Optimization and characterization of surface treated *Lagenaria siceraria* fiber and its reinforcement effect on epoxy composites

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Abstract

Purpose – This paper aims to investigate the resultant optimal ultimate tensile strength, elongation, flexural strength and modulus, compression strength and impact strength of fabricated alkali-treated Lagenaria siceraria fiber (LSF)-reinforced polymer matrix composite by optimizing input factors and microstructural characterization by influencing fiber length, fiber concentration and treatment condition of LSF.

Design/methodology/approach – The fabrication of LSF-reinforced composite specimens involved surface treatment followed by custom experimental design using a simple hand layup process. The wear analysis was performed by a multi-tribotester TR25 machine, and the developed model was validated by using statistical software Design Expert V.8 and analysis of variance (ANOVA). The surface morphology of the sample was also analyzed by field emission scanning electron microscopy.

Findings – The alkali treatment for LSFs had reduced the hemicellulose, and enhanced mechanical performance was observed for 30 wt.% concentration of *L. siceraria* in epoxy resin. Thermogravimetric analysis revealed thermal stability up to 245°C; microstructure revealed fiber entanglements in case of longer fiber length and compression strength reduction; and the surface-treated fiber composites exhibited reduced occurrences of defects and enhanced matrix–fiber bonding. Enhanced mechanical performances were observed, namely, ultimate tensile strength of 17.072 MPa, elongation of 1.847%, flexural strength of 50.4 MPa, flexural modulus of 3,376.31 GPa, compression strength of 52.154 MPa and impact strength of 0.53 joules.

Originality/value – The novel approach of optimizing and characterizing alkali surface-treated LSF-reinforced epoxy matrix composite was explored, varying fiber length and concentrations for specimens by empirical relations and experimental design to obtain optimal performance validated by ANOVA. Enhanced properties were obtained for: 7 mm fiber length and 30 wt.% concentration of fiber in the composite for alkali-treated fiber.

Keywords *Lagenaria siceraria*, Alkali treatment, Natural fibers, Epoxy matrix, Optimization, Mechanical properties, Optimization techniques, Epoxy resins

Paper type Research paper

1. Introduction

The ever-increasing requirement for environment conscious materials and their applications are ever increasing, especially in light of the pandemic situation around the globe. With the slowing momentum of the mainstay materials supply chain around the world, the long-expected stimulus for adopting sustainable materials and solutions has arrived. This lack of impetus has catalyzed increasing applications of ready-to-procure natural fibers such as jute, flax and coconut to name a few. Plant fibers are naturally bestowed with mechanical properties comparable to synthetics in terms of reinforcement ability although with lesser processing advantages (Leao *et al.*, 2006). The industrial demand for natural fiber composites has increased in recent years with the major applications being in the social sectors with large public interactions acting as an incitement to adopt natural fiber products. Several studies have explored the fabrication of polymer matrix composites (PMCs) by natural fiber reinforcement with varying parameters such as the content of fiber, fiber length, methods of surface treatment and eventual characterizations of performance of the fabricated composite (Prasad *et al.*, 2021). Natural fibers such as hemp, jute and cotton were mixed with poly-lactic acid and were tested for their mechanical performance. The PLA composite with hemp exhibited enhanced tensile strength, while the composite reinforced with jute produced exhibited improved Young's

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modulus (Liao *et al.*, 2020). The automotive industry has also recently concentrated more on adopting natural-fiber-based composites for interior materials and upholstery moving away from traditional leather. These composites integrated palm, banana, sisal, coir fibers and exhibit improved performance and are hard wearing (Vidal *et al.*, 2021). *Lagenaria siceraria* (LS) or common bottle gourd is a vegetable plant that thrives on most ecosystems. Its remnants are abundant and mostly discarded without any use cases. But natural fibers such as LS fiber (LSF) are generally susceptible to hydrophobic interaction with synthetic matrices, thus limiting their structural applications (Nagappan *et al.*, 2021). This is mainly attributed to the presence of hydrophilic hemicellulose. This issue can be rectified by the chemical treatment of fibers. This treatment involves a range of fiber treatments such as alkalization, bleaching, acetylation and silane treating to chemically modify the surface for better fiber–matrix interaction (Jamshaid *et al.*, 2020). Mercerization is an alkali treating method by which the natural fiber is immersed in a concentrated solution such as sodium hydroxide. The solution reacts with the hemicellulose and other hydrophilic compounds such as lignin and wax, which surround the fiber surface, and removes them effectively. This results in fiber that has a certain roughness on the surface enabling the fibers to interact better with the matrices (Mohana Krishnudu *et al.*, 2020). Chemical modification of natural fibers directly enhances the mechanical properties of the composite. The effect of abaca fibers immersed in varying concentrations of sodium hydroxide solutions resulted in their increased crystalline nature, tensile and Young's modulus in contrast to the untreated fibers with increased interfacial shear strength (Malik *et al.*, 2021). Studies showed that control of weight percentage concentration of sodium hydroxide is almost ideal in treating natural fibers to remove excessive content of moisture while simultaneously increasing interfacial bonding strength (Mrajji *et al.*, 2021). Recent studies have largely concentrated on determining optimal input conditions for fabrication and treating natural fiber PMCs (Kumar *et al.*, 2020). Studies involved optimization of hemicellulose, lignin and wax removal from sisal fiber with varying alkali concentrations by use of different experimental designs such as Box–Behnken, Central composite or even custom experimental designs to identify optimal conditions for fabrication (Gurukarthik Babu *et al.*, 2019). Response surface methodology was used in some cases to optimize the treatment of natural fibers for maximum cellulose concentration to enable better bonding.

In the present study, the novel approach of optimizing the input factors of fabrication and characterizing surface-treated LSF-reinforced epoxy matrix composite was investigated. The LSFs were surface treated, and by the following custom experimental design, the specimens were fabricated by simple hand layup process. The input fiber length and concentrations were varied in fabricating 19 experimental specimens. Empirical relations were developed; the design was validated for adequacy by analysis of variance (ANOVA). The experimental specimens were characterized for mechanical properties, and microstructural investigation and their respective improvements were evaluated.

2. Materials and methods

2.1 Lagenaria siceraria fiber and extraction

LSF is obtained from the commonly available bottle gourd vegetable known for its medicinal and nutritious efficacy. The stem of the LS plant was separated from its leaves and was rinsed with water to eliminate impurities. The skin of the plant stems is used to extract the natural fiber by the process of water-retting. The elongated plant stems were sliced into pieces of 15 cm in length and were submerged in water for a 14-day period to eliminate the presence of microbes (Rozyanty *et al.*, 2021). The stems were separated from their fibers and were rinsed with distilled water and were subsequently dried under the sun for 72 h to remove existing moisture.

The dried fibers are then subjected to a mercerization process in which the natural LSF is immersed in a strong solution to induce enlargement within the fibers to create structural changes to the microstructure and physical properties of the fiber (Mrajji *et al.*, 2021). The dried fibers were soaked in sodium hydroxide solutions with weight/volume (w/v) concentrations varying from 2% to 15% and different temperature levels ranging from 25°C to 70°C. The fibers are treated for a period of 6 h. Subsequently, the treated LSFs were rinsed with an acetic acid solution of 1% volume concentration and then with distilled water and were dried in a hot-air oven for 24 h at 60°C. The dried fibers were then ultimately cooled and placed in airtight storage for analysis.

2.2 Determination of chemical composition of Lagenaria siceraria fibers

The extracted natural LSF is a composite comprising cellulose, hemicelluloses, wax, lignin and ash content. Natural fibers are typically reinforced by cellulose microfibrils with hemicelluloses and lignin being matrix elements. It is desirable to have a higher content of cellulose reinforcement to improve the tensile strength of the fiber. Hemicelluloses are amorphous and non-homogenous and thus they transfer stresses to microfibrils in event of high stressed applications leading to reduced mechanical performance. Thus, it is inferred that to have enhanced strength, it is imperative for hemicelluloses to be lower in content than cellulose (Vinayagamoorthy *et al.*, 2019).

The density of the LSFs was measured per ASTM-D-792 standard for density and specific gravity method. The Mettler Toledo XS205 laboratory balance was used to measure the density of the extracted fibers; the amount of wax present in the fiber is measured by Conrad's procedure. The ash content of the LSF is the measure of its mineral content. The composition of ash is generally used to infer the origin of fibers and relate to the type of treatment to be applied to the fibers. It is determined to level of ash content present within the fibers is determined per ASTM E1755-01 standard where a defined amount of LSF was burned in a non-ferrous crucible until complete incineration. The amount of leftover ash residue is termed ash content (Adeyi *et al.*, 2021). The moisture content of the LSF was determined by subjecting the fibers to an oven (Model Memmert HCP 108) with an air-conditioned environment. The fiber bundles were successively introduced at a constantly maintained temperature of 25°C with a relative humidity of 60%–90%. The fiber specimens were weighed at regular periods and moisture content determined. The measured chemical composition of untreated LSF and treated LSF are listed in Table 1.

The microstructure of the treated and untreated LSFs was analyzed for surface properties by scanning electron

micrographs. The micrograph of the untreated LSF reveals the presence of foreign matters trapped within the fiber prior to alkali treatment (mercerization). Nevertheless, subsequent to alkali treatment, the surface of the LSF can be observed to exhibit a rough texture in comparison to the untreated LSF devoid of foreign matter. Thus, it can be inferred that the mercerization process removes hemicellulose away from the surface of the LSF. This effectively enhances the wettability between the surface of the LSF and the epoxy matrix.

2.3 Preparation of polymer matrix composites

The present study involves the investigation of a PMC reinforced with LSF. The treated LSF is used as reinforcement on an epoxy-based PMC. The epoxy resin used in this study is epoxy Araldite LY-556, which serves as the matrix material. The hardener used in the study was used to cure the epoxy resin is Aradur HY-951, which is a low viscous unfilled epoxy resin casting hardener. The epoxy matrix of the composite was toughened by using a hydroxy-terminated silicon modified polyurethane oligomer. The epoxy resin was modified by mixing epoxy resin and 5 wt.% of hydroxy-terminated silicon modified polyurethane oligomer. During the modification, no catalysts or solvents were used. The process of modification lasted for upto 1 h at standard room temperature (25°C). The epoxy resin and the predetermined amount of oligomer were both weighed on a container and stirred together by a magnetic stirrer for a duration of 1 h at standard room temperature (25°C).

The untreated and treated LSFs were sliced into varying sizes of 3, 5, 7 and 9 mm. The ratio of epoxy and hardener mixture was 10:1, and a total of 19 PMC specimens were fabricated with the previously cut varying length LSFs as reinforcement. The fabricated PMCs had dimensions of 300 mm length, 300 mm width and a thickness of 3 mm. The PMCs were fabricated by varying LSF weight concentrations such as 15, 20, 25, 30 and 35. The PMCs were fabricated by a simple hand layup process and were subsequently compression molded for 3 h with a pressure of 80 MPa under 80°C. The composite sheet is then extracted from the cured mold by application of wax. The PMCs were subsequently cut per ASTM testing standards for mechanical properties characterization.

2.4 Experimental design

The LSF-reinforced epoxy PMC was fabricated based on a custom experimental design. The design was prepared, analyzed in Minitab V.16 software. The design of experiments involved three input varying factors such as fiber length (mm), fiber concentration weight percentage (%) and condition of the fiber (treated or untreated). The output responses were ultimate tensile strength (MPa), elongation of the specimen (%), flexural strength (MPa), flexural modulus (GPa), compressive strength (MPa) and impact strength (joules). The custom design of experiment had a total of 19 trial runs. Statistical analysis by developed regression equations was performed to determine the output responses and their adequacy was verified by ANOVA. The input factors and output responses in the experimental trial are tabulated in Table 2.

Table 1 Chemical mixture of the untreated and treated *Lagenaria siceraria* fiber

Fiber	Cellulose (%)	Hemicellulose (%)	Lignin (%)	Wax (%)	Moisture (%)	Ash (%)	Density (g/cm^3)
Untreated LSF							
Raw LSF	79.91	12.69	7.62	0.31	9.96	0.92	1.216
Jute	64.4	12	11.8	0.7	1.1	–	1.3
Flax	64.1	16.7	2.0	1.5–3.3	3.9	–	1.5
Hemp	68	15	10	0.8	–	–	1.47
Kenaf	31–72	20.3–21.5	8–19	–	–	–	1.45
Ramie	68.6–85	13–16.7	0.5–0.7	0.3	7.5–17	–	1.5
P. foetida	77.96	12.63	10.47	0.35	9.54	0.94	1.328
A. leucophloea	68.09	13.60	17.73	0.55	8.83	–	1.385
I. staphylina	72.76	13.6	19.56	1.51	8.28	1.4	1.40
Treated LSF							
Raw LSF	79.91	12.69	7.62	0.31	9.96	–	1.216
2% ATLSF	82.63	8.41	7.37	0.18	8.94	–	1.219
4% ATLSF	86.49	4.23	6.98	0.2	8.23	–	1.223
6% ATLSF	84.21	3.18	6.08	0.08	8.56	–	1.227
8% ATLSF	85.33	2.2	5.42	0.02	9.12	–	1.232
10% ATLSF	83.98	–	5.13	–	9.47	–	1.238
15% ATLSF	80.12	–	4.42	–	9.84	–	1.237
Jute	64.4	12.0	11.8	0.7	1.1	–	1.3
Flax	64.1	16.7	2.0	1.5–3.3	3.9	–	1.5
Hemp	68.0	15	10.0	0.8	–	–	1.47
Kenaf	31–72	20.3–21.5	8–19	–	–	–	–
Ramie	68.6–85	13–16.7	0.5–0.7	0.3	7.5–17		1.5
P. foetida	77.96	12.63	10.47	0.35	9.54	–	1.328

Note: ATLSF = Alkali Treated Lagenaria Sicereria fiber

2.5 Developing empirical relationship by RSM

Response surface methodology (RSM) is a mathematical methodology used to develop regression models to analyze problems concerning the effects of input parameters on desired output response of an experiment (Hassan *et al.*, 2019). In the present investigation, the output responses of the PMC fabrication involved are the ultimate tensile strength, elongation, flexural strength and modulus, compression strength and impact strength of LSF-reinforced epoxy PMC specimens. The input parameters of fabrication are the length of LSF (A), the weight percentage of LSF (B) and condition of LSF (C). These output responses are dependent on the input conditions and, therefore, are functions of the input parameters, and are expressed as:

$$(\text{Ultimate Tensile Strength, Elongation, Flexural Strength, Flexural Modulus, Compressive Strength, Impact Strength}) = f\{A, B, C\} \quad (1)$$

Second-order regression equations were developed as empirical relations with the three factors for each output response:

$$\text{Ultimate Tensile Strength} = +16.08 + 0.3849A - 0.1041B - 0.7688C - 0.1031AB - 3564AC - 0.7378BC - 0.7876A^2 - 1.41B^2 \text{ MPa} \quad (2)$$

$$\text{Elongation} = +1.69 + 0.0514A + 0.0229B - 0.1009C - 0.0033AB - 0.0333AC - 0.0598BC - 0.1450A^2 - 0.2104B^2\,(\%) \quad (3)$$

$$\text{Flexural Strength} = +46.35 + 1.17A + 0.2692B - 2.40C - 0.5289AB - 0.2913AC - 1.68BC - 4.98A^2 - 5.74B^2 \text{ MPa} \quad (4)$$

$$\text{Flexural Modulus} = +3050.34 + 108.11A - 46.68B - 255.43C - 90.61AB - 4.72AC - 269.72BC - 513.78A^2 - 655.29B^2\text{GPa} \quad (5)$$

$$\text{Compressive Strength} = +47.53 + 1.78A + 0.1696B - 3.47C - 0.8564AB - 0.0073AC - 2.10BC - 6.73A^2 - 7.28B^2 \text{ MPa} \quad (6)$$

$$\text{Impact Strength} = +0.4879 + 0.0233A - 0.0038B - 0.0322C - 0.0017AB - 0.0057AC - 0.0195BC - 0.0779A^2 - 0.0850\,B^2 \text{ joules} \quad (7)$$

Table 2 Experimental plan using DOE

Run	Factor 1 A: fiber length mm	Factor 2 B: fiber weight (%)	Factor 3 C: condition	Response 1 Ultimate tensile strength MPa	Response 2 Elongation (%)	Response 3 Flexural strength MPa	Response 4 Flexural modulus GPa	Response 5 Compressive strength MPa	Response 6 Impact strength Joules
1	9	20	Untreated	14.684	1.475	40.85	2,635.38	40.076	0.41
2	5	25	Treated	16.564	1.737	47.95	3,254.53	50.91	0.51
3	3	25	Treated	15.364	1.532	40.94	2,673.26	40.437	0.41
4	3	35	Treated	14.431	1.425	38.5	2,215.44	37.832	0.33
5	9	35	Treated	15.875	1.573	39.45	2,327.43	38.752	0.39
6	5	25	Treated	16.564	1.737	45.95	3,054.53	47.91	0.48
7	5	35	Untreated	13.042	1.356	36.83	1,875.17	34.444	0.36
8	7	15	Untreated	14.927	1.424	40	2,519.22	39.537	0.4
9	9	15	Treated	14.735	1.421	37.26	2,000.22	36.439	0.36
10	7	15	Untreated	14.927	1.424	40	2,519.22	39.537	0.4
11	3	25	Untreated	14.657	1.405	37.98	2,162.75	36.732	0.37
12	9	30	Untreated	13.643	1.364	36.41	1,835.42	34.975	0.34
13	3	15	Treated	13.237	1.312	36	1,810.24	34.172	0.33
14	5	35	Untreated	13.042	1.306	35.83	1,750.17	33.444	0.32
15	7	35	Untreated	13.028	1.442	39.42	3,054.12	48.154	0.59
16	3	25	Untreated	14.657	1.505	39.98	2,362.75	36.732	0.37
17	7	30	Treated	17.072	1.847	50.4	3,376.31	52.154	0.53
18	5	20	Untreated	14.437	1.437	38.74	2,270.38	37.236	0.38
19	5	15	Treated	14.352	1.442	39	2,300.23	38.835	0.39

The polynomial equations were used to predict the PMC specimens' output responses.

2.6 Characterizations of polymer matrix composites

The fabricated PMCs were characterized for their mechanical properties such as ultimate tensile strength (MPa), elongation of the specimen (%), flexural strength (MPa), flexural modulus (GPa), compressive strength (MPa) and impact strength (joules) by standard test procedures. The test specimens were cut per ASTM standards for mechanical characterization. Ultimate tensile strength and percentage of elongation of the PMCs were tested by a universal testing machine and calculated per ASTM D3039 standard. The flexural strength and flexural modulus of the specimens were determined by the three-point bending method per ASTM D790 test standard by use of the universal testing machine. The PMC specimens were also impacted strength tested per ASTM D256 with the Izod impact testing procedure. The compression strength was determined by applying compression load to the PMC specimens placed in a specially designed compressive load-inducing fixture per ASTM D341 standard (Mohana Krishnudu *et al.*, 2020). The metallurgical properties of the PMC specimens were determined by analyzing micrographs obtained by scanning electron microscopy (SEM). The surfaces of the specimens were subjected to sputtering prior to the SEM process to circumvent the accumulation of electrical charge on the test surface.

2.7 Thermogravimetric analysis

The thermogravimetric analysis (TGA) was performed as per ASTM E1131 standard. The TGA Q500 from TA Instruments was the testing equipment used to perform TGA where 10 g of the sample weight was subjected to heating from room temperature to 800°C under a heating rate of 20°C/min. The nitrogen was used as the atmospheric gas and was pumped under a flow rate of 10 mL/min.

3. Result and discussion

3.1 Analysis of variance

The ANOVA was used to determine the adequacy of the developed empirical relations with a confidence interval of 95%. The developed model was considered to be valid on the condition that the *F*-ratio was found to be lower than the *F*-value in the F-table (Kumar *et al.*, 2020). RSM was used to determine the significant input factors affecting the mechanical properties of the PMC. The *F*-value for the ANOVA model developed was a significant 22.58, with a chance of 0.021%. For the lack of fit, the *F*-value obtained was an insignificant 11.74. The co-efficients used to determine the significant factor were: "R-Sq" (determination co-efficient) and "adj R-Sq" (adjusted co-efficient). The value of R-Sq depended on the variation of dependent variable depicted by variations, and for adj R-Sq, the values were set by significant terms. The estimated values are 0.9690, 0.9463, 0.9221, 0.9323, 0.9144 and 0.9225 for R-Sq and adj R-Sq 0.9522, 0.9463, 0.9251, 0.9377, 0.9120 and 0.9283 for ultimate tensile strength, elongation, flexural strength, flexural modulus, compressive strength and impact strength, respectively.

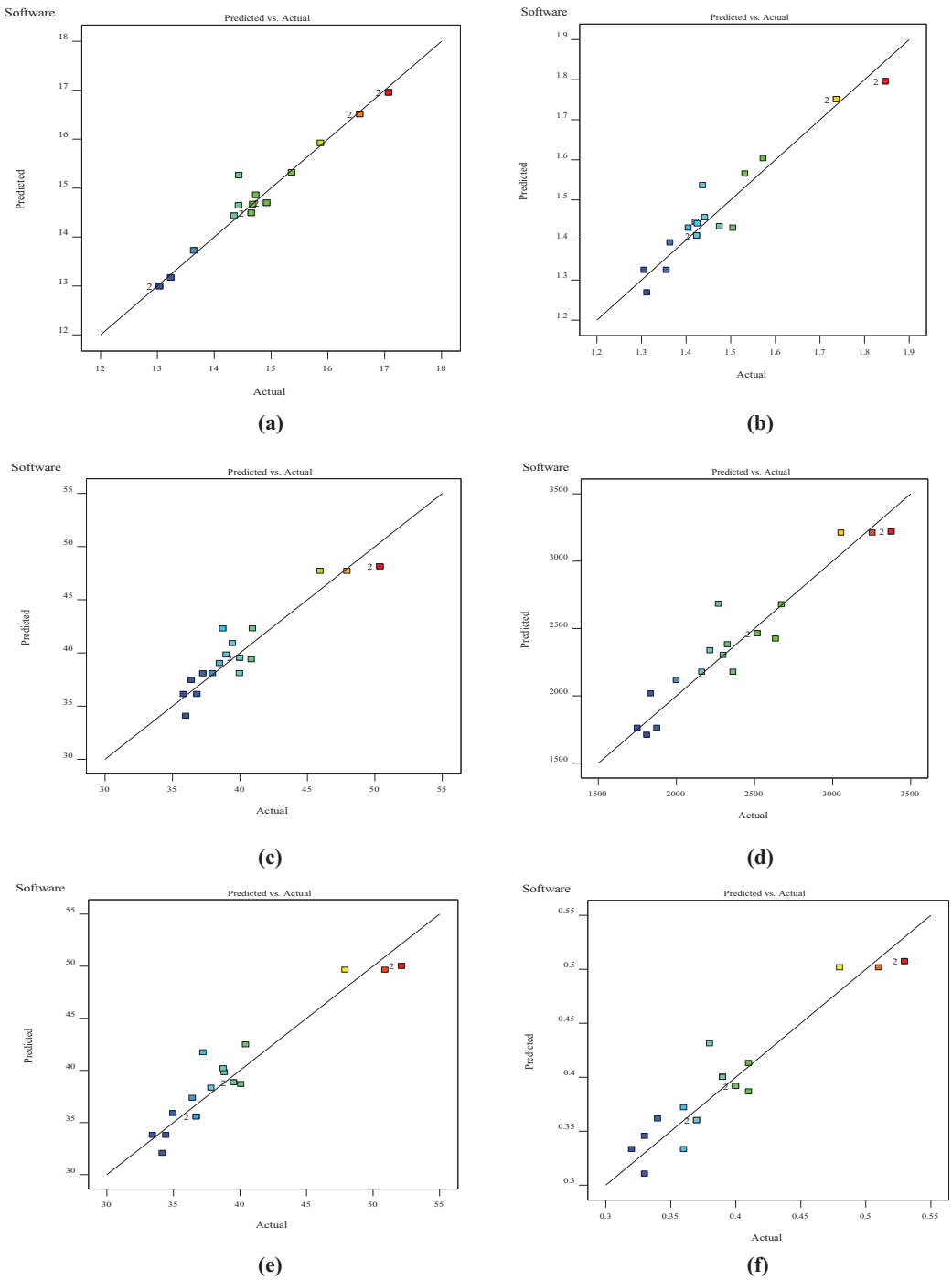
The residual probability of output responses was illustrated in Figure 1(a)–(f). The residuals plotted along a straight line between the actual and predicted values can be inferred. The residuals can be seen to be scattered near the slope of the plot indicating the accuracy of the predicted values of the output responses. The distribution of errors is closely related to verifying the validity adequacy of the empirical relations.

3.2 Effect of process parameters on all mechanical properties

The influencing input factors and their subsequent level of effect on the output responses of the alkali-treated LSF-reinforced epoxy PMC were inferred from Figure 2(a)–(f). The input factors of the fabrication process are LSF length (mm) and LSF concentration weight percentage (%). These input factors were represented by the curves A, B and C in the perturbation plots in Figure 2(a)–(f), respectively. The perturbation plots represent the trends of output responses against deviation from reference points (coded units). The length of LSF is instrumental in bolstering the directional properties of the composite, and the amount of weight percentage of the LSF plays important role in strengthening the matrix of the PMC. The parametric curves exhibiting sharp curvature in the perturbation plots represent the most influencing parameters for the respective output responses. As can be inferred from Figure 2(a), 2(b) and 2(d), curve B clearly exhibits the highest curvature indicating that the weight percentage of LSF in the PMC is significant toward the enhancement of ultimate tensile strength, elongation and flexural modulus. Although the variation in curvature between parametric curves A and B are less obvious in cases of Figure 2(c), (e) and to a certain extent in Figure 2(f), it was being established that the weight percentage of LS concentration influences the composites' flexural strength, compression strength and impact strength. It is established that the enhancement in mechanical properties is influenced by the fiber length with longer fibers providing stimulus to a rapid increase in output responses alongside a linear increase in fiber weight percentage concentration. These increases are seen until it reaches a plateau at 0.00 coded units in the perturbation plots of the respective properties and diminishes beyond that point. The peak values represent the optimal fiber length and concentration factor levels that are studied.

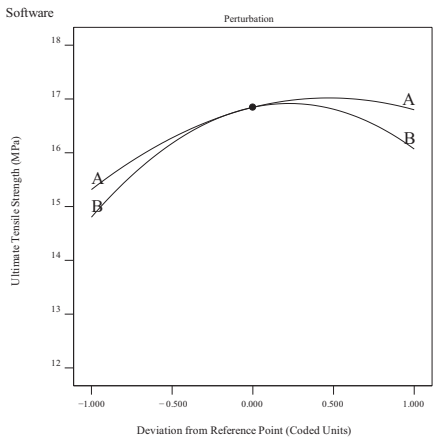
The results of the mechanical properties of the LSF-reinforced PMC are shown in Figure 3(a)–(f). The tensile, elongation, flexural, compression and impact properties are found to be greatly influenced by the fiber concentration and fiber length. As can be seen from the two- and three-dimensional plots marking the trends of the respective mechanical properties versus the input factors, the enhancement follows a slope with the curvatures of respective plots peaking at the optimal levels of input parameters. The measurable input parameters such as fiber length and fiber weight concentration were represented as plot points for the resultant values for the 17 trials. The tensile strength in Figure 3(a) is observed to be strongly affected by the fiber weight concentration with the peak value of 17.072 MPa reaching at 7 wt.% concentration. This is the resultant effect of improved matrix–fiber adhesion (Azeez *et al.*, 2020), which accompanies increased fiber concentration of and fiber length

Figure 1 Actual versus predicted (a) ultimate tensile strength; (b) elongation; (c) flexural strength; (d) flexural modulus; (e) compressive strength and (f) impact strength

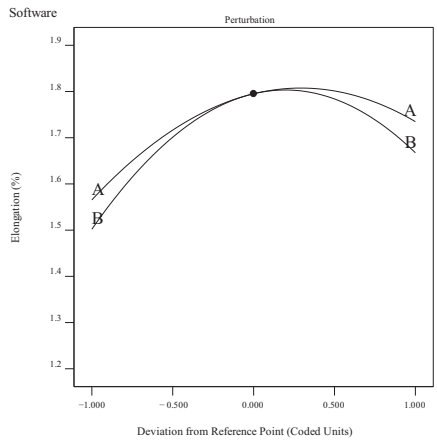


of 17 mm. The tensile strength falls beyond this critical level because of the increased interference in the bonding of matrix and fiber reinforcement. A similar trend is observed in the case of enhancement in elongation in Figure 3(b) where the peak value of 1.847% is observed at 7 mm fiber length and 30 wt.% concentration. With a high level of fiber length, the molecular weight matrix increases contributing to better ductility. The fiber fraction is found to greatly influence the flexural properties of the LSF-reinforced PMC. The plots in Figure 3(c) and 3(d) illustrate the peak flexural strength of 50.4 MPa and 3,376.31

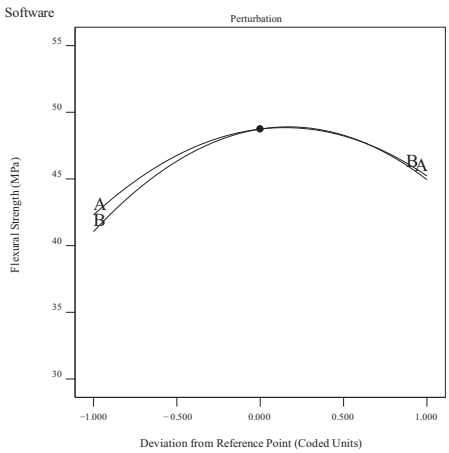
Figure 2 Perturbation plots of output responses: (a) ultimate tensile strength; (b) elongation; (c) flexural strength; (d) flexural modulus; (e) compressive strength and (f) impact strength



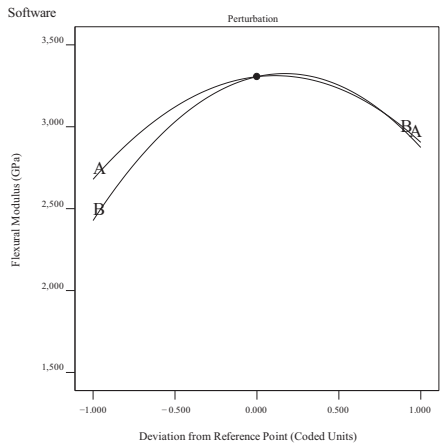
(a)



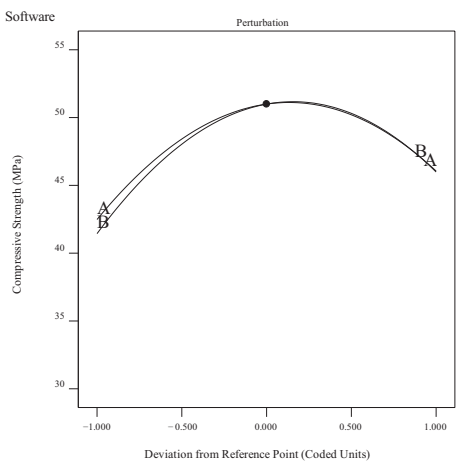
(b)



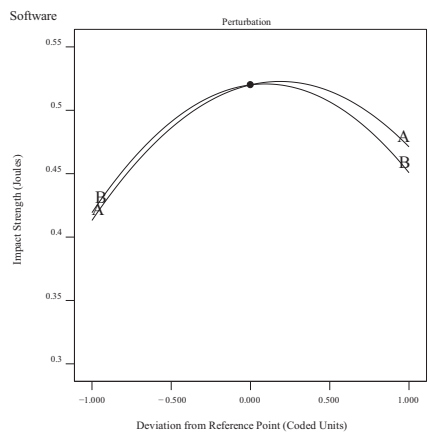
(c)



(d)

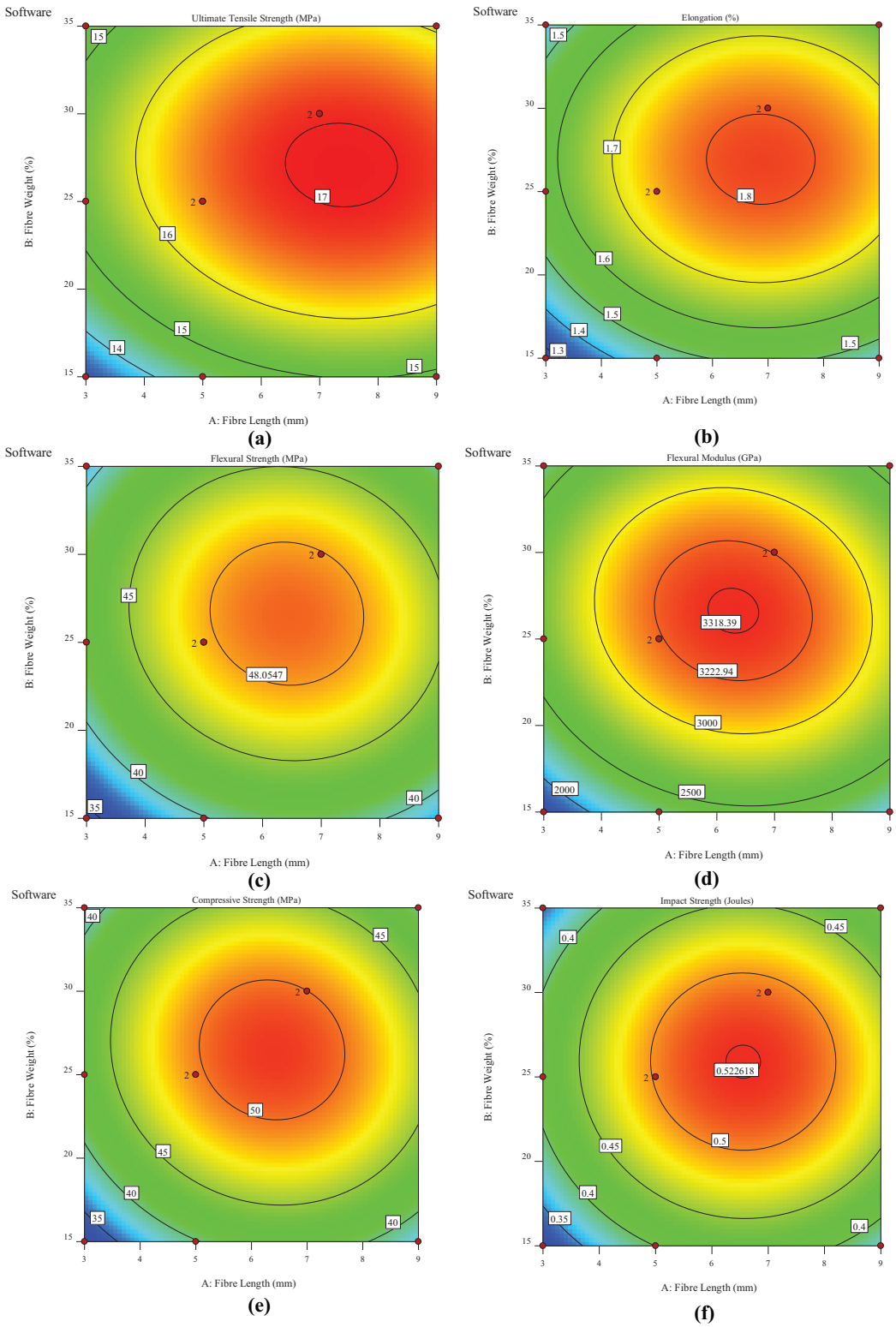


(e)



(f)

Figure 3 Influence of length and fiber weight concentration on (a) tensile strength; (b) elongation; (c) flexural strength; (d) flexural modulus; (e) compression and (f) impact strength



GPa observed for a PMC specimen with alkali-treated fiber 17 mm in length and with 30 wt.% concentration. This is because of the effect of the increased adhesive nature of the PMC matrix leading to improved tensile and flexural properties. It can be attributed to the reinforcement fibers of this trial specimen being optimal and offering transfer of stress between the epoxy matrix and the LSF. At a lower concentration of fibers, the epoxy matrix is not held enough by the LSFs and results in the formation of localized strains at low stress within the matrix (Yaghoobi and Fereidoon, 2019). However, it is observed with increasing fiber concentration, the flexural properties decrease, presumably caused by the entanglement of LSFs in the composite. This is evident in the result of compression strength in Figure 3(e) where the peak compression strength is obtained 52.154 MPa with the same fiber length and concentration. The increasing fiber concentrations improve the ability of the composite to distribute stress evenly leading to increased composite stiffness. Beyond 30 wt.% concentration, the compression strength diminishes because of irregularities in fiber packing and deviant polymer phases. This leads to a reduction in in-plane stiffness and thereby reducing the compression strength thereafter. The impact strength observed in Figure 3(f) is lower when fiber length and concentration are lower and increases with their corresponding increase and reaches a peak value of 0.53 joules at 7 mm and 30% fiber length and weight concentration, respectively. Past these values, the increased matrix–fiber interaction at higher fiber lengths and concentration leads to a reduction of impact strength (Prasad *et al.*, 2021).

3.3 Optimization

The range of input factors for optimal mechanical properties for treated and untreated LSFs are illustrated in Figure 4(a) and (b). The fiber length and concentrations range from 5.97 to 8.86 mm and 26.20% to 15.61%, respectively, for alkali-treated fiber, whereas for untreated fiber, the fiber length and concentrations range from 6 to 8.64 mm and 25.59% to 15.88%, respectively. It was observed that the optimal type of fiber treatment was alkali-treated LSF with 7 mm fiber length and 30% fiber concentration from the optimization.

3.4 Thermal deterioration

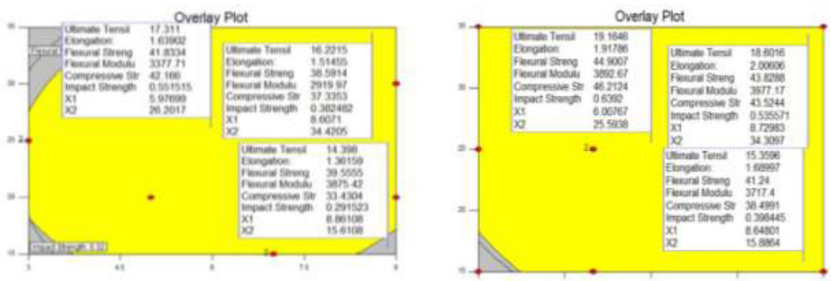
The reduction of the composite material was studied under a gradual increase in material heating. Figure 5 illustrates that the known composite sample weight underwent steady heating, and a subsequent weight loss of 5% was observed at a temperature of 245°C. The steady heating was continued until 800°C where it was observed that when the residual mass was 4.87 wt.%, a massive drop in weight was observed at a temperature of 580°C. It also corresponded to the differential thermal analysis curve. It was observed that the reduction had occurred at the high temperatures illustrating the increased thermal stability. This was caused by the bond between the epoxy matrix and LSFs.

3.5 Microstructural analysis and fractography

The microstructural and fractographical analysis of the treated and untreated LSF PMC specimens can be seen in Figure 6(a)–(h). The micrographs of the specimens are analyzed and characterized for their respective tensile, flexural, compression and impact properties. The surface of the fractured specimen in Figure 6(a) shows good adhesion between the fiber–matrix and resulting fiber bundles being closely stacked. The alkali-treated fiber bundles exhibit better surface adhesion because of the presence of a certain amount of surface roughness. As a result, the LSF induces improved tensile strength in the PMC specimen. The fracture surface observed after the tensile test (ASTM D3039) in Figure 6(b) exhibits the fiber breakage during the test, which apparently illustrates the relatively uneven bonding between the fiber and matrix. The fiber breakage is typically characterized by the kink bands exhibited by singular fibers, and the failures of fiber surfaces were observed to be transverse. The peak ultimate tensile strength and elongation for the treated composite with 7 mm fiber length and 30 wt.% fiber weight were observed to be 17.072 MPa and 1.847%. As reported in a study of *Muntingia calabura* bark fiber composite (Vinod *et al.*, 2021), the treatment of the fibers had led to the removal of moisture content, thereby enhancing the tensile strength of the composite in the present work.

The flexural tested PMC specimens exhibited strong crystalline nature, which correlates with the tensile performance of the fiber–matrix adhesion (Figure 6(c)). The rich matrix adhesion is observed, and the fracture of the fiber observed exhibits longitudinal splitting. In comparison to another *Cordia dichotoma* natural fiber (Madhusudhan Reddy *et al.*, 2018), which exhibited flexural strength of 347 MPa in an epoxy matrix composite, the LSF-reinforced epoxy composite had exhibited 50.4 MPa flexural strength. Although it is low in comparison, it is the highest among the investigated composite

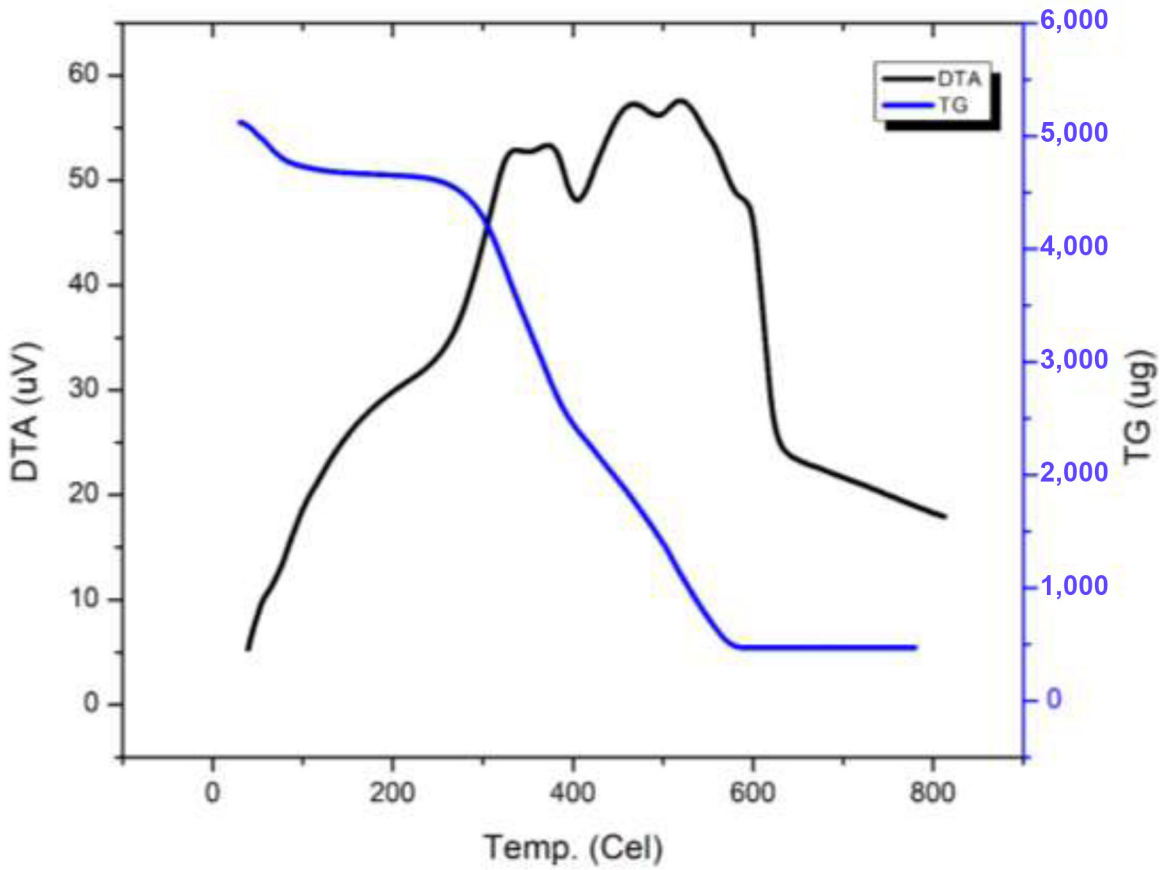
Figure 4 Overlay plot for optimized input factors



Treated PMC materials

Untreated PMC materials

Figure 5 TGA graph



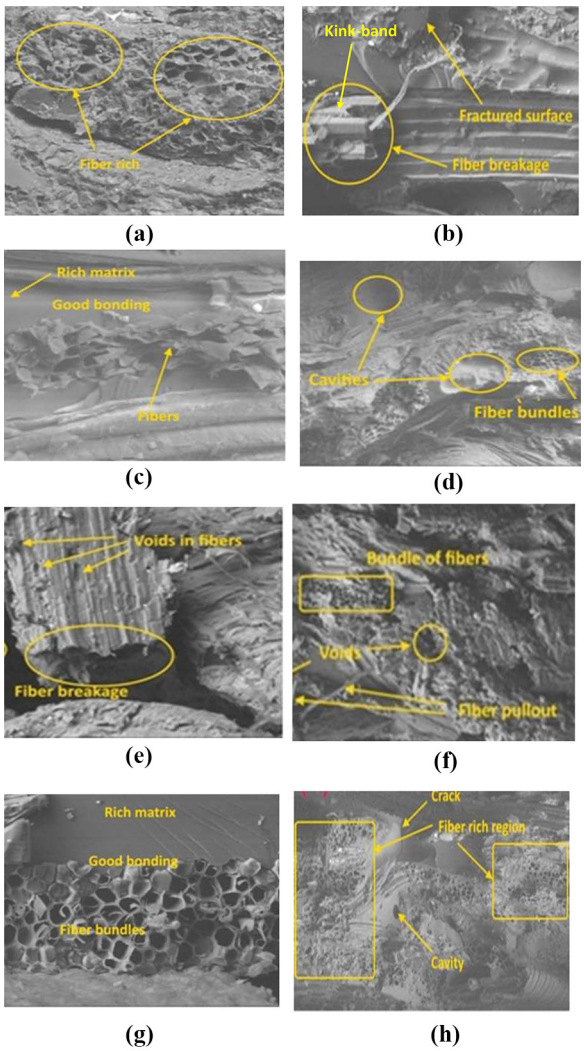
specimens. The untreated LSFs seen in Figure 6(d) exhibit fiber bundles fracture, which is a result of insufficient adhesion. This is also apparent in the formation of cavities within the matrix, where the presence of LSF is naught, thus the applied stress is not transmitted within the fiber reinforcements. In the case of Figure 6(e), the specimen treated exhibits fiber breakage during compression test and diminutive amounts of void defects present are seen. The fracture is linear with kink bands observed in the fibrils indicating compression failure (Jamshaid *et al.*, 2020). The untreated specimen compression fracture in Figure 6(f) highlights the major defects present within the matrix of the PMC. The relatively large void defects are responsible for premature microcracking in the matrix, which combines into splitting failure. The failure observed in alkali-treated LSF impact tested PMC specimen from Figure 6(g) is observed to be similar in failure geometry to the specimen exhibited in Figure 6(a), which is because of the optimal fiber length and concentration achieved during the experimental and optimization trials. The peak impact strength of 0.53 joules was observed for the treated specimen with 7 mm fiber length and 30 wt.% fiber weight. As in the case of an existing study of a *Coccinia indica* fiber-infused epoxy composite (Bhuvaneshwaran *et al.*, 2019), the enhancement of impact strength in the present work is attributed to the greater adhesion between the fiber and the matrix, while the presence of natural fiber at each crack propagation path is responsible for absorbing impact energy. The untreated LSF impact tested PMC specimen in Figure 6(h) exhibits the presence of cavities between the fiber–matrix interface and is the primary cause of the impact failure of the specimen. The impact strength is lower in this case because of the high level of adhesion of fiber–matrix (Azeez *et al.*, 2020).

4. Conclusion

In the present investigation, the optimization of alkali-treated LSF-reinforced PMC was explored. The PMC was subjected to mechanical characterizations to assess the strengthening efficacy of treated LSF reinforcement on epoxy-based composite. Tensile, flexural, compression and impact tests were conducted per ASTM standards and the results were observed. Optimization was performed to identify the optimal input factors in the fabrication of the LSF-reinforced PMC and were compared with the experimental trials to verify the validity of the experimental design. The results obtained from the investigation are:

- The extracted LSFs prior to alkali treatment exhibited higher levels of hemicellulose. This is detrimental to the distribution of stresses within the composite structure. The alkali-treated fibers exhibited much lowered presence of hemicellulose, lignin, wax and improved levels of cellulose, which enables better fiber–matrix bonding during fabrication of composite.

Figure 6 Alkali-treated and untreated specimen of tensile strength, flexural strength and modulus, compression and impact strength



Notes: (a) Tensile alkali-treated PMC specimen; (b) tensile untreated PMC specimen; (c) flexural alkali-treated PMC specimen; (d) flexural untreated PMC specimen; (e) compression alkali-treated PMC specimen; (f) compression untreated PMC specimen; (g) impact alkali-treated PMC specimen; and (h) impact untreated PMC specimen

- The PMC was fabricated based on custom experimental design and involved 19 specimens with varying input factors: LSF length, fiber weight concentration and condition of fiber (treated or untreated). These specimens were subjected to experimental trials to characterize for mechanical properties: ultimate tensile strength, elongation, flexural strength and modulus, compression strength and impact strength.
- The specimen from experimental trial 17 exhibited enhanced mechanical performance such as ultimate tensile strength of 17.072 MPa, elongation of 1.847%, flexural strength of 50.4 MPa, flexural modulus of 3,376.31 GPa, compression strength of 52.154 MPa and impact strength of 0.53 joules. These enhanced properties were observed for input factors: 7 mm fiber length, 30 wt. % concentration of fiber in the composite and condition of fiber was alkali treated.
- Subsequent optimization results indicated concurrence with the experimental results, and the optimized input factors were validated as the input factors of experimental trial 17.
- The TGA analysis revealed the thermal stability of the LSFs is thermally stable up to 245°C.
- The microstructural and fractographical analysis of the tested specimen revealed the significance of fiber concentration in enhancing the strength of the fiber in coordination with the corresponding fiber length. It was also found that higher fiber lengths do not necessarily improve the strength of the fiber as they become susceptible to entanglement, which directly influences the decrease in compression strength of the composite.
- The surface-treated fiber-reinforced composites fared better because of decreased instances of defects such as voids and cracks, whereas the untreated fibers exhibited poor matrix–fiber adhesion, premature fiber bundle breakage and microcracking.

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