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Mechanical properties of carbon-aramid hybrid fiber-reinforced epoxy/poly(vinyl butyral) composites

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Abstract

Currently, it is important to manufacture fiber-reinforced epoxy matrix composites with high impact resistance besides their high strength and modulus values for industries such as automotive, aerospace, and aviation due to the brittle structure of epoxy. In this regard, we found the attractive results of the 0.5 wt% poly(vinyl butyral) (PVB)-containing epoxy blend in terms of strength, Young's modulus, and impact resistance. These results substantially motivated us to manufacture fiber-reinforced advanced epoxy/PVB matrix composites with 30–60 vol% carbon-aramid fiber ratios. Flexural (three-point bending) and tensile tests were performed to obtain strength and modulus values by measuring the force required to break the fiber-reinforced composite specimens and elongation at break points. Interlaminar shear strength tests were performed by the short beam bending method by measuring the resistance of the composite to delamination. The Charpy impact test was used to measure the energy absorbed during crack formation and fracture propagation. The composites with PVB were generally superior to those without PVB (EPCs). Two types of findings were observed. First, PVB increased the tensile and flexural strength values substantially for the 30 and 40 vol% ratios, but the modulus values slightly decreased. Second, PVB also substantially increased the modulus values for the 50 vol% ratio besides the strength values. It was thought that this result could be attributed to the increase in the compatibility of the fiber/matrix for the 50 vol% ratio. These decreases for EPCs could be derived from the micro-cracks and weaker interface between fiber and matrix.

KEYWORDS

carbon-aramid hybrid fibers, epoxy/PVB blend matrix, fiber-reinforced composites, poly (vinyl butyral)

1 | INTRODUCTION

Polymer matrix composites have extensively been used in various fields such as the aerospace, automotive, marine, and defense industries. Their common advantages are high corrosion resistance, light weight, and highly dielectric characteristics. Epoxies have been the mainstream polymer materials for applications like coatings, adhesives, composite laminates, and structural components. Epoxy resins as matrix materials are widely used in the production of fiber-reinforced composites due to their wetting ability and adhesion to fibers, low shrinkage, considerable cohesion strength, adequate dielectric characteristics, and high strength and stiffness.[1,2] However, they have fallen behind compared to other engineering polymers due to some of their shortcomings such as brittleness or notch sensitivity.[3] Brittleness is the most important shortcoming of epoxy materials, and this is caused by their highly crosslinking structure. This behavior has limited their usage areas in composite constructions such as automobile body sections, aircraft interior panels, and the wings and radome of airplanes. For example, an epoxy matrix was used in the radomes of airplanes, and due to the high-speed bird impacts on the radome of the aircraft, cracks and breaks occurred at the nose of the aeroplane.[4] For these reasons, a considerable effort has been made to increase the toughness values of epoxies.[5,6] Recently, epoxy toughness enhancement has been achieved by the introduction of rubber modifiers, active diluents, or thermoplastic polymers. Unfortunately, the usage of rubber modifiers has led to a notable decrease in Young's modulus and tensile strength values and a dramatic increase in viscosity. It is well-known that epoxy–high strength thermoplastic blends have been widely used in high-tech industries. Hence, expensive high-strength thermoplastics are used to eliminate this brittle behavior of epoxy, and these thermoplastics include polysulfone, phenoxy resin polyethersulfone, poly(arylene ether ketone), and polyetherimide (PEI). For its usage with epoxies as thermoplastic polymers, poly(vinyl butyral) (PVB) is a prominent material among thermoplastics in terms of its mechanical properties and low cost.

PVB is the most common glass lamination inter-layer due to its strong adhesion, high impact resistance, high flexibility, excellent optical clarity, UV and heat stability, and moisture insensitivity.[7,8] Its commercial name is BUTVAR in the industry, and this material has also been used in applications such as wood finishes, adhesives, textile coatings, toners and printing inks, thick films, ceramic binders, wire enamels, surface coatings, and wash primers.[7,8] Furthermore, PVB can be used to increase interfacial strength and the compatibility of fibers such as glass, carbon, and Kevlar with the matrix. Hence, PVB has an attractive potential for usage in laminated composite applications.[9–11]

Up until now, many studies have been carried out and are in progress to increase the mechanical properties of epoxy composites. Most of these studies in the literature have involved compromises in terms of mechanical properties. While fillers or reinforcing additives increase stiffness and strength values, toughness decreases.[12]

In the relevant literature, the strengthening of the matrix attracts attention as one of main approaches to improving mechanical properties. Rigid nanoparticles or microparticles such as graphite, TiO_2, $CaCO_3$, CNT, silica, and Al_2O_3 have been shown to increase the mechanical properties of these materials.[13,14]

Vijayan et al.[14] reported epoxy nanocomposites filled with TiO_2 and TiO_2/Halloysite (HNT) hybrid particles. First, they presented decreases of 8% and 16% in the tensile strength and toughness of the epoxy nanocomposites with a 5 phr TiO_2 filler, respectively. Then, they presented increases of 40%, 44%, and 157% in the tensile strength, Young's modulus, and toughness of the epoxy nanocomposites with a 5 phr TiO_2/HNT hybrid filler, respectively. This TiO_2/HNT hybrid filler was produced by a hydrothermal reaction, and spherical TiO_2 particles were transformed into a tubular structure surrounding HNT clay tubular particles. While fractured surfaces of the epoxy nanocomposites filled with TiO_2 particles presented deep craters and trenches, those of the epoxy nanocomposites filled with TiO_2/HNT hybrid particles presented fairly uniform filler dispersion without craters and trenches. These results showed that TiO_2/HNT hybrid particles could prevent undesirable agglomeration.

He et al.[15] investigated the compressive strength of nano-calcium carbonate/epoxy and treated nano-calcium carbonate/epoxy casts. They treated nano-calcium carbonate with silane coupling agents and increased the compatibility and interfacial strength of the matrix and the nanoparticles. Therefore, they obtained an improvement of 14.1% in compressive strength and an improvement of 106.3% in total fracture work due to the nano-calcium carbonate treated with silane coupling agents.

Burkov and Eremin[13] studied carbon-fiber-reinforced epoxy composites modified by single-wall nanotubes (SWCNT) and micro milled carbon fiber fillers. They presented maximal increases of 8.6% and 14% in the tensile strength and flexural strength of the carbon-fiber-reinforced epoxy composites with 0.3 wt% SWCNT particles, respectively. They stated that this strengthening behavior could have been derived from the improved shear-related interfacial properties by the addition of the SWCNT particles.

Vinay et al.[1] examined the effects of Al_2O_3 nanofillers (0–3 wt%) in basalt/epoxy composites that were manufactured. They reported that while ultimate tensile strength decreased by 19% in the material with Al_2O_3 nanofillers, flexural strength increased by 31%. The authors thought that while the brittle nature of Al_2O_3 nanofillers affected the tensile strength of the material adversely, the good interaction between the fillers and the epoxy improved its flexural strength. However, high ratios caused agglomeration due to the high surface energy of the Al_2O_3 nanofillers.

Tiwari et al.[10] studied epoxy composites filled with fly ash (FA) particles (obtained from Indian soil chulha) in the range of 0–50 wt% (constituents; Silica (50.5%), Alumina (23.6%), Fe_2O_3 (11.5%), in addition to CaO, MgO,

SO_3, and other elements). They reported maximal increases of about 80% and 56% in respectively the tensile strength and Izod impact of the fly ash particle-reinforced epoxy composites. They stated that the interfacial bonding of the FA particles with the matrix provided the substantial transfer of the stresses through the intermolecular structures and promoting higher overall strength and impact strength. Moreover, the agglomeration of the FA particles was also observed in scanning electron microscopy (SEM) morphologies. It was stated that these larger FA particles contributed to an increase in impact resistance by preventing cracking growth.

It is known that PVB polymers have been extensively used in the literature for many applications due to their cost-effectiveness, strong binding, flexibility, and optical clarity. The blending of low-modulus polymers generally reduces strength and modulus values. PVB is a multifunctional material. It consists of poly(vinyl butyral), poly(vinyl alcohol), and poly(vinyl acetate) that have different functional groups. It has been used for various purposes due to its flexibility and strong binding properties. The blending of PVB with polymeric materials generally increases impact strength, especially.[11]

Hofmann and Lee[16] reported the modification of polymers by using poly(vinyl butyral)-based additives. They stated that PVB pellets could act as impact modifiers and processing aids when blended at low ratios into a variety of engineering resins. The residual hydroxyl groups in PVB provide active surface sites to improve adhesion to other materials such as nanoparticles and fibers. The adhesion of PVB has been found to enhance stiffness/compatibility when glass fibers and minerals are added into engineering resins.

Nadler et al.[17] prepared MWCNT/epoxy composites with various functionalized MWCNT particles. These were OH-CNT and CNT functionalized with a commercial acrylic copolymer with pigment affine and amine (Disperbyk 2070) and a commercial polymer (PVB, Kuruay B60H). Disperbyk 2070 and PVB were bonded to MWCNT non-covalently. The highest mechanical improvements were found for the nanocomposite with 0.5 wt% PVB-CNT. PVB also increased the usage ratio of CNT. The researchers reported increases of 32% and 29% in flexural modulus and strength, respectively. PVB-CNT presented an excellent fracture surface with much more homogenous particle distribution. PVB provided high interfacial strength between the CNT and the epoxy matrix by its functional groups.

In our previous study,[11] we had carried out an analysis on PVB-containing epoxy blends. Transparent and homogeneous PVB solutions that varied from 0.5%. to 4 wt% in reinforcement ratios were stirred with epoxy resins. The 0.5 wt% PVB-containing epoxy blend (EPVB0.5) exhibited the most promising values in terms of strength and impact resistance. EPVB0.5 showed increases of 25% and 45% in tensile strength and impact resistance, respectively.

As another issue, it is well-known that epoxy matrices have been reinforced using various synthetic fibers such as carbon, glass, and aramid fibers to improve their mechanical properties. Moreover, some nanoparticles and microparticles were used to increase the interfacial strength and compatibility between the matrix and fibers such as CNTs, graphene oxide (GO), and reduced graphene oxide (rGO).

Sanchez et al.[18] presented multiscale carbon fiber/epoxy composites with non-functionalized CNT and amino-functionalized CNT particles (0.1–0.3 wt%). They reported increases of about 12% and 8% in the flexural strength and modulus, respectively, of carbon fiber/epoxy composites with amino-functionalized CNT particles. These values were larger than those with non-functionalized CNT. This situation was attributed to the formation of stronger interfaces than those in non-functionalized CNT/epoxy nanocomposites. Therefore, amino-functionalized CNT particles provided a better load transfer from the matrix to the CNT.

Fan et al.[19] prepared glass fiber-reinforced epoxy composites by the injection of MWCNT/epoxy suspensions into glass fiber mats and investigated the interlaminar shear stress (ILSS) values of the composites. The ILSS results increased from 32.5 to 38.5 MPa by the integration of MWCNT at 1%.

Adak et al.[20] investigated the hybridization effects of hydrazine-reduced graphene oxide (rGO) on the interlaminar shear stress (ILSS) and impact strength of woven carbon fiber/epoxy composites. They reported respective increases of 84% and 100% in the ILSS and impact strength of the woven carbon fiber/epoxy composites with rGO particles. The fracture surfaces of the composites with rGO particles showed the compatible dispersion of the rGO in the epoxy matrix and the branched fracture of the specimens. The authors reported that rGO particles can be used as matrix modifiers to provide resistance to matrix fracture and fiber fracture.

The main objective of this study is to develop novel fiber-reinforced composites using an Epoxy/PVB blend as a matrix. This is because the favorable effects of PVB in epoxy resins were observed in our previous study in terms of increasing the strength and impact resistance of the pure epoxy. It is known that PVB has various and important functional groups such as vinyl butyral, vinyl alcohol, and vinyl acetate.[21] It was considered that these functional groups would increase the compatibility and interfacial strength between the fiber and the matrix like reported in the studies conducted by Sanchez,[18] Fan,[19] and Adak.[20] In addition to these, Nadler et al.[17] mentioned the high compatibility of PVB with epoxy and CNT particles. Therefore, it was expected that PVB would show high compatibility with epoxy and carbon fiber besides its compatibility with glass fibers. Moreover, PVB has high compatibility with aramid fibers, and it is widely used as a matrix in ballistic applications.[22] In light of this information, it was decided to use carbon-aramid hybrid fiber to reinforce the Epoxy/PVB blend matrix. Thus, carbon-aramid hybrid fiber fabric-reinforced epoxy matrix (EPCs) and epoxy/PVB matrix (EPVBCs) composites were prepared at various volumetric fiber fabric ratios (30%, 40%, 50%, and 60%) in this study. The produced specimens were tested in tensile and flexural tests. Flexural (three-point bending method) and tensile tests were used to measure the force required to break the fiber-reinforced composite specimens and elongation at break points to determine the strength and modulus values. Interlaminar shear strength tests (ILSS) were used to measure the resistance of the composites to delamination by the short beam bending method. Unnotched Charpy impact tests were used to measure the energy absorbed during crack formation and the fracture propagation of the composites. The fracture surfaces of the Epoxy/PVB blends and fiber-reinforced composite specimens were morphologically characterized by SEM. Considering the results, the EPVBCs were generally superior to the EPCs. Two types of findings were observed. First, PVB increased the tensile and flexural strength values of the composites substantially up to the ratio of 50 vol%, but the modulus values slightly decreased up to this ratio. Second, PVB also substantially increased the modulus values for the ratio of 50 vol%, besides the strength values. It was thought that this result could be attributed to the increase in the compatibility of the fiber and the matrix for the ratio of 50 vol%. This was because the strength values of the EPCs showed decreases for 50 and 60 vol%. It was considered that these decreases could have originated from the micro-cracks. In general, the composite with PVB at the ratio of 50 vol% (EPVBC50) and the composite with PVB at the ratio 40 vol% (EPVBC40) were prominent with their improved mechanical properties.

2 | EXPERIMENTAL

2.1 | Materials

The epoxy resin EPIKOTE 828 based on a diglycidyl ether of bisphenol-A-based reactive resin and its curing agent EPIKURE F205 hardener were purchased from Hexion. BDMA (Benzyl dimethyl amine) accelerator was purchased from Aldrich. Butvar B-90 that was used as a flexible thermoplastic and poly(vinyl butyral) (PVB) were purchased from Eastman. Ethanol was used as a solvent for PVB. Teflon was used as the mold material. An ultrasonic bath and a homogenizer were used for stirring. Carbon-aramid hybrid 2 × 2 twill woven fiber fabric (185 g/m^2) was purchased from a local corporation.

2.2 | Preparation of carbon-aramid hybrid fiber-reinforced epoxy/PVB composites

In our previous study,[9] epoxy/PVB blends were prepared using the solution casting method, and their mechanical properties were investigated. These mechanical properties are shown in Table 1. The 0.5 wt% PVB-containing epoxy blend showed increases in both tensile strength and Izod impact strength by 25% and 47%, respectively. Based on this result, carbon-aramid hybrid fiber fabric-reinforced 0.5 wt% PVB-containing epoxy matrix composites (EPVBCs) were manufactured by hand lay-up method that was applied by cold-press with 10 bars into closed mold.

For this study, PVB granules were firstly dissolved in the ethanol solvent. The PVB solution was first mechanically and then ultrasonically stirred. After the stirring process, a homogeneous and clear solution was obtained. This solution was stirred with the epoxy resin first mechanically and then ultrasonically. A homogeneous and clear resin solution was obtained. After this process, the curing agent was stirred mechanically into the epoxy/PVB resin solution stoichiometrically. The prepared epoxy/PVB resin solution was applied to carbon-aramid hybrid fibers by hand lay-up method that applied by cold-press with 10 bars into closed mold. A brush and a roller were used to impregnate the fiber fabric with the solutions. The carbon-aramid hybrid fiber fabrics were cut into a certain size to obtain different volumetric ratios, including 30%, 40%, 50%, and 60%. During composite production, the first ply was laid onto a thin release agent into a steel mold, and the resin solution mixture was distributed uniformly on the ply. The ply was brushed and rolled by using the resin solution. Then, the other plies were stacked up and aligned with each other and impregnated with the same method in the steel closed mold. After this implementation, a cold-press with 10 bars was applied onto the closed mold, and the material was cured at room temperature for 24 h.

2.3 | Measurements

Three samples were prepared for each material. The analyses were conducted based on the average results. The

TABLE 1 Mechanical properties of epoxy/poly(vinyl butyral) blends in the previous study.

Samples	Young modulus (MPa)	Failure strain (%)	Tensile strength (MPa)	Izod impact (kJ/m^2)
EPOXY	2962	1.1	28.5	9.05
EPVB0.5	1786	2.7	36.1	13.30
EPVB1	1127	2.6	21.7	7.48
EPVB1.5	1280	3.3	24	7.00
EPVB2	774	5	15	3.30

coefficients of variation (CoV) are presented in figures and tables. CoV is the ratio of the standard deviation and the mean of the results of a sample multiplied by 100 that is given in Equation 1. In this equation, s is the standard deviation, and μ is the mean of the values of a sample.

$$\mathrm{CoV} = \frac{\mathrm{s}}{\mu} \times 100. \tag{1}$$

CoV is a measure of relative event dispersion and is generally used for comparing relative risk. Moreover, it can also be used to evaluate quantitative likelihood or probability distribution. CoV measures the spread of values like standard deviation. While standard deviation measures distance from the mean, CoV measures the ratio of the standard deviation to the mean. The samples were characterized mechanically by tensile and flexural (three-point bending) tests. These tensile and flexural tests were performed according to ASTM D3039 and TS EN-ISO 178, respectively. Moreover, unnotched Charpy impact strength and ILSS tests were performed according to EN-ISO 179-1 and ASTM D2344, respectively.

The dimensions of the tensile test specimens were adjusted to 15 mm (width) × 130 mm (gauge length). The dimensions of the flexural test specimens were adjusted to 15 mm (width) × 80 mm (length). The dimensions of the unnotched Charpy test specimens were adjusted to 10 mm (width) × 60 mm (length). The dimensions of the ILSS test specimens were adjusted to 10 mm (width) × 16 mm (length). The thickness of these specimens varied according to their fiber reinforcement ratios. The average thickness values of these specimens were approximately 4, 4.5, 5, and 6 mm for the volumetric ratios of 30%, 40%, 50%, and 60%, respectively. Moreover, all composite specimens were prepared in rectangular shapes. The carbon-aramid hybrid fiber fabric (2 × 2 Twill) of the composite laminates was cut symmetrically in “0/45/0/45/ 45/0/45/0”.

The Charpy impact tests were performed using an Instron CEAST 9050 Charpy impact tester with a 25J hammer. The composite laminates were prepared in unnotched and rectangular shapes. Thus, the Charpy test was conducted in the unnotched form according to the EN-ISO 179-1 standard. The Charpy tester was a pendulum tester. The unnotched composite specimens in rectangular shape were taken at its mid-length supported by the horizontal plane at both ends and impacted by a pendulum with a known weight, swinging from a known height. By means of swinging of the pendulum, the composite specimens were impacted and broken. In this procedure, the amount of total energy required during the crack formation and fracture propagation of the composites was determined according to Equation 2. The total energy is given by:

$$\mu_{\text{total}} = \mathrm{mg}\left[h_0 - h_f\right], \tag{2}$$

where μ total is the total energy, m is the mass of the pendulum, g is the gravitational mass of acceleration, h_0 is the initial height of the pendulum, and h_f is the final height of the pendulum.

The flexural tests were carried out using an Instron model 4505 device and the Series IX acquisition software. This three-point flexure test was used to evaluate the flexural strength and flexural modulus of the composite specimens. The flexural test was carried out at a crosshead speed of 2 mm/min. In the three-point flexure test procedure, flexural strength and flexural modulus were calculated according to the formulations in Equations 3 and 4.

$$\sigma = \frac{3PL}{2bd^2}, \tag{3}$$

$$E = \frac{L^3 m}{4bd^3}, \tag{4}$$

In these formulations, σ is the stress of the composite specimen at mid-span (N/m^2), P is the load at a given point on the load-deflection curve (N), L is the support span, b is the width of the tested specimen, d is the thickness of the tested specimen, E is the modulus of elasticity in bending (N/mm^2), and m is the slope of the tangent to the initial straight-line portion of the load-deflection curve (N/mm).

The tensile tests were carried out using an Instron model 4505 device and the Series IX acquisition software as in the flexural test. This tensile test was used to evaluate the flexural strength and flexural modulus of the composite specimens. The test was carried out at a crosshead speed of 5 mm/min. In the tensile test procedure, tensile strength and tensile modulus were calculated according to the formulations in Equations 5 and 6.

$$\sigma = \frac{P}{A}, \tag{5}$$

$$E = \frac{\sigma}{\epsilon}. \tag{6}$$

The tensile tests measured the force required to break the fiber-reinforced composite specimen and elongation at break points to determine the strength and modulus values. In these formulations, σ is the stress of the composite specimen, P is the load at a given point on the load-deflection curve (N), A is the cross-sectional area of the composite specimen (mm^2), E is the modulus of elasticity in tensile (N/mm^2), ϵ is the strain of the composite specimen, and these values represent the elongation of the composite specimens.

The ILSS test measured the resistance of the composites to delamination by the short beam bending method. The support span in the specimen thickness of the ILSS test is much smaller than that of the three-point flexure test. This method provides a great opportunity to obtain large amounts of shear stresses in composite specimens. It can be defined as the short three-point bending test. This way, a shear stress fracture can be obtained in brittle matrix materials. Therefore, shear stress strength was calculated according to Equation 7.

$$F^{\mathrm{sbs}} = \frac{3P_{\mathrm{m}}}{4bh} \tag{7}$$

In this formulation, F^{sbs} is the short-beam strength (MPa), P_{m} is the maximum load observed during the test (N), b is the width of the tested specimen (mm), and h is the thickness of the tested specimen (mm).

3 | RESULTS AND DISCUSSION

PVB is considered to be an acetal and is formed by the reaction of an aldehyde and alcohol. However, PVB does not consist of only polyvinyl butyral. The structure of PVB is built up of approximately 80% polyvinyl butyral, 18%–23% polyvinyl alcohol (PVOH), and a maximum of 2.5% polyvinyl acetate. This structure is shown in Figure 1. The relative amounts of these segments are controlled, but they are generally randomly distributed through the molecular chain. The properties of the polymers can be optimized by controlling the ratios of the three segments. PVB has a structure that is highly compatible with fibers and other polymer materials due to the high amount of functional groups on its polymer backbone as shown in Figure 1. It has strong adhesive properties and has been used in the sizing processes of glass, Kevlar, and carbon fibers.[17,19]

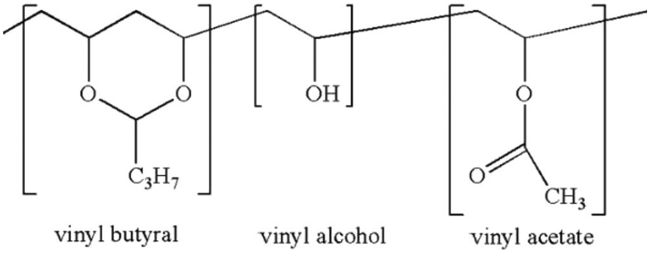

FIGURE 1 Structure of poly(vinyl butyral).

In our previous study,[11] this flexible material was blended into epoxy resin to increase the impact strength of the epoxy. In the results of the tests, it was found that EPVB0.5 showed increases of 25% and 45% in the tensile and Izod impact strength tests, respectively. Furthermore, its Young's modulus value was favorable.[11] Therefore, it was thought that fiber-reinforced epoxy/PVB composites would present high strength, impact resistance, and modulus values compared to fiber-reinforced pure epoxy composites.

3.1 | Flexural, tensile, and ILSS properties

The strength and modulus results of CF-reinforced polymer matrix composites in flexural and tensile conditions are affected by the ratio of voids and CF in composites, interfacial properties between the fiber and the matrix, and the mechanical properties of the materials constituting the composite. The cross-sectional views of the CF fiber-reinforced polymer matrix composites that are given in the section of SEM morphologies showed the strong composite structure. Thus, it was deduced that the CF fabric reinforcement was impregnated with the epoxy and the epoxy/PVB matrix at sufficient levels. Moreover, the EPVBCs generally showed stronger composite structures, as well as higher interfacial strength values between the CF and the matrix according to their SEM morphologies and mechanical results. In particular, the cross-sectional SEM morphology of EPVBC40 after the flexural test had less delamination.

While PVB has a higher toughness than epoxy, the modulus and strength values of PVB are lower than those of pure epoxy. For these reasons, it can be expected that the modulus and strength values of composites with PVB would be lower. In this study, PVB created a compatible interface between the CF and the epoxy matrix due to its favorable functional groups. Therefore, this compatible interface increased the transferring ability of forces from the matrix to the CF. This situation increased the strength and modulus values of the EPCs, especially EPC40 and EPC50. The incorporation of PVB into the EPCs substantially increased their overall mechanical and impact strength properties.

In this study, EPVBC40 and EPVBC50 showed improved mechanical properties overall. EPVBC40 presented improved tensile, flexural, and impact resistance results. In comparison to EPC40 (without PVB addition), almost all properties of EPVBC40 showed significant improvement, except for tensile and flexural modulus values. Additionally, the flexural and tensile modulus values of EPVBC40 were favorable for a fiber-reinforced advanced composite. The tensile and flexural strength values of EPVBC40 increased respectively by 3.3% and 10% in comparison to those of EPC40. In addition to these results, the highest increases were observed in impact resistance values. While the impact resistance value of EPC40 was 143.6 MPa, the impact resistance value of EPVBC40 was 184.6 MPa. There was a substantial increase of 29%. Strength values showed decreases for the samples with higher fiber ratios than 40%. This result could be attributed to the increase in the amount of cracks in the matrix. For example, the tensile and flexural strength values of EPC40 were as low as 29% and 14.5% in comparison to those of EPC50. On the other hand, the tensile and flexural strength values of EPVBC40 were as low as 9% and 2% in comparison to those of EPVBC50. The epoxy/PVB matrix limited these decreases in the samples with a reinforcement ratio of 50%. In addition to this behavior, the tensile and flexural modulus values of EPVBC50 showed increases by 44% and 27% in comparison to EPVBC40.

The substantial effect of PVB can be observed for both the prominent epoxy/PVB blend (EPVB0.5) and carbon-aramid hybrid fiber fabric-reinforced epoxy/PVB composite in Table 2. This table shows the results of the comparison of the pure epoxy, the prominent epoxy/PVB blend, the prominent fiber-reinforced epoxy/PVB composite (EPVBC40 and EPVBC50), and the prominent fiber-reinforced epoxy composites (EPC40 and EPC50).

Figures 2 and 3 that are representative drawings show the relationship between micro-cracks and PVB. Figure 2 reveals that the amount of micro-cracks increased along with the increasing fiber fabric ratios. The mechanical results were compatible with this behavior. Figure 3 shows the behavior of PVB towards the micro-cracks on the composites and presents the morphological change of PVB with the change in the fiber fabric ratios. This morphological change affected the distribution and shape of PVB in the fiber-reinforced epoxy matrix. The homogeneous distribution, size, and shape of PVB prevented the growth of micro-cracks. Therefore, the increasing amount of micro-cracks with the increase in the reinforcement fiber ratio did not affect the mechanical results adversely. Considering these parameters, EPVBC40 and EPVBC50 showed the optimum PVB size and distribution values. Moreover, the morphology of PVB was somewhat elliptical for EPVBC50. It was thought that this behavior of PVB affected the mechanical properties more positively. All PVB behaviors for EPVBC40 and EPVBC50 also increased the interfacial attractions between the fiber and the matrix. SEM morphologies presented compatible results with these comments and discussions.

The effect of the carbon-aramid hybrid fiber fabric on the epoxy and epoxy/PVB matrices was remarkable.

TABLE 2 Comparison of the mechanical properties of pure epoxy, prominent epoxy/poly(vinyl butyral) (PVB) blend, prominent fiber-reinforced epoxy, and epoxy/PVB composites.

Samples	Tensile strength (MPa)	Young's modulus (GPa)	Flexural modulus (GPa)	Flexural strength (MPa)	ILSS (MPa)	Impact strength (KJ/m^2)
NEAT EPOXY	28.5	2.96	–	–	–	9.05 (ASTM D4812-99)
EPVB0.5	36.1	1.78	–	–	–	13.3 (ASTM D4812-99)
EPC40	513	34.5	27.6	351	44	143.6 (ISO 179-1)
EPC50	343	28	20	300	28	78.6 (ISO 179-1)
EPVBC40	530	28.9	24.4	386	54	184.6 (ISO 179-1)
EPVBC50	480	41.6	31.1	378	48	110 (ISO 179-1)

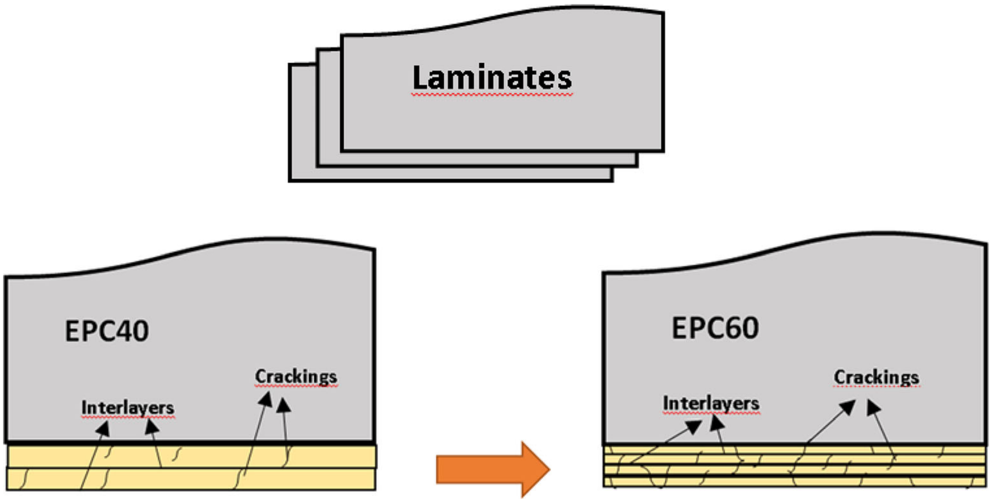

FIGURE 2 Representative micro-cracks amount in interlayer scheme of the composite laminates.

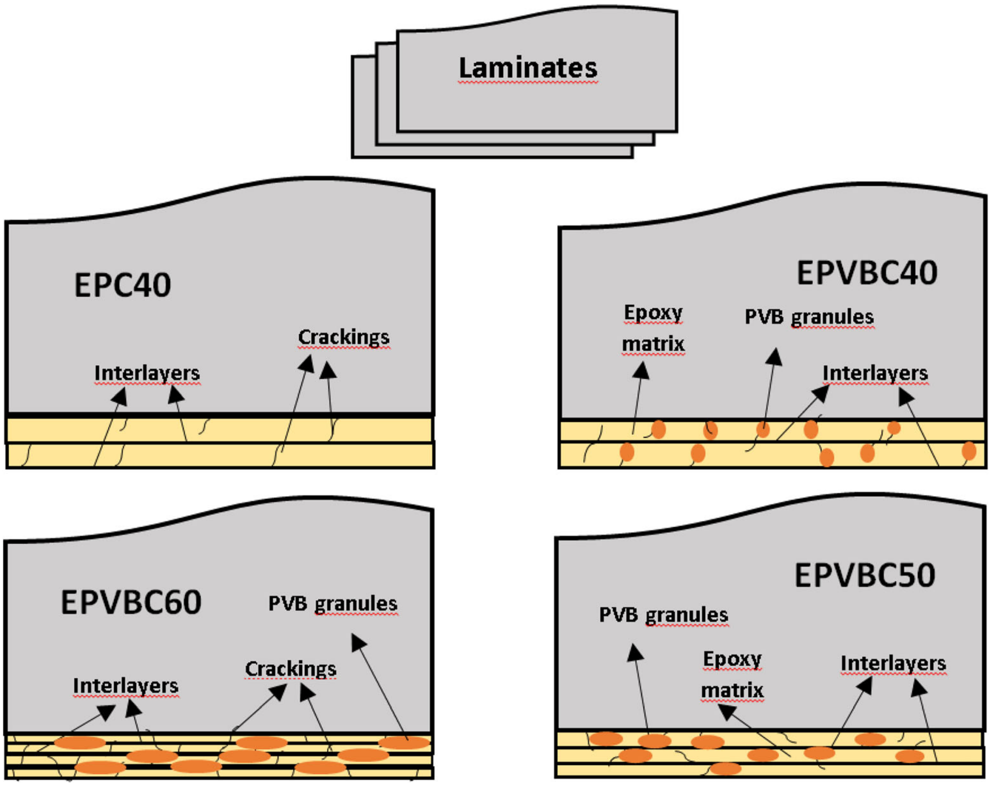

FIGURE 3 Poly(vinyl butyral) (PVB) granule behavior scheme in interlayers of carbon-aramid hybrid fiber-reinforced laminates.

TABLE 3 Tensile and impact test results of fiber-reinforced epoxy and epoxy/poly(vinyl butyral) composites.

Samples	Young's Modulus (GPa)	CoV	Tensile Strength (MPa)	CoV	Impact test(kJ/m^2)	CoV
EPC30	28	19.6	375	6.4	94.0	6.6
EPC40	34.5	13.2	513	2.4	143.6	3.5
EPC50	28	15.7	343	7.3	78.6	6.4
EPC60	30	11.9	299	6.6	56	15
EPVBC30	27	13.2	400	4.5	116.4	6.1
EPVBC40	28.9	16.6	530	2.9	184.6	3.2
EPVBC50	41.6	9.7	480	2.8	110	5.9
EPVBC60	31.4	12.9	380	3.5	86.3	6.9

TABLE 4 Flexural and interlaminar shear stress (ILSS) test results of fiber-reinforced epoxy and epoxy/poly(vinyl butyral) composites.

Samples	Flexural modulus (GPa)	CoV	Flexural strength (MPa)	CoV	ILSS (MPa)	CoV
EPC30	22.8	24.7	341	4.5	45	5.6
EPC40	27.6	18	351	2.6	44	9
EPC50	20	22	300	4.8	28	11.1
EPC60	28.1	13.7	314	4.3	21	19
EPVBC30	21.68	17.2	345.1	3.5	50	8.2
EPVBC40	24.4	15.8	386	2.6	54	6.5
EPVBC50	31.1	12.9	378	2.9	48	5.2
EPVBC60	2.4	88.6	89.1	12.2	NA	NA

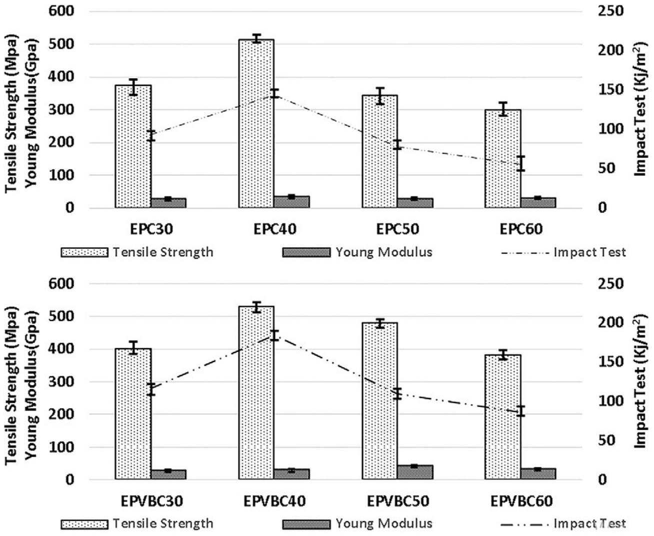

FIGURE 4 Changing trends of tensile and impact strength test results of carbon-aramid hybrid fiber fabric-reinforced epoxy matrix (EPCs) and epoxy/PVB matrix (EPVBCs).

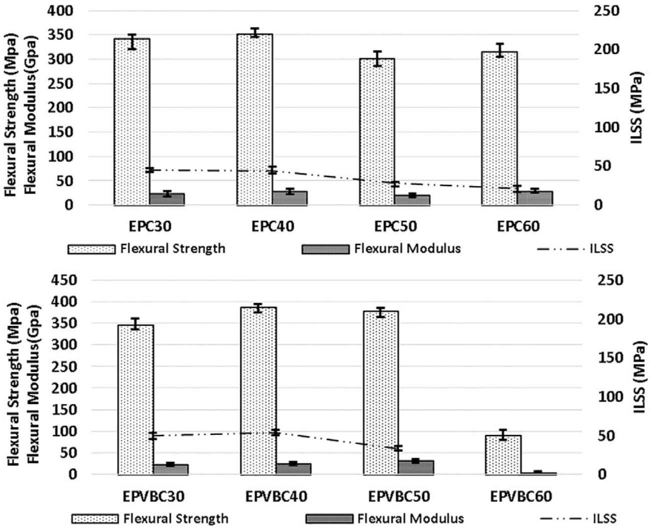

FIGURE 5 Changing trends of flexural and interlaminar shear stress (ILSS) test results of carbon-aramid hybrid fiber fabric-reinforced epoxy matrix (EPCs) and epoxy/PVB matrix (EPVBCs).

Tables 3 and 4 present the tensile and flexural test results, impact strength, and ILSS values for EPCs and EPVBCs. Moreover, the increasing or decreasing trends of EPCs and EPVBCs can be observed with the changes in the hybrid fiber fabric ratios in Figures 4 and 5. In the results of EPCs, the strength values increased up to a certain limit of the hybrid fiber ratio. While the tensile and flexural test results of EPCs increased up to the ratio of 40%, the tensile and flexural modulus results decreased with the increasing ratios. EPC40 showed the most favorable values among EPCs in terms of tensile and flexural strength, modulus, and impact resistance. In comparison to EPC30, the results of EPC40 had increases by 28%, 37%, and 53% for Young's modulus, tensile strength, and impact strength, respectively. However, EPC50 and EPC60 showed significant decreasing trends for all mechanical properties in comparison to EPC40. These decreases could be explained by the hand-layup method for ratios of 50% and above.

EPVBCs showed a similar increasing trend to EPCs. The flexural and tensile strength values of EPVBCs had an increasing trend up to the fiber ratio of 40%. The highest impact resistance result of EPVBCs was also obtained for EPVBC40. EPVBC40 showed increases by 32% and 12% respectively for tensile and flexural strength in comparison to EPVBC30. The improvement in impact resistance was 59%. Additionally, the flexural and tensile modulus values of EPVBCs had an increasing trend up to the fiber ratio of 50%. The flexural and tensile modulus showed increases by 47% and 54% from the ratios of 30%–50%, respectively. However, EPVBC60 showed a substantial decrease in the tensile and flexural results. Especially the flexural test results failed to match an average fiber-reinforced composite. These results of EPVBC60 could not be acceptable for a fiber-reinforced epoxy composite. This is because its flexural modulus and flexural strength decreased by 90% and 75% in comparison to EPVBC30. In addition to this, interlaminar shear stress (ILSS) results could not even be obtained, and the layers were easily separated from each other due to the weakness of the interlayers. The ILSS result of EPVBC60 confirmed this substantial decrease.

The flexural and ILSS results showed an increasing trend up to the reinforcement ratio of 40% for EPCs and EPVBCs. According to the results of the comparison of EPVBCs to EPCs for the same ratios, EPVBCs generally had higher strength and impact resistance values. Furthermore, EPCs had higher modulus values, except EPVBC50. EPVBC50 interestingly showed the highest modulus values. On the other hand, EPVBC60 did not achieve an average composite's performance due to its very weak interfacial bonding between the fiber and the matrix. It was thought that PVB granules were replaced in the interfacial areas between the plies, because the interlaminar distance between plies decrease with an increase in fiber ratios. Figure 3 shows the scheme of this behavior. Additionally, EPVBCs showed higher ILSS values than EPCs. Thus, it was thought that PVB increased the bonding strength between the layers. Overall, the presence of PVB increased the mechanical and impact strength values. The mechanical properties were found compatible with the increasing or decreasing trends of ILSS values. It is well-known that ILSS results are related to mechanical properties, especially flexural properties.

3.2 | Impact resistance properties

As well-known, there are three mechanisms that enhance the impact strength properties and fracture toughness of polymer matrix materials when rubber particles are incorporated into them.

- The deformation of the rubber particle phase dissipates the impact energy
- The cavitation in the rubber particles dissipates the impact energy, leads to enhanced impact strength
- Micro or nano rubber particles that are homogeneously dispersed prevent the growth of cracks and behave as crack inhibitors

In this study, PVBs showed rubber-like behavior in that they improved impact resistance. It was thought that PVB particles mostly presented the second and third mechanisms above in the fiber-reinforced composites. Spherical and nodular shapes of PVB particles can be easily seen in the SEM morphologies for both blends and fiber-reinforced composites. It was thought that these spherical PVB particles served as crack inhibitors in both blends and fiber-reinforced composites. Moreover, these particles behaved more effectively in terms of impact resistance at the fiber reinforcement ratios of 40% and 50 vol%. PVB particles were dispersed more homogeneously for EPVBC40 and EPVBC50. In addition to this, the shape of PVB particles transformed into an elliptical formation for the ratios of 50% and 60 vol%. The optimum elliptical shape led to the occurrence of more effective crack inhibition behaviors. It was thought that the maximum impact strength increase that was found in EPVBC50 could be attributed to this behavior of PVB particles. However, this situation caused agglomeration for the ratio of 60 vol%. It was thought that the low impact resistance value of EPVBC60 could be associated with this agglomeration. Moreover, cavitation occurrence in PVB particles was observed. This behavior can be easily seen in epoxy/PVB blends in Figure 6. It was considered that this approach could be also effective in improving the impact resistance of fiber-reinforced composites.[23] On the other hand, the attractive functional groups of PVB are highly effective on the interfacial strength and compatibility between matrix materials and fibers. It was expected that this behavior could increase interlaminar fracture toughness as reported in Park's study[24] on core shell rubber incorporated into CF-reinforced epoxy composites.

The presence of PVB and the fiber fabric provided important changes in the properties. As known, an increase of the fiber ratio improves mechanical properties such as strength and modulus values. The presence of PVB led to a compatible behavior with the reinforcement of the fiber fabric. However, this behavior started to change for higher fiber ratios than 50%. Figure 3 shows the scheme of the behavior of PVB in the interlayers towards transverse cracking processes during the tests. It was thought that these transverse cracking cases could have occurred much more frequently for the composites with higher fiber ratios than 50% as shown in Figure 2. Moreover, it was thought that the PVB agglomerates became elliptical in shape.

These elliptically shaped particulates were longer in parallel to the fibers and prevented to the formation and growth of micro cracks (transverse cracks) on the surfaces of the fibers. Therefore, EPVBC50 could provide higher modulus values than EPVBC40 in addition to its improved strength values. However, these PVB agglomerates became much more elliptical in shape for EPVBC60, and these agglomerates infiltrated into the interlaminar areas as shown in Figure 3. The substantial decrease in mechanical properties for EPVBC60 could be attributed to this behavior of PVB particulates.

3.3 | Comparison of the results to reinforcement studies about epoxy-based composites in the literature

In comparison to the results of reinforcement studies about epoxy-based composites in the literature, the

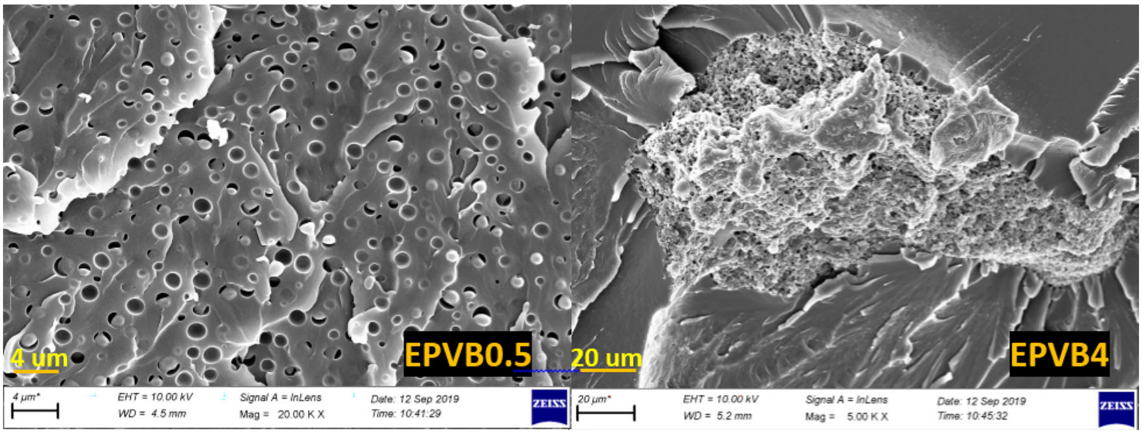

FIGURE 6 Fracture surface morphologies of EPVB0.5 and EPVB4.

EPVBC40 and EPVBC50 specimens in this study showed much more enhanced mechanical and impact resistance properties. The properties of EPVBC40 and EPVBC50 in this study and those in some prominent studies in the literature are given in Table 5. Moreover, information is also given about the blend in our previous study (EPVB0.5) in Table 5. In general, Table 5 presents various prominent reinforcement and toughening particles or fillers incorporated into fiber (mostly CF)-reinforced epoxy composites by various methods. Sprenger[25] prepared a hybrid reinforcement system by using nanosilica and CTBN (carboxyl-terminated butadiene–acrylonitrile) rubber particles. This hybrid system was incorporated into both pure epoxy and CF-reinforced epoxy. This system increased the G_{1c} fracture toughness for both pure epoxy and CF reinforced epoxy. However, it improved the pure epoxy much more than the CF-reinforced epoxy. Sprenger[25] also cited the results reported by Zeng et al.[23] that were related to a reinforcement and toughening hybrid system. For this hybrid system, 6 wt% nanosilica and 6 wt% CSR (core shell rubber) were used, and this system was incorporated into CF-reinforced epoxy composites.

While this hybrid system showed an increase of 125% for G_{1c} fracture toughness, it showed a decrease of 6% for the modulus of elasticity. Burkov and Eremin[13] prepared CF-reinforced epoxy composites into which milled micro-carbon fiber and SWCNT (single-wall carbon nanotubes) were incorporated. While the 0.5 wt% milled micro-carbon fiber-incorporated CF-reinforced epoxy composites showed increases of 1% and 3% in tensile strength and modulus, respectively, the increase in impact resistance was 2%. While the 0.3 wt% SWCNT-incorporated CF-reinforced epoxy composites showed an increase of 8% in tensile strength, the tensile modulus and the impact resistance showed decreases of 5% and 6%, respectively. Park et al.[24] incorporated CSR particles into CF-reinforced epoxy composites and obtained substantial increases in tensile strength, tensile modulus, and impact resistance, respectively. While the composites with 11.4 wt% CSR showed increases of 104%, 31%, and 17% in tensile strength, tensile modulus, and impact resistance, those with 19.4 wt% CSR showed increases of 3%, 44%, and 87% in tensile strength, tensile modulus, and impact resistance, respectively. Qu et al.[26] reported CF-reinforced 0.2 wt% graphene oxide (GO)-modified epoxy composites. These composites showed increases of 6% and 10% in flexural strength and modulus, respectively. Shan et al.[27] prepared CF-reinforced epoxy composites with MWCNT and EP20 (epoxy) solutions. These solutions were prepared with ethanol and sprayed separately onto CF fabric surfaces. The authors then impregnated the epoxy matrix into the sprayed CF fabric. By this method, they achieved increases of 3% and 4% in flexural strength and modulus, respectively. Moreover, they obtained increases of 25% and 17% in G_{Ic} and G_{IIc} fracture toughness values. Considering the studies whose results are shown in Table 5, the EPVBC40 and EPVBC50 specimens in this study presented more favorable results. It was found that 0.5 wt% PVB was much more effective for reinforcing and toughening both the pure epoxy and CF-reinforced epoxy composites. This effect was particularly more pronounced in the EPVBC40 and EPVBC50 specimens. In the comparisons of these composites to EPC40 and EPC50, substantial increases were observed in terms of mechanical properties and impact resistance. EPVBC50 showed substantial increases of 26%, 56%, and 40% in tensile strength, tensile modulus, and impact resistance, respectively. Moreover, it showed respective increases of 40% and 49% in tensile strength and tensile modulus. A similar effect was observed in Park et al.'s study[24] in terms of mechanical and impact resistance properties. It was thought that PVB and core shell rubber particles demonstrated a similar mechanism between the epoxy matrix and the CF fabric.

TABLE 5 Comparison of the mechanical properties of epoxy/PVB matrix (EPVBCs) to the properties of other materials reported in similar reinforcement studies about epoxy-based composites in the literature.

			Changes as %					
Samples	**Reference**	**Product detail**	**Tensile strength**	**Tensile modulus**	**Flexural strength**	**Flexural modulus**	**Impact strength**	**Fracture Toughness (G_{IIC}, G_{IIC})**
EPVBC40	This study	Carbon fiber (40 vol%) reinforced 0.5 wt% PVB/epoxy blend matrix composite	+3	−16	+10	−11	+29	–
EPVBC50	This study	Carbon fiber (50 vol%) reinforced 0.5 wt% PVB/epoxy blend matrix composite	+40	+49	+26	+56	+40	–
Glass fiber DGEBA/amine (epoxy)	[25]	Glass fiber-reinforced epoxy filled with 4.1 wt% hybrid nanosilica and CTBN (carboxyl-terminated butadiene–acrylonitrile) rubber	–	–	–	–	–	+15 (G_{IC})
Carbon fiber/epoxy with hybrid nanosilica and CSR	[23]	Carbon fiber-reinforced epoxy filled with hybrid 6 wt% nanosilica and 6 wt% CSR (core shell rubber)	–	−6	–	–	–	+125 (G_{IC})
Carbon fiber/epoxy with carbon micro filler	[13]	Carbon fiber-reinforced epoxy filled with 0.5 wt% milled micro-carbon fiber (100 micron length)	+1	+3	–	–	+2 (V notched Charpy)	–
Carbon fiber/epoxy with SWCNT	[13]	Carbon fiber-reinforced epoxy filled with 0.3 wt% SWCNT (single wall carbon nanotubes)	+8	−5	–	–	+6 (V notched Charpy)	–
Carbon fiber/epoxy with CSR	[24]	Carbon fiber-reinforced epoxy filled with 11.4 wt% CSR (core shell rubber)	+104	+31	–	–	+17 (Izod)	–
Carbon fiber/epoxy with CSR	[24]	Carbon fiber-reinforced epoxy filled with 19.4 wt% CSR (core shell rubber)	+3	+44	–	–	+87 (Izod)	–
Carbon fiber/GO-modified epoxy	[26]	0.2 wt% Graphene oxide (GO) modified epoxy reinforced by carbon fiber	–	–	+6 (three-point bend)	+10 (three-point bend)	–	–
	[27]	MWCNT (0.3–1) wt% and E20 (epoxy) solutions were sprayed	+2	−3		+4 (three-point bend)	–	+25 (G_{IC}) +17 (G_{IIC})

TABLE 5 (Continued)

Samples	Reference	Product detail	Changes as %					
			Tensile strength	Tensile modulus	Flexural strength	Flexural modulus	Impact strength	Fracture Toughness (G_{IIc}, G_{IIc})
Carbon fiber/epoxy with MWCNT by developed spraying process		on CF fabrics as a coating and epoxy matrix (E51) integrated into CF fabric			+3 (three-point bend)			

Note: "– "means decrease in properties. "+" means increase in properties.

3.4 | SEM morphologies

The SEM morphologies in Figure 6 show PVB particulates in epoxy/PVB on the fracture surfaces. The 0.5% PVB-containing epoxy blend (EPVB0.5) presented a homogeneous distribution. PVB particulates were round and spherical. The 4% PVB-containing epoxy blend (EPVB4) showed agglomerations. The PVB agglomerates could have reduced the degree of crosslinking in the epoxy bonds, and this situation could have affected mechanical properties negatively. This behavior was also observed in our previous study.[11] Therefore, EPVB0.5 was used as the matrix for the carbon-aramid hybrid fiber-reinforced composites in this study. Moreover, EPVB0.5 showed rough fracture surfaces that were related to a toughening mechanism compared to the pure epoxy.[28]

Figure 7 shows the SEM morphologies of the fracture surfaces of the flexural test samples after the tests. The cross-sections of the carbon-aramid hybrid fiber-reinforced composite laminates in the thickness direction are shown after the tests. These SEM morphologies revealed complex breakages such as delamination, matrix cracking, fiber-matrix debonding, and fiber breakage. EPVBC60 showed a largely delaminated area at both the compressive and tensile sides of the composite. On the contrary, the fractured specimens of EPVBC40 and EPC40 had much less delaminated areas. A largely delaminated area and severe matrix cracking in the fractured EPVBC60 indicated poor fiber–matrix interfacial bonding. On the other hand, fewer delaminated areas and less matrix cracking in EPVBC40 indicated improved interfacial bonding that may have been restricted by fiber–matrix debonding, fiber pull-out, and ultimate delamination.[29] Moreover, considering the interfaces of the fiber-PVB-containing epoxy matrix and the fiber-epoxy matrix, it was observed that the fiber-PVB-containing epoxy matrix showed more improved interfaces. While fractures were seen along the interface for EPC40, there was almost no fracture along the interface for EPVBC40.

While PVB particulates affected the fiber-reinforced composites positively for EPVBC30, EPVBC40, and EPVBC50, they created adverse effects for EPVBC60. The shape of PVB particulates changed for EPVBC60, and they became more agglomerated between the interlayers as shown in Figure 7. These particulates caused some void spaces between the interlayers. Therefore, microcracking could easily form and grow. This situation showed the very low interlaminar strength of the material. The PVB particulates in EPVBC40 showed more homogeneous and spherical structures. These particulates prevented the growth of micro-cracks and increased the compatibility between the fiber and the matrix.

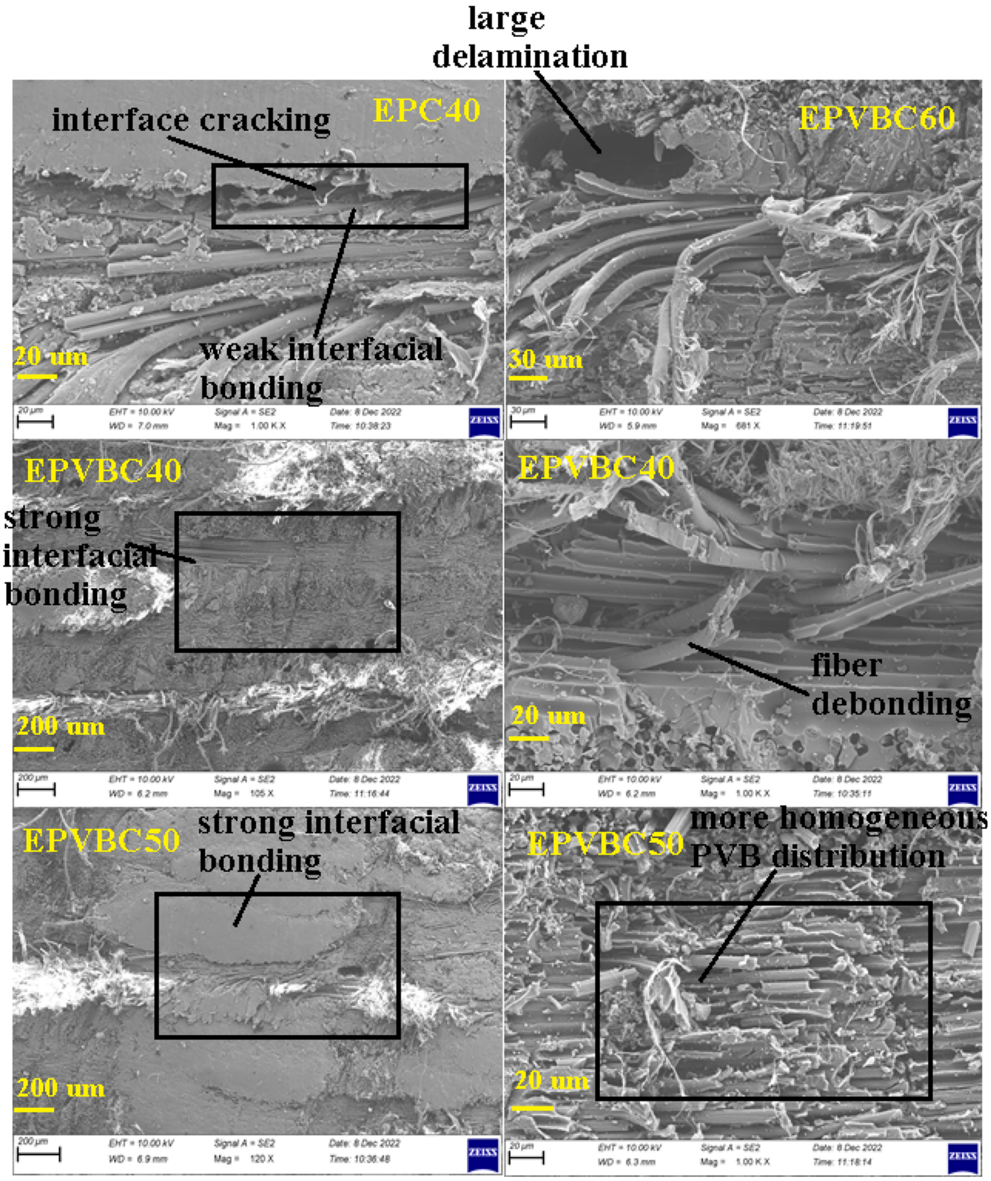

FIGURE 7 Cross-sections of carbon-aramid hybrid fiber-reinforced composite laminates in the thickness direction after the flexural tests.

These developments also improved strength and impact resistance values. The trends of ILSS, tensile, flexural, and impact resistance test results were highly compatible with the SEM morphologies of the tested samples.[17,30]

Currently, fiber-reinforced epoxy composites require high impact resistance without any adverse effects on their mechanical properties such as strength and modulus values, especially for the aviation, automobile, and military industries. The composites with the Epoxy/PVB blend matrix in this study including EPVBC40 and EPVCB50 presented substantial increases in strength, modulus, and impact resistance results. These advanced composites seemed to be important candidates for engineering applications in the automobile and aviation sectors. In addition to these, if these epoxy/PVB matrix materials were reinforced by nanofillers together with CF fabrics such as CNTs, graphene, graphene oxide, silica, and titanium oxide particles, more favorable results could be obtained. Moreover, it is expected that more advanced methods such as VARTM, autoclave, and vacuum infusion can increase the effectiveness of PVB in fiber-reinforced epoxy matrix materials. These methods can make PVB powders more effective at higher fiber reinforcement ratios.

4 | CONCLUSION

In this study, we prepared a series of carbon-aramid hybrid fiber fabric-reinforced epoxy and epoxy/PVB matrix composites (EPCs and EPVBCs) in the fiber addition ratio range of 30–60%. The weight ratio of PVB in the epoxy resin was adjusted 0.5% due to the impressive effect of this ratio in our previous study. Tensile, flexural, ILSS, and impact strength test results were obtained. The EPVB0.5 sample showed significant increases in impact strength and tensile strength values in our previous study, and it was used as the matrix for the carbon-aramid hybrid fiber fabric-reinforced composites in this study. EPVBCs showed increases in modulus values as well as strength and impact strength values. Moreover, the ILSS tests of EPVBCs presented highly improved results. Overall, EPVBC40 and EPVBC50 showed the most prominent and improved results. They also presented substantial superior mechanical properties compared to EPC40 and EPC50. It was thought that this result could be attributed to the increase in the compatibility of the fiber/matrix and higher interfacial strength between fiber and matrix. However, EPVBC60 showed sharp decreases in terms of mechanical properties. In comparison to EPC60, the behavior of PVB particulates affected the composite with the fiber ratio of 60% very adversely and large delamination was observed between fiber and matrix. It was thought that EPVBC40 and EPVBC50 are important thermoplastic composite candidates for the potential applications with high strength, modulus and impact resistance in automobile and aviation sectors, especially. They can also replace with some composite materials in some applications such as automobile body sections, aircraft interior panels, and the wings and radome of airplanes. It is thought that epoxy/PVB matrix will substantially contribute to most of fibers such as carbon, glass and aramid. In the future, to obtain more favorable results, the epoxy/PVB matrix can be used in hybrid systems by using nanofillers such as CNTs, graphene oxides, silica particles, and titanium oxide particles. Furthermore, the epoxy/PVB matrix can also be more effective at higher fiber reinforcement ratios by more advanced composite processing methods such as autoclave and VARTM.

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DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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