the SS curve and/or dynamic viscoelasticity. In those cases, it is recommended to measure at a slower speed for artificial muscle applications.

- By using highly conductive materials, it is possible to improve the performance of the DE actuator and the performance of the DE power generator.

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