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ORIGINAL ARTICLE

Studies on mechanical and water absorption behavior of biofiber-reinforced epoxy biocomposites added with seashell, eggshell, and coconut biofillers

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Abstract

The present study emphasizes the mechanical characteristics and water uptake behavior of seashell, eggshell, and coconut fillers added with sisal, kenaf, and pineapple leaf fiber-reinforced epoxy composites. The present study compares the difference in mechanical performance between filler-based composites with only fiber-based composites. The weight proportion of fillers and fiber reinforcement collectively were 30% by weight, and epoxy was 70% by weight in all prepared specimens. According to the results of the experimental findings, the inclusion of biofillers with fiber and hybridization of fibers gives a reduction in void content as sisal/epoxy/seashell composite shows a minimum 2.09% void content than other specimens. Hybrid pineapple/sisal/kenaf/epoxy composite absorbs minimum water content during the water immersion test. Kenaf/epoxy/seashell composite exhibits a maximum tensile strength of 72.25 MPa, and kenaf/epoxy/eggshell composite achieved a maximum value of tensile modulus at 30.49 GPa as compared to other developed composite specimens. While flexural strength was maximum for sisal/epoxy/eggshell composite at 257.25 MPa, flexural modulus was maximum for kenaf/epoxy/eggshell composite at 68.4 MPa. Sisal/epoxy/coconut composite achieved a maximum impact strength of 0.9 J as compared to all developed composite specimens. Scan electron microscopy (SEM) reveals the mechanism of fiber/matrix debonding, fiber fracture, and fracture of matrix after mechanical testing.

Keywords Biocomposites · Epoxy · Mechanical properties · Water absorption · Scan electron microscopy

1 Introduction

Composite materials were originally developed in the early 1900s, with resins acting as a matrix and carbon fiber acting as reinforcement. Fiber-reinforced composites were prominent in military applications during WWII due to their lightweight. However, due to their poor strength, reinforcing was applied to improve their mechanical properties. Many types of synthetic fibers, such as carbon, aramid, and basalt, were manufactured for a longer period after WWII. Carbon fiber-reinforced composites, on the other hand, are widely employed in a wide range of applications due to their lightweight. Some of the applications of composite materials include interior and exterior design in architecture, automotive and transportation, aerospace industries, electrical industries, home appliances, sports and leisure, marine, and corrosive environments [1].

Various types of reinforcement and polymeric matrix are mixed to form a newer material known as polymer composite material. Most of the time, the reinforcement serves as a carrier element, delivering strength in a certain direction while guarding against harmful environmental conditions. The strengthening phase can be used by researchers to improve the resilience of their composites. Fibers and different filler particles are commonly utilized to strengthen composites. The two types of fibers that can be discovered are artificial and synthetic fibers. A technique generated synthetic fiber, which has superior mechanical qualities. The biggest downside of synthetic fiber is that it is not biodegradable, which has environmental consequences. Glass, Kevlar, aramid, and carbon are types of synthetic fibers [2–4].

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To address environmental concerns, natural fiber was created as an alternative to synthetic fibers. Plants, animals, and minerals are used to extract the natural raw materials which serve as biofibers and biofillers in composite materials. Biofibers and biofillers have several advantages, including their availability, degradability, non-toxicity, low cost, and simplicity of handling. Sisal, ramie, pineapple, kenaf, flax, hemp, jute, and sisal are examples of natural fibers [5, 6]. Composites made of natural fibers are utilized in every field of engineering from structural to non-structural components. Various automobile components such as dashboards, seat-liners, and interior parts have the potential application of biocomposite materials. These biocomposites have a vital role in household products such as trays, utensils, and luggage containers. Defense applications such as bulletproof jackets, lightweight stretcher, and armor handles have important applications of biocomposite materials [7, 8].

The filler comes in the form of flakes or powder to strengthen the composite materials. The three most frequent forms of filling are organic filling, inorganic filling, and metal filling. Particle size has the greatest influence on the mechanical characteristics of composites [9]. Micro and biofillers are used in the polymer matrix. At this step, both fiber and filler are employed in polymer composites, with fiber enhancing strength and filler improving module. To deliver superior mechanical qualities than traditional materials, fiber and filler composite polymers should be properly chosen [10].

Research conducted for the inclusion of eggshell nanoparticles with fiber-based epoxy composites considerably changes the mechanical, thermal, and water uptake properties of developed composites [5]. The authors concluded that the incorporation of eggshell with hemp fiber composite achieved enhanced tensile, flexural, and impact strength as compared to hemp fiber-based composite. It was feasible to assess how *Tamarindus indica* L. seed powder and the hybridization of hemp and natural fibers influenced the characteristics of polymer nanocomposites by varying the concentration (in weight %) of both natural fibers. Because of the apparent compatibility between the matrix and the fibers, composites containing 40% JF and 10% HF had lower void content [6]. The findings indicate that the inclusion of HP has a substantial effect on the properties of *Coccinia grandis* fiber-reinforced composites (CGFRCs) and that HP may be a feasible nanofiller for most polymer matrix composites [7, 8].

In this research work, plant-based fibers like pineapple, kenaf, sisal, and fibers are utilized as a reinforcement for polymeric materials. Sisal fiber is a natural fiber with high strength and modulus, simple accessibility, cheap cost, great durability, and recyclability, as well as low maintenance, wear, and tear. It has a high-water absorption capacity [9–11]. Sisal fiber is made from plant leaves. It is often obtained by machine decortications. The strands are typically creamy white, 80–120 cm long, and 0.2–0.4 mm in diameter, with a shiny aspect. Brazil produces around 3,000,000 tons per year, followed by China, Mexico, Tanzania, Kenya, and Madagascar. Sisal is an agave family member; the commercially produced species is *Agave sisalana* [12–15]. The plants live for 7 to 12 years before producing a flower stalk 4 to 6 m tall before dying. Sisal is a xerophytic plant, which means it can thrive in arid areas but not well in poorly drained soil. The average lifespan of a sisal plant is 15–18 years. Sisal is typically harvested once a year, but if the soil and climate conditions allow, it can be harvested three times in 2 years [15–18]. Kenaf fiber is both robust and delicate to the touch. Kenaf has a breaking strength comparable to low-grade jute, and it is only marginally degraded when wet. Kenaf fibers feature a long staple, allowing to produce highly fine and robust yarn. It may be lightweight or heavyweight. Kenaf fiber has low flexibility and is delicate in nature. Pineapple leaf fibers function as typical cellulosic fibers derived from plants. It aims to boost the tensile strength volume in polymer matrix composites by up to 30%. Total deformation was also demonstrated to increase in volume by 10%. This result demonstrates a dramatic improvement in tensile strength. As the volumetric proportion of pineapple leaf fibers increases, so does its elastic modulus. It has been demonstrated that the epoxy matrix is the best for forming a composite synthetic fiber matrix [18–25]. Seashell, eggshell, and coconut fillers were used as a filler in the form of powder in this study. And all selected fillers have good mixing with epoxy polymer matrix and enhanced the interfacial adhesion between all selected fibers and epoxy polymer matrix which enhanced the overall structural stability and strength of the fabricated composite specimen [25–28].

In the present study, attempts have been made to evaluate the performance of polymeric materials reinforced with pineapple-, kenaf-, and sisal fiber-reinforced epoxy composites. An approach of the addition of biofillers (eggshell, seashell, and coconut shell) was added with all selected biofibers to fabricate the filler-based polymer composites. According to previous literature, no study was available which focused on the potential incorporation of these biofillers on the performance of natural fiber-based polymer composites. Mechanical performance, water uptake, and density calculation of all these developed composites were examined in the present study. Table 1 illustrates the abbreviation of developed composite specimens.

2 Materials and methods

All the selected filler materials (coconut, eggshell, and seashell) were used in sizes from 10 to 2400 nm. The chemical composition of eggshells was 1% magnesium carbonate, 1% calcium phosphate, 4% organic matter, and 94% calcium

Table 1 Abbreviations for developed composite specimens

Abbreviations	Full form
PE	Pineapple/epoxy
SE	Sisal/epoxy
KE	Kenaf/epoxy
PES	Pineapple/epoxy/seashell
PEE	Pineapple/epoxy/eggshell
PEC	Pineapple/epoxy/coconut
SES	Sisal/epoxy/seashell
SEE	Sisal/epoxy/eggshell
SEC	Sisal/epoxy/coconut
KES	Kenaf/epoxy/seashell
KEE	Kenaf/epoxy/eggshell
KEC	Kenaf/epoxy/coconut
PSKE	Pineapple/sisal/kenaf/epoxy

Table 2 Properties of epoxy polymer used for polymer composites [4]

Types of properties	Values
Viscosity	10,000–12,000 mPa*s
Density	1.15–1.20 g/cm^3 at 25 °C
Specific gravity	1.12 g/cc at 25 °C
Appearance	A milky white liquid
Solid content percent	84%

carbonate [11]. The chemical composition of seashell powder contains calcium carbonate ($CaCO_3$) in two forms (aragonite and calcite). The most leading parts in seashell powder are carbon (C), oxygen (O), and calcium (Ca). Coconut powder contains hemicelluloses, cellulose, and lignin in their chemical composition [11]. Selected raw fillers were extracted from the wastages and then crushed using a mixer and grinder and converted into nano-range using a ball milling machine. Plant fibers (pineapple, kenaf, and sisal) were used as a reinforcement. All the fiber were supplied by Go Green Products, Alwarthirunagar, Chennai, India, 600087.

Epoxy (Araldite LY 556) is used as a polymer matrix which is based on bisphenol-A. All the properties of the epoxy polymer matrix are represented in Table 2. Table 3 shows concentrations of the fibers, fillers, and epoxy resin to prepare the specimen for testing. Figures 1, 2, 3, and 4 represent the percentage of different types of fillers, fibers, epoxy, and developed composite specimens.

2.1 Processing

A wooden closed mold was prepared to perform the closed mold hand lay-up technique for polymeric sample preparation. The inner surfaces of the mold were covered by plastic films, and silica gel was sprayed over the plastic films which eliminated the sticking behavior of the epoxy resin with the plastic film covered inner wall of the mold. After successful preparation of the mold, filler particles were mixed with epoxy resin. The filler mixed epoxy and neat epoxy were spread over the surface of the mold and then the fiber mat was covered with the layer of epoxy. The layers of epoxy resin mixed with filler and fiber mats were repeated up to the desired thickness or percentages of resin, filler, and mat of prepared composite specimen [4]. After that, the prepared specimen was pressed by the weight and kept inside the mold for curing at least for 36 h. After curing, the mold was opened and a prepared composite specimen was out from the mold.

Table 3 Materials and their concentrations used for the development of composite material

Composite	PALF	Ramie	Sisal	Kenaf	Eggshell	Seashell	Walnut shell	Coconut endocarp	Bio-epoxy
PRSE	10%	10%	10%	0%	0%	0%	0%	0%	70%
PES	15%	0%	0%	0%	0%	15%	0%	0%	70%
KES	0%	0%	0%	15%	0%	15%	0%	0%	70%
SEC	0%	0%	15%	0%	0%	0%	0%	15%	70%
SES	0%	0%	15%	0%	0%	15%	0%	0%	70%
KEC	0%	0%	0%	15%	0%	0%	0%	15%	70%
SE	0%	0%	30%	0%	0%	0%	0%	0%	70%
PEE	15%	0%	0%	0%	15%	0%	0%	0%	70%
KEE	0%	0%	0%	15%	15%	0%	0%	0%	70%
SWE	0%	0%	15%	0%	0%	0%	15%	0%	70%
PALF	15%	0%	0%	0%	0%	0%	0%	15%	70%
SEN	0%	0%	30%	0%	0%	0%	0%	0%	70%
SEE	0%	0%	15%	0%	15%	0%	0%	0%	70%
PALFE	30%	0%	0%	0%	0%	0%	0%	0%	70%

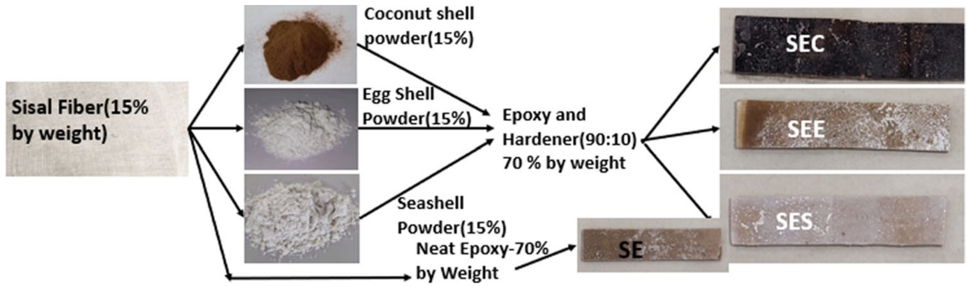


Fig. 1 Sisal fiber-based composite with different proportions of filler materials

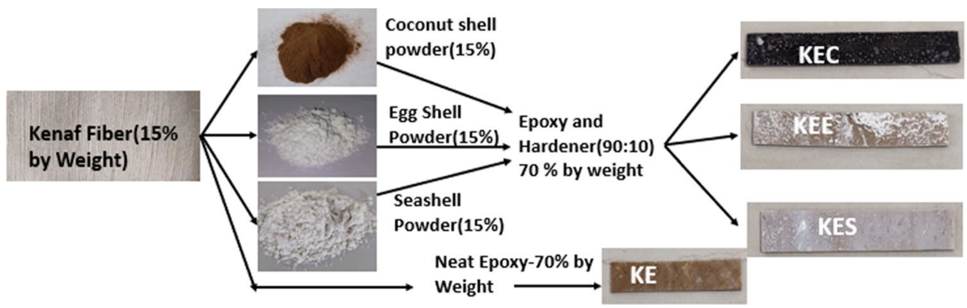


Fig. 2 Kenaf fiber-based composite with different proportions of filler materials

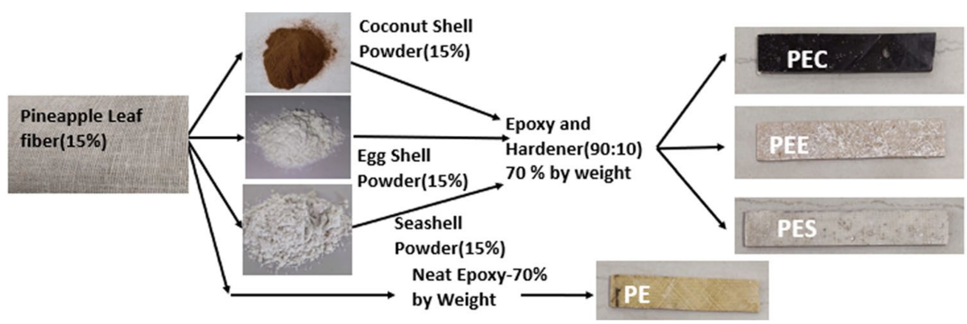


Fig. 3 Pineapple fiber-based composite with different proportions of filler materials

Fig. 4 Pineapple, sisal, kenaf fiber-based hybrid composite

2.2 Density calculation

Experimental density calculation was conducted by Archimedes' principle, and theoretical density was calculated by the standard formula as shown in Eq. 1. Differences between experimental and theoretical densities were calculated to find out the voids present in the prepared composite specimen using Eq. 2 [29].

The equation used for the calculation of theoretical density is shown in Eq. 1.

$$\frac{1}{\rho_{\text{sample}}} = \frac{W_{\text{fiber}}}{\rho_{\text{fiber}}} + \frac{W_{\text{matrix}}}{\rho_{\text{matrix}}} + \frac{W_{\text{filler particles}}}{\rho_{\text{filler particles}}} \tag{1}$$

In Eq. 1, ρ denotes the density and W denotes the weight fraction.

$$\text{Void content} = \frac{\text{theoretical density} - \text{experimental density}}{\text{theoretical density}} \times 100\% \tag{2}$$

2.3 Water absorption behavior

A water immersion test was carried out to calculate the moisture content absorbed by the prepared composite specimen during their real applications. All the prepared composite specimens go for weight measurement prior to water immersion, and again, weight measurement is carried out after water immersion of specimens for a specific interval of time. The water uptake measurements of the specimens were assessed by using rectangular samples of similar sizes. The water immersion test was carried out according to ASTM D5229 standards for all the prepared specimens.

2.4 Hardness

The resistance to permanent indentation was estimated by hardness testing of all fabricated specimens. Material hardness determines the suitability of prepared specimens for various loading applications. It serves as a key indicator of resistance to factors like friction wear and erosion caused by steam. Shore-D hardness testing was conducted to determine the hardness values.

2.5 Surface roughness

Analyzing the surface roughness of the specimen aids in identifying the uneven surface of the prepared specimen. This examination is valuable for assessing the smoothness of the specimen's surface, which in turn helps determine its wear rate. A rough surface typically results in a higher wear rate compared to a smoother one. The surface roughness measurement was conducted using a TJD520 digital surface roughness tester.

2.6 Tensile strength

Tensile testing on the composite is needed to find out its capacity to withstand tensile stress or force prior to failure. This testing grants a valuable understanding into the composite's ultimate tensile strength, tensile modulus (Young's modulus), and elongation at break. All these are the important constraints to be measured while using a material in most applications. The tensile test was performed using an Instron 5952 machine at a crosshead speed of 2 mm/min, with samples prepared in accordance with ASTM 3039 standards. All the experiments of tensile test were performed at Bhaskracharya College of Polymer Science, University of Delhi.

2.7 Flexural strength

Flexural testing on the material accedes data on its flexural strength and flexural modulus. This testing method establishes the maximum bending stress the material can endure before failure, a critical factor in assessing its suitability for applications involving bending forces. Flexural testing of the composites was carried out using an Instron 5952 machine at a crosshead speed of 2 mm/min, with samples prepared following ASTM D790 standards.

2.8 Impact strength

The impact test assesses the composite's impact strength, indicating its capability to withstand sudden loads or impacts, crucial for applications requiring shock absorption. This parameter is particularly significant for materials utilized in environments with consistent shock requirements. Impact tests were performed on unnotched specimens using the Charpy impact test setup employed at Shree Mata Vaishno Devi University Katra. The impact test machine conducted the experiments of the impact test with a maximum impact energy of 50 J and a striking velocity of 3 m/s.

2.9 Morphological analysis

Fracture surfaces of specimens after mechanical tests were examined using scan electron microscopy (S-3700 NUltra Large VP-SEM). All prepared polymeric samples passed through the gold coating to improve the conductivity before capturing the micrographs of fractured surfaces.

3 Results and discussion

3.1 Experimentation of density

The experimental density of all the prepared composite specimens is calculated using small-sized samples [25 mm × 25 mm (length × breadth)]. These samples were first weighed using a simple weighing machine. The volume of each sample was then measured using Archimedes' principle, by immersing the sample in a beaker filled with water and noting the rise in water volume. This rise in volume was used to determine the sample's volume. The measured weight and calculated volume were then used to find the experimental density of the biocomposite samples [29]. Based on actual density, the difference between actual density and theoretical density using the formula in Equation 2 calculates the percentage void content. The findings of density calculations and void percentage show that the incorporation of biofillers (coconut, eggshell, and seashell) reduced the void percentage in prepared specimens, and a hybrid composite of pineapple, sisal, and kenaf fiber also shows a lower void percentage as compared to single fiber-based composites. All prepared composite specimens achieved less than 10% of void content which imparts the suitability of the selected hand lay-up technique of prepared fillers and fibers-based polymer composites as shown in Table 4.

3.2 Water absorption capacity

The rate at which the weight of the samples increased due to the absorption of water was measured at regular intervals and noted. The weight variation was calculated using Eq. 3.

$$\text{Percentage change in weight}(\%) = \frac{m_2 - m_1}{m_1} \times 100\% \quad (3)$$

Here,

m1—initial mass of the sample in grams, and.

m2—mass of the sample after certain intervals in grams.

The data collected from the water immersion test is shown in Fig. 5. From the figure, it is clearly visible that SEE shows maximum water uptake followed by PEE, SE, SEC, and KE respectively. The least amount of water is absorbed by PSKE followed by KES and SES respectively. It is also clear that the ability to absorb water was saturated at around 120 square roots of minutes for all developed specimens. The hydrophilic nature of all used fillers and fibers imparts the water absorption behavior to all developed specimens, and the hydrophobic nature of epoxy polymer resists the water absorption [30]. In the case of filler-based composite specimens, especially, coconut filler-based composite specimens absorbed a higher amount of water during the initial stage of the water immersion test from 0 to 40 h. This high amount of absorption of coconut filler is due to the high hydrophilic nature of coconut filler. The lowest water uptake of a hybrid composite of pineapple-, sisal-, and kenaf fiber-reinforced epoxy composite was due to the different surface properties of the three fibers [30, 31]. Similar studies were already performed by previous researchers on different types of biofiller- and biofiber-reinforced polymeric composites. Radhakrishnan et al. [1] performed the water immersion behavior of jute and flax reinforced with epoxy composites filled with eggshell and coconut shell as a nano-powder. The authors investigated that coconut filler-based composite absorbed a higher amount of water as compared to other specimens due to the more hydrophilic and freer hydroxy group present in the cellulose content of coconut filler. Abdel-Rahim and Mohammed [32] investigated the water uptake study of eggshell filler epoxy polymer composites. The authors concluded the filling of epoxy achieved better tensile strength but imparted a lower water absorption capacity to developed specimens among other prepared specimens. A higher amount of water uptake during application affects the loading behavior of the developed composite which is a very crucial factor that will be considered in the loading application of fiber- and filler-based composites.

Table 4 Percentage of void content based on theoretical and actual densities of all prepared specimens

Types of composites	Theoretical density (Kg/Cu Mt)	Actual density (Kg/Cu Mt)	Void content (%)
SE	1.28	1.2221	5.7893
PSKE	1.8282	1.7759	5.2203
KES	1.3158	1.2818	3.3953
SEC	1.25	1.2195	3.0477
PES	1.858	1.8047	5.3227
KEE	1.232	1.2025	2.9461
PEE	1.43	1.3907	3.9228
PE	2.2	2.1607	6.0498
KE	1.9583	1.8978	5.5622
PEC	1.3571	1.3211	3.5963
KEC	1.4643	1.4236	4.0651
SEE	1.275	1.2431	3.184
SES	1.1	1.0790	2.0996

3.3 Hardness

The results of the hardness test conducted on the composite samples are shown in Fig. 6. From the figure, PSKE hybrid composite has the highest hardness value of 76.5 Shore-D. The lowest value of hardness was achieved by SE composites at 25 Shore-D. PE composite shows the second highest value of hardness at 73 Shore-D. Other prepared specimens such as PES, KES, SEC, SES, KEC, KE, PEE, KEE, PEC, and SEE achieved hardness values of 45.5, 51.25, 45.5, 51, 54, 57.25, 53.25, 42.5, 45, and 66.25 Shore-D. Results of hardness show that the incorporation of biofillers reduces

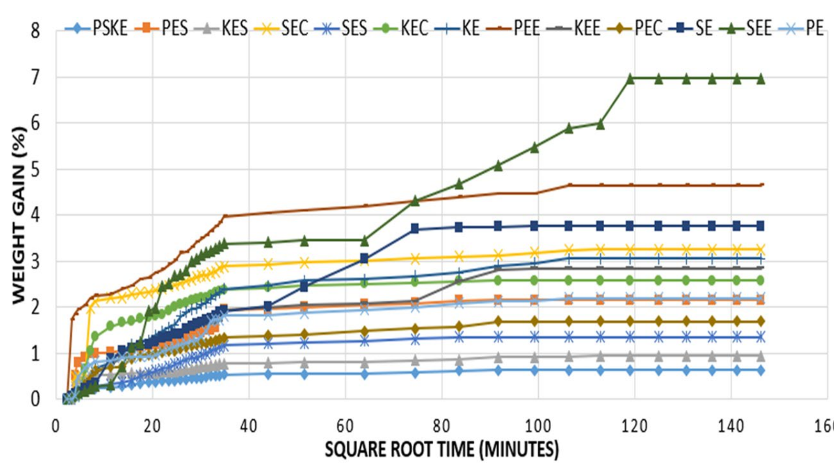


Fig. 5 Experimental investigation of water immersion test of all developed specimens

the hardness values as compared to only fiber-based composite specimens. And hybridization of three fibers (pineapple, sisal, and kenaf) reinforced with epoxy achieved the highest value of hardness due to the different properties of fibers and their debonding with epoxy polymer. It also shows that a higher amount of fiber percentage with epoxy polymer attained the highest value of hardness. Yontar et al. [15] studied the effect of green-produced silver nanoparticles on the properties of PVA composite. The author found that the addition of Ag nanoparticles into the composite led to the improvement in the hardness of the composite. The hardness value was found to be increased by 2.3 times. Chaudhary et al. [30] manufactured jute-, hemp-, and flax-reinforced epoxy composites and their hybrid composites. The authors concluded that hybridization of jute, hemp, and flax fibers with epoxy achieved the highest value of Shore-D hardness as compared to single fiber-based epoxy composites. Experimentation of hardness value stands developed specimens in various sliding and loading applications in various industries.

3.4 Surface roughness

All the prepared specimens were for surface roughness test and values are shown in Table 5. Surface roughness test helps to identify the pattern of deformation of prepared specimens during wear and tear application. Surface roughness also helps to detect the irregularities at joining two specimens together. The joining of different specimens plays a vital role in the assembly of the final products. Coconut filler-based epoxy composites attained high surface roughness as compared to other made composite specimens. KEC composite shows the highest surface roughness of 0.063 µm. Other coconut filler-based composites (SEC and PEC) show 0.057- and 0.054-µm surface roughness. The rough surface of coconut fillers imparts the rough surface to the prepared specimen, while the smooth surface of seashells imparts the lowest value of surface roughness in the developed specimen. PES composite shows the lowest value of surface roughness at 0.021 µm as compared to all other prepared composite specimens. Other seashell filler-based composites (SES and KES) show 0.022- and 0.024-µm surface roughness. The incorporation of fillers with fiber reinforcement reduces the surface roughness value as compared to a single fiber-based composite specimen. Hybridization of pineapple/sisal/kenaf/fiber-based epoxy composite shows 0.044 µm. Various authors found similar results in their research work. Jena and Kumar [33] studied the effect of clam shell filler on the surface properties of a prepared composite specimen. The authors concluded that the incorporation of clam shell with glass fiber/polymer lowers the surface roughness value. Similar findings were shown by Krishna et al. [34] for seashell and glass fiber-added epoxy-based polymer matrix

Table 5 Experimental value of surface roughness of all prepared specimens

Type of composite	Surface roughness (Ra)
Pineapple/epoxy	0.033 ± 0.011 µm
Sisal/epoxy	0.044 ± 0.012 µm
Kenaf/epoxy	0.022 ± 0.004 µm
Pineapple/epoxy/seashell	0.021 ± 0.002 µm
Pineapple/epoxy/eggshell	0.025 ± 0.001 µm
Pineapple/epoxy/coconut	0.054 ± 0.002 µm
Sisal/epoxy/seashell	0.022 ± 0.001 µm
Sisal/epoxy/eggshell	0.037 ± 0.012 µm
Sisal/epoxy/coconut	0.057 ± 0.012 µm
Kenaf/epoxy/seashell	0.024 ± 0.001 µm
Kenaf/epoxy/eggshell	0.026 ± 0.012 µm
Kenaf/epoxy/coconut	0.063 ± 0.001 µm
Pineapple/sisal/kenaf/epoxy	0.044 ± 0.012 µm

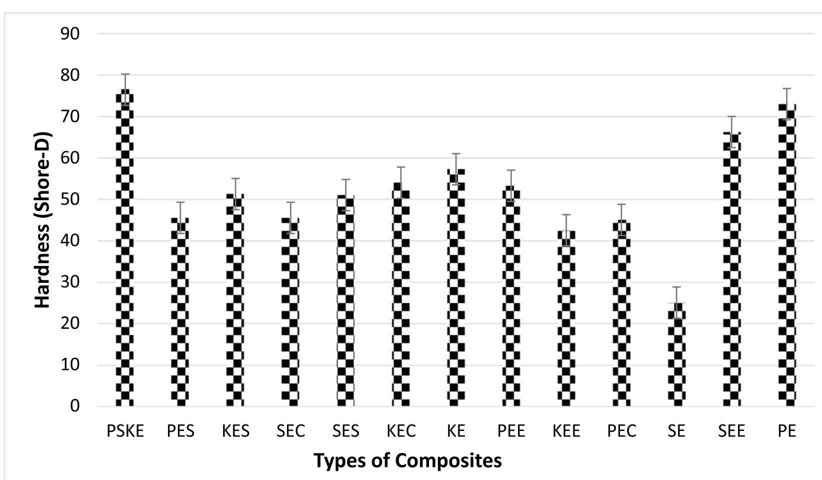


Fig. 6 Experimental findings of hardness (Shore-D) of all developed specimens

composite. The authors concluded that merging seashell fillers with fibers provides a better surface finish to prepared specimens. A better surface finish of prepared specimens helps the uniform setup of these specimens in different fixtures of mechanical testing performed in this study.

3.5 Tensile strength

The outcomes derived from the tensile testing (tensile strength, Young's modulus, and elongation at break) of all prepared composite specimens are shown in Figs. 7, 8, and 9. Results of the tensile test displayed that the incorporation of biofillers (coconut, eggshell, and seashell) increased the tensile strength of the prepared composite specimens as compared to only fiber-reinforced epoxy composite specimens. And hybridization of pineapple, kenaf, and sisal fibers with epoxy composite achieved a higher tensile strength value than single fiber-based epoxy composite specimens. In all used fillers, seashell filler with fiber-reinforced epoxy showed higher tensile strength than other filler- and fiber-reinforced epoxies. KES composite achieved the highest tensile strength of 72.25 MPa and lowest tensile strength was 21.72 MPa of SE composite. Other prepared composite specimens (PSKE, PES, SEC, SES, KEC, KE, PEE, KEE, PEC, SEE, and PE) show 60.17, 62.14, 47.33, 58.56, 53.46, 39.19, 42.89, 67.52, 48.88, 37.62, and 28.80 MPa. The incorporation of fillers eliminates the possibilities of voids inside the composite structure and fills the gaps between the fiber and matrix interface. The proper filling of the interface provides a better debonding between the fiber and matrix interface which provides overall structural stability and improved tensile strength during tensile test than only fiber-reinforced polymer composite. In used fillers, seashell filler-based composite already shows better hardness which helps to sustain the permanent failure of the composite specimen during tensile test. Several numbers of authors performed their research on filler- and fiber-based composite and the effect of fillers on the characteristics of the developed composite specimen. Prabhudass et al. [35] studied the effect of MWCNT-filled bamboo/kenaf-reinforced epoxy nanocomposite. The author found that after the addition of MWCNT nanofiller into the composite, the tensile strength of the composite increased by 3.7%. Singh et al. [36] studied the effect of the addition of silica on the mechanical properties of hemp/sisal-reinforced epoxy composite. The authors found that with the addition of silica nanoparticle into the composite, the mechanical properties of the composite increased. The highest increase in tensile strength

Fig. 7 Experimental findings of tensile strength of all developed specimens

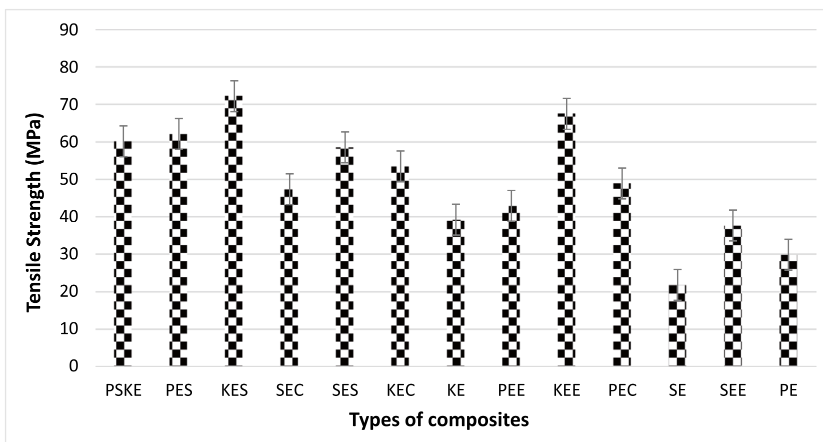
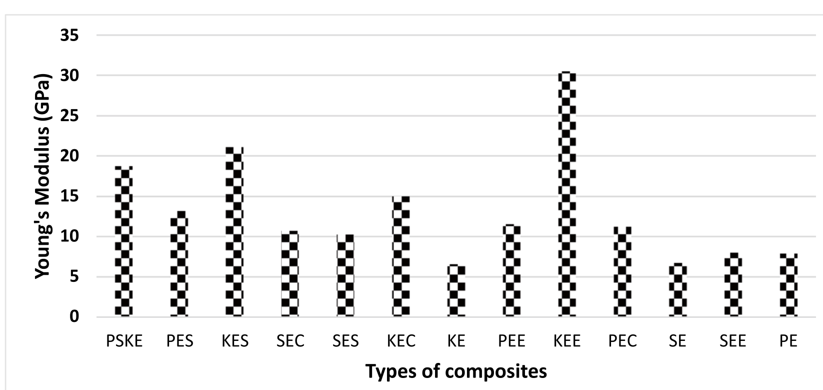


Fig. 8 Experimental findings of Young's modulus of all developed specimens



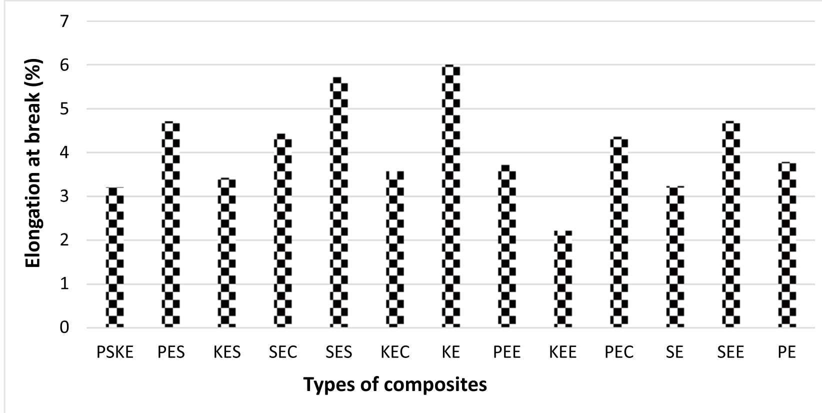


Fig. 9 Experimental findings of elongation at break of all developed specimens

was seen at 2 wt% addition of silica into the composite. On increasing the amount of silica added into the composite, the properties deteriorated.

Young's modulus was calculated, and the results are shown in Fig. 8. Like tensile strength, KEE and KES composite shows the highest and second highest Young's modulus of 30.49 and 21.07 GPa. Hybridization of pineapple, kenaf, and sisal fiber with epoxy composite achieved the third highest value of Young's modulus at 18.72 GPa. The lowest Young's modulus was achieved by KE and SE composites at 6.63 and 6.71 GPa. Experimental findings display that the hybridization of fibers and incorporation of biofiller enhance the values of Young's modulus like tensile strength values of all prepared specimens. Young's modulus increases when the internal resistance applied by the molecular structure increases and the strain during the application of force is reduced. So, the hybridization of fibers and incorporation of biofillers eliminates the possibilities of air entrapments, voids, and irregularities in the internal structure of the prepared composite specimen.

The maximum value of elongation at break was achieved by KES composite at 6% as shown in Fig. 9. A higher value of elongation reduces the value of Young's modulus which is clearly visible in the minimum value of Young's modulus of KE composite as other developed composite specimens. Similarly, the minimum value of elongation at break was achieved by KEE composite specimen at 2.2%, and this minimum value of elongation provides the maximum Young's modulus to KEE composite specimen as discussed in Young's modulus section. Other prepared specimens (PSKE, PES, KES, SEC, SES, KEC, PEE, PEC, SE, SEE, and PE) show the value of Young's modulus at 3.2, 4.7, 3.4, 4.4, 5.7, 3.5, 3.7, 4.3, 3.2, 4.7, and 3.7%. Researchers found different findings for elongation at break for nanofiller-based polymeric composites. Madhu et al. [37] studied the effect of Ca_2SiO_4 nanofiller on the properties of glass/silk fiber-reinforced epoxy composite. The author found that the addition of nanofiller improved the elongation at the break of the composite. The composite with 3 wt% of nanofiller showed the best elongation at break. Shunmugasundaram et al. [38] studied the effect of carbon nanotube and graphene nanofiller on the properties of neem fiber/epoxy composite. The addition of 4 wt% of graphene in the composite improved the tensile strength by 16.38% while the addition of 4wt% of carbon nanotube improved the tensile strength by 15.13%.

After the tensile test, all the fractured surfaces of specimens were examined using scan electron microscopy (SEM) shown in Fig. 10. Some fracture mechanisms are present in the micrographs like debondings between fiber and matrix phase, fiber fractures, fiber and fillers breakage from matrix, etc. In filler-based composites, SEM micrographs clearly show the distribution of fillers which are bonded with fiber bundles and reduce the possibilities of void content. Fillers are completely distributed with fiber and matrix phase and imparting better bonding between fiber matrix phase which provides the overall structural stability to the tested composite specimen.

3.6 Flexural strength

The flexural testing results for the bi-composite samples are presented in Figs. 11 and 12. All prepared composite specimens were tested under a 3-point bending test to evaluate the flexural strength. Among all the prepared composite specimens, sisal fiber-based composite specimen shows a higher value of flexural strength. The lowest value of hardness value displayed in the hardness results above shows the flexible and ductile behavior of the sisal-based epoxy composites. And findings of flexural strength clearly display the incorporation of fillers with fibers enhancing the flexural strength than single fiber-based composite specimens, and the hybridization of pineapple, kenaf, and sisal fiber with epoxy provides comparable results as compared to filler- and fiber-based composites due to different surface behaviors to different fiber mats. SEE composite shows the highest value of flexural strength of 257.25 MPa,

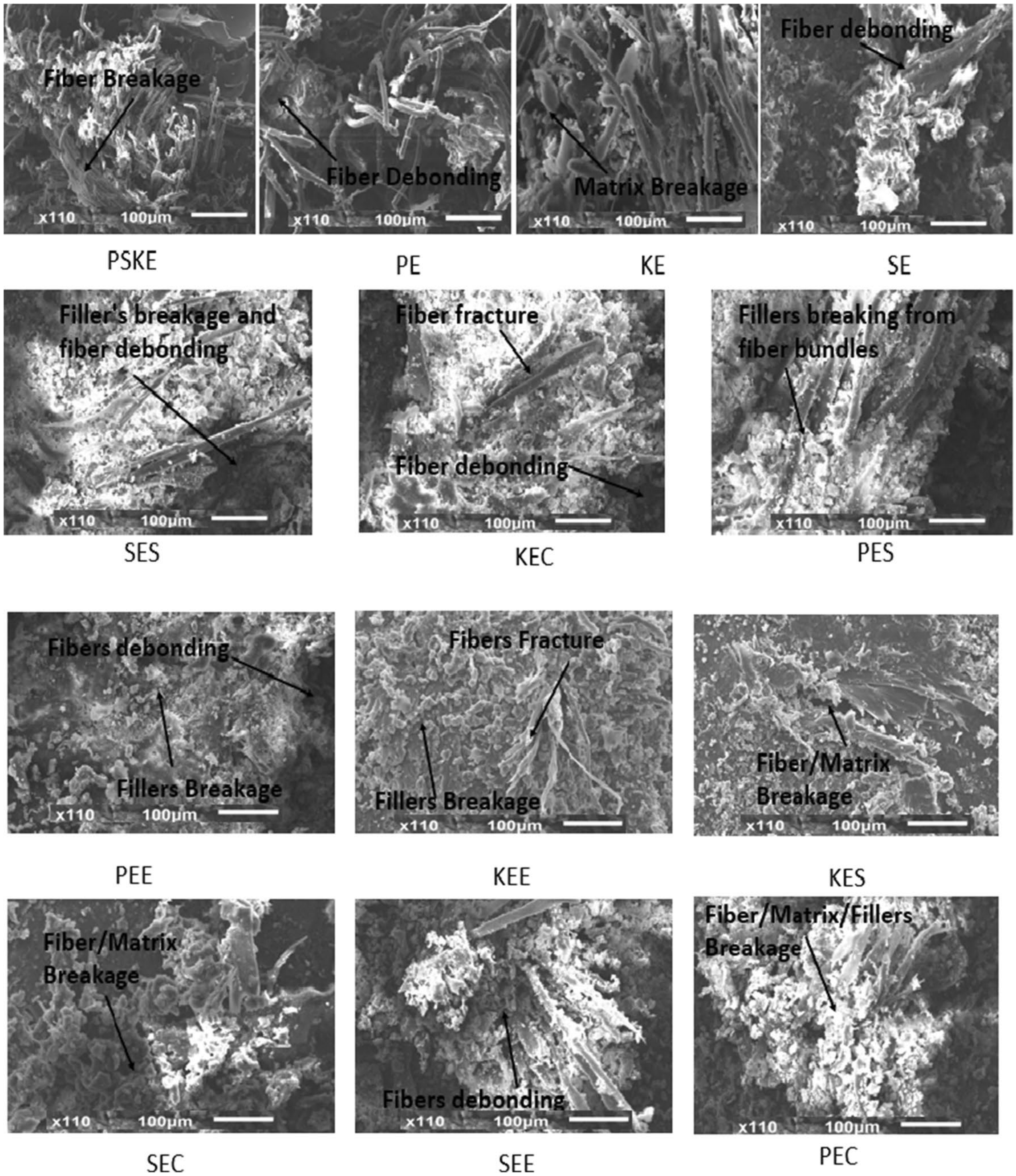


Fig. 10 SEM images of prepared composite specimens after tensile test

and KES, SEC, and SES composites achieved the second highest and comparable flexural strength of 198.6, 193.8, and 195.7 MPa, while PEE composite achieved the lowest flexural strength at 85.5 MPA and hybrid composite of PSKE shows 169.5 MPa. Other prepared specimens (PES, KEC, KE, KEE, PEC, SE, and PE) achieved 139.5, 119.6, 138, 151.5, 109.3, 112 and 108 MPa. Based on surface properties, kenaf and pineapple fibers are rougher fiber than sisal fiber and the incorporation of biofillers with sisal fiber enhanced the interfacial adhesion between sisal fiber and epoxy resin which imparts the flexible nature of sisal/filler/epoxy composite specimens. This nature of flexibility with ductile nature enhanced the flexural strength of sisal-based composite specimens. In previous research work, authors have investigated fiber- and filler-based epoxy composites. Bellairu et al. [39] studied the application of the mixture-design technique in the addition of nanofiller into the polymer composite and the changes in the mechanical properties of the composite after the addition of the nanofiller. The author found that the addition of

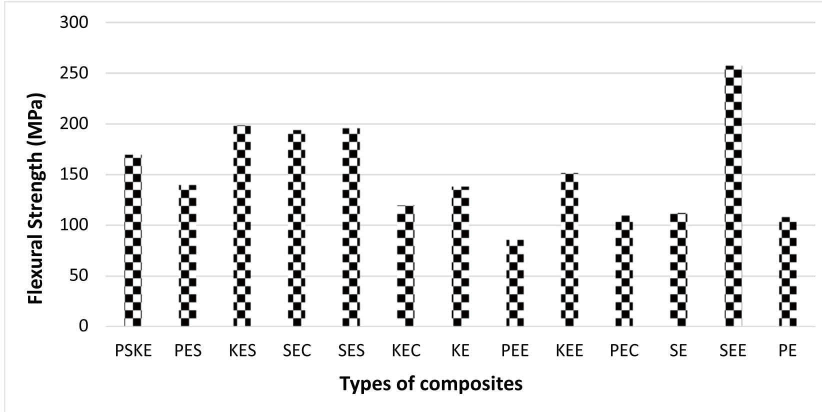


Fig. 11 Experimental findings of flexural strength of all developed specimens

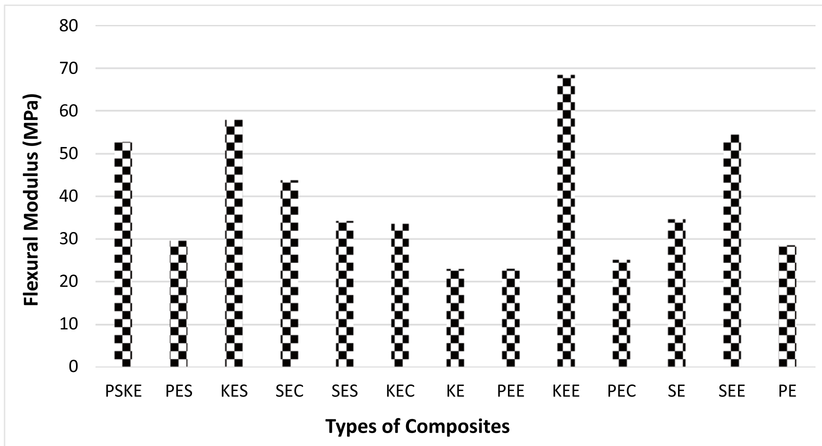


Fig. 12 Experimental findings of the flexural modulus of all developed specimens

MWCNT nanofiller into Catala fiber/epoxy composite led to an improvement in the flexural strength of the composite significantly. Chaturvedi et al. [40] studied the effect of the addition of carbon nanotube fillers on the properties of fly ash/epoxy-based polymer composite. The authors found that the flexural strength of the nanocomposite increased with an increase in the amount of carbon nanotube added. This was found to be due to the high dispersion of the carbon nanotube in the epoxy matrix which led to a weakening of the Vander Waals force, hence improving the flexural strength of the composite.

Flexural modulus results also demonstrate that the incorporation of fillers with fiber-reinforced epoxy composite delivers better results than fiber-based epoxy composites. The highest value of flexural modulus was attained by KEE composite at 68.41 MPa. The lowest value of flexural modulus was attained by KE and PEE composites at 23. PSKE hybrid composite and KES specimen show the second highest and comparable flexural modulus of 57.7 and 57.9 MPa. Other prepared specimens (PES, SEC, SES, KEC, PEC, SE, SEE, and PE) exhibit 29.5, 43.7, 34.2, 33.5, 25, 36.6, 54.4, and 28.5 MPa respectively.

After the flexural test, all fractured surfaces of the specimens were checked using scanning electron microscopy (SEM) to evaluate the cause of fracture as shown in Fig. 13. The micrographs revealed various fracture mechanisms, such as debonding between the fiber and matrix phases, fiber fractures, and the breakage of fibers and fillers from the matrix. In filler-based composites, the SEM images clearly display the distribution of fillers bonded with fiber bundles, which reduces the likelihood of void content. The incorporation of fillers improves the fiber/matrix interfacial strength which gives flexibility to the composite specimen. This flexibility resists the deformation during bending of the specimen in a 3-point bending test and imparts good bending strength.

3.7 Impact strength

The results attained from the impact tests for all prepared composite specimens are shown in Fig. 14. Table 6 represents the impact strength values of each prepared specimen. Like tensile strength and flexural strength, merging of fillers with fibers improved the value of impact strength than only

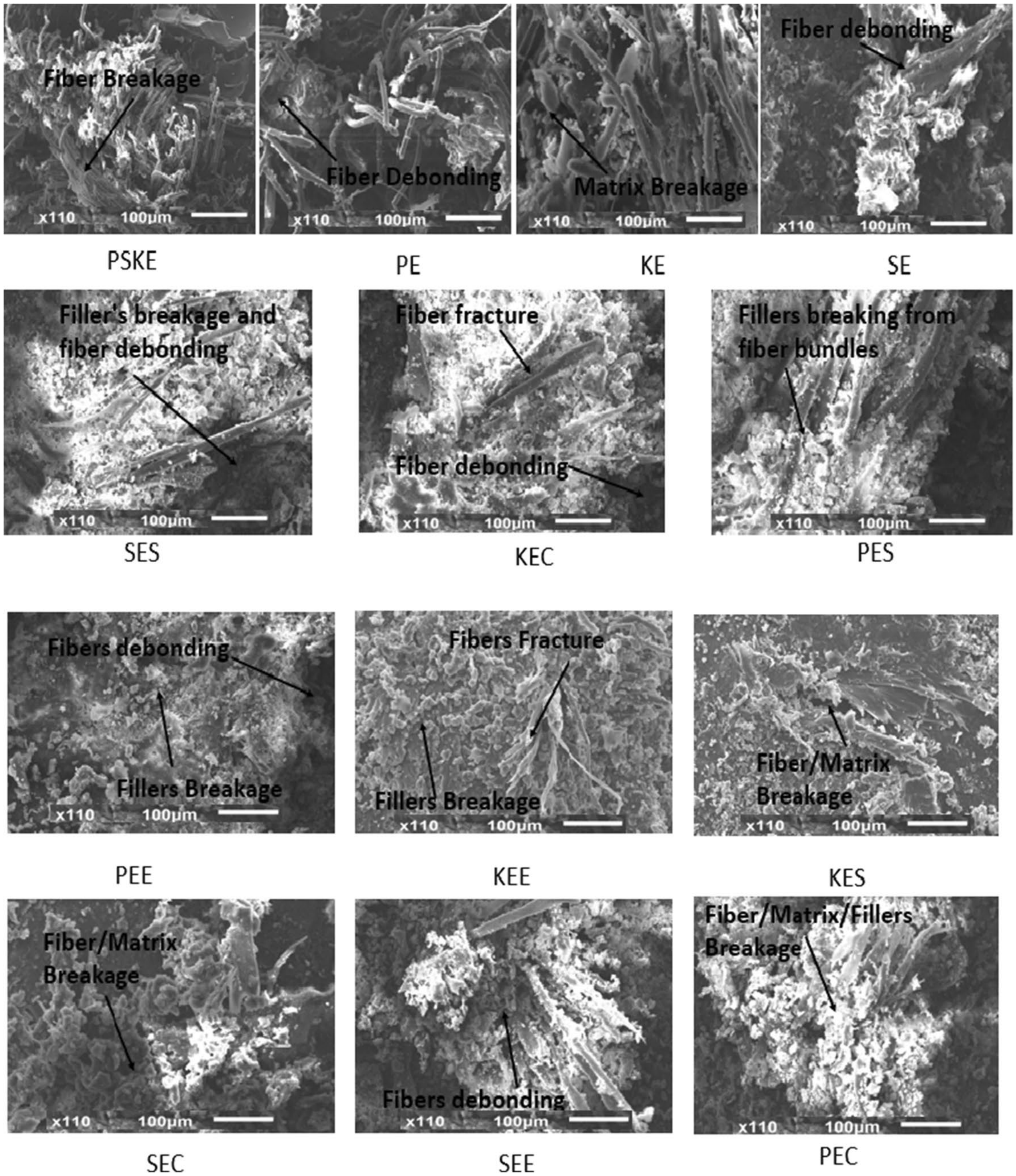


Fig. 13 SEM images of prepared composite specimens after flexural test

fiber-reinforced polymer composites. Among all the prepared composite specimens, sisal fiber-based composites with a combination of coconut, eggshell, and seashell fillers achieved higher impact strength than other fiber-based composites. And coconut filler with sisal fiber-reinforced epoxy achieved the highest value of impact strength at 0.9 J. Rough surface properties of coconut enhanced the wettability between fiber and matrix interface, and ductile behavior of sisal fiber with brittle behavior of epoxy resin imparts the higher absorbance during sudden load of impact test. PEE composite shows the lowest value of impact strength at 0.1 J. The second highest impact strength was attained by SES and SEE at 0.6 J, while PSKE, PES, and KES composite specimens achieved a similar impact strength of 0.2 J. Other prepared specimens (KEC, KE, KEE, SE, and PE) attained the impact strength of 0.3, 0.4, 0.5, 0.3, and 0.3 J. Previous researchers performed their research work on the effect of various fillers on the impact strength of fiber-reinforced composites. Venkatesh et al. [13] studied the effect of the addition of MWCNT/nano-bagasse nanofiller on the mechanical properties of E-glass/epoxy composite. The

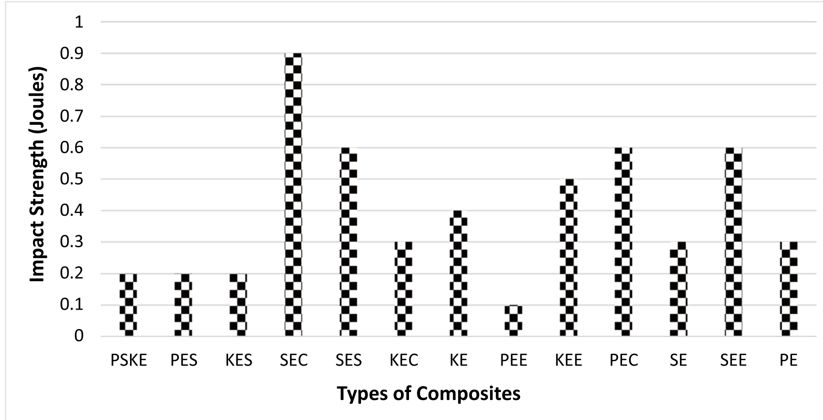


Fig. 14 Experimental findings of impact strength of all developed specimens

Table 6 Impact strength of all fabricated specimens

Type of composite	Impact strength (Joules)
Pineapple/epoxy	0.3 J
Sisal/epoxy	0.3 J
Kenaf/epoxy	0.4 J
Pineapple/epoxy/seashell	0.2 J
Pineapple/epoxy/eggshell	0.1 J
Pineapple/epoxy/coconut	0.6 J
Sisal/epoxy/seashell	0.6 J
Sisal/epoxy/eggshell	0.6 J
Sisal/epoxy/coconut	0.9 J
Kenaf/epoxy/seashell	0.2 J
Kenaf/epoxy/eggshell	0.5 J
Kenaf/epoxy/coconut	0.3 J
Pineapple/sisal/kenaf/epoxy	0.2 J

authors found that the addition of the nanofiller into the composite led to an improvement in the impact strength of the composite. The improvement was maximum in the nanocomposite with seven layers of the composite and 1 wt% of CNT nanofiller. Mayakannan [14] studied the effect of the addition of silica nanoparticles into the PALF/sisal fiber-based polymer composite. The authors found that adding the silica nanoparticle into the composite improved the mechanical properties of the composite. The mechanical properties were found to be maximum in the case of the C-type hybrid composite created by the author.

4 Conclusion

In the present study, a detailed discussion about the effect of fillers (coconut, eggshell, and seashell) on the mechanical performance of developed composites was discussed, and all biofiller-based composites were compared with single fiber-based composites and hybrid composite of pineapple-, sisal-, and kenaf fiber-reinforced epoxy composites. Based on the findings, the following conclusions are made.

1. The incorporation of biofillers with fibers and epoxy polymers eliminates the percentage of void content, and seashell-filled composite specimens show a minimum percentage of void content than all other prepared composite specimens.
2. The water absorption study displays the behavior of the water uptake capacity of all prepared specimens with respect to time. Due to the hydrophilic nature of fibers and fillers, all prepared specimens absorb the water content. The high hydrophilic nature of coconut filler endows the high-water absorption to the coconut filler-based composite specimens.
3. Discoveries of tensile strength exhibit that seashell and eggshell-filled composite specimens achieved the highest tensile strength and Young's modulus as compared to other prepared specimens. Seashell also imparts good elongation at break to composite specimens, while hybridization of fibers (pineapple, sisal, and kenaf) provides the highest hardness than other prepared specimens. In the tensile test, KES composite shows the highest tensile strength of 72.25 MPa and the lowest tensile strength achieved by SE composite at 21.72 MPa.
4. Outcomes of flexural strength and flexural modulus exhibit that incorporated fillers enhance the value of flexural strength of all prepared specimens and eggshell work as an effective filler for enhancing the flexural strength of all prepared specimens. Among all the developed specimens, sisal- and eggshell-based epoxy composite achieved the maximum value of flexural strength of 257.25 MPa.
5. Findings of impact strength reveal that sisal fiber with all three fillers (pineapple, sisal, and kenaf) displays a higher impact strength. Reducing the percentage of void

content in filler-based composite exhibits a higher value of impact strength as compared to only fiber-based composites. Sisal fiber-based composites with a combination of coconut, eggshell, and seashell fillers show higher impact strength than other fiber- and filler-based composites. Coconut/sisal/epoxy composite achieved the highest value of impact strength at 0.9 J.

The testing results clearly show that composite specimens without fiber reinforcement have significantly lower mechanical performance compared to filler- and fiber-based composite specimens. The addition of fillers and hybridization of fibers highlight the crucial role of fillers and fibers in enhancing both the strength and interfacial adhesion of the prepared specimens. This study, along with previous research, suggests that the strength of composite samples can be further improved by increasing the amount of fiber and filler reinforcement to a certain extent. Future research should focus on using different natural fibers as reinforcement and natural materials as biofillers to further explore this area. Different weight proportions of fibers/fillers and chemical modification of fibers/fillers should be explored in this area.

Author contribution Bodhisatwa Seal: writing—original manuscript; Vijay Chaudhary, Susmita Dey Sadhu: ideas, conceptualization, supervision, reviewing, and editing.

Data availability Not applicable.

Declarations

Ethical approval Not applicable.

Competing interests The authors declare no competing interests.

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