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ORIGINAL PAPER

# Fabrication and Characterization of Acrylonitrile Butadiene Rubber and Stitched E-Glass Fibre Tailored Nano-Silica Epoxy Resin Composite

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**Abstract**
In this research, the role of acrylonitrile butadiene rubber (ABR) and nano-silica along with stitched E-glass fibre in epoxy composite was investigated. The main objective of this present study is how the additions of ABR and nano-silica particle influences in the mechanical, drop load impact and fracture toughness properties of glass-epoxy composite. The layering of rubber sheet for improving the reinforcing effect is a novel approach in reinforced plastic. The composites were prepared using various stacking sequence of ABR and E-glass fibre with nano-silica toughened epoxy resin. The composites were characterized using ASTM standards. According to mechanical properties the composite designation $FAF_2$ symmetric arrangement gives better overall (rank of 97) properties. Similarly, the composite designation $AFA_2$ gives very higher penetration resistance against the fast moving impactor. The fracture toughness behaviour of the composite shows higher energy release rate of 3.8 $MJ/m^2$ and fracture toughness of 22 MPa for 3 vol.% of nano-silica particle dispersed FAF epoxy composite. The SEM micrographs show improved adhesion of 3 vol.% nano-silica with fibre and ABR in composites. The penetration improved rubber based epoxy composites could be used in armour based device, automobile body parts and domestic product manufacturing industries.

**Keywords** Composite · Rubber · Nano-silica · Mechanical properties · Drop load impact and fracture toughness

## 1 Introduction

Rubber is an elastomeric material having good elastic properties and energy absorption behaviour. The use of rubber is drastically increased in the current engineering trend [1]. Lot of engineering applications are required either natural rubber or synthetic rubber reinforcements based on the output properties required [2]. Polymer matrix composites also widely used material in engineering having lot of scope and applications in industrial, automobile and domestic applications [3, 4]. The polymer matrix composites even used in high load bearing applications [5]. In general for high load bearing applications, the polymer matrix may reinforce with high stiffness long continuous fibres and nano or micro particles [6]. Recent years many researchers used to add metallic or aramid fibres along with primary fibre to improve the sudden load bearing and other mechanical properties. Moreover, some researchers have researched intra-ply format of fibre 1 and fibre 2 as a combined form to get improved output in mechanical and sudden load bearing properties [7]. Some researchers have done studies by reinforcing the fibre 1 and fibre 2 or fibre 1 and metallic meshes as various stacking sequenced form to improve the load bearing capabilities and other mechanical properties. Arun prakash et al. [8] studied the effect of adding E-glass fibre, aluminium and stainless steel-wire mesh as an alternating layered composite to improve the low-velocity drop load impact resistance. According to their statement placing metallic wire-meshes may improve the sudden loading characteristics provided with good interfacial adhesion.

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Dinesh et al. [9] studied the effect of adding stainless steel wire-mesh along with pineapple fibre in the epoxy matrix. Their research study revealed that the addition of SS wire-mesh greatly influenced in the load bearing capabilities of bio oil toughened epoxy resin composite. Similarly, Gokul et al. [10] researched the combined effect of E-glass and Kevlar fibre in the epoxy resin matrix. The outcome of the study revealed that the addition of Kevlar fibre along with glass fibre as an intra-ply format improved the low velocity impact damage behaviour of epoxy resin composite. Arul et al. [11] investigated the effect of adding SiC wire-mesh along with three different types of natural fibres to improve the low velocity impact behaviour of natural fibre composite. According to their conclusion, the presence of SiC wire-mesh greatly supports in the improvement of mechanical and drop load impact damage behaviours by suppressing the micro cracks formation. Lokesh et al. [12] researched the effect of adding waste tyre rubber particle of 150 μm into epoxy resin. They concluded that the addition of waste rubber particles into epoxy resin increased the toughness of matrix and improves the mechanical and wear behaviour. Ozdemir et al. [13] investigated the effect of adding rubber particles into CFRP composites. The study of the author's revealed that the addition of rubber particles increases the fracture toughness and interlaminar shear strength behaviour of CFRP-epoxy composite. Based on the previous literatures many researchers have studied the low velocity impact behaviour of epoxy composites by incorporating metallic wire-meshes and mixed form of two different fibres. But very few researchers only have taken natural or synthetic rubber as a reinforcing element instead of wire-meshes and other engineering fibres. Based on this research gap the current research is aimed such a way that to utilize an ABR sheet of lower thickness to acts as reinforcing element along with E-glass fibre and nano-silica particle to improve the mechanical, fracture toughness and drop load impact behaviour of epoxy composite. In another hand the nano-silica particle could select as matrix toughening agent since nano-silica has a tendency to improve adhesion with matrix, improve mechanical properties of composites, having high specific heat capacity, which inturn improves the thermal properties and abundantly available resource. Parthipan et al. [14] revealed in their research study that the addition of nano-silica particle into epoxy resin improved the mechanical and adhesion behaviour of reinforcement and matrix. Similarly, Thendral et al. [15] investigated the role of nano-silica particle in bio compatible PBAT composite film. The authors concluded that the addition of nano-silica particles improved the anti-microbial and tensile strength of bio compatible film. Thus the role of nano-silica in the composite application is very high and essential in many dimensions. The proposed composites could be prepared using hand layup method and characterized using ASTM standards [16]. These drop load impact damage preventing epoxy composites could be used in automobile, defence and domestic applications where high damping strength of material properties are required [17].

## 2 Experimental Part

### 2.1 Materials

Table 1 shows the basic properties of research materials used in this present study. All the materials used in this present study are in as-received form without any surface-treatment.

### 2.2 Composite Making

The ABR/glass fibre-reinforced nano-silica toughened epoxy composites are prepared using hand layup method. In this, the composites are prepared via two step process. At the first step, the nano-silica particles are mixed with epoxy resin and mixed thoroughly until a complete colloidal solution takes place. At second step the colloidal solution is poured into the wax pre-coated rubber mould and allowed to fill. The ABR and E-glass fibre of totally 3 layers with a volume of 40% was laid with different stacking sequence. Finally, the composites were cured at room temperature for 48 h and removed from the mould. The mould removed composite plates are then inspected for any process damages. Table 2 shows the various epoxy composites fabricated using different stacking sequence of ABR and stitched glass fibre [18].

### 2.3 Test Sample Preparation

The cured epoxy and its composites are subjected to strength evaluation by ASTM standards. An abrasive water jet machine (KMT Water jets, S 3015, Germany) was used to cut the samples for various tests proposed. The process parameters while cutting the composites are as follows. Jet operating pressure of 200 psi, an abrasive flow rate of 0.4 g/s, nozzle tip diameter of 1.1 mm and garnet size of 80 mesh were used as parameters [19].

## 3 Characterizations

The proposed ABR and E-glass fibre-reinforced epoxy resin hybrid composite was evaluated for mechanical, drop load impact and wear properties. The tensile, flexural, interlaminar shear strength and fracture toughness were evaluated using a 10 T universal testing machine, INSTRON 4855 series, UK using ASTM D-638, D-3039, D-790 and D-5045. The impact test was carried out using a 20 J impact tester using ASTM D 256. The drop energy absorption and deformation analysis were examined using a drop load impactor following ASTM D-4762 using a drop load impactor INSTRON-9000,

**Table 1** Research materials used in study

| Material Name | Density (g/cm3) | Molecular weight (g/mol) | supplier |
|---|---|---|---|
| Epoxy resin (LY556, Araldite) | 1.18 | 190.01 | Huntsman, India, Ltd |
| TETA (HY951, Araldite) | 1.05 | 146.20 | Huntsman, India, Ltd |
| ABR (1.2 mm thickness) | 1.00 | 53.06 | Metro composite R&D, Centre, India |
| E-glass fibre | 2.54 | – | Metro composite R&D, Centre, India |
| Nano-silica (10-20 nm) | 2.40 | 59.96 | Sigma Aldrich, USA |

USA with the mass of 2Kg, striker diameter of 2.5 cm and velocity of 1.3 m/s. The SEM micro graphs were scanned using HITACHI S 1500, JAPAN. The samples were sputtered using gold before capturing images.

## 4 Results and Discussion

### 4.1 Mechanical Properties

Table 3 shows the tensile, flexural, impact and ILSS behaviour of epoxy and the hybrid composites fabricated. The best composite in all properties was selected based on the normalized strength factor method followed by ranking. The strength factor was calculated by finding normalized strength in each category and taking average of the same [20]. Based on the rank the composite designation $FAF_2$ gives overall best result. The overall ranking for this composite is 97/100, which indicates the composite posses balanced performance in all properties such as tensile, flexural, impact, ILSS and fracture toughness.

It is observed that the pure epoxy and its composites give significant results in tensile, flexural, impact and ILSS behaviour. It is noted that the pure epoxy resin gives 63 MPa, 90 MPa and 0.4 J for tensile, flexural and impact toughness respectively. Further addition of E-glass fibre into epoxy resin improved the properties. The improvement of 55%, 43% and 89% were observed for tensile, flexural and izod impact toughness. This improvement is because of the uniform load sharing behaviour of E-glass fibre in epoxy resin. The fibre receives a large load from the matrix and reduces the stress intensity factor in the matrix phase. Thus the plastic deformation never occurs at early loads [21]. It is further noted that the addition of ABR rubber of 10 vol.% along with 1 and 3 vol.% of nano-silica particles into glass-epoxy composite further modified the mechanical properties. The tensile strength of composite is reduced marginally due to the low tensile strength ABR rubber addition along with E-glass fibre. On compare, with $FAF_1$ the $FAF_2$ composite designation gives improved tensile strength. The presence of 3 vol.% of nano-silica particles greatly supports load bearing properties and also improves the adhesion of reinforcements with matrix [22]. Thus little improvement in tensile strength is observed for composite designation $FAF_2$.

It is noted that in flexural and impact properties the addition of 10 vol.% of ABR and 1, 3 vol.% of nano-silica particle gives improved properties. The improvement of 5% and 8% in flexural strength similarly, 12% and 30% in Izod impact toughness were observed for composite designation $FAF_1$ and $FAF_2$ respectively. This improvement is because of high toughness ABR and high load sharing nano-silica presence in the epoxy matrix. The presence of ABR neatly stretches during the bending load. Similarly, in sudden impact load, the energy absorption rate of ABR is higher than that of glass fibre-epoxy matrix. Thus lot of energy is observed by the composite and gives improved impact toughness [23]. Moreover, the presence of nano-silica gives higher bonding strength (adhesion) between reinforcements and matrix. The presence of nano-silica also ensures the suppression of micro cracks, which are likely to pop-up during loading condition.

**Table 2** Composites fabricated and tested

| Composite Designation | Epoxy (vol.%) | ABR (vol.%) | E-glass fibre (vol.%) | Nano-silica (vol.%) |
|---|---|---|---|---|
| E | 100 | – | – | – |
| FFF | 70 | – | 30 | – |
| $FAF_1$ | 69 | 10 | 20 | 1 |
| $FAF_2$ | 67 | 10 | 20 | 3 |
| $AFA_1$ | 69 | 20 | 10 | 1 |
| $AFA_2$ | 67 | 20 | 10 | 3 |

F-Fibre; A-ABR.

**Table 3** Strength factor of composites

| Composite Designation | T. Strength (MPa) | Nor. T. Strength % | F. Strength (MPa) | Nor. F. Strength % | Izod impact (J) | N. Izod impact % | ILSS (MPa) | N. ILSS % | Strength factor |
|---|---|---|---|---|---|---|---|---|---|
| E | 63 | **45** | **90** | **53** | 0.4 | 7 | – | – | 35 |
| FFF | 141 | 100 | 157 | 92 | 3.8 | 70 | 24 | 100 | 91 |
| $FAF_1$ | 124 | 88 | 165 | 96 | 4.3 | 79 | 21 | 88 | 88 |
| $FAF_2$ | 130 | 92 | 171 | 100 | 5.4 | 100 | 23 | 96 | **97** |
| $AFA_1$ | 118 | 84 | 162 | 95 | 4.8 | 89 | 19 | 79 | 87 |
| $AFA_2$ | 126 | 89 | 167 | 96 | 5.2 | 96 | 22 | 92 | 93 |

T-tensile; F-flexural; N-Normalized.

The presence of micro cracks never grow-up, thus improved flexural and izod impact toughness is observed [24]. It is noted that further addition of large volume (20vol. %) of ABR into epoxy resin along with E-glass fibre gives significant results in mechanical properties. It is observed that the composite contains 20 vol.% of ABR and nano-silica of 1, 3 vol.% into epoxy resin gives significantly improved results in mechanical properties. It is observed that the tensile strength of 118 and 126Mpa were observed for composite designation $AFA_1$ and $AFA_2$ respectively. On comparing with pure epoxy resin this is much improved value but when comparing with FFF composite designation this bit lesser. This is because of low tensile modulus ABR addition into epoxy resin. Even then the addition of 3 vol.% of nano-silica improves the tensile strength. This phenomenon is the reason for improved adhesion of silica particles with fibre and matrix, which inturn improves the smooth load transfer [25].

It is noted that the addition of 20 vol.% of ABR and up to 3 vol.% of nano silica particle improves the flexural strength and impact toughness. The high energy absorbing ABR and micro load sharing nano-silica is the cause of these improvements. On comparing with $FAF_1$ and $FAF_2$ the flexural and izod properties were lower. This phenomenon is the reason of poly phase load sharing phenomenon in epoxy matrix [26]. It is further noted that the ILSS of $FAF_1$, $FAF_2$, $AFA_1$ and $AFA_2$ composite designation gives marginally lower ILSS value. The FFF composite designation gives higher ILSS of 24 MPa. This reduction in the hybrid composites is the reason of adding multi phase reinforcements in the epoxy resin. These multi phase reinforcements (FAF and AFA) reduce the interfacial strength and led the composite to deform plastically by triggering interfacial cracking. It is noted that the 3 vol.% of nano-silica particle in both FAF and AFA gives considerably better result than 1 vol.% of nano-silica in FAF and AFA designations. This is because of high adhesion behaviour of nano-silica, which is greatly supports in interfacial bonding and transfer the load smoothly [27]. Figure 1 shows the SEM fractography images. The pure epoxy resin shows almost flat fractured portion after the tensile test, which indicates high brittleness of casted epoxy resin. Figure 1(b) shows moderate fibre pullout in epoxy matrix of FFF composite designation. Figure 1(c) shows highly reacted phase of ABR and E-glass fibre. There are more shear cubs in matrix, which indicates improvement in toughness of composite by the addition of ABR and nano-silica [28]. Figure 1(d) shows the fractograph of interfacial de-bonded ABR and matrix surface, which indicates poor adhesion of ABR with matrix. The presence of nano-silica improved the adhesion but still it is not adequate to maintain higher interfacial strength.

## 4.2 Drop Load Impact Analysis

Figure 2 shows the time vs. Energy absorption graph of various composites fabricated. It is observed that the pure epoxy resin gives very lower energy absorption, which is equal to 0.7 J. This lower energy absorption is the cause of high brittle nature of pure epoxy resin. It is observed that the addition of E-glass fibre of 30 vol.% into epoxy resin improved the energy absorption. The highest energy absorption of 13.5 J was observed for FFF composite designation. This improvement is because of effective load sharing and high load bearing capability of E-glass fibre in epoxy resin composite [29]. It is further observed that the addition of 10 vol.% of ABR and 20 vol.% of E-glass fibre along with 1 and 3 vol.% of nano-silica particle improved the energy absorption and penetration resistance. Highest energy absorption of 14.2 J and 16.8 J is observed for composite designations $AFA_1$ and $AFA_2$ respectively. This improvement is because of high toughness ABR and nano-silica presence along with E-glass fibre in epoxy resin composite. The presence of ABR improves the energy absorption rate by stretching out freely, while the nano-silica particle improves the adhesion behaviour. Being flexible nature of ABR the sudden load is uniformly shared in all portion of rubber thus the stress intensity factor is reduces. The uniformly distributed load further transferred to E-glass fibre and micro crack suppresser nano-silica. Since the nano-silica is good for adhesion improvement it used to improve the adhesion between reinforcements and matrix chemically. The adhesion improved reinforcements with-stand for large applied

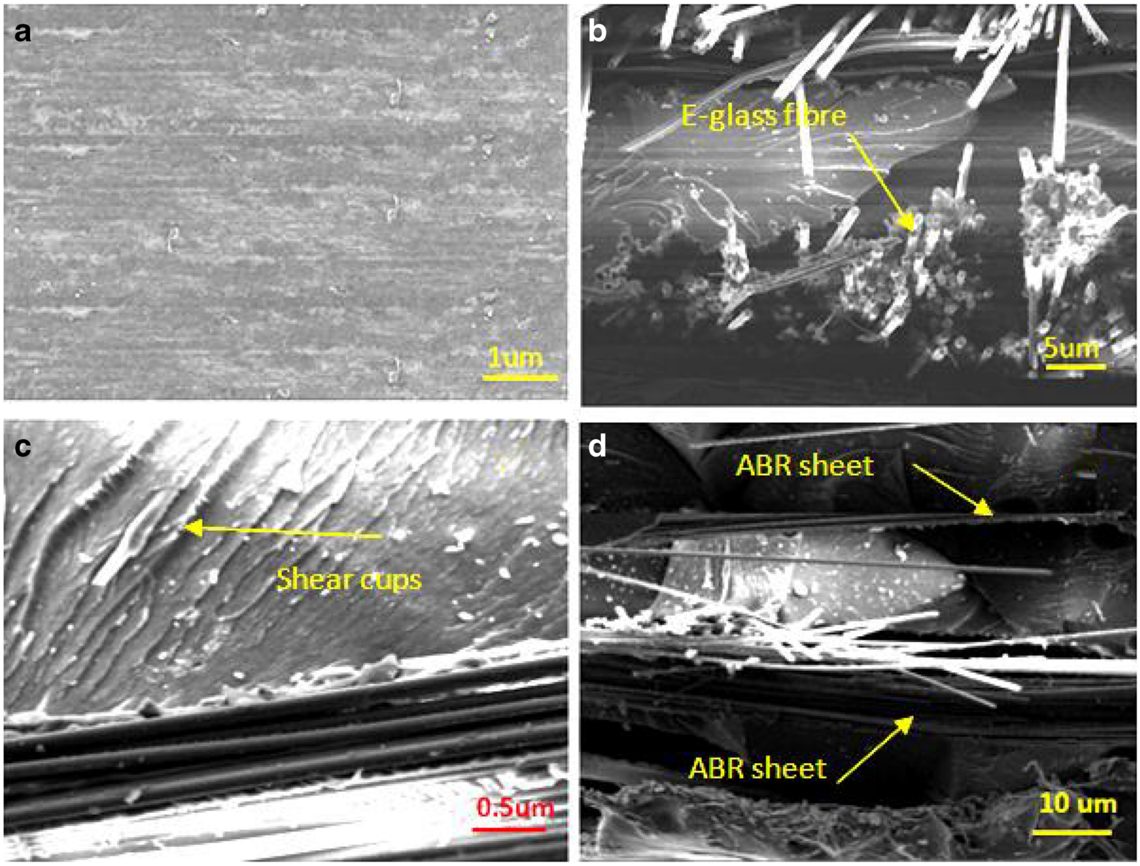


**Fig. 1** SEM fractograph of (a) pure epoxy resin, (b) FFF, (c) $FAF_2$ and (d) $AFA_2$ composite designation

load and prevent interfacial de-bonding. Thus improved impact energy absorption is observed [30].

It is observed that further increasing ABR volume up to 20% and nano-silica of 1 and 3 vol.% along with glass fibre in epoxy resin composites gives improved energy absorption. The Improvement of 18 J and 19.8 J were observed for composite designations $AFA_1$ and $AFA_2$ respectively. This high energy absorption is the cause of the presence of large volume

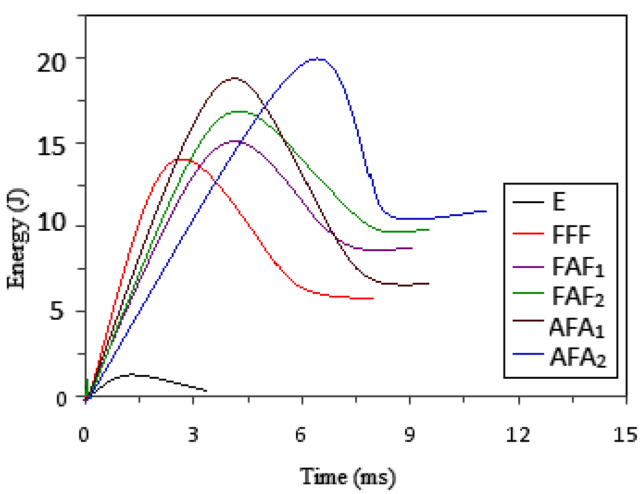


**Fig. 2** Energy absorption behaviour of composites

of ABR sheet along with glass fibre. The high stretch-ability of ABR in epoxy matrix receives large load and retains the composite tougher. Moreover, the addition of nano-silica acts as an adhesion agent between ABR and fibre with epoxy resin. The adhesion improved composites are less prone for interfacial de-bonding phenomenon, which inturn improves the energy absorption [31]. Thus addition of ABR and nano-silica particle in glass-epoxy composites greatly supports in energy absorption behaviour of composite. Figure 3 shows the impacting and rear side damage of composites exposed to sudden impact load. It is observed that in FFF composite designation the penetration is observed in rear end, which indicates poor load absorbing behaviour and penetration resistance of composite. But the composite designation $FAF_1$ and $FAF_2$ shows high penetration resistance against sudden imapctor. There is a bit impression of impactor at the rear end of this composite, which indicates improved energy absorption. This improvement is the reason of presence of high toughness ABR presence between two layers of E-glass fibres and receives large energy. Similarly, the composites $AFA_1$ and $AFA_2$ shows highly improved energy absorption than $FAF_1$ and $FAF_2$. The presence of large volume (20 vol.%) of ABR receives most of the kinetic energy of the impactor and arrest the penetration to the rear end. There is no impression at the rear end of these composite designations, which

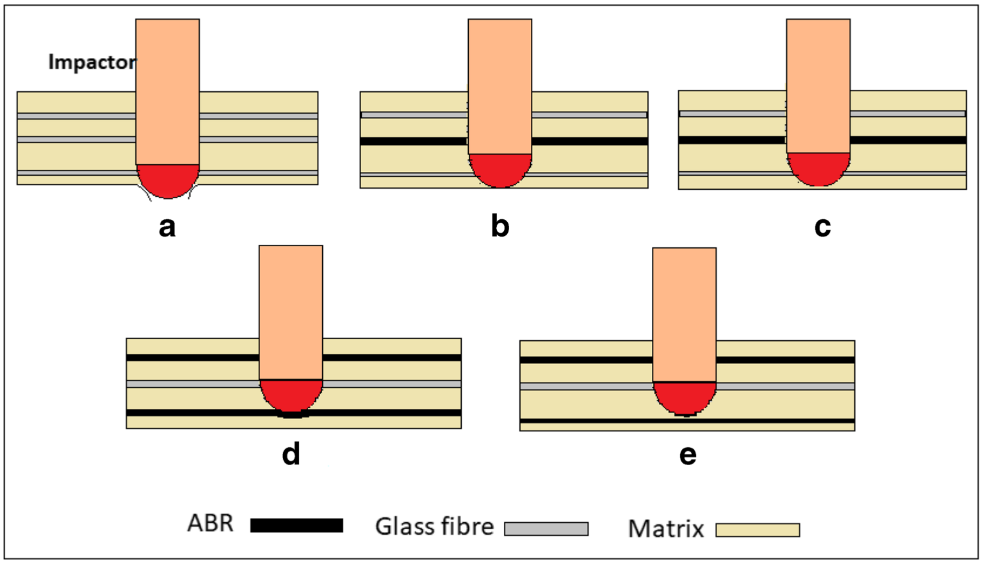


**Fig. 3** Drop load impact image of (a) FFF, (b) $FAF_1$, (c) $FAF_2$, (d) $AFA_1$ and (e) $AFA_2$ composite designation

confirms the improved toughness of the composites. The impactor is stuck with bottom layered ABR and lost full energy. Thus there is no indentation is received in the penetrating side of composite [32].

### 4.3 Fracture Toughness Behaviour

Table 4 gives the fracture toughness and energy release rate of epoxy and its composites. It is noted that the pure epoxy gives fracture toughness of 19.4 MPa.$\sqrt{\mathbf{m}}$ with energy release rate of 0.568 MJ/m$^2$. This value is significantly less since epoxy contains no micro crack suppressing mechanism within it and having higher stress intensity factor at the micro cracks' tip. The high brittle epoxy resin has more micro flaws they used to grow up when the epoxy sheet is subjected for the tear load [33]. It is further noted that the addition of 30 vol.% of E-glass fibre in epoxy resin composite improves the fracture toughness and energy release rate. The improvement of 26 and 30% was observed on comparing with pure epoxy resin. This improvement is because of presence of glass fibre in matrix, which absorbs the applied load and reduces the propagation of internal micro cracks. Moreover it absorbs large energy and reduced the stress intensity factor on matrix. Thus improvement in fracture toughness and energy release rate is observed. It is further observed that the presence of 10 vol.% of ABR along with glass fibre marginally reduces the fracture toughness. This reduction is attributes to the flexible rubber molecules, which could not bear the crack propagation within rubber molecules and finally deform completely. Once the rubber layer fractured the shear load on remaining composite dominates more and makes the composite to deform plastically. The presence of nano-silica further improved the fracture toughness and energy release rate. The composite designations $FAF_1$ and $FAF_2$ give very marginal decrement on comparing with FFF composite designation. The addition of ABR slightly reduces the fracture toughness and energy release rate but the addition of nano-silica particles compensate the lost strength and improved the fracture toughness and energy release rate [34]. The composite designation $FAF_2$ gives fracture toughness of 25.6 MPa.$\sqrt{\mathbf{m}}$ whereas the FFF designation gives 26.1 MPa.$\sqrt{\mathbf{m}}$. This result is almost closer to the FFF composite designation.

Similar results were observed in $AFA_1$ and $AFA_2$ composite designations too. The addition of 20 vol.% of ABR reduced the fracture toughness but the inclusion of nano-silica improved the strength. The 3 vol.% of nano-silica along with 20 vol.% of ABR gives the fracture toughness of 24.2 MPa.$\sqrt{\mathbf{m}}$ and energy release

**Table 4** Fracture toughness results

| Composite designation | Maximum load N | Young's modulus (E) MPa | Fracture Toughness ($K_1c$) MPa.$\sqrt{\mathbf{m}}$ | Energy Release Rate ($G_1c$) MJ/m$^2$ |
|---|---|---|---|---|
| E | 450.0 ± 0.16 | 614.0 ± 0.18 | 19.4 ± 0.12 | 0.568 ± 0.071 |
| FFF | 477.0 ± 0.12 | 697.0 ± 0.15 | 26.1 ± 0.12 | 0.808 ± 0.075 |
| $FAF_1$ | 463.9 ± 0.47 | 659.2 ± 0.68 | 23.6 ± 0.09 | 0.743 ± 0.022 |
| $FAF_2$ | 470.2 ± 0.34 | 675.6 ± 0.71 | 25.6 ± 0.17 | 0.761 ± 0.007 |
| $AFA_1$ | 449.0 ± 0.17 | 630.0 ± 0.34 | 22.1 ± 0.19 | 0.757 ± 0.071 |
| $AFA_2$ | 468.4 ± 0.12 | 662.8 ± 0.18 | 24.2 ± 0.81 | 0.734 ± 0.095 |

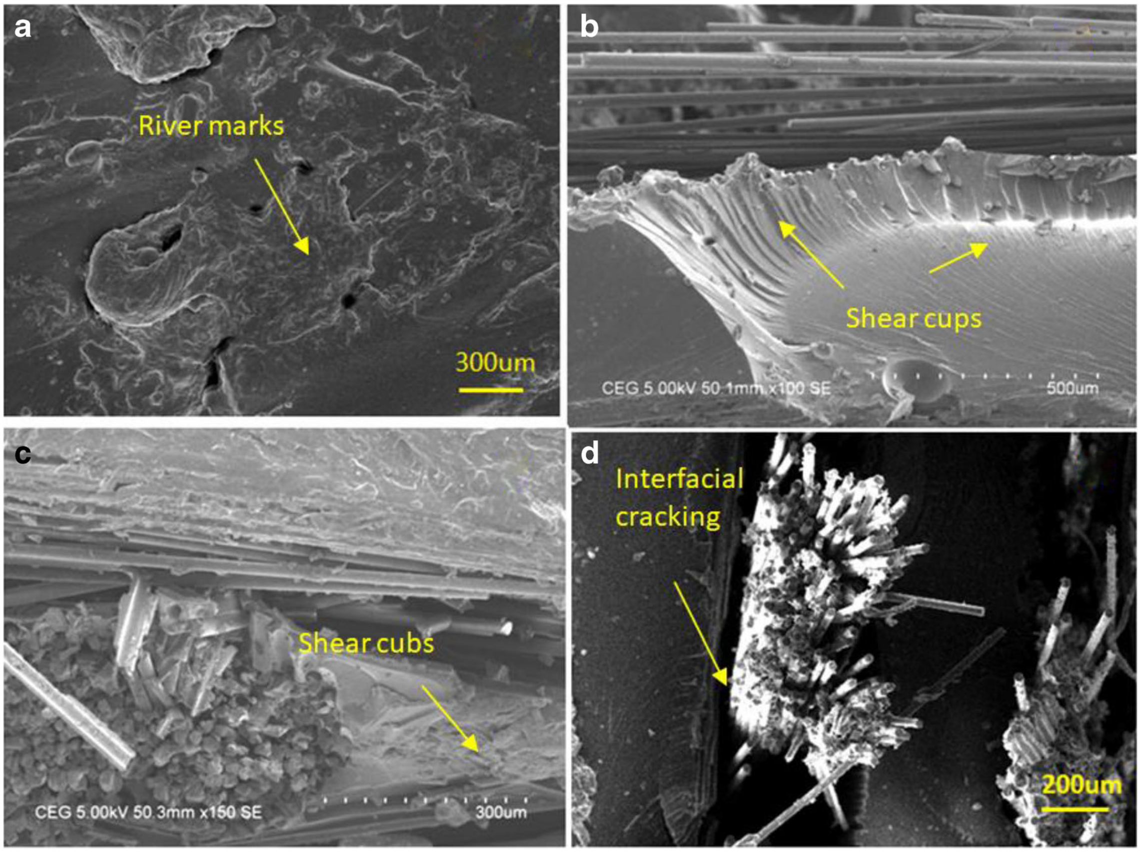


**Fig. 4** SEM images of fracture toughness tested (a) epoxy resin; (b and c) FFF composite and (d) $FAF_2$ composite designation

rate of 0.734 MJ/m$^2$. The presence of nano-silica improves the adhesion of reinforcements with resin and reduces the stress intensity factor, which inturn suppresses the micro flaws does not propagate. Thus improvement in fracture toughness and energy release rate is observed. Hence the addition of ABR marginally reduces the fracture toughness and energy release rate but addition of nano-silica particle further improves the lost strength. Figure 4 shows the SEM fractograph of fracture toughness composite specimens. The pure epoxy resin shows (fig. 4a) brittle flat fracture and more river marks, which confirms the high brittle nature of the matrix. Figure 4(b and c) shows the fractograph of FFF composite specimen after the fracture toughness test. The fractured portion illustrates more shear cubs and striations, which indicates the suppression of micro-cracks and fracture [35]. Figure 4(d) shows the fractograph of $FAF_2$ composite designation. The fractured portion shows interfacial cracking and debonding, which confirms that the fracture happened due to the presence of lower tear strength of ABR [36].

## 5 Conclusion

This present study dealt the effect of adding acrylonitrile butadiene rubber and nano-silica particle into glass-epoxy composite in mechanical, drop load impact energy absorption and fracture toughness behaviour. The composites were prepared using reinforcements ABR, E-glass fibre and nano-silica with appropriate quantities. The composites were prepared using hand layup method and no significant defects were observed due to ABR addition. Following are the summary of this present study.

- The strength factor results show highest rank of 97/100 for $FAF_2$ composite designation. The presence of 10 vol.% of rubber and 3 vol.% of nano-silica particle gives overall improved effect in tensile, flexural, impact and ILSS. A highest tensile strength of 141 MPa was observed for composite designation FFF.
- The drop load impact results show that the composite designation $AFA_2$ gives highest penetration resistant against the sudden load. Maximum energy absorption of 19.8 J was observed for $AFA_2$ composite designation.
- The fracture toughness results show that the addition of ABR marginally reduced the fracture toughness and energy release rate. Lowest fracture toughness of 22.1 MPa.$\sqrt{\mathbf{m}}$ was observed for composite designation $AFA_1$ whereas the addition of nano-silica particle improves the fracture toughness and energy release rate further, which is close to the fibre-reinforced epoxy composite.

- Thus the addition of acrylonitrile butadiene rubber and nano-silica particle greatly supports to improve the mechanical and drop load impact behaviour of epoxy composite with considerably improved fracture toughness and energy release rate.
- Being high toughness and impact resistance, these composites could be used in defence product manufacturing, automobile door panels, aircrafts body, sports equipments, structural members and domestic product manufacturing.

## Compliance with Ethical Standards

**Conflict of Interest** All authors hereby confirmed that this article does not have any conflict of interest.

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