ZULFAQAR Journal of Defence Science, Engineering & Technology
e-ISSN: 2773-5281
Vol. 6, Issue 1 (2023)
DOI: https://doi.org/10.58247/jdset-2023-0601-04
Journal homepage: https://zulfaqarjdset.upnm.edu.my

IMPACT OF NANOFILLERS ON DIELECTRIC PROPERTIES OF POLYURETHANE ELASTOMERS: FROM SYNTHESIS TO FUTURE PROSPECTS

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ARTICLE INFO

ARTICLE HISTORY
Received: 01-12-2022
Revised: 20-02-2023
Accepted: 01-04-2023
Published: 30-06-2023

KEYWORDS
Dielectric elastomer
Polyurethane
Graphene
Barium titrates
Generator

ABSTRACT

Dielectric elastomer (DE) devices are soft or flexible capacitors, composed of a thin elastomeric membrane sandwiched between two compliant electrodes, that can transduce electrical to mechanical energy, and vice versa. Polyurethane possesses outstanding dielectric properties which can be applied in developing DE with excellent performance. Nevertheless, it still needs some modification to improve the dielectric properties to ensure their practicality in the industry. Researchers throughout the years had worked on several nanofillers such as graphene and barium titrate to enhance the dielectric performance of polyurethane elastomer to ensure their applicability in the field. This paper discusses the basic principle of dielectric elastomer as generator and actuator, the popular nanofillers used to enhance polyurethane dielectric constant, and the suitable application for polyurethane elastomer.

1.0 INTRODUCTION

Dielectric elastomer (DE) was discovered since the earliest day of electricity when James Maxwell conceived the effect on dielectric materials known as Maxwell stress in his work on the foundation of electromagnetic theory [1]. Back in the 1990s, DE started to attract researchers' attention when several research papers and discoveries for potential applications were blooming. This may be due to the unique characteristic of DEs that can convert electrical energy to mechanical energy, and vice versa, making them versatile to be applied in various industries. There are various suitable polymers to act as a dielectric elastomer such as acrylic [2–5], natural rubber [6], silicone rubber [7–9], poly (vinylidene fluoride) (PVDF) [10–11] and polyurethane.

Polyurethane has become a promising material to be adapted as a dielectric elastomer due to its large force outputs and high dielectric constant, allowing them to be actuated at the lower electric field. Paul and Pei [12] in their literature investigated materials that are suitable to be applied as DE in terms of their mechanical and electrical properties. Some of the material is pre-strained to obtain their best performance. Figure 1 indicates the attributes of the best state of acrylic, silicone rubber and polyurethane elastomer. As compared to acrylic and silicone, polyurethane is still an atrocious elastomer material with poor actuation behaviour and low energy density. Besides, it has a high Young's modulus which negatively affects the electromechanical properties of the material. The microstructure of polyurethane, especially the hydrogen bonds between hard segments leading to a high number of Young's modulus [13]. Other than that, there is a large number of N-H/C=O hydrogen bonds in polyurethane limiting the mobility of the polarized groups and the dipole polarizability of polyurethane which also restricts the dielectric constant of polyurethane [14]. Besides, polyurethane also possesses low breakdown strength which limits its allowable working electric field. Despite that, polyurethane possesses the highest dielectric constant with the lowest dielectric loss, making it a reasonable material

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to be applied as a dielectric elastomer. However, enhancement is still necessary to make polyurethane a favourable dielectric elastomer. Figure 1 shows the comparison of polyurethane, silicone, and acrylic attributes.

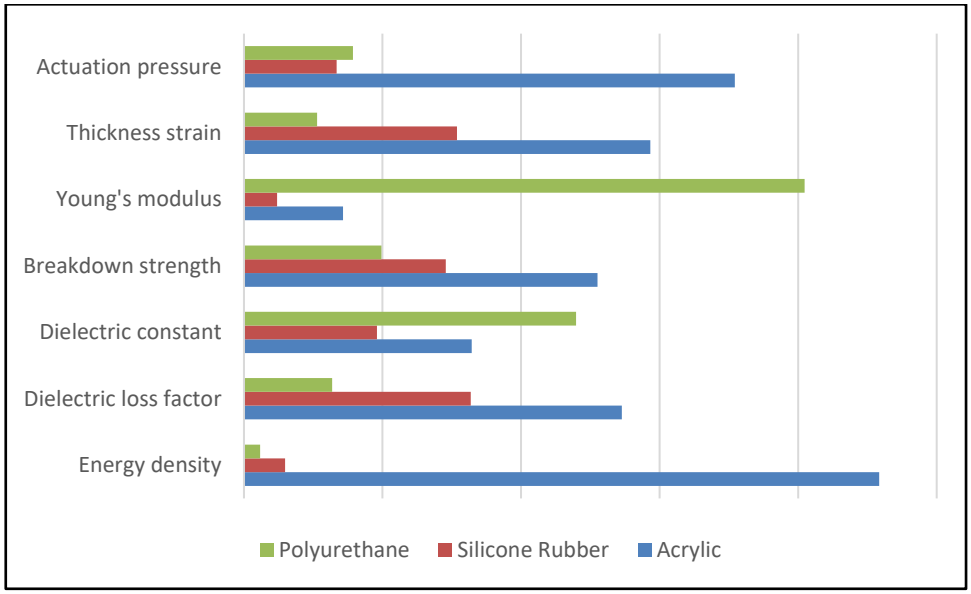

Figure 1. Comparison of polyurethane, silicone, and acrylic attributes [12]

This paper will briefly explain the principle of dielectric elastomer, while further explanation on the nanofillers used in the enhancement of polyurethane elastomer's dielectric properties. Discuss on the potential application of the enhanced polyurethane elastomer will also be done afterward.

2.0 DIELECTRIC ELASTOMER THEORY

Dielectric elastomer (DE) is an interesting mechanism that can act as both actuator and generator. DE applies the basic transduction principle in which the electrical energy stored in capacitors, U, increases with the material's capacitance, C. Given by:

$$U = \frac{1}{2} CV^2 \tag{1}$$

and

$$C = \varepsilon_o \varepsilon_r \frac{A}{z} \tag{2}$$

where V is the voltage, ε_o is the permittivity of free space, ε_r is the relative permittivity, A is the surface area and z is the distance of the electrodes or the thickness of the membrane. Dielectric elastomer generators (DEG) are variable capacitors in which the elastomer is sandwiched between two electrodes as illustrated in Figure 2. Electrical energy can be produced from a stretched, charged DEG by releasing its mechanical deformation while maintaining the charge on its electrodes [15]. Specifically, the operating principle of a DEG in terms of a cyclic sequence of electromechanical transformations can be illustrated in Figure 3. The operating principle of DEG subjected to uniform stretches on the plane of the electrodes can be explained through the following phases:

Phase 1 (P1) - As the DEG initially dwells in a relaxed state, where its capacitance is minimum, external loads make it expand and lead it to a stretched state, where the capacitance is maximum. No charge is present on the DEG during this phase.

Phase 2 (P2) - Charge Q is deposited on the electrodes, leading the DEG to a stretched and charged state, where the capacitance is constant. This phase is called "priming", which utilises an amount of electrical energy being spent to charge the device.

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Phase 3 (P3) - As the charge on the DEG is held constant, the external loads and the DE elastic stresses work against the electrostatic charge, taking the DEG back to a state with minimum capacitance, Cmin. During this generation phase, the external forces are converted into electrostatic energy and stored in the DEG electric field.

Phase (P4) - The DEG is finally held in the minimum capacitance configuration and discharged, and the stored electrostatic energy is harvested. The net amount of generated electrical energy is the difference between the energy recovered during the discharging phase (P4) and spent during priming (P2).

Figure 2. Dielectric elastomer generator (DEG) illustration

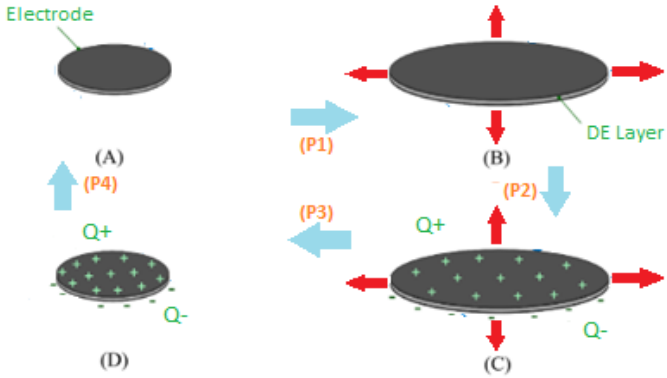

Figure 3. Operating principle of dielectric elastomer generator (DEG)

On the other hand, the dielectric elastomer actuator (DEA) working principle is just the opposite of the DEG. Instead of stretching and relaxing to produce electricity, DEA is the polymer that stretches and relaxes when being electrified. The basic principle of DEA is shown in Figure 4.

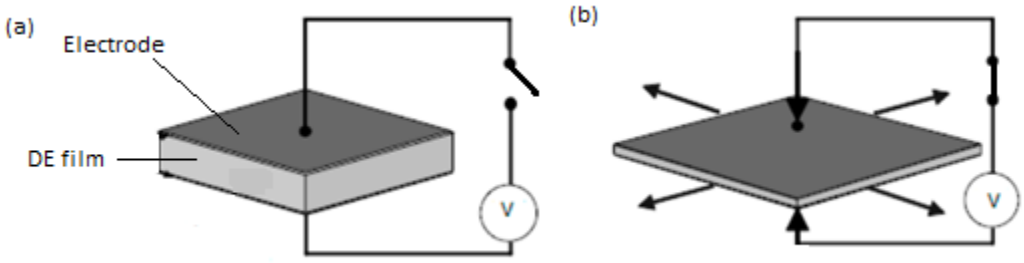

Figure 4. Basic principle of dielectric elastomer actuator (DEA)

When the voltage is applied to the electrode, the charge will be built at each side, causing attraction force in between the opposite charges which induces compression of elastomer film in the thickness direction. However, the incompressible nature of elastomers resulting in increases in the area in other planes, facilitated by the repulsion of the same charges. As the thickness of the elastomer films reduces, the conversion of electrical energy into mechanical energy in the form of electrostatic pressure given by the Maxwell pressure (σ) Eqn. (3):

$$\sigma = \varepsilon_o \varepsilon_r \left(\frac{V}{d}\right)^2 \tag{3}$$

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where V is the applied voltage, ε_o is the permittivity of free space, ε_r is the relative permittivity, and d is the distance of the electrodes or the thickness of elastomer film.

As for DEG, the dielectric permittivity of the polymer is very significant in DEA performance. It should be able to endure large voltages, for the actuation to be as large as possible. In addition, they should have high dielectric strength and low dielectric losses to maximize efficiency and avoid premature failures [16]. Eqn. (4) represents the electro-mechanical sensitivity of the DE and as it presented, a polymer with high dielectric permittivity and low Young's modulus, Y is desired to ensure the high electro-mechanical sensitivity.

$$\beta = \frac{\varepsilon_r}{Y} \tag{4}$$

3.0 NANOFILLERS USED TO ENHANCED POLYURETHANE ELASTOMER DIELECTRIC PROPERTIES

Increasing dielectric permittivity of polyurethane elastomer produces a direct increase in its capacitance, which helps in developing superior DEG and DEA. A conventional approach to do so is by creating elastomer composites. The idea is to combine high dielectric permittivity and excellent mechanical values of certain nanofillers with polyurethane. Several types of nanofillers have been proven to enhance polyurethane elastomer dielectric performance over the years, which are the carbon-based nanofillers, ceramic-based nanofillers, and hybrid nanofillers.

3.1 Carbon-Based Nanofillers

Due to its extraordinary properties, graphene has attracted tremendous attention in the field of polymer nanocomposites. Graphene can induce a considerable improvement in mechanical, thermal, and electrical properties of the resulting graphene–polymer nanocomposites at very low loading contents. It possesses high electrical and thermal conductivities, a high surface-to-volume ratio, and excellent mechanical properties [17–18]. Other than graphene, carbon nanotube (CNT) is another ideal material in polymer nanocomposites due to its high flexibility, high aspect ratio, and low mass density. However, the limitation of CNT is its uniformed dispersion because of the high surface energy and tendency to agglomerate in the bulk of the polymer and elastomer matrix [19]. Despite that, it is still one of the most favourable fillers for researchers over the years. Table 1 shows the dielectric constant of polyurethane composite with graphene and carbon nanotube fillers at various loadings.

Table 1. Dielectric constant of polyurethane composite with carbon-based nanofillers at various loadings

Carbon-based filler	Filler loadings (wt%)	Dielectric constant (at 1 kHz)	Reference
Pure Graphene	0.5	6.0	[20]
	1.5	9.0	
	3.0	13	
Graphene Oxide	0.5	10	
	1.5	12	
	3.0	16	
Thermally Reduced Graphene Oxide	0.5	16	[21]
	1.0	209	
	2.0	1875	
Thermally exfoliated and annealed graphene (TRG)/polyurethane composites Filler	5.5	377	[22]
Carbon Nanotube	0.5	8.0	[23]
	1.0	8.3	
	1.5	9.2	
	2.0	9.5	
Nitric Acid-treated Carbon Nanotube	0.5	8.7	
	1.0	9.0	
	1.5	9.3	
	2.0	9.4	
Silane-treated Carbon Nanotube	0.5	8.8	

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Carbon-based filler	Filler loadings (wt%)	Dielectric constant (at 1 kHz)	Reference
	1.0	8.8	
	1.5	9.1	
	2.0	9.4	

Polyurethane possesses many hydrogen bonds, in which the NH group acts as the donor, and either the carbonyl group from the hard segment or the ester oxygen group from the soft segment acts as the acceptor. These hydrogen bonds limit the mobility of the polarized groups of PU chains, thus limiting the dipole orientation polarization ability of PU, and restricting the increase in dielectric permittivity of PU. By adding functionalized graphene, the hydrogen can be disrupted which results in an increase in the dipole orientation polarization ability of PU chains, thus the dielectric performance of PU. As shown in Table 1, the dielectric constant of polyurethane increases with the loading of the graphene fillers. In comparison to pure graphene, graphene oxide can produce polyurethane elastomer with a higher dielectric constant [20]. This is due to many oxygens' functional groups (C-O-C, C-OH, and C=O) that enable graphene oxide to form homogenous colloidal suspensions in organic or aqueous solvents. The strong interaction between graphene oxide and polyurethane could prevent the direct connection of graphene oxide, subsequently resulting in a low dielectric loss. However, the oxidation of graphite could also lead to severe disruption of the graphite structure, whereby the dielectric constant will decrease. Thus, researchers were moving toward reducing the graphene oxide by thermally or chemically treating the graphene oxide.

Liu et al [21] thermally reduced the graphene oxide by in situ partial thermal reductions of graphene nanosheet-polyurethane composites at 180°C. The result from the X-ray photoelectron spectroscope (XPS) shows that the C/O ratio of the thermally reduced graphene oxide is higher than the pure graphene oxide, indicating the removal of the oxygen-containing group from the composite. Therefore, the graphite network is restored, which increases the interfacial polarization of the reduced graphene oxide and thus increases the number of charge carriers accumulated at the interface between the nanofillers and the PU matrix. The remaining oxygen functional group helps in disrupting the hydrogen bonding between the PU chain and enhances the interaction between the nanofillers with PU through hydrogen bonding, leading to the coating of the PU on reduced graphene oxide, thus creating suppression of the leakage current. All these factors are responsible for creating the large increases of dielectric constant in very low loadings (2 wt%) which is up to 1875 at 10kHz. Another method was used by Bansala et al [22] in the effort to reduce the oxygen content within the graphene oxide. They exfoliated the graphene oxide thermally at 200°C and annealed them at 800°C. The XRD confirmed the reduction of graphene oxide through this method. The polyurethane-reduced graphene oxide elastomer was fabricated, and it resulted in the improvement of the dielectric constant of the polyurethane elastomer. At 1 kHz, the fabricated elastomer recorded a dielectric constant of 377 at 5.5 wt% loadings.

Other than graphene, the carbon nanotube is another popular carbon-based nanofillers used to enhance the dielectric properties of polyurethane elastomer. Tayfun et al [23] synthesized carbon nanotube with polyurethane and did some surface modification towards the carbon nanotube using nitric acid and also silane treatment. The results show the increment of the dielectric constant for all types of carbon nanotube fillers, with the dielectric constant increasing with the content of the filler. The addition of carbon nanotube filler increases the free volume of the polymer structure, creating easier orientation of dipole groups along the field which causes an increase in the dielectric constant. However, there is no significant difference in terms of a dielectric constant between the pristine carbon nanotube filler and the modified carbon nanotube filler.

3.2 Ceramic-Based Nanofillers

Inorganic filler such as ceramic possesses high permittivity values which, in principle, should be able to increase the dielectric constant of a given elastomer while maintaining its dielectric nature when synthesized together. Several research incorporate other inorganic fillers such as titanium oxide (TiO) [24–26], barium titrate (BT) [27–28], zinc oxide [29–30] and strontium titanate (ST) [31] to enhance the dielectric of the elastomer.

Calcium cooper titanate (CCTO) provides interesting features when being incorporated with polyurethane matrix. Wan et al [32] investigated the dielectric properties of CCTO filled polyurethane

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composites. The dielectric permittivity values of CCTO with 10, 20, 30, and 40 were determined as ~13, 17, 26, and 33, respectively, which correspond to 63%, 113%, 225%, and 313% higher than that of neat TPU respectively at 1 kHz. Increasing the CCTO content in the composite caused a smaller distance between inclusions that led to more effective dipole interactions and higher charge storage capability. Other than that, zinc oxide (ZnO) is also a promising nanofiller that can be used to enhance polyurethane dielectric properties. Kaur et al [33] reported the dielectric constant of the polyurethane increased with the ZnO doping concentration. At 1 kHz, the dielectric constant of ZnO-polyurethane elastomer can reach up to 7 at a 15% doping concentration of ZnO. Another ceramic-based filler that provides excellent dielectric constant improvement is calcium copper titanate (CCTO).

3.3 Hybrid Nanofillers

A hybrid nanofiller means a combination of two or more types of nanofillers. As discussed earlier, both carbon-based and ceramic-based nanofillers can enhance the dielectric performance of polyurethane elastomer. However, both nanofillers have their disadvantages. The main drawback with carbon-based nanofiller is the high energy dissipation associated within the nanofillers due to high current leakage resulting from direct contact of nanofillers and high mobility of charge carriers [34]. Nanofillers such as graphene, carbon black and carbon nanotube tend to easily stack and agglomerate, forming conductive pathways, and thus diminishing the dielectric strength [35–37]. On the other hand, ceramic nanofillers usually require high loading fractions to effectively improve ε and thus produce a large increase in elastic modulus. Hence, the resulting composites usually suffer from a loss of flexibility and processability. Hybrid nanofillers such as a combination of carbon-based and ceramic-based nanofillers can overcome these problems as each nanofillers can complement each other.

Chen et al [38] took an initiative by fabricating a hybrid polyurethane-elastomer with reduced graphene oxide-titanium dioxide (RGO-TiO2) functionalized filler and polyurethane matrix and comparing them with the reduced graphene oxide-polyurethane composite (RGO-PU). Under SEM characterization, phase separation was observed in RGO-PU, attributed to the poor dispersion and serious graphene aggregation. On the contrary, hybrid nanocomposites seem to be homogeneously distributed in the polymer matrix. Thus, the dielectric constant of the hybrid-polyurethane elastomer is higher than the RGO-PU elastomer at the same weight percentage. Figure 5 shows the dielectric constant of RGO-PU elastomer and RGO-TiO2-PU elastomer at a 1:1 ratio (1G1T-PU) at various temperatures and 3 wt%.

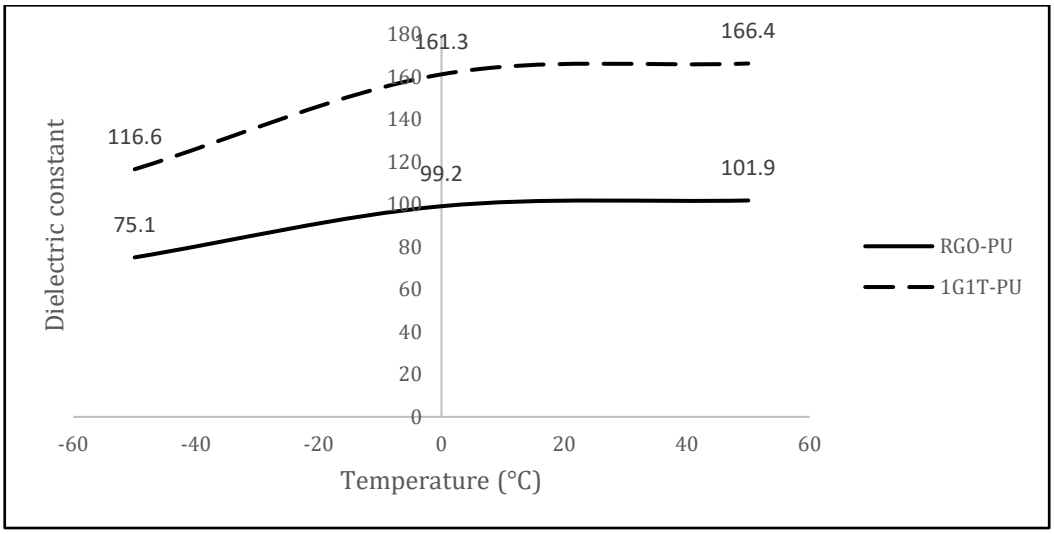

Figure 5. Dielectric constant of RGO-PU elastomer and RGO-TiO2-PU elastomer at various temperatures (at 3 wt%) [38]

Chen et al [39] investigated the dielectric properties of PU dielectric elastomer with carbon nanotube (CNT) and graphene (GRN) nanofillers. CNT-PU, GRN-PU, 1C1G-PU (1CNT: 1GRN), 1C2G-PU (1CNT: 2GRN), and 2C1G-PU (2CNT: 1GRN) at different weight percentages (0.5, 1.0, 1.5, 2.0 and 2.5 wt%) were prepared and examined. The results from the scanning electron microscope (SEM) showed that the fracture surface of the hybrid composites is very smooth, indicating the homogenous dispersion of the nanofillers. Consequently, hybrid composite possesses a higher dielectric constant. 1C1G-PU composites

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possess the highest dielectric constant (≈ 275 at 1 kHz) compared to other composites. For the CNT-PU, the aggregation of CNT still serves as the electrode material to form a micro-capacitor network in the matrix for increasing the dielectric constant. However, the number of micro-capacitors is restricted because of the aggregation of CNT. As to GRN-PU composites, there is an increment of dielectric constant as well, but not as significant. As to hybrid filler PU composites, the mixture of hybrids improves the dispersibility of GRN and CNT in the polymeric matrix which allows the construction of more micro-capacitors in GRN-CNT@PU composites, leading to a higher dielectric constant.

4.0 POTENTIAL APPLICATION OF POLYURETHANE DE

High dielectric performance of PU elastomer makes it very reliable to be used as both DEG and DEA. This part will underline a few applications that are suitable for the excellence of PU elastomer.

4.1 Energy Harvesting

DE are highly versatile and can work as both actuators and generator, due to its properties of high energy density, large deformation and good electromechanical conversion efficiency that can open various possible applications in the industry and one of them is the energy harvesting technologies [40], [41]. Pioneered by Pelrine et al. in 2001 [42], basic principles of using DEs for-energy harvesting are set and since then, numerous methods have been investigated. Moretti et al [43] investigated the application of DE in wave-energy converter (WEC) technologies with an oscillating water column (OWC) system. The structure of the OWC system consists of a partially submerged hollow structure, with an upper part forming an air chamber and an immersed part opened to the sea action as illustrated in Figure 6. OWC system utilizes the wave-induced pressure oscillations to induce the reciprocating motion of the water column inside the chamber, causing compression and expansion of the air entrapped in the chamber.

DE is suggested to be used as the power take-off (PTO) system that converts the pneumatic power into beneficial electrical energy. This paper reported the result of the generation test on the prototype that shows the application of DE in the OWC system provides encouraging results for possible larger-scale energy harvesting applications. However, further fundamental steps are required for DEGs to become a viable technology for wave energy PTO systems including the synthesis of advanced dielectric materials with enhanced dielectric strength in which PU elastomer gives a very brilliant performance.

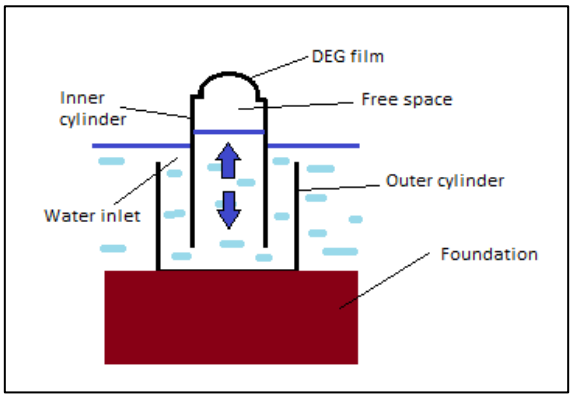

Figure 6. Illustration of oscillation water column (OWC) system [43]

Other than a wave-energy converter, another potential energy harvesting system that can apply DEs in its system is a wind-power generator or wind turbine. Zhang et al [44] utilized Vibro-impact DEG (VI DEG) system in wind energy harvester by proposing a model with VI DEG. Acrylic membranes were embedded into a two-blade turbine to harvest wind-induced rotational energy from low-speed wind environments. The result is outstanding compared to other unconventional wind energy harvesters. It is found that the proposed system can achieve better maximal output power up to 0.7125 mW under a wind speed of 3.99 ms-1. This proves the suitability and workability of DE in energy harvesting applications.

Zhang et al [45] proposed a novel contact-type DEG which can harvest energy from two dielectric elastomer membranes (DEMs) under a regular contact-type displacement excitation. The energy harvesting performance of the CT DEG in simulated contact environments is investigated and within the

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correct excitation, system structure, pre-stretched ratio and input voltage, the system can produce an output voltage 10 times higher than the input voltage, and the system output power can be achieved as high as 22.94 mW under a given displacement excitation.

4.2 Artificial And Robotic Muscle

Another application that suits polyurethane elastomer attributes as a DEA is the artificial and robotic muscle which has been investigated and explored in a few works of literature [46–49]. In the marine robotic industry, DEA has been vastly investigated to be used in soft robotic field for marine application. According to Christianson et al, traditional underwater robots are usually driven by propellers or jet thrusters, which generate considerable noise and vibration. This additional noise is especially problematic when studying elusive animals or when studying underwater acoustics. Nevertheless, they consume a large amount of power [50]. Shintake et al. [51] stated that DEAs are compliant, highly responsive, efficient and exhibit large actuation strokes, which make them suitable to be applied for biomimetic underwater robots. Gu et al. [52] also stated that DEAs usually have the approximate density to the water, which makes it easier to operate with natural buoyancy.

Otherwise, a significant amount of power will be consumed by maintaining the equilibrium of the robots underwater. Godaba et al [53] in their paper discussed regarding marine jellyfish robots as unmanned underwater vehicles for valuable functions, such as study of marine life, exploration of sea and seabed conditions, monitoring of ocean currents, detection of ocean intruders, etc. Traditional actuators such as electric motors and electromagnetic actuators have been popularly employed to achieve jellyfish-like morphology and movement for easier navigation. Although traditional actuators can achieve jellyfish-like morphology and movement, there are still weaknesses such as small deformation, consumption of large electric energy, low energy density, high noise, and slow response. This is something that can be overcome with dielectric elastomers actuators (DEAs) and has been agreed upon in other literature as well.

4.3 Soft Sensors

As a polymer that possesses a high dielectric constant even at a pristine state, polyurethane elastomer can be developed into a high sensitivity soft sensor with some enhancement such as adding functionalized fillers. One of the applications of soft sensors is to safely monitor soft movements or interactions with humans. It is used to measure strain, pressure, force, light, humidity, and temperature like human skin, which has advantageous properties such as flexibility, stretchability, high sensitivity, and technological compatibility with a large area. The sensitivity of the elastomer sensors highly depends on the dielectric constant. The higher the dielectric constant, the higher the sensitivity of the dielectric elastomer sensor [54–56]. Ren et al [57] proposed a facile, high-performance, and cost-effective strain sensor using the carbon nanotubes (CNTs)/thermoplastic polyurethane (TPU) with an aligned wave-like structure. The strain sensor is prepared through the electrospinning technique and the sensing properties of CNTs/TPU mats in vertical and parallel directions were investigated. The results show that the aligned CNTs/TPU fibrous mats in the vertical direction possessed an ultra-high stretchability (900%) and excellent durability (10,000 cycles at the strain of 200%). An ultra-low detection limit (0.5%) and fast response time of 70 ms were also achieved, exhibiting a favourable sensitivity. In this study, the strain sensor was proposed to be applied in monitoring human motion such as cheek bulging and phonation and even vigorous motion like leg squatting and elbow bending.

Zhuang et al [58] proposed a flexible thermoplastic polyurethane (TPU)/CNT polymer composite for selective laser sintering (SLS) processing pressure sensors. TPU/CNT composites were prepared with different conductivity and piezoresistive properties. Piezoresistive performance and percolation theory results prove that the composite shows the best pressure sensing ability of 0.549 kPa-1 for 17-240 kPa pressure at 0.25 wt% CNT-containing TPU/CNT composite, and it was successfully used as a sensor to detect plantar pressure distribution in a human foot. Ke and his team [59] fabricated pressure sensors using TPU matrix and hybrid-nanofillers of CNT and graphene nanoplatelets (GNP) at various compositions to tune the composite dielectric properties. In a composition of CNS and GNP at a mass ratio of 3:1, the composite sensor shows the highest pressure-sensitivity of 2.05 MPa-1 for 0-1.2 MPa pressure, compared with 0.18 MPa-1 for neat TPU, enabling potential wearable pressure sensor applications.

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5.0 CONCLUSION

Polyurethane is an interesting dielectric material that possesses both excellent dielectric and mechanical properties. With a proper enhancement method, the polyurethane-based dielectric elastomer can achieve even better properties that can fulfil the demands of the industry. Functionalized fillers such as graphene, carbon nanotubes and barium titrate are favourable nanofillers to enhance dielectric elastomer performance. Future study on improving the polyurethane matrix should also be established. Although the nanofillers are proven to be effective in improving the DEs dielectric performance, there are still few drawbacks from its application in terms of electrical conductivity and mechanical performance. Thus, the enhancement of the matrix itself may help in improving the DE performance, reducing on the reliability towards nanofillers properties.

6.0 ACKNOWLEDGMENT

The authors fully acknowledged Ministry of Higher Education (MOHE) and National Defence University of Malaysia (NDUM) for this research feasible. The authors also would like to thank the technical staff in every institution involved for their assistant.

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