MULTIFUNCTIONAL COMPOSITES FOR ENERGY STORAGE

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ABSTRACT

Due to the increasing greenhouse gas emissions and gradual run out of fossil fuels, there is a growing concern on the environmental protections and global energy demands in the world. Therefore, new energy storage technologies have been continuously developed to be integrated with renewable energy systems in recent years. On the other hand, advanced composites have become more and more popular in automotive and aerospace industry because of their significant advantages such as the high specific strength to weight ratio and non-corrosion properties. Thus, the cost and energy for transportation, maintenance and fabrication of composite structures with a complexed shape could be reduced. Todays, research interests in developing multifunctional materials in order to reduce the fuel and energy consumption have increased significantly. Therefore, this paper is focused on the development of the multifunctional energy storage systems. The concept of multifunctional energy storage systems, followed by the introduction of structural dielectric capacitors are introduced. Then, the experimental findings, in terms of the improvements on mechanical and electrical properties of the structural dielectric capacitors, conducted by other researchers are given. In addition, it has been proven that the structural dielectric capacitors could maintain their capacitive function under a mechanical loading. Lastly, existing challenges that would be faced in the realization of the structural dielectric capacitors are discussed.

1 INTRODUCTION

Fuel consumptions and CO_2 emissions are major concerns in the entire aerospace and automotive industry. To reduce the use of fuel for flying and on-road vehicles, more lightweight designs such as using advanced composites for structural parts have been adopted in recent years. The significances of being lightweight, mechanically strong and resistant to corrosion make the advanced composites very popular for structural applications. Usually, they are made by strong fibre cushioned with weak matrix to form a new material with a high stiffness-to-weight ratio. Matrices including polyester, vinyl-ester and epoxy, and fibre reinforcements such as glass fibres, carbon fibres and natural fibres are commonly used for the advanced composites. Apart from the transportation sector, the advanced composites are also employed for supporting components for energy storage systems, such as fly-wheels systems, to improve the overall efficiency through the reduction of their mass and ultimately enhancing the mechanical performance.

On the other hand, the environmental issues mentioned above also motivate the rapid development of renewable energy technologies in a conjunction with energy storage systems. In general, traditional energy storage systems are heavy, corrosive and easily damaged, resulting in short lifetimes. For instance, lead acid batteries are commonly used in industries because they are the most economical for large power applications, in where weight is less concerned. However, they are not environmentally friendly due to the corrosive electrolytes and lead used as constituents. In addition, the applications of the lead acid batteries are restricted in the automotive industry because they are large and heavy. Therefore, more research works have appeared along the line of innovating new advanced energy storage technologies.

Recently, the creation of multifunctional materials opens a new research area in the advanced energy storage systems [1]. The multifunctional material is defined as a material with an integration of structural and non-structural function. The combination of functions and properties into an individual structure offers significant advantages of weight and energy saving. The non-structural functions can be optical, electrical, magnetic, thermal etc. Thus, the multifunctional materials have a great potential to be applied in many areas, for examples healthcare, security, energy, and packaging and aerospace. These kinds of materials would bring engineered systems to a higher level with a better functionality and adaptability in the future. Therefore, the realization of the multifunctional materials raises the greatest attention in different research and engineering fields.

From the multifunctional perspective, carbon fibre reinforced composite (CFRP) plays a crucial role in developing a load-carrying structure with the capacitive function. The attraction of using carbon materials to replace conventional conductive materials as electrodes is arisen from their unique combination of high electrical conductivity and surface area range, as well as their comparably stability in different chemical solutions, which are acidic, basic or aprotic, and various thermal conditions [2, 3]. Embedded battery composites (EBC) and structural dielectric capacitors are typical examples of electrical energy storage technologies by using CFRP [4-6].

The purpose of this paper is to summarize the state of knowledge on the multifunctional materials for structural dielectric capacitors. The experimental results shown in literatures and common methodologies to optimize the overall performance of the multifunctional materials are also presented. Moreover, the technical challenges in the development of the structural dielectric capacitors are discussed at the end of this paper.

2 MULTIFUNCTIONAL ENERGY STORAGE SYSTEMS

Conventional dielectric capacitors have lower energy densities as compared to batteries and supercapacitors. However, they have higher power densities and longer lifetimes than those of batteries and supercapacitors because there is no redox reaction but only electron transmissions involved in the charging and discharging process of the dielectric capacitors. Moreover, with the development of the multifunctional composites, using CFRP for electrodes in the dielectric capacitors allows electronics to be integrated into structures, leading to a space saving [7]. As a result, multiple carbon fibre layers in the structural dielectric capacitors could provide a more extensive capacitance than conventional energy storage methods.

The energy and power densities of energy storage systems are important parameters in the electrical characterization. Most of the research works have been focused on achieving the high capacity-to-volume ratios of the conventional batteries and capacitors, rather than investigating their mechanical performances. Since the multifunctional composites are proposed for structural applications, the mechanical performance is becoming as important as their electrical performance. Therefore, both mechanical and electrical properties of the multifunctional composites should be investigated to analyze their multifunctional efficiencies. The multifunctional efficiency (η_{mf}) is the boundary of defining a mass-saving multifunctional design, as shown in Equation (1) [5]. In the analysis, the electrical efficiency (η_e) is the ratio of the specific energy density of the multifunctional composite to that of the conventional capacitor while the mechanical efficiency (η_s) of the multifunctional composite is the ratio of the specific tensile property of the multifunctional composite to that of the conventional structure. If the multifunctional efficiency exceeds unity, there would be a weight reduction achieved by the new system as compared to the conventional system, which consists of capacitive and structural components. In the following sections, the introduction of the structural dielectric capacitors and the overviews on modifying their mechanical and electrical properties are given.

$$\eta_{mf} = \eta_e + \eta_s > 1 \tag{1}$$

3 STRUCTURAL DIELECTRIC CAPACITORS

In a dielectric capacitor, electrostatic energy is stored in the form of electric charges at two or more electrically conductive plates, which are separated by a dielectric material, presented in Fig. 1. The purposes of the dielectric separator are to avoid any contact between two conductive plates and to obtain a high capacitance due to the dielectric polarization. As mentioned above, only electron transmission takes place in the charging and discharging process of the dielectric capacitors. As a result, their cycle life and recharge function are longer and faster as compared to a conventional battery [8]. Furthermore, electrical energy can be stored in dielectric capacitors over long charging time and discharged within a short period of time in a controlled condition [9]. Since the energy density of a traditional dielectric capacitor is relatively low, it is usually used for flattening short-term variations in a flow of electrical energy rather than a bulk energy storage [10]. The capacitance is highly depended on an overlapping area of conductive plates and the separation between them [9]. The theoretical capacitance and energy density can be calculated from Equation (2) and (3) respectively. With the development of the multifunctional composites, more energy can be stored in the structural dielectric capacitors and the multifunctional characters could expand their applications to the automotive industry and unmanned aerial vehicles (UAV).

$$C_{theoretical} = \frac{\varepsilon_r \varepsilon_0 A}{d} \tag{2}$$

where ε_0 is an absolute permittivity equals to 8.86 x 10^{-12} (F/m); ε_r is a relative dielectric constant of separator ($\varepsilon_r \geq 1$); A is an overlapping area of two electrodes (m^2) and d is a dielectric thickness (m).

$$\bar{\Gamma}_{SC} = \frac{\frac{1}{2} C V^2}{m_{SC}} \tag{3}$$

where C is a capacitance of a capacitor (F); V is an electric breakdown voltage (V); m_{SC} is the total mass of a capacitor (kg).

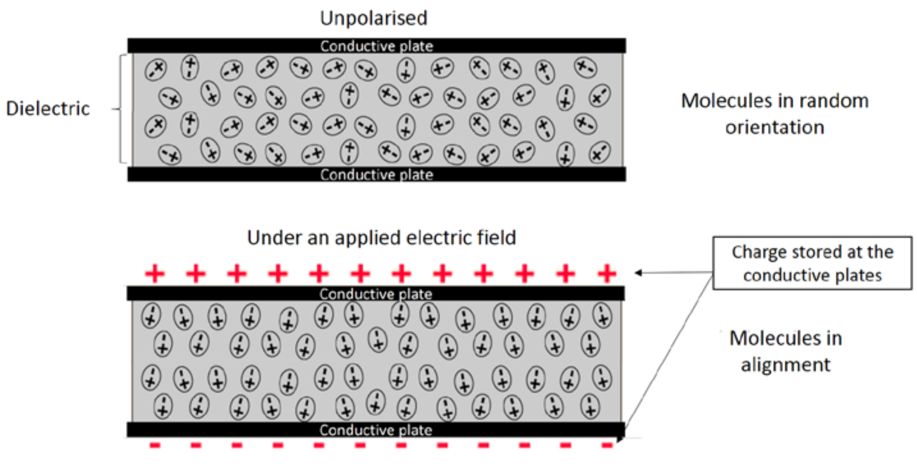

Figure 1: Configuration of a conventional capacitor

In most structural dielectric capacitors, CFRP is adopted to serve as electrically conductive plates (electrodes) as well as a structural support. In previous studies, two different existing structural dielectric capacitor configurations, shown in Fig. 2, were investigated. For a glass fibre insulating capacitor, epoxy matrix are used as a binder as well as a dielectric while a glass fibre weave acts as an insulator; for a polymer film dielectric capacitor, epoxy functions as a binder and a polymer thin film used as a dielectric. The main difference between an insulator and dielectric is their ability of polarization, which influents the dielectric constant of a material. The former is usually used as an electric obstruction due to its low electrical conductivity; the latter is used as an indication of the energy storage capacity due to its high polarizability. This kind of lightweight design incredibly reduces energy consumptions in operations. Although the energy density of the structural dielectric capacitors is relatively low as compared to other structural energy storage systems such as embedded battery composites [4, 11] and structural supercapacitors [12], a high consistency of constituents in an entire structure and the use of the structural epoxy give a better mechanical performance. Thus, it could aim to be a load-carry composite for energy storage applications such as advanced structures of automobiles and unmanned aircraft vehicles [7, 13].

The potential of the structural dielectric capacitors for load-bearing composites have been demonstrated since 1999 [14]. However, materials such as epoxy and glass fibres utilized in major studies have relatively low dielectric constants, and consequently low energy density and multifunctional efficiency. To optimize the overall performance of the structural dielectric capacitors, both dielectric permittivity and bonding strength between electrodes and dielectrics have to be modified. In the following section, both developed and possible improvements on the mechanical and electrical properties of the structural dielectric capacitors are discussed separately, followed by mechanical influences on the electrical performance of the structural dielectric capacitors.

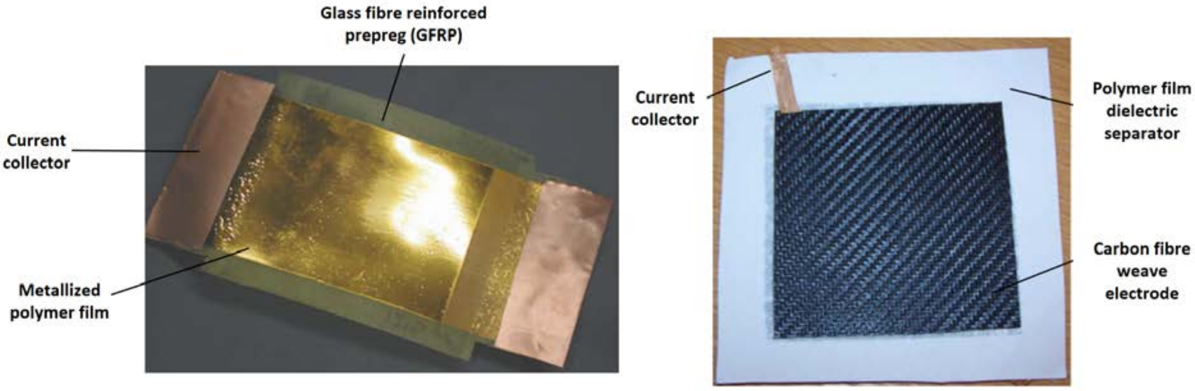

Figure 2: Common configurations of current structural dielectric capacitors: Glass fibre insulating capacitor (left) and polymer film dielectric capacitor (right) [15, 16]

3.1 Mechanical properties

Interlaminar shear strength (ILSS) and tensile properties are the most important factors in the structural characterization of this kind of multifunctional composite. Since the structural dielectric capacitors are made of laminates, delamination is the major failure mode and restricts their mechanical properties, especially the tensile properties. Therefore, a good interfacial adhesion is one of the most important factors in providing an excellent stress distribution between constituents of the structural dielectric capacitors to achieve the high mechanical performance.

Carlson et al. [17] carried out mechanical characterizations on the PET-film based structural dielectric capacitors with different dielectric thicknesses, shown in Table 1. It is obvious that there is a great knockdown in the tensile stiffness and ultimate tensile strength (UTS) of specimens as compared to the reference value of CFRP. Meanwhile, a decreasing trend of tensile stiffness with increasing the dielectric thickness is also observed. It may be due to the presence of a weak polymer film between CFRP electrodes and the delamination behavior between electrodes and dielectric separators, shown in Fig. 3. Besides, a similar study on investigating the influence of interleaving films on the mechanical properties of CFRP was implemented by Kim and Lee [18] and they pointed out that the reduction in tensile strengths was caused by decreasing the fibre volume fractions of the interleaved specimens, which led to the low mechanical performance.

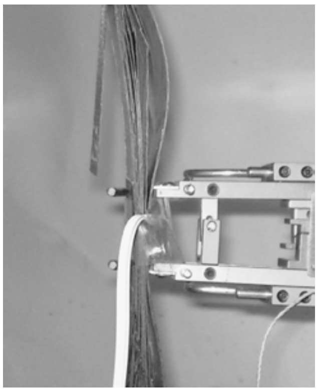

Figure 3: Failure modes of a tensile specimen [17]

Yavirach et al. [19] focused on studying the shear strengths of composites and polymers with different plasma treatments including oxygen, argon, nitrogen and a mixture of helium and nitrogen plasma groups. They showed that polar functional groups could be generated on the polymer surface, leading to the increases in surface wettability and tensile-shear bond strength. For the structural dielectric capacitors, Carlson et al. [6, 16, 17] tried to modify the interfacial interaction between CFRP electrodes and dielectrics by using a N_2 plasma activation. From Table 1, there is an obvious drop in ILSS values of the interleaved specimens as compared to the reference CFRP because of the poor adhesion property between CFRP electrodes and the PET film. However, there is no significant improvement on ILSS of the PET film specimens before and after the plasma treatment. It may be concluded that the N_2 plasma activation is not a proper method to modify the surface properties of the PET film. Since the interfacial bond between electrodes and dielectrics is a critical point in developing the structural dielectric capacitors, other possible surface treatments should be developed to enhance the surface adhesion between two phases, resulting in the high mechanical performance without any compromise on the electrical properties.

Dielectric thickness (mm)	Plasma treatment	Young's modulus (GPa)	Ultimate tensile strength (MPa)	ILSS (MPa)
50µm	No	42.7	354	29.5
50µm	Yes	42.5	320	32.0
75µm	No	44.6	377	30.6
75µm	Yes	41.7	344	30.7
125µm	No	36.5	317	32.5
125µm	Yes	37.8	339	31.8
Reference CFRP	-	56.1	631	54.4

Table 1: Mechanical properties before and after the plasma treatment of the PET-film based structural dielectric capacitors [17]

3.2 Electrical properties

Electrical improvements in the structural dielectric capacitors can be divided into two parts: improving the electrode properties and dielectric properties. To study the effects of electrode characteristics, O'Brien et al. [20] carried out a comprehensive evaluation on the electrical properties by using thin metallized films of paper, polyimide (PI) and biaxially oriented polypropylene (BOPP) for the structural electrodes and GFRP as the dielectric, shown in Fig. 4. The metallization of either Al, Zn or Zn-Al alloy in various thicknesses from 4.3 to 12 nm was applied on the electrode films. A correlation was found that the energy density increased with increasing the surface resistivity of the metallized film. In addition, Chen [21] indicated that the electric breakdown strength (EBD) could be enhanced with an appropriate surface resistivity by balancing the capability of self-healing and electrical conductivity in the metallized electrodes. Therefore, it is proven that thickness and surface resistivity of electrodes are key parameters on the electrical performance. It is also expected that the fibre volume fraction of CFRP electrodes used in another type of the structural dielectric capacitors could be a factor to achieve the high electrical performance.

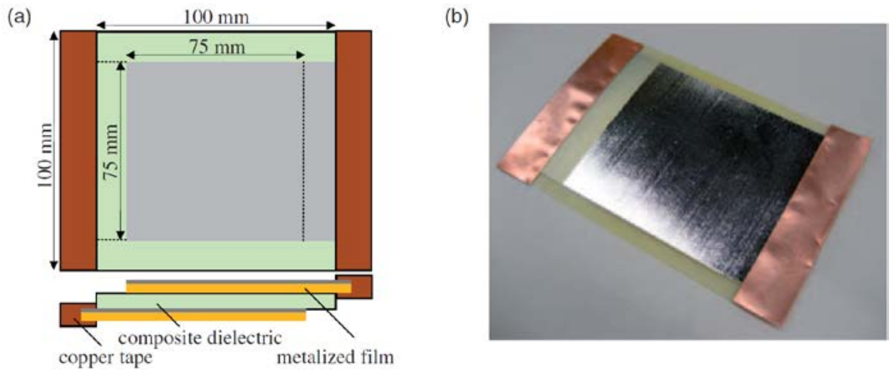

Figure 4: Configuration of capacitor using metalized polymer films (left) and a woven glass/ FR4 epoxy prepreg sample (right) [5]

Besides, the selections of dielectric materials and thicknesses are also important for the electrical properties. At the beginning, Luo and Chung [7] demonstrated the feasibility of using CFRP electrodes and different papers as dielectric separators in the structural dielectric capacitors. Later, Carlson et al. [16] and O'Brien et al. [5] developed other multifunctional dielectric capacitors by using various polymer thin films and GFRP as dielectrics respectively. Their experimental values of the electrical properties of the structural dielectric capacitors are presented in Table 2. From the table, it is observed that the average energy density of polymer films based structural capacitors is almost 80 times more than that of papers. This is because of the superior EBD of polymer films as compared to impregnated papers. In addition, although all GFRP specimens have lower values in both capacitance and EBD than those of the PET film specimen, their energy density (refer to Equation (3)) are higher as compared to the PET film specimen. This may be attributed to the extremely thin metallized electrodes used, resulting in a low density of this kind of structural dielectric capacitor.

Furthermore, Carlson et al. [17] studied the electrical properties of the PET-films based structural dielectric capacitors in various dielectric thicknesses (50, 75 and 125 μm). Table 3 shows that experimental capacitances decreases with increasing the thickness of dielectric separators. In addition, the 125μm PET-film specimen has the highest breakdown voltage but the lowest EBD as compared to 50μm and 75μm PET-film specimens because the volume of the 125μm PET-film specimen is larger than those of the 50μm and 75μm PET-film specimens. As a result, there is a high possibility of having flaws, which initiate an earlier dielectric breakdown, in the 125μm PET-film specimen. Although the capacitance of the 125μm PET-film sample is the lowest, its specific energy density is still the highest as compared to the 50μm and 75μm PET-film specimens due to the compensation from the relatively high breakdown voltage.

Another way of modifying the dielectric properties is to introduce nanoparticles into the dielectric materials. Lovell [22] studied the effect of reinforced fillers on dielectric properties of epoxy polymers and determined that the mismatch of permittivity between fillers and matrix reduced EBD due to the field enhancement. Therefore, the addition of nanoparticles into the matrix-phase of reinforced composites can rematch the dielectric properties between reinforcement and matrix, and consequently eliminate the local intensification, which initiates the dielectric breakdown. Moreover, dielectric permittivity of the polymer matrix can be increased by the addition of nanoparticles, which have high dielectric constants, resulting in the high electrical performance. Typical nanoparticles including Al_2O_3, TiO_2, $BaTiO_3$, and carbon nanotubes (CNT) are commonly used for enhancing dielectric permittivity of epoxy-based nanocomposites [23-32]. Therefore, it shows the potential of using nanoparticles to improve the electrical performance of the structural dielectric capacitors.

Electrode material	Dielectric material	Thickness (mm)	Capacitance (μF/ m^2)	Electric breakdown strength (kV/mm)	Energy density (J/g)	Ref.
CFRP	Paper (40 g/m^2)	0.071	0.712	7.75	0.00013	[16]
CFRP	Paper (80 g/m^2)	0.089	2.466	8.76	0.00088	[16]
CFRP	Paper (150 g/m^2)	0.173	0.766	9.77	0.00120	[16]
CFRP	PA film	0.050	0.868	156	0.03400	[16]
CFRP	PET film	0.019	1.860	337	0.05200	[16]
CFRP	PC film	0.155	0.206	183#	0.08900#	[16]
Metallized polymer film	GFRP (106) (V_f = 20.6%)	0.140	0.286	164	0.34	[5]
Metallized polymer film	GFRP (1080) (V_f = 27.9%)	0.160	0.256	152	0.28	[5]
Metallized polymer film	GFRP (2116) (V_f = 33.4%)	0.150	0.293	165	0.34	[5]
Metallized polymer film	GFRP (120) (V_f = 49.5%)	0.110	0.444	63	0.05	[5]
Metallized polymer film	GFRP (7781) (V_f = 61.6%)	0.220	0.231	107	0.17	[5]

The samples had not failed at maximum voltage in the breakdown tests

Table 2: Summary of results for various structural capacitors

Dielectric thickness (μm)	Capacitance (μF/ m^2)	Breakdown voltage (kV)	Electric breakdown strength (kV/mm)	Energy density (J/g)
50μm	0.447	14.6	292	0.06
75μm	0.300	22.4	299	0.08
125μm	0.193	29.4	235	0.09

Table 3: Electrical properties of the PET-film based dielectric capacitors [17]

3.3 Mechanical influences on electrical performance

The structural dielectric capacitors are the utilization of capacitive elements as the load-bearing structure. As a result, mechanical influences, such as tensile and compression loadings, in real-life applications may alter the capacitive function. It is therefore important to analyze the effect of the mechanical stresses on the electrical performance.

Nishijima and Hara [33] carried out a comprehensive study to investigate the dielectric properties of insulants, which were GFRP and PET, under compressive and tensile stress testing, as presented in Fig. 5. In the compressive stress test, a mechanical stress was applied in the thickness direction of the insulants. EBD firstly increases with increasing the compressive loading and then decreases after reaching a maximum point. It is suggested that the decrease in EBD at the high stress level was caused by the formation of voids and cracks. In the tensile loading test, both EBD of GFRP and PET decreased with increasing the stress level. The difference on EBD between compressive and tensile stresses may relate to the change of modes of crack initiation and propagation. This is because the cracks, which are perpendicular to the applied stress direction, grow easier under a tensile stress than those under a compressive loading. Consequently, the average EBD in the tensile loading condition is lower than that in the compressive loading condition.

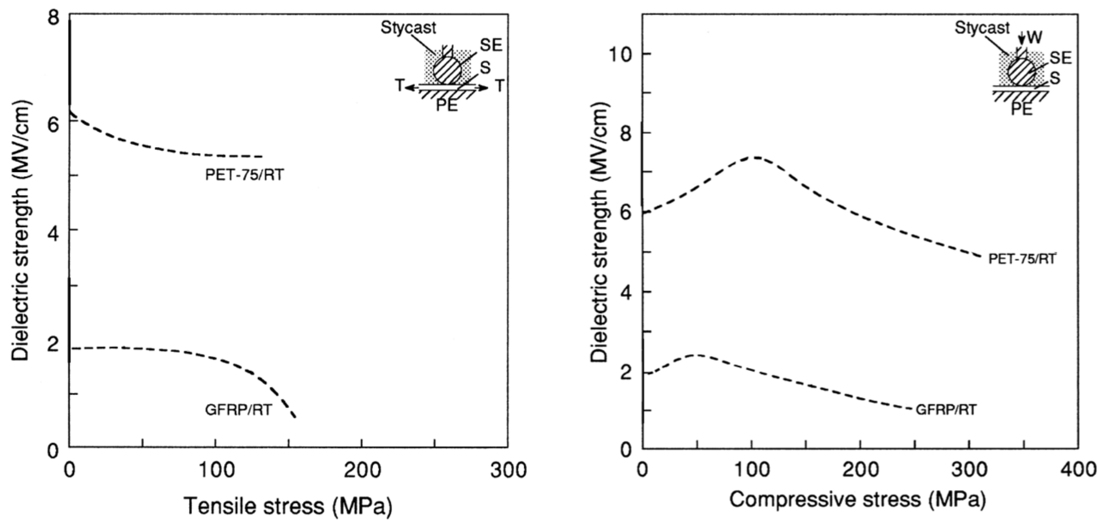

Figure 5: Effects of tensile and compressive stresses on EBD at room temperature [33]

In addition, Carlson et al. [34] studied the effect of mechanical loads on capacitance of the PET-film based structural dielectric capacitors. A force was applied in a tensional direction of the specimens to introduce intralaminar matrix cracks inside CFRP electrodes and measurements of capacitance were taken before, during and after the tensile loading. The electrical performance of undamaged and damaged specimens were analyzed, recorded in Table 4. The capacitance of specimens at the loaded state is 50% less than that of undamaged specimens because air, which has a lower relative dielectric constant than polymer, filled in those opening matrix cracks. Unexpectedly, the capacitance of the unloaded specimens is recovered and even higher than that of undamaged specimens. It may be attributed to a permanent plastic deformation, which led to a reduction in the dielectric thickness, under a tensile loading. More importantly, this result shows that the structural dielectric capacitors can retain the electrical functions in the loading condition.

Dielectric	Capacitance (μF/ m²)		
	Before loading	Under loading	After loading
50μm PET-film NaOH treated	0.008	0.004	0.011

Table 4: Measurement of capacitance in different states [76]

4 Challenges and further developments

For the GFRP based structural dielectric capacitors (refer to Fig. 2), epoxy, which is a crosslinking polymer, acts as a dielectric and supportive material. Therefore, the structural properties could be enhanced at the high crosslink degree. However, there is a loss in the polarizability due to the formation of large epoxy chains, which would be more difficult to shift their equilibrium positions to cause the dielectric polarization, resulting in the low energy and power density. The optimal degree of cross-linkage of cured composites could be obtained by comparing multifunctional efficiencies (refer to Equation (1)) of different sets of specimens.

Besides, the energy density of the structural dielectric capacitors is relatively low as compared to other structural energy storage systems. To achieve the high electrical performance, the alternative approach is to introduce nanoparticles into a matrix as a secondary reinforcement for matching the dielectric properties between reinforcement and matrix. It may also improve the mechanical properties such as interlaminar fracture toughness and compression-after-impact [35].

Furthermore, delamination is one of the most concerned failures in the laminated composites and the strength of CFRP is limited by the interfacial properties between laminates [35]. For both GFRP and polymer film based structural dielectric capacitors, the interfacial bond between laminates is the key parameter to maximize the mechanical performance. The introduction of nanoparticles into resin can improve interlaminar strengths of GFRP based structural dielectric capacitors [36]. Meanwhile, other advanced surface treatments should be developed to enhance the interfacial properties between laminates of the polymer film based structural dielectric capacitors without any loss in dielectric properties.

5 CONCLUSION

The multifunctional materials have been continuously developed with the aim to reduce the energy and fuel consumption. The advanced energy storage system made by CFRP is one of the typical examples of realizing the multifunctional materials. In this paper, the introduction of the structural dielectric capacitors, followed by their states of understanding are given. Moreover, the research works in terms of the improvements on mechanical and electrical properties of the structural dielectric capacitors, performed by other researchers are presented. Lastly, the scientific challenges and further development of the structural dielectric capacitors are analyzed.

To realize the structural dielectric capacitors, the most difficult part is to fabricate the multifunctional dielectrics for maximizing the mechanical and electrical properties, especially the interfacial properties, of the structural dielectric capacitors. However, the mechanical and electrical requirements of the multifunctional materials could be different in different applications. Therefore, the criterion of selecting materials for electrodes and dielectrics, including the mechanical and electrical properties, lifetime, cost and ease of fabrication of composites, should be taken into considerations comprehensively in every single application. Last but not least, it is expected that there would be more new multifunctional materials to be developed in the future to bring engineering fields to a higher level.

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