Tensile, Compressive, and Shear Properties of UHMWPE/Carbon Hybrid Fiber Reinforced Polymer Composites with a Novel Liquid Thermoplastic Resin, Elium®

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ABSTRACT

In virtue of recyclability purposes, efficient and ease of fabrication, higher impact and damage tolerance properties, a new liquid Methyl Methacrylate (MMA) thermoplastic resin, Elium® 188, has been recently developed. This new resin can replace (epoxy) thermoset-based resins for the fabrication of laminated composites at room temperature with equivalent in-plane and superior out-of-plane mechanical properties. In this study, plain weave ultra-high molecule weight polyethylene fiber (UHMWPE), carbon fiber, and their hybrid systems are fabricated by Vacuum Assisted Resin Infusion (VARI) method at ambient temperature. ASTM standard tests of tensile, compression, and shear have been conducted to determine the mechanical properties of the laminates with the aim of comparing the results with those of thermosets. Fractographic analyses are performed to have a better understanding of the behavior of these new thermoplastic-based laminates. The test results are also validated by the Mechanics of Structure Genome (MSG). Results show the mechanical properties of thermoplastic-based laminates are comparable to those thermoset-based but with the aforementioned merits, yielding more applications in the industry. These datasets can be used as a reference for researchers working in theoretical studies such as impacts or damage simulations.

KEYWORDS: *Vacuum Assisted Resin Infusion (VARI); Thermoplastic Resin (Elium); Mechanical Properties; Woven Composites, Fractography*

1 INTRODUCTION

The use of fiber-reinforced polymer composites (FRPCs), due to their excellent mechanical and geometrical properties such as high strength to weight ratio and ease of manufacturing, has gained a great deal of attention in many industries, namely aerospace, mechanical, marine, to name but a few (Kazemi et al. 2018). Currently, the most applicable FRPC laminates are fabricated with thermosetting organic resins, which exhibit great mechanical properties. Nonetheless, these thermoset-based laminates have the significant disadvantage of poor out-of-plane properties, as they exhibit brittle behavior that can increase the susceptibility of delamination in those structures, especially in impact applications (Matadi Boumbimba et al. 2017).

In recent years, the requirements for material recyclability has prompted the development of thermoplastic laminates. However, for the development of such a thermoplastic resin, fabrication and mechanical characteristics should also be considered. ARKEMA has been able to develop a novel liquid thermoplastic resin, polymerizing at room temperature to satisfy manufacturing requirements and furthermore, improving the out-of-plane response of FRPCs (Kinvi-Dossou et al. 2018). The woven FRPC thermoplastic-based laminates are assumed to enhance impact energy absorption (Reid and Zhou 2000) and to show equivalent in-plane properties (Aitharaju and Averill 1999), compared to unidirectional FRPCs. Among woven laminates, UHMWPE, glass fibers, and carbon fibers are being widely used, thanks to their exceptional mechanical properties. UHMWPE fibers, due to their extremely high molecular mass, provide; exceptional impact strength and abrasion resistance, special processing characteristics, good resistance toward chemicals and wear, high energy dissipation and are lightweight. Its ability to absorb energy is 1.8 times that of carbon fiber, 2.6 times that of aramid fiber, and 33 times that of glass fibers. On the other hand, they also suffer from several drawbacks, such as low surface energy and poor creep and heat resistance. Therefore, when combined with carbon fibers, UHMWPE fibers can achieve higher performance, as they possess excellent properties such as higher stiffness, better corrosion resistance, thermal and electrical conductivity.

Regarding traditional thermoplastic-based FRPC laminate, there are quite a few studies in the literature. However, for this new liquid Methylmethacrylate (MMA) thermoplastic matrix, Elium® 188, most of the studies are focused on the out-of-plane mechanical characteristics, such as the low-velocity impact response by Bhudolia and Joshi (Bhudolia and Joshi 2018). Boufaida et al. (Boufaida et al. 2015) analyzed the influence of different fiber surface treatments on the mechanical properties of plain weave glass fiber in (Elium) thermoplastic-based composites. (Shanmugam et al. 2019) surveyed two different surface treatment approaches for improving bonding strength between this thermoplastic resin, Elium, and titanium alloy plate by multi-step anodization and single-step micro arc oxidation methods.

With the recent introduction of Elium® 188, most of the investigative work has been to determine the impact characteristics of FRPC laminates, mainly consisting of glass fibers. However, there are much fewer comprehensive studies of in-plane mechanical properties for thermoplastic-based FRPC laminates and their hybrid systems, both experimentally and theoretically. Hence, a more comprehensive study of these thermoplastic-based laminates is needed, as these data are cornerstones for designers who deal with theoretical works, such as impact and damage simulations.

2 EXPERIMENTAL PROCEDURE

The reinforcements used in the current study are plain weave UHMWPE and carbon fibers. UHMWPE fibers have been bought from two different companies to compare the results. The first UHMWPE plain weave fiber is from SOVETL™, and the other one is QuantaFlex™ from QUANTUMETA. Regarding carbon fibers, plain weave HexForce 282 from Hexcel Corporation is used. The resin used in this work is a low viscosity (Viscosity Brookfield LVF #260 rpm of 100 mPa.s) thermoplastic liquid resin (Elium® 188) recently developed by ARKEMA. This thermoplastic resin is suitable for the infusion process and can be cured at room temperature with the major advantages of being post-thermoformable, recyclable, and offering new possibilities for composite/composite or composite/metal assemblies. The curing time depends on the ambient temperature and the size of the laminate. Surprisingly, at high temperature of 80 °C, Elium® 188 resin cures in just 5 minutes. The properties of a 4 mm unfilled resin casting of Elium are given in Table 1 based on ARKEMA's technical datasheet. In addition, tensile tests for pure Elium are conducted based on ASTM standards to compare the results with the data from the technical data sheet, which is provided in Table 1.

Table 1. Properties of a 4 mm unfilled resin casting

Resin	Hardener	Mixing Ratio	Density (gr/cm^3)	Tensile Strength (MPa)	Tensile Modulus (MPa)	Compression Strength (MPa)	Fracture Toughness ,K_{1c} ($MPa.m^{0.5}$)	Flexural Modulus (MPa)
Elium 188	Benzoyl Peroxide	2%	1.01	76	3300	130	1.2	3250

Before fabricating the composite samples, pure Elium samples are fabricated for comparing the results provided by the manufacturer and to have a better understanding of the behavior of composite laminates in the standard tests and in the failure mode analyses. To do so, ASTM D638 is followed for tensile test, which consists of dumbbell-shaped (dog bone) samples. For Elium® 188 thermoplastic resin, 2% Benzoyl Peroxide (BPO) is used as a hardener or initiator for polymerization. This amount can be extended to 3%, based on the desirable curing time. This resin cures less than an hour in room temperature. Regarding composite laminates, vacuum assisted resin infusion (VARI) method is followed. The kit used for this process consists of a vacuum pump, pressure pot, inlet and outlet hoses for infusion of resin, peel plies, breather, mesh flow, spiral tube, to name but a few (Fig. 1). VARI involves the vacuum injection of a low viscosity resin into composite laminates. Thanks to the high permeability of woven fabrics, infusion processes are normally used for fabrication. Regarding hybrid systems, two layup systems are considered, the first one has carbon fibers on top and bottom and UHMWPE fibers in the middle Fig. 1 (b - left), the other has UHMWPE fibers on top and bottom and carbon fibers in the middle Fig. 1 (b – right).

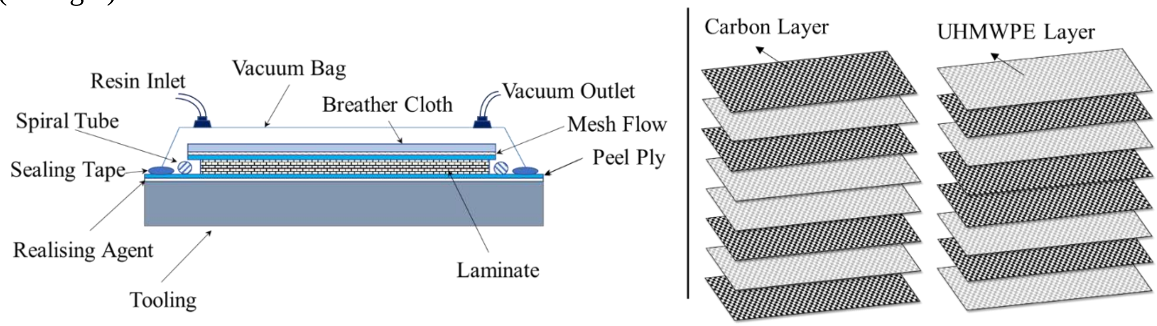

Fig. 1. Schematic of Vacuum Assisted Resin Infusion (VARI) (a), Hybrid lamination layups (b)

After curing, based on the ASTM standards for tensile D3039, compression D6641, and shear D5379 tests, the samples are cut via abrasive waterjet cutting machine and tested. The thickness and the fiber volume fraction of the laminates are provided in Table 2.

Table 2. Laminate code, layup, and thickness of each laminate in this study

Laminate code	Layup*	Thickness (mm)	Average Fiber Volume
1	$(C)_8$	1.57 ± 0.02	0.58 ± 0.03
2	$(C/PEQ)_{2S}$	2.23 ± 0.02	0.54 ± 0.03
3	$(C/PES)_{2S}$	2.27 ± 0.03	0.50 ± 0.02
4	$(PES/C)_{2S}$	2.27 ± 0.03	0.49 ± 0.02
5	$(PEQ)_8$	2.88 ± 0.03	0.50 ± 0.02
6	$(PES)_8$	2.95 ± 0.04	0.47 ± 0.02

*C: Carbon Plain Weave Fiber
PES: UHMWPE Plain weave fiber from SOVETL
PEQ: UHMWPE Plain weave fiber from QuantaFlex

3 EXPERIMENTAL RESULTS

The tensile properties of laminates and Elium are shown in Fig. 2. As can be seen, the figure consists of seven different curves, six for the laminates and one for pure Elium as a reference to compare the results. For pure Elium, the material behavior initially is linear and then enters into non-linear behavior due to to its visco elastic property. By dividing the ultimate load by the gauge length area, the ultimate strength obtained is between 68 to 75 MPa, which is comparable with the ultimate strength provided in the manufacturer datasheet. Young's modulus value also agrees well with the datasheet. Concerning tensile behavior of the laminates, in small displacements (strains), a nonlinear regime can be seen, followed by a linear behavior. This has roots in the thermoplastic nature of the resin, which gradually becomes less effective when the effect of reinforcements dominate. This linear behavior continues until the material experiences a brittle failure. The failure is more brittle for laminates that contain carbon fibers and is more plastic for UHMWPE laminates. Comparing the diagrams in Fig. 2, the strength and Young's modulus for laminate 1 (C_8) are much higher than others, with the average value of $E \approx 57.9$ GPa and strength of $\sigma \approx 720$ MPa. These values are comparable with those of epoxy-based carbon laminates. For comparison, the same carbon fabric with Sikafloor epoxy is fabricated and tested, which shows equivalent Young's modulus of $E \approx 58$ GPa and tensile strength of $\sigma \approx 710$ MPa. For hybrid systems, the samples containing QuantaFlexTM UHMWPE fibers show higher stiffness and strength in comparison to SOVETL laminates. Although two different UHMWPE fiber laminates with the same density and almost the same surface weight are fabricated and tested, the stiffness and strength for PEP-A172 (QuantaFlexTM) laminate are higher than those for SOVETL one. This can be attributed to higher surface weight and larger unit cells and yarn spacing for PEP-A172 fabric, providing better resin impregnation. As a result, better bonding properties results in lower void estimations and higher mechanical properties. However, comparing the results of UHMWPE $(PES)_8$ and $(PEQ)_8$, laminates with pure Elium show Young's moduli close to that of pure Elium. This may be due to poor bonding properties between UHMWPE fibers and Elium, and/or poor fibers quality (, as their mechanical properties are not provided in the manufacturer datasheet). Regarding hybrid systems with the same fibers and different layups, the results are almost the same for in-plane mechanical properties in tension. Hence, the stacking sequence as provided in Fig. 1 does not have a significant effect on the tensile results (laminate 3 $(C/PES)_{2S}$ and 4 $(PES/C)_{2S}$).

As the failure characteristics of the new thermoplastic-based laminates have not been comprehensively investigated, Scanning Electron Microscopy (SEM, JEOL-6390) is used for analyses of the failure modes. Images show the fractography of laminates 1 (C_8) and 5 $(PES)_8$ in tension, showing a complete brittle failure for laminate 1 (C_8) and a plastic one for laminate 5 $(PES)_8$. In Fig. 2 (a), delamination between different carbon laminate layers as well as warp and weft yarns can be seen. It describes the sequence of failure of carbon fiber starting by crack propagation, debonding, and fracture. These interfacial cracks prevent the force to be transferred in a proper way, resulting in matrix debonding, which finally causes carbon fibers to pull-out. Fig. 2 (b) illustrates fracture in fiber, which has a non-planar and irregular surface with a serrated aspect; no necking shape was observed. In contrast to the failure modes of laminate 1 (C_8), laminate 5 $(PES)_8$ shows a completely different failure regime in tension. In Fig. 2 (c), (weft) fibers pull-out are obvious, being significantly elongated without the brittle pattern that appears in carbon. The fracture in (weft) fiber occurred far from the warp fibers, which shows the necking and elongation prior to fracture. In (weft) fibers, some twisting can be seen, which can because of the preparation of the SEM sample or breakage of some fibers during the test. Fig. 2 (d) shows necking and shrinkage in the diameter of UHMWPE fibers, reaching a complete failure and consequent fracture, which represents the plastic behavior of polyethylene fibers.

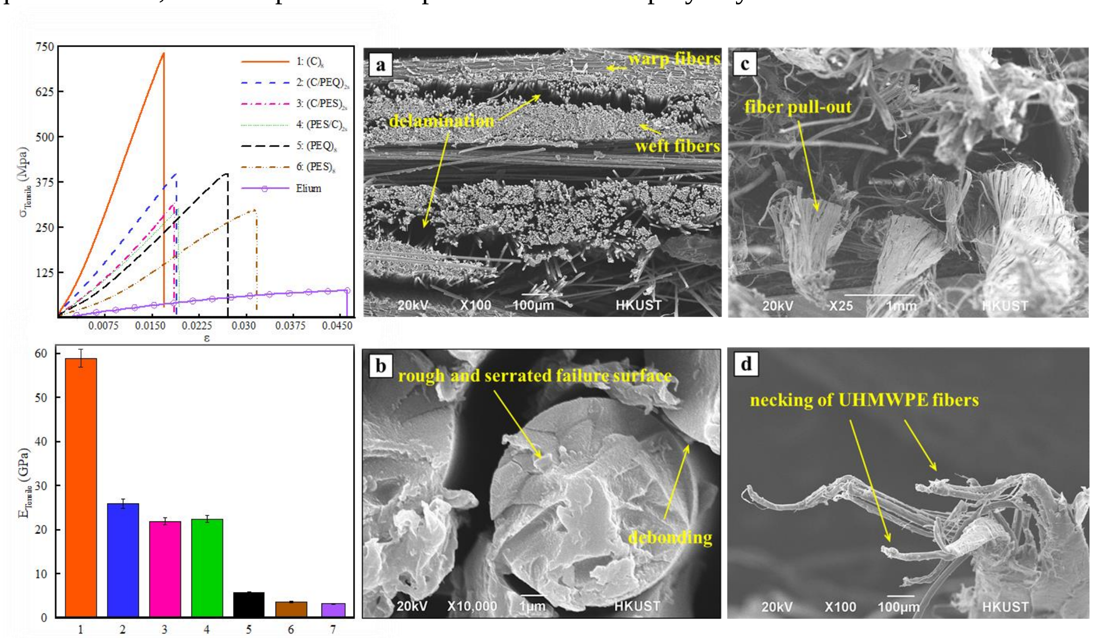

Fig. 2. Tensile stress-strain curve and Young's moduli values, fractography of laminate 1 (C_8) failure in tension: warp and weft failures (a), carbon fiber breakage surface (b), and fractography of failure in tension for laminate 5 $(PEQ)_8$: weft fiber pull-out (c), and necking of UHMWPE fibers (d)

Regarding FRPC laminates, ultimate compression results are usually much lower than those in tension; however, Young's moduli are almost the same. To validate this, test results in compression are presented in Fig. 3, which in a similar way to tension, show the highest stiffness for carbon fiber laminates and its hybrid systems and the lowest values for UHMWPE laminates. Concerning the compression stress-strain curve in Fig. 3, three different zones (regions) can be seen. In the first zone, the response is nearly elastic (linear), where compressive Young's moduli are calculated. In the second zone, the material undergoes a non-linear behavior, which this non-linearity has to do with matrix micro-cracking and micro-plasticity, resulting in a decrease in compressive Young's modulus. In the third region, where compressive ultimate strength is achieved, the material fails. However, this failure is brittle for high stiffness materials like laminates 1 (C_8) to laminate 4 $(PES/C)_{2S}$ (containing carbon fibers). For laminates, 5 $(PES)_8$ and 6 $(PEQ)_8$ that consist of only UHMWPE fibers, the breakage does not happen, thanks to the plastic behavior of fibers.

Regarding failure analyses, as the material undergoes a combined end- and shear-loading, a shear failure is expected, which can also be combined with compression. To show this, laminate 1 (C_8) failure is considered in Fig. 3 (a, and b), showing the material failure in shear mode (45°). Fig. 3 (a) shows the failed area along the weft and warp yarns. In Fig. 3 (b), fracture surface for carbon fibers in shear failure mode are depicted. As a result of this failure, the laminate is fractured/broken into two separate parts with 45° fractured surfaces. However, for other hybrid or UHMWPE laminates, this brittle failure response was not observed, as UHMWPE fibers show a plastic response, (Fig. 3 (c, and d)). During compression, UHMWPE fibers rotate and take on a "hook" shape, Fig. 3 (d). Hence, fiber breakage is not the main failure mode in this case. As a result, crack initiation, propagation, as well as debonding between fibers and resin are considered to be the main failure modes that happen gradually, resulting in a gradual decrease of the loading. Moreover, as the fibers rotate, they undergo a severe deformation. Consequently, cracks appear in these fibers, Fig. 3 (c). This phenomenon causes a gradual fiber failure, another reason for the decrease in compression the load and strength. In UHMWPE laminates, the failures for different fibers happen at different intervals, justifying the non-linearity in the compression stress-strain curve.

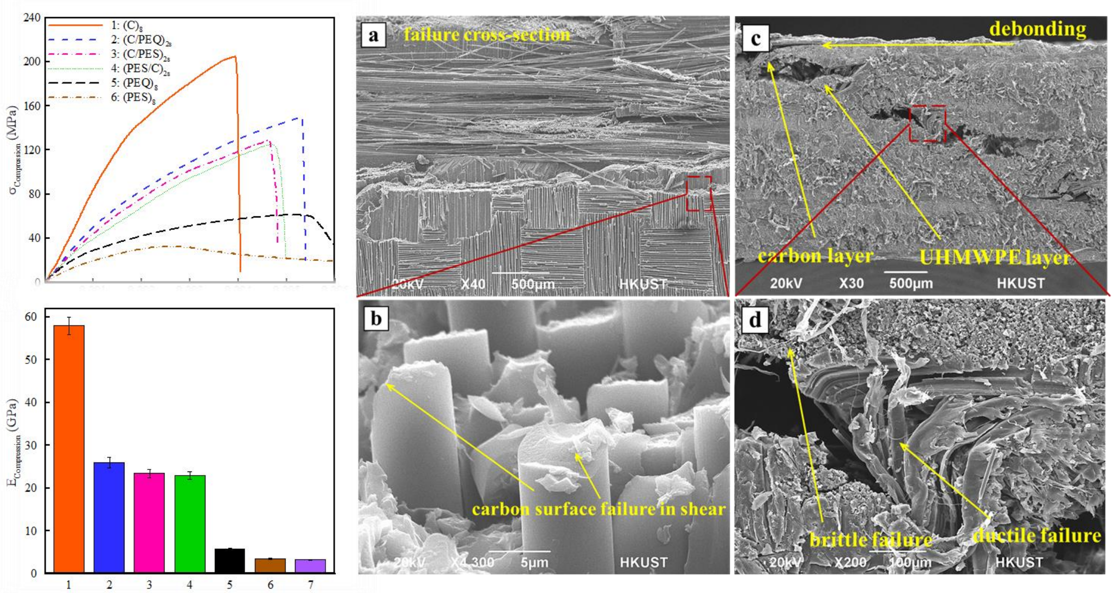

Fig. 3. Compression stress-strain curve and Young's moduli values, fractography of laminate 1 (C_8) failure in compression: warp and weft failures (a), carbon single fiber failure cross-section (b), and fractography of failure in compression for laminate 2 $(C/PEQ)_{2S}$: (c) and (d)

In woven or 0/90 laminates the shear ultimate strength may be lower than the maximum force attained during the test and the fibers may reorient following shear failure, allowing the fibers to carry much more force. This reorientation usually occurs in composites with tough matrix materials that are very nonlinear in shear or in composite laminates containing off-axis fibers. In these cases, the shear failure force can often be determined by correlating visual observation of failure in the test section with a force drop or by a significant change in the slope of the force-displacement plot. However, like in this study, materials may deform to such an extent that shear failure does not occur at all; rather the specimen ultimately fails in a mixed-mode failure. In this situation, the response of the material can be categorized into four different zones. First, in low strains, the response is linearly elastic (stage 1). After that, larger strains result in matrix micro-plasticity and micro-cracking, which cause non-linearity and a decrease in the shear modulus of the material (stage 2). For larger strains, the initiation and propagation of these micro-cracks are slowed down due to crack saturation, which provides a more linear behavior (stage 3). If more loading is applied, the material fails at very high strains due to the breakage of the fibers (stage 4). However, as the laminates tested here entered a mixed-mode zone, due to the limitation in the fixture (arm) movement, the 4th failure zone (stage 4) was not achieved. In such a case, as provided in ASTM D5379, to avoid reporting of results that are not representative of shear strength, this test method terminates data reporting at an engineering shear strain of 5 % (an approximate value at the end of stage 3). The results regarding the shear strengths, shear moduli are presented in Fig. 4. For comparison, carbon fabrics are fabricated with Sikafloor epoxy and tested. These values for shear modulus and ultimate strength are $G \approx 5.0$ GPa and $\sigma \approx 80$ MPa, respectively.

In contrast to epoxy-based composites, which show a brittle failure (i.e. fracture) in the V-notch section, Elium-based composite laminates do not show such a response due to the visco elastic behavior of the resin. Fig. 4 illustrates SEM images of laminate 5 $(PEQ)_8$, Multiple cracks initiated and propagated along the V-notch (both top and bottom), resulting in partial delamination around the cracks. For laminate 1 $(C)_8$, after initiation and propagation of the cracks, fibers suffered a brittle and sudden breakage. However, for laminate 5 $(PEQ)_8$, due to the plastic behavior of the UHMWPE, fiber disorientation, as well as shrinkage in fibers diameter are observed.

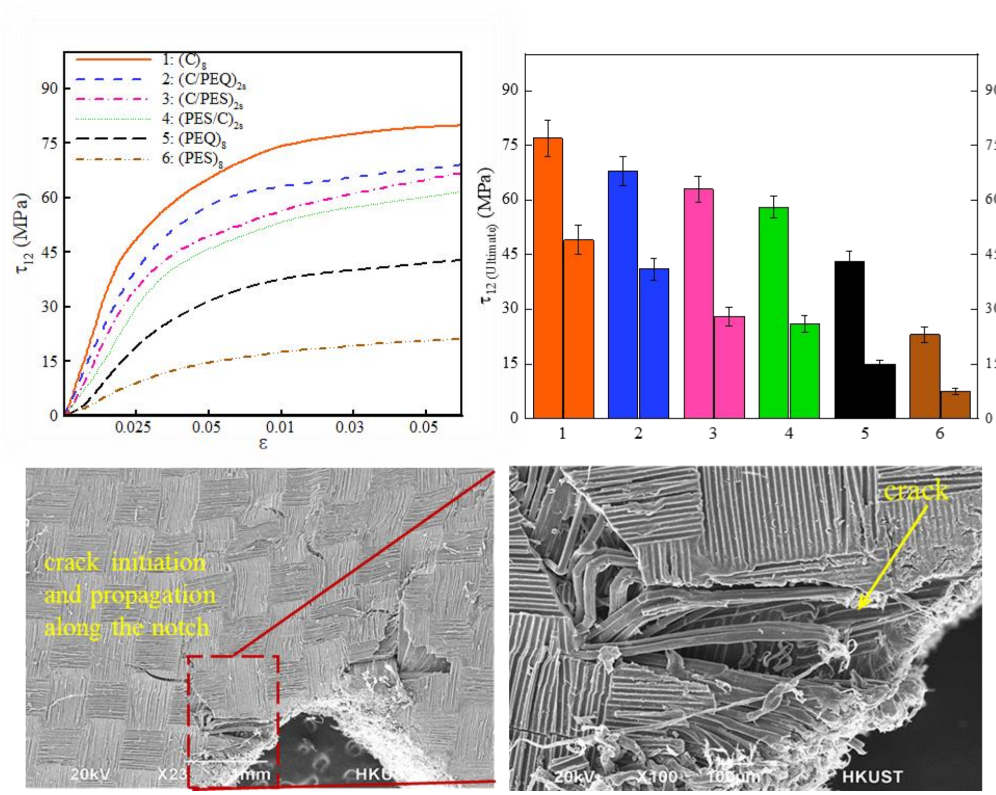

Fig. 4. Shear stress-strain curve, moduli, and ultimate shear strengths for different laminates, and fractography and microscopic features of laminates laminate 2 $(C/PEQ)_{2S}$ in shear near the V-notch section

4 THEORETICAL RESULTS

To verify the experimental results, the Mechanics of Structure Genome (MSG) is applied (Liu et al. 2017). The Structure genome (SG) can be defined as the smallest mathematical building block of a structure, which serves as the analysis domain within MSG. SG can be a one-dimensional (1D), 2D or 3D domain that depends on the heterogeneity of the structure. In MSG, a homogenized model can be formulated by minimizing the information loss between the original heterogeneous body and the homogenized one (Kinvi-Dossou et al. 2018). Here, one of the laminates, that is laminate 1 (C_8), is chosen for this end. The periodicity of the fabric-repeating unit is T = 2.0mm, Fig. 5(a). From SEM measurements, the section shape of a fiber tow is considered as an elliptic with a major diameter and minor diameter of 1.5mm and 0.13mm, respectively (Fig. 5 (b)). The diameter of fibers in the tow randomly varies between 6 and 7 µm (Fig. 5 (c)). At the microscopic scale, the yarns are regarded as a unidirectional composite that consists of a volume fraction of carbon fibers around 80%, embedded in Elium resin. Within the MSG, 2D unit cells are sufficient to calculate the effective properties of transverse isotropic yarns. Hexagonal packing is adopted here, Fig. 5(d). The Young's modulus and the Poisson's ratio of carbon fibers are respectively E_f = 220.0 GPa and ν_f = 0.22, and Elium Young's modulus and Poisson's ratio are E_m = 3.60 GPa and ν_m = 0.37, respectively as tested experimentally. Table 3 provided the effective elastic properties of the laminate 1 (C_8) composite yarn, as computed using the MSG and their comparison with the results of Rule of Mixture.

At the mesoscopic scale, the unit cell considered for the simulation is a 2×2 yarns embedded in Elium. Texgen software was then used to generate a voxel mesh file with a local material orientation for each element in Abaqus format (Lin et al. 2011). 50×50×20 hexahedral quadratic elements (C3D8) have been generated. The engineering constants agree with experimental results in the plane of the plate. However, the findings are different for the interlaminar shear modulus G_{13}. The experimental interlaminar shear modulus was estimated through a beam theory on a 16 ply laminate. The effect of plies number on this predicted value has not been investigated. The main advantage of our MSG-based model is its ability to simultaneously estimate the fives independents components of the elastic properties tensor through only one computation, with a relatively low CPU time.

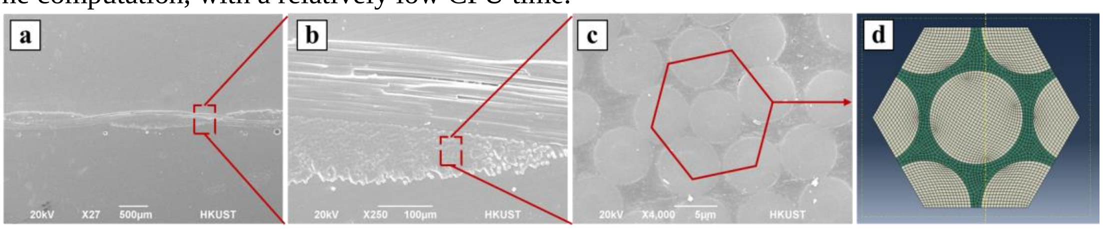

Fig. 5. Through-thickness SEM observations yarn's undulations (a, and b), hexagonal packing (c), (d) 2D mesh RVE

Table 3. Effective properties of the composite yarn for laminate 1 (C_8)

	SGM	Rule of Mixtures (Reddy)
E_1 (GPa)	182.22	182.26
$E_2 = E_3$ (GPa)	32.51	15.59
G_{23} (GPa)	12.06	-
$G_{13} = G_{12}$ (GPa)	10.57	4.84
ν_{23}	0.24	-
$\nu_{13} = \nu_{12}$	0.34	0.35

Table 4. Macro homogenization results and experimental data for laminate 1 (C_8)

	Numerical	Experimental
$E_1 = E_2$ (GPa)	57.5	57.9
E_3 (GPa)	13.8	-
G_{12} (GPa)	5.09	5.00
$G_{23} = G_{13}$ (GPa)	3.17	4.63
ν_{23}	0.11	0.08
$\nu_{13}=\nu_{12}$	0.36	-

5 CONCLUSION

In this study, new thermoplastic-based FRPC laminates consisting of plain weave UHMWPE, carbon fiber, and their hybrid systems with different layups were fabricated by Vacuum Assisted Resin Infusion (VARI) method at ambient temperature. ASTM standard tests in tensile, compression, and shear were performed to obtain the mechanical properties, followed by fractography of the failure modes by Scanning Electron Microscope (SEM) to have a better understanding of these new thermoplastic-based laminates behavior. With comparing the results of this study with those in thermoset-based FRPC laminates, Elium can be a competitive resin to replace traditional resins for the fabrication of composite structures, with the advantages of; better recyclability, higher impact and damage tolerance properties and, efficient and ease of fabrication. The experiment results are validated by Mechanics of Structure Genome (MSG) approach, which provides a more comprehensive data set for researchers to apply the data for theoretical studies in different applications such as impact and damage mechanisms.

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