Jurnal
Teknologi

Full Paper

EFFECT OF POLYVINYLIDENE FLUORIDE LOADINGS ON THE MECHANICAL PROPERTIES AND BURNING RATE OF EPOXY COMPOSITES

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Article history
Received
10 April 2023
Received in revised form
21 August 2024
Accepted
3 November 2024
Published Online
26 June 2025

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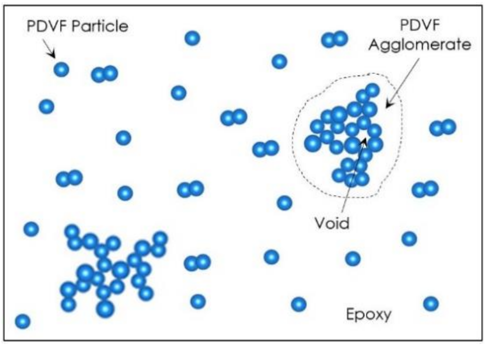
Graphical abstract



Abstract

Polyvinylidene fluoride (PVDF) has excellent mechanical, electrical, and thermal properties, making it suitable for a wide range of industrial and technological applications. PVDF-filled epoxy composites were prepared at 1% to 10% loadings via the casting method to improve the mechanical and thermal properties of epoxy. Tensile strength, tensile modulus, flexural strength, and flexural modulus are the lowest at 1% PVDF filler due to insufficient reinforcement by low filler loading. Besides, the presence of high void content resulting in a drop of tensile strength and flexural strength to lower than the epoxy alone. Strength and modulus increased at 3% and 5% loading, then decline again at much higher loadings. At high loading of PVDF, the strength and modulus are reduced due to the filler agglomerations which are supported by the density and percentage of void content. Besides causing voids inside the composites system, filler agglomerations also act as stress concentration sites.

Keywords: Polyvinylidene fluoride, epoxy resin, tensile properties, flexural properties, filler loadings

Abstrak

Polyvinylidene fluoride (PVDF) mempunyai sifat mekanikal, pengalir electrik dan haba yang sangat baik, menjadikannya sesuai untuk pelbagai aplikasi industri dan teknologi. Komposit epoksi yang terisi PVDF disediakan melalui kaedah tuangan di mana jumlah PVDF yang digunakan adalah di antara 1% hingga 10% untuk menambah baik sifat mekanikal dan pengalir haba epoksi. Kekuatan tegangan, modulus tegangan, kekuatan lentur dan modulus lentur adalah yang paling rendah pada kandungan 1% PVDF disebabkan oleh kesan pengukuhan yang tidak mencukupi oleh jumlah PVDF yang rendah. Selain itu, kehadiran kandungan rongga yang tinggi mengakibatkan penurunan kekuatan tegangan dan lentur menjadi lebih rendah daripada epoksi sahaja. Kekuatan dan modulus meningkat pada kandungan 3% dan 5%, kemudian menurun lagi pada kandungan yang lebih tinggi. Untuk komposit dengan kandungan tinggi PVDF, kekuatan dan modulus berkurang disebabkan oleh aglomerasi PVDF yang disokong oleh data ketumpatan dan peratusan kandungan rongga. Selain menyebabkan rongga di dalam sistem komposit, aglomerasi PVDF juga bertindak sebagai tapak tumpuan tekanan.

Kata kunci: Polivinilidena fluorida, resin epoksi, sifat tegangan, sifar lentur, jumlah pengisi.

1.0 INTRODUCTION

Epoxy composites are widely used in various applications, such as aerospace, automotive, marine, and construction industries, due to their unique combination of properties. The specific properties of an epoxy composite can be tailored by adjusting the type and amount of reinforcing material, as well as the processing parameters used to form the composite material [1, 2]. However, the 3D cross-linking network structure of cured epoxy resin is intrinsically brittle, which lowers its wear resistance. Particulate-filled epoxy composites are a type of composite material that consists of epoxy resin as the matrix and various types of particles as the filler. The addition of particulate particles as reinforcement in the epoxy resin can enhance the mechanical, thermal, and electrical properties of the resulting composites. The size, shape, distribution, and concentration of the particles, as well as the type of epoxy resin used, will affect the attributes of particulate-filled epoxy composites. The curing conditions, like temperature and pressure, can also have an impact on the composite's qualities. Epoxy composites with particulate fillers are used in a variety of industries, including aerospace, automotive, electrical, and biomedical ones. Among other things, they are employed in the production of structural elements, electrical insulators, heat sinks, and medical implants [3, 4].

Polyvinylidene fluoride (PVDF) is a polar polymer with excellent mechanical, electrical, and thermal properties, making it a popular material for a wide range of industrial and technological applications. PVDF is known for its exceptional chemical resistance, UV stability, and resistance to flames, making it a preferred material for harsh environments. Moreover, PVDF has exceptional electrical insulation, piezoelectric characteristics, and great mechanical strength, stiffness, and toughness [5, 6].

Many PVDF-based products, including pipes, films, coatings, and fibres, have been created as a result of these distinctive features. PVDF is a semi-crystalline polymer with a repeating unit of $-CH_2-CF_2-$. The polarity of the fluorine atoms and the carbon-fluorine bond gives PVDF its unique combination of properties [7]. Depending on the processing circumstances, PVDF can exist in a variety of forms, including amorphous phases and alpha, beta, and gamma crystalline phases. The different crystalline phases of PVDF result in different mechanical and thermal properties, which can be tailored for specific applications. PVDF is frequently used as a filler in polymer composites to enhance the final material's mechanical and physical qualities. The stiffness, toughness, and wear resistance of the composite material can be improved with the addition of PVDF particles to the polymer matrix [8, 9]. The use of PVDF fillers has been explored in various polymer matrices, such as epoxy, polyurethane, and polyethylene, for various applications, such as coatings, adhesives, and composites [10-12].

PVDF is widely used for microwave absorption, supercapacitor, and many other fields due to its higher dielectric constant electroactive behaviour compared to other polymeric materials [13, 14]. Apart from that, PVDF finds applications as a flame-retardant additive in the thermoplastic industry. Fluorinated polyolefin PVDF is categorised as UL94 V-0 indicating that it is non-flammable and self-extinguishing. PVDF, with the combination of other additives such as potassium diphenyl sulfone sulfonate and poly (aminopropyl/phenyl silsesquioxane), has been used as flame retardant additives in PC [15] where the improvement in flame retardancy of PC compositions arose from the synergistic interaction of three additives. The synergistic interaction increases the flame retardancy of the PC by reducing the activation energy of decomposition and increasing the thermal decomposition rate to promote the formation of an insulating carbon layer or char on the surface. FTIR analysis of the resulting char showed a highly cross-linking aromatic ester and ether which inhibited the supply of flammable gas and heat transfer.

In a particulate-filled composite, factor such as filler concentration is one of the factors influencing the end properties of the composites, besides filler size and shape, filler-matrix interaction, surface treatment, and others. Dielectric permittivity of the filled epoxy composites with 3.5 vol%, 5 vol%, 7.5 vol% and 10 vol% Barium Titanate, $BaTiO_3$ increase with increasing $BaTiO_3$ content [16], increase in tensile strength, flexural strength, erosive wear resistance, and viscoelastic stiffness of epoxy composites with the increasing amount of wood apple shell particles [17], 12.81 times enhancement in the thermal conductivity along the direction of the magnetic field with the incorporation of 1 – 9 vol% of nickel-coated copper nanowires [18], and others. However, the correlation between the filler loading and the properties of the composites was not as simple. Other parameters such as interfacial interaction, stress transfer mechanism, and filler dispersion can be great influencers on the properties of the composites. Highest tensile properties and flexural properties of the untreated palm kernel cake/epoxy composite obtained at 30% loading. At the same percentage of filler loading, alkali treated palm kernel cake composites exhibited even higher tensile and flexural properties due to the improvement in the interfacial adhesion [19].

Herein, we have reported the inclusion of PVDF powder as a filler reinforcement in the epoxy matrix at a range of filler loading. The tensile properties, flexural properties and burning rate were analyzed as the effect of increasing PVDF loadings. The density, void content and water absorption were also discussed as to support the findings.

2.0 METHODOLOGY

Polyvinylidene fluoride Kynar 761 white powder was purchased from Arkema Inc. with approximately 5 micrometer particle size and bulk density of approximately 0.29 g/cm^3. Epoxy CP362 Diglycidyl Ether of Bisphenol A (DGEBA) and aliphatic amine hardener with a 100:50 mixing ratio were purchased from Oriental Option Sdn. Bhd. with initial viscosity of 13000 cps and 400 cps, respectively. PVDF was incorporated into the epoxy resin at a loading range of 1, 3, 5, 7, 9, 10 wt% and mechanically stirred for 5 minutes at 450 rpm. Hardener was then added to the mixture to initiate the crosslinking process by mechanically stirring for 10 minutes at 450 rpm. The mixture was poured into the mould of 150 mm x 150 mm and cured for 24 hours at room temperature. All formulations were calculated and weighted to produce samples of 5 mm thickness.

Density measurement was performed on the Shimadzu UX420S solid density meter using 20 mm x 20 mm samples. Five replications of each loading were carried out. A water absorption test was conducted using 20 mm x 20 mm samples immersed in the water for 24 hours. The horizontal and 45-degree burning test was carried out using 150 mm x 20 mm samples where the samples were held using a retort stand at their respective position. The distance between the flame and the free-end sample is kept at 2 cm. The samples were exposed to the flame for 60 seconds. The burning time was recorded as soon as the flame was removed until the burning reached the 1 cm mark on the samples. Tensile test and flexural test were conducted using a Shimadzu universal testing machine with a tensile and 3-point bending setup. Both tests were carried out on 150 mm x 20 mm samples at a constant crosshead speed of 5 mm/min at room temperature. Five replications of each loading were carried out. The fractured composite samples were examined using a Digital Microscope Keyence VHX-6000.

3.0 RESULTS AND DISCUSSION

Figure 1 and Table 1 shows the graph and values of measured density and percentage of void content of PVDF/Epoxy composites, respectively. Theoretically, when the PVDF loadings increased, the density of the filled composites should be increased since the volume fraction of the filler which has a higher density than the epoxy is increasing. However, according to Figure 1, the density of the PVDF-filled epoxy fluctuates especially at filler loading ranging from 1% to 7% where the values are close to the unfilled epoxy. The apparent drops in density are obtained at 9% and 10% loading, which could be attributed to increased void formation or agglomeration of PVDF particles. When the filler content is high, PVDF particles might agglomerate, creating larger voids and disrupting the composite's uniform structure, thus reducing the overall density.

Further analysis of the density was carried out by calculating the percentage of void content using Equations 1, 2 and 3 as shown. Equation 1 shows the formula used to calculate the theoretical density (ρ_{th}) and Equations 2 and 3 show the formula used to calculate the percentage of void content (%Void).

$$\rho_{th} = (\rho_m \cdot Vf_m) + (\rho_f \cdot Vf_f) \quad \text{(Equation 1)}$$

$$Vf_{void} = (\rho_{th} - \rho_{ac})/\rho_{th} \quad \text{(Equation 2)}$$

$$\%Void = Vf_{void} \times 100 \quad \text{(Equation 3)}$$

where ρ_m is the density of the epoxy resin (1.16 g/cm^3), Vf_m is the volume fraction of the matrix, ρ_f is the density of the PVDF (1.78 g/cm^3), Vf_f is the volume fraction of the filler, Vf_{void} is the void volume fraction, ρ_{th} is the theoretical density of the composites and ρ_{ac} is the actual or measured density of the composites.

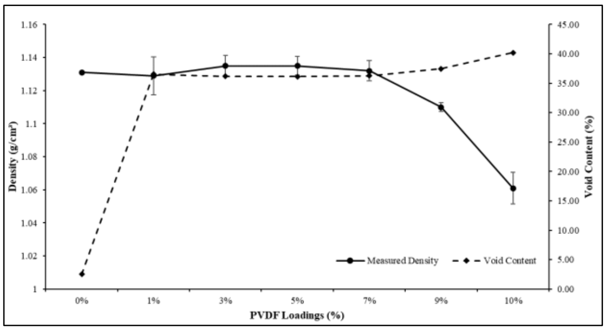


Figure 1 Measured density and the percentage of void content of PVDF/Epoxy composites

Table 1 Values of measured density and the percentage of void content of PVDF/Epoxy composites

PVDF Loadings (%)	Measured Density (g/cm^3)	Void Content (%)
0	1.131	2.50
1	1.129	36.55
3	1.135	36.17
5	1.135	36.12
7	1.132	36.25
9	1.110	37.44
10	1.061	40.18

Roughly, the percentage of void content of the composites can be said, to increase as the filler loading increased. At 1% to 7% loadings, consistent values of void content are obtained. An increasing curve was then obtained at 9% and 10% loadings. The inclusion of filler in the epoxy system can contribute to the presence of void space in the composite, which can cause a reduction in the density. Voids can exist in the epoxy system in at least two ways [20]. First is during the mixing of epoxy resin and the filler where the stirring process will cause a vortex to form in the mixture which creates a localized area of low-pressure causing air to be drawn into the mixture. This will result in the formation of bubbles that will exist in the cured epoxy system if not removed. Action has been taken to minimize the voids by lightly spraying acetone on the surface of the samples before the mixture achieves the gel time. However, since the samples were cured at room temperature, the bubbles located at the bottom and middle of the samples might not be affected by the action and maintained in the cured composites.

Secondly, increasing PVDF loading resulted in increased formation of void content. As the filler concentration increases, the likelihood of stronger interactions between the filler particles grows, resulting in the formation of clusters. PVDF is a highly inert and stable material. Since there is no surface treatment done on the PVDF particles, there is a high possibility of lower interfacial interaction with the epoxy resin due to its inertness. In general, the inclusion of filler in a composite can have both positive and negative impacts on the properties of the composite. On the positive side, the inclusion of filler can result in the enhancement of mechanical properties such as higher stiffness and strength due to the ability of the filler to reinforce the matrix. On the downside, it can result in filler agglomeration which can reduce the adhesion between the filler and the resin resulting in a weaker composite material. Agglomerated filler particles are clusters of individual filler particles that have bonded together as a result of their mutually attractive forces. These clusters of fillers can create voids or spaces within the composites, especially if they are not fully dispersed or if the filler loading is high. This is the second way voids can exist in cured composites. Filler agglomerates also cause inefficient stress transfer from the matrix to the filler particles due to poor filler-matrix interfacial adhesion.

Moreover, increasing PVDF loading leads to an increase in the viscosity of the epoxy resin. When fillers are added to the epoxy resin, they will occupy spaces in between the epoxy molecules which will create a crowded and obstructed environment, thus increasing the viscosity of the resin. During the curing stage, a portion of bubbles or voids inside the composites will normally move upward and escape through the surface. However, since the viscosity of the resin is high, the bubbles especially the small ones are facing a high resistance to moving upward causing them to trap inside the cured composites [21].

Figure 2 shows the rate of water absorption of PVDF/Epoxy composites. In general, the rate of water absorption is increasing with increasing filler loading where the unfilled epoxy exhibited the lowest rate. PVDF is a hydrophobic material which repels water and is not easily wetted by it. Thus, the increasing amount of filler inside the epoxy system should have led to a decrease in the rate of water absorption. The presence of microvoids and pores in the composites system act as pathways for water to penetrate the composites, hence increasing the rate of water absorption [22, 23].

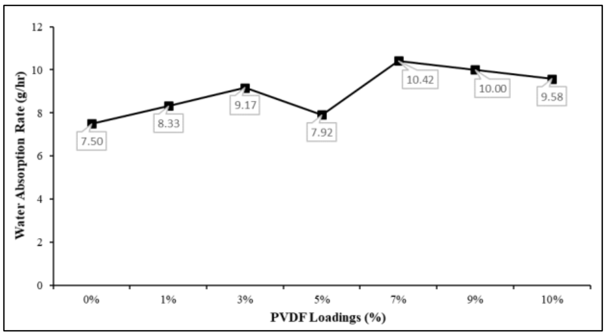


Figure 2 Water absorption rate of PVDF/Epoxy composites

The rate of the burning test is presented in Figure 3 and Table 2, whereas in general, it can be said that the horizontal test showed a higher burning rate compared to the 45-degree burning test. Burning rate is a measure of how quickly a material burns or combusts when exposed to a flame or heat source and is typically expressed in units of length per time. In a horizontal burning test, the sample is placed horizontally and exposed to the vertical flame, causing the flame to spread over the surface of the sample. This test configuration can lead to a higher supply of oxygen for burning which will contribute to more efficient combustion, thus increasing the burning rate.

From the aspect of filler loading, 3% loading showed the lowest burning rate for both test configurations. PVDF Kynar 761 is given the highest classifications (V-O) and 44% LOI (Limiting Oxygen Index), indicating that it is non-flammable and self-extinguishing [24, 25]. Hence, the inclusion of PVDF into the epoxy matrix system has the potential to reduce the burning rate where the PVDF particles will absorb the heat, reduce the heat transfer and prolong the heat propagation through the material. However, the presence of voids inside the composite system may have disturbed the effectiveness of PVDF in slowing down the heat transfer, as occurred in the samples of higher PVDF loadings. Voids can serve as pathways for the propagation of flames which will accelerate the heat transfer and increase the burning rate.

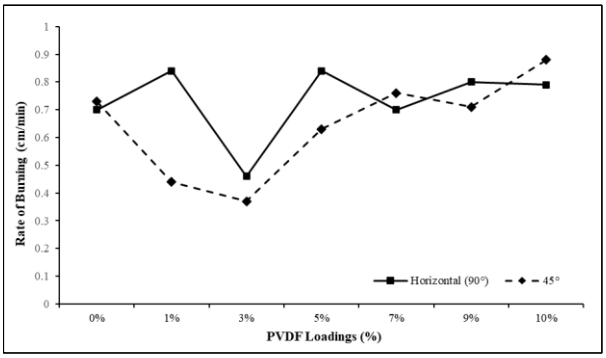


Figure 3 Graph of burning rate of PVDF/Epoxy composites

Table 2 Burning Rate of PVDF/Epoxy composites

PVDF Loadings (%)	Rate of Burning (cm/min)	
	Horizontal (90°)	-45°
0	0.70	0.73
1	0.84	0.44
3	0.46	0.37
5	0.84	0.63
7	0.70	0.76
9	0.80	0.71
10	0.79	0.88

Tensile strength, Young's modulus and tensile strain of the PVDF/Epoxy composites are shown in Figure 4 and Table 3.

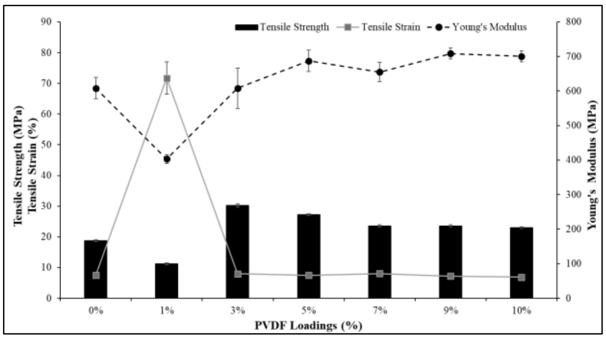


Figure 4 Graph of tensile strength, Young's modulus, and tensile strain of PVDF/Epoxy composites

Table 3 Tensile strength, Young's modulus, and tensile strain of PVDF/Epoxy composites

PVDF Loadings (%)	Tensile Strength (MPa)	Young's Modulus (MPa)	Tensile Strain (%)
0	18.76	608.27	7.50
1	11.22	402.99	71.72
3	30.23	607.86	7.98
5	27.25	687.39	7.48
7	23.59	654.85	8.07
9	23.57	708.34	7.21
10	22.97	700.47	6.87

According to the graph, the tensile strength of the composites declines by about 40% with the addition of 1% of PVDF filler. As the loading increased to 3%, the tensile strength increases by 61% compared to the unfilled epoxy. The inclusion of 1% PDVF filler might be too low to provide sufficient reinforcement effect, besides the presence of high void content resulting in the drop of tensile strength to even lower than the unfilled samples [26, 27].

As the loading increased to 3%, the tensile strength increased. As the filler loading was further increased, the tensile strength shows a gradual decreasing pattern. At high loading of PVDF, the tensile strength of the composites might be reduced due to the filler agglomerations which are supported by the density and percentage of void content. Besides causing voids inside the composites system, filler agglomerations also act as stress concentration sites. When the composites were subjected to stress, given its tension or compression, the agglomerations regions experience higher stresses and strains compared to the surrounding matrix, causing the formation of stress concentration sites. Stress concentration sites are the locality in the composites system where the stresses are significantly higher than the average stress. A premature failure such as cracking or fracture can initiate from these points and propagate to the adjacent area. The presence of voids inside the composite system can worsen the situation where the voids will fasten the propagation of the cracks [28-30].

As tensile strength dropped at 1% loading due to insufficient reinforcement and void content, the tensile strain at the same loading increased by approximately 89%. Lack of reinforcement results in a more compliant matrix which can undergo more deformation under tensile stress. This is supported by the drop in Young's modulus as shown in Figure 4 indicating a decrease in the stiffness or rigidity of the composite structure.

Further increase in the filler loadings to 3 and 5% shows a declining trend of tensile strain due to the restricted chain mobility by the PVDF's particles. This is also in line with the increasing trend of Young's modulus where the restricted chain mobility prevents the molecular chains from sliding past one another resulting in a stiffer composite. In general, the addition of filler in the composites at the optimum loading will increase the tensile strength and modulus since the filler particles will hinder the molecular chain mobility causing the composites to be stiffer. This situation will result in the reduction of tensile strain due to the inability of the chains to move and reorient to accommodate the applied stress. However, at high filler loading with a high tendency of filler agglomerations, the interfacial adhesion that significantly contributes to the strength and modulus of the composite will be disrupted. The presence of microvoids, either due to external factors or the filler agglomerations, can make the situation worse.

The trend observed for the flexural strength, flexural modulus and flexural strain was similar to the trend obtained for tensile properties. Referring to Figure 5 and Table 4, the incorporation of 1% PVDF caused a 43% and 50% drop in the flexural strength and flexural modulus, respectively. The possible reason for the decline is the insufficient amount of PVDF to impart the reinforcement to the composites and also due to the high void content.

As the amount of filler increased to 3% and 5%, the flexural strength and modulus increased to even higher than the unfilled sample. But then, as the filler loading was further increased to 7, 9 and 10%, both flexural strength and modulus declined. Based on the study done by H. Wang *et al.* [31], the flexural strength of PVDF/epoxy composites is found to be inversely proportional to the PVDF content, implying that the flexural strength of PVDF/epoxy composites is reduced when there is an increment in the PVDF content.

However, in this study, the highest flexural strength and modulus were achieved at 5% filler loading. In flexural testing or three-point bend testing, the sample undergoes bending deformation, with the top surface of the sample experiencing compression and the bottom surface experiencing tension. The situation results in the development of internal stresses within the material, which can lead to the initiation and propagation of cracks. 10% filled composites exhibited the lowest flexural strength and modulus, and then closely followed by the 9% filled composites. This might be contributed by the high percentage of void content present inside the composites system supported by the lower values of density results.

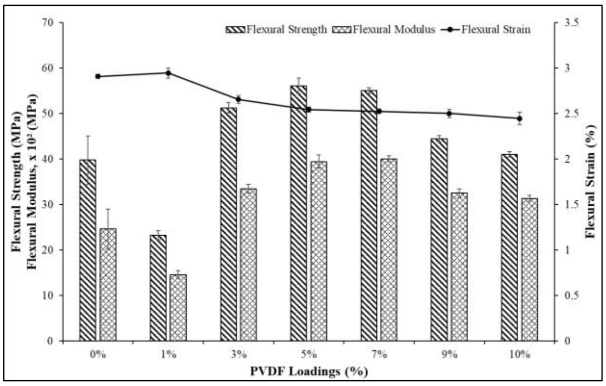


Figure 5 Graph of flexural strength, flexural modulus, and flexural strain of PVDF/Epoxy composites

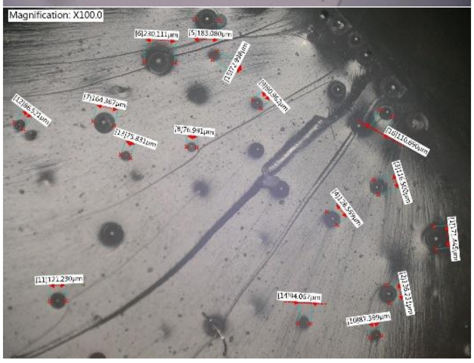
Table 4 Flexural strength, flexural modulus, and flexural strain of PVDF/Epoxy composites

PVDF Loadings (%)	Flexural Strength (MPa)	Flexural Modulus $x10^2$ (MPa)	Flexural Strain (%)
0	39.77	24.64	2.91
1	23.22	14.60	2.95
3	51.28	33.49	2.66
5	56.09	39.46	2.54
7	55.06	40.07	2.53
9	44.45	32.49	2.50
10	41.00	31.37	2.45

In addition, the possibility of filler agglomerations incurring in the composites of a high amount of filler was also high since PVDF was incorporated into the epoxy without any prior surface modification. Thus, the interaction between filler-filler is expected to be higher than the filler-matrix, especially at high filler loading. The filler agglomerations and void will act as stress concentration sites and the propagation of crack will also become faster. The decline of flexural strain started from 3 % filler loading and gradually decreased as the loading increased. The presence of PVDF particles among the epoxy three-dimensional (3D) crosslinked network structure restricted the mobility of the polymer chains and increased the stiffness of the composite, thus leading to a reduction in the flexural strain. In this network, the crosslink chains are interconnected in all three dimensions, creating a rigid and stable structure. When PVDF particles are added to the epoxy matrix, they occupy space within the 3D network. These particles physically obstruct the movement of epoxy polymer chains.

Table 5 presents the micrographs of the samples. The unfilled epoxy exhibits minimal void presence. However, as the filler loadings increase, both the percentage of void content and the size of the voids increase, as evidenced by the micrographs. The void size ranges from 76 to 230 µm for 1% loading, 89 to 160 µm for 3% loading, 71 to 171 µm for 5% loading, 103 to 500 µm for 7% loading, 150 to 294 µm for 9% loading, and 253 to 474 µm for 10% loading. Notably, at loadings between 5% and 10%, white accumulated areas are visible in the micrographs, indicating the agglomeration of PVDF particles, with the white areas expanding as filler loadings increase.

Table 5 Micrographs of the PVDF/Epoxy composites

PVDF Loading (%)	Micrographs (x100 Magnification)
0	
1	
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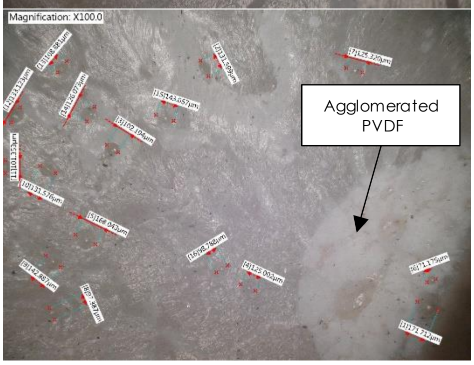
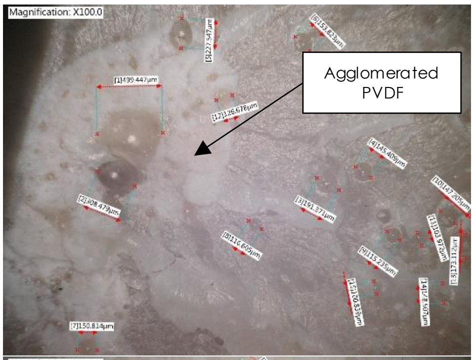
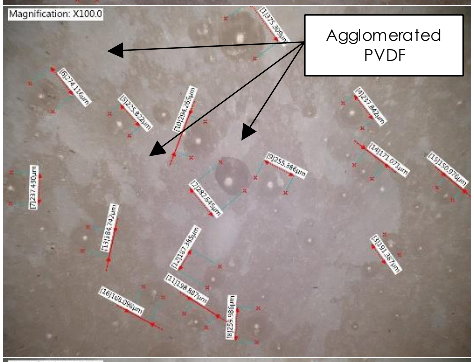
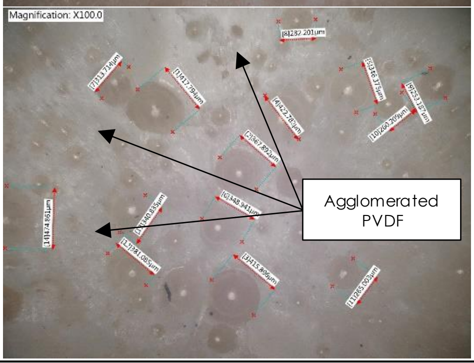
5	

7	
9	
10	

4.0 CONCLUSION

The inclusion of PVDF powder which possesses excellent mechanical and thermal properties in the epoxy resin at a loading range of 1% to 10% showed a potential enhancement in the tensile properties, flexural properties, and thermal stability. At low loading of 1%, the tensile and flexural dropped lower than the unfilled epoxy which might be contributed by the insufficient reinforcement effect, besides the presence of high void content. 3% filler loading can be concluded as the optimum loading in this study since its shows high tensile strength, tensile modulus, flexural strength, and flexural modulus, besides exhibiting the slowest burning rate for both horizontal and 45-degree test configurations. 9% and 10% filler loading seem to be excessive since the tensile and flexural properties of the composites dropped and the burning rate is high. The factor that might contribute to these is the formation of filler agglomerates which not just act as stress concentration sites but also increase the percentage of void contents.

Acknowledgement

The authors gratefully acknowledge the funding for this research provided under grant RACER/1/2019/STG07/UITM//2. They also extend their thanks to the Centre for Chemical Synthesis and Polymer Technology, the Institute of Science, and the Faculty of Applied Sciences at Universiti Teknologi MARA for their support and the provision of research facilities.

Conflicts of Interest

The author(s) declare(s) that there is no conflict of interest regarding the publication of this paper.

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