








## Research Article

# Sugarcane Bagasse-Derived Activated Carbon- (AC-) Epoxy Vitrimer Biocomposite: Thermomechanical and Self-Healing Performance

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Vitrimeric materials have emerged as fascinating and sustainable materials owing to their malleability, reprocessability, and recyclability. Sustainable vitrimeric materials can be prepared by reinforcing polymeric matrix with bioderived fillers. In the current work, a sustainable vitrimer is prepared by incorporating biomass-derived activated carbon (AC) filler into the epoxy matrix to achieve enhanced thermal and mechanical properties. Thus, prepared biocomposite vitrimers demonstrate a lower-temperature self-healing (70°C for 5 min) via disulfide exchanges, compared to the pristine epoxy vitrimers (80°C for 5 min). Significantly, the self-healing performances have been studied extensively with the flexural studies; and changes in material healing efficiency have been demonstrated based on the observed changes in modulus.

## 1. Introduction

Conventional thermosetting polymers have been applied in various fields like automobile, construction, aviation, wind turbines, and coatings [1]. The last decade has witnessed exciting progress concerning the design and development of structural polymeric materials, necessary to keep up with the challenging tasks that scientists all over the world have

to face for the future. Although many thermosetting composites have been effectively designed to ensure good mechanical performance and self-responsive ability [2–5], the usage of thermoset material is restricted due to their poor reusability and reprocessability. This is due to their extremely high crosslink density, which restricts chain exchanges and therefore decreases malleable nature. On account of this, adequate research on malleable thermoset materials has been carried

out in the last few years [6]. Previously reported reprocessable thermosets *via* Diels-Alder chemistry have demonstrated (dissociative covalent adaptive network) their exchanges in a disruptive way, wherein network crosslink density has changed abruptly [7]. In 2011, Montarnal and coworkers [8] introduced reprocessable thermoset “vitri-mer,” which extended the existing classification, as well as entailed fixed crosslink density via exchangeable covalent adaptive network formations. Owing to fixed network integrity, vitri-mer materials behave like a silica glass former. Depending on the temperature range, they can also manifest the behavior of a viscoelastic fluid. They are characterized by a special viscoelasticity regime, which denotes a topology freezing-point temperature ( $T_v$ ), which can be situated below or above the glass transition temperature ( $T_g$ ). Also, the established  $T_v$  has been extrapolated through Arrhenius and Maxwell equations; hence, it was measured after viscosity reaches  $10^{12}$  Pa·s [8].

Moreover, many thermoset vitrimers have been introduced with different chemistries like transesterification [9], transcarbonylation [10], transamination [11], disulfide linkages [12], and imine exchanges [6]. Transesterification-based vitri-mer system has been majorly discussed (with or without catalyst), due to their simple preparation and effective covalent exchange mechanism [13–15]. The reported malleable thermoset vitrimers are all recyclable/reprocessable, and some of them have exhibited self-healing performances. Vitrimers having intrinsic self-healing are more attractive than those based on extrinsic self-healing mechanisms in which the healing can be activated via covalent network exchanges [16]. In particular, low-temperature self-healing demonstrated disulfide exchange-assisted vitri-mer materials which have been reported adequately, to acquire the processable vitri-mer system with intrinsic self-healing properties [17, 18]. Further, the different chemistry involvement encourages the vitri-mer material with prevailed properties for a real-time application (such as soft electronics, 3D printing, and adhesives) [19, 20].

Since the development of vitri-mer studies, various kinds of fillers have been incorporated in epoxy/vitri-meric matrices, where better thermal and mechanical properties were achieved with effect of nanofiller addition. In particular, the addition of carbon allotrope-based fillers in a vitri-meric system resulted in prominent mechanical and self-healing properties [21]. In recent times, bioderived monomers have been included in vitri-mer studies to prepare sustainable vitri-mer materials [15, 22–24], where like sustainable bio-based polymers have gained huge interest to aspire the green environment [25]. Bio-based vitrimers have been developed from different derivatives like lignin, fructose, and soybean oil [26]. However, their thermal and mechanical performances were found to be comparatively lower than the synthetically prepared vitri-meric materials. To overcome these weaknesses, the addition of fillers into the matrix has been realized and exhibited notable sustainability and improved thermal/mechanical properties. Recently, bio-based epoxy/cashew nutshell liquid vitri-mer was demonstrated with CNTs, where the inclusion of CNT in the vitri-mer was helpful to acclaim NIR light/low-temperature shape recovery and transesterification exchanges. Significantly, an addition of 0.3 wt% CNT

resulted in a prominent shape memory via thermal (at 60°C) and near-infrared (NIR) light [27]. Furthermore, transcarbonation exchange-promoted vitrimers were studied with cellulose paper, where the paper has introduced hydrogen bonds with polycarbonate covalent bonds. Also, the performed vitri-mer composite described the prudent self-healing and shape memory properties [28].

In addition to achieve more sustainable composites, sugarcane bagasse-derived activated carbon/bio-based fillers have been unified in an epoxy vitri-mer system to achieve a low-temperature self-healing material (via the reduction of  $T_g$ ). Inquisitively, sugarcane bagasse-derived activated carbon-(AC-) epoxy vitri-mer biocomposites including their thermo-mechanical and self-healing properties have not been explored yet. In the effort to prepare sustainable bio-based vitrimers, bio-filler involvement in vitrimers has been of great demand and required much attention by the researchers; thus, herein, we present an epoxy vitri-mer biocomposite sugarcane bagasse-derived chemically activated carbon (AC) as biofiller. Owing to this, the biocomposite vitri-mer is designed with aromatic disulfide crosslink-assisted self-healing; also, different concentrations of AC are used to optimize chemical and mechanical properties of epoxy vitri-mer biocomposites. The influence of AC on the glass transition temperature ( $T_g$ ) of the matrix has also been studied. The self-healing efficiency (based on a covalently adaptive network) through flexural studies and active carbon dependent changes on healing and thermo-mechanical performances have been eloquently addressed.

## 2. Experimental Section

**2.1. Material and Methods.** Bisphenol A diglycidyl ether (BADGE) (340.41 g/mol) resin, 40 wt% phosphoric acid (BDH grade), and 2-aminophenyl disulfide (AFD) (248.37 g/mol) hardener were used as received from Sigma-Aldrich.

**2.2. Preparation of Epoxy Vitri-mer Biocomposites.** Activated carbon was prepared from sugarcane refuses, based on the literature (supporting information (SI) (available here)) [29]. The prepared AC was dispersed in ethanol (150 mg in 10 mL) via ultrasonication for 30 min. Subsequently, finely dispersed activated carbon in different loadings (EP-x%; x-0 (p-pristine), 0.1, 0.2, 0.5, 1, and 2) was added in BADGE resin and heated at 80°C under vacuum condition till ethanol evaporation. Thereafter, a stoichiometric ratio of hardener 2-aminophenyl disulfide (AFD) was added and stirred at the same temperature for 15 min. Finally, the degassed mixture was kept in a silicon mold and cured at 150°C for 5 h.

**2.3. Preparation of Conventional Thermoset Epoxy Biocomposite.** To evaluate the effect of AFD incorporation onto self-healing properties, AFD free activated carbon-based conventional thermoset epoxy biocomposites were also prepared. Activated carbon-dispersed ethanol solution was added in BADGE (500 mg, 1.47 mmol) resin at 80°C temperature with stirring. After evaporation of ethanol, weighed DETA (60 mg, 0.5 mmol) was added and stirred at 40°C for 15 min. Then, the mixture was poured in a silicon mold

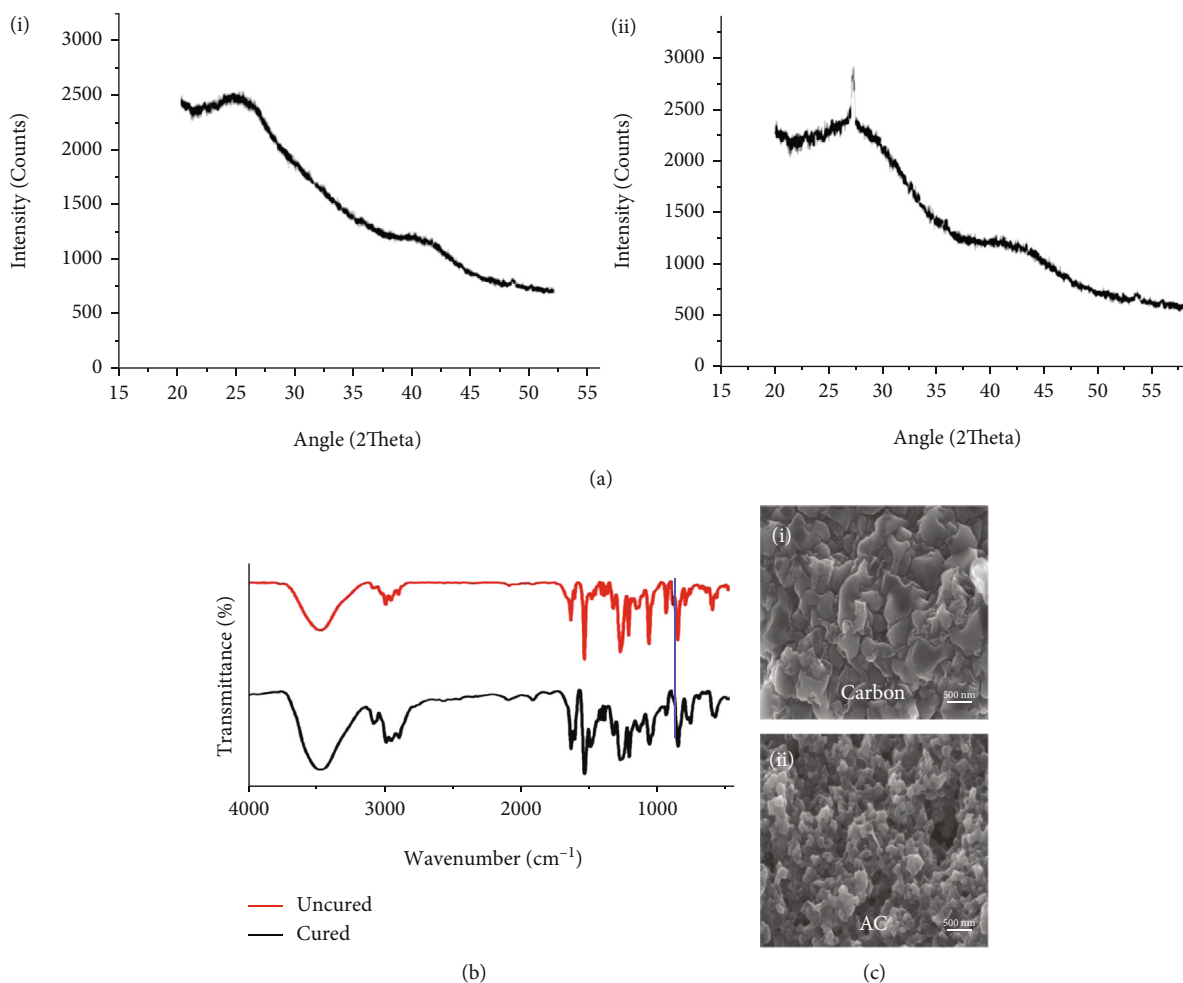


FIGURE 1: (a) XRD pattern of activated carbon (AC). (b) FT-IR spectra for uncured and cured epoxy. (c) SEM images for biomass-derived (i) carbon and (ii) activated carbon.

and kept at room temperature for 24 h. The obtained specimens were involved in different studies.

**2.4. Characterization Techniques.** X-ray diffraction spectroscopy was analyzed through D8 ADVANCE ECO–Bruker to identify the activated carbon. Scanning electron microscopy (SEM; Quanta FEG 200) analysis was used to observe the surface of activated carbon. The epoxy curing was observed through FT-IR (Frontier FT-IR/FIR, PerkinElmer) spectra analysis.  $T_g$  was characterized using thermomechanical analyzer (TA-Q400 EM) dimensional change experiments. In TA-Q400 EM, a three-point bending test was performed to address the storage modulus, loss modulus, stress relaxation, and stress-strain behavior of the rectangular specimens ( $15 \times 5 \times 0.5$  mm). All dimensional change, storage modulus, and loss modulus experiments were performed through temperature ranges from 40°C to 120°C using heating rates of 10°C/min with 50 mL/min nitrogen purge gas flow and 0.02 N force. In the stress relaxation study,  $1 \times 10^{-3}$  N pre-loaded force was applied to straighten the specimen. During the test, the required temperature and 1% strain were applied, and then, relaxation modulus was evaluated with respect to time. Stress-strain experiments were performed

in strain ramp mode with 0.02 N force and evaluated strain at 40°C isothermal temperature.

### 3. Results and Discussions

**3.1. Material Characterization.** Biomass-derived carbon and AC were investigated by XRD analysis to identify their formation from biomass (Figure 1(a), (i) and (ii)). The performed XRD for biomass-derived carbon has resulted in a broad peak around 26°, where the absence of sharp peaks denoted the amorphous structure. Subsequently, the performed AC exhibited that narrow peaks have indicated the regularity of crystalline structure and layer integral formation with adequate surface volume [30, 31]. Altogether, the observed peak at 26.27° is a characteristic of graphite flake-like hexagonal structure, and therefore, AC can also be termed as assemblies of defective graphene [21]. Thus, the chemically activated carbon (comprised of graphite and  $\pi$  electron-connected graphene layer) exhibits an intense porosity, which has an increase in surface area, despite that it deforms the graphene layer structure [32, 33]. SEM analysis shows the sugarcane bagasse-derived carbon and activated carbon surface, in which chemically (phosphoric acid) activated carbon highlights pores on its

TABLE 1: Glass transition temperature of epoxy nanocomposites.

Samples	EP-p	EP-0.1	Epoxy-AC (EP-x)			EP-2
			EP-0.2	EP-0.5	EP-1	
Glass transition temperature ( $T_g$ )	64	63	60	59.2	59	59.8

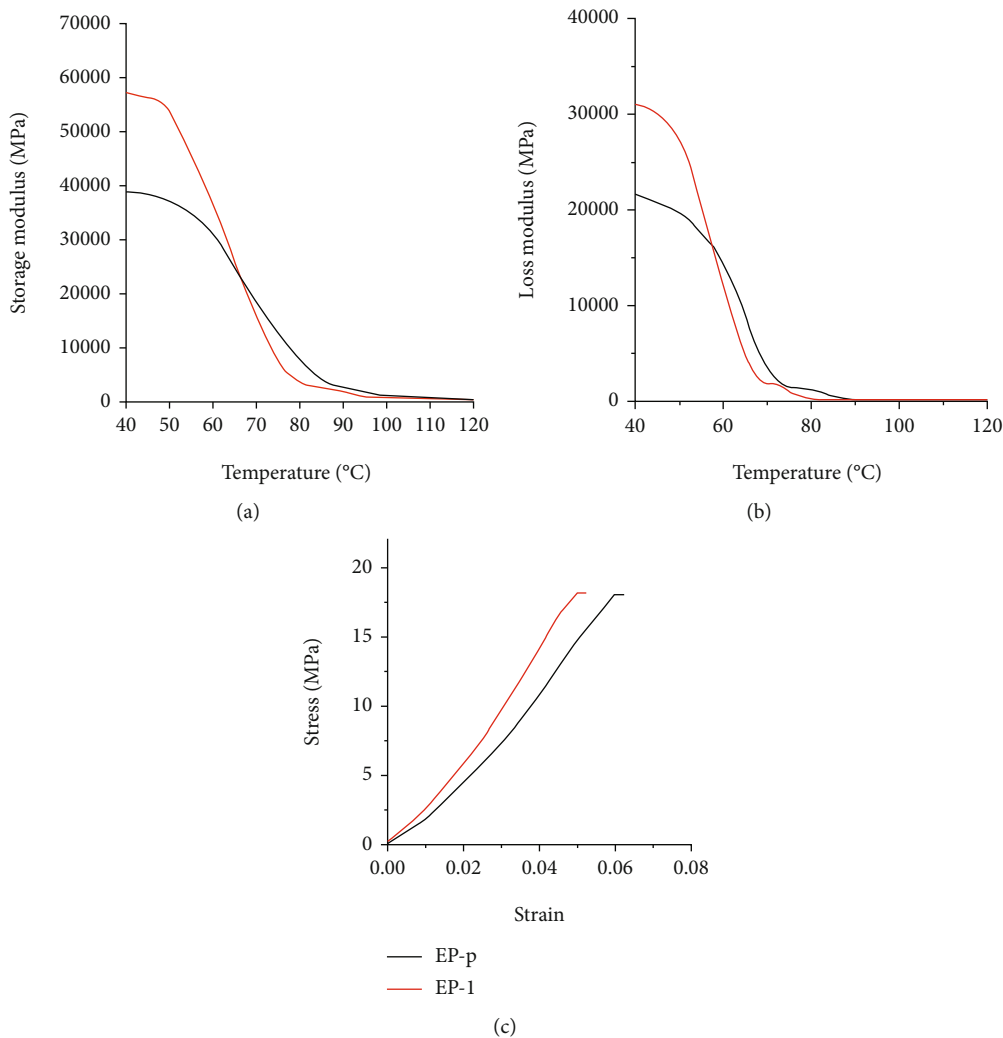


FIGURE 2: (a) Storage modulus, (b) loss modulus, and (c) flexural stress-strain curve for epoxy and biocomposite vitrimers.

surface (Figure 1(c), (i) and (ii)). These low-volume pores confer to AC the capability to extend the surface area for chemical reactions and are helpful to attain progressive chain exchanges. FT-IR analysis was performed to analyze the curing of epoxy vitrimer biocomposites, where the diminution of oxirane ring ( $915\text{ cm}^{-1}$ ) was observed and denoted a complete curing of epoxy (Figure 1(b)) [34]. The epoxy curing was noted at the interval of every one hour using FT-IR analysis, and it was found that the curing was complete after 5 h (see Figure S1).

**3.2. Thermomechanical Properties.** Thermomechanical properties of AC-epoxy vitrimer biocomposites were investigated through a thermomechanical analyzer, which allows the determination of their  $T_g$  and dynamic mechanical perfor-

mance. The determined  $T_g$  for the pristine epoxy vitrimer and the AC-epoxy vitrimer biocomposites loaded with different percentages of AC filler are tabulated in Table 1 (see Figure S2). From the observed results, material EP-1 denotes the lowest  $T_g$  among the investigated epoxy samples. A reduction in  $T_g$  was observed for all biocomposites, which could be due to the free volume space between matrix and nanofillers [35], and thus enhances the chain mobility helpful to achieve low-temperature self-healing properties. However, further addition of filler leads to slight increase in  $T_g$ , which could be due to agglomeration of nanofillers in epoxy matrix [36, 37].

Storage modulus and loss modulus of the pristine epoxy vitrimer and AC-epoxy vitrimer biocomposites were observed to understand the temperature-dependent viscoelasticity

TABLE 2: Dynamic properties of epoxy for epoxy and biocomposite vitrimers.

Samples	Storage modulus (GPa)	Flexural strength (MPa)	Flexural strain at break (mm/mm)	Flexural modulus (GPa)	Activation energy ( $E_a$ ) (kJ/mol)	Topology transition temperature ( $T_v$ ) ( $^{\circ}$ C)	Before healing (GPa)	After healing (GPa)	
								1 <sup>st</sup>	2 <sup>nd</sup>
EP-p	39.7	18.0	0.06	32.1	59	19	32.1	23.4	19.5
EP-1	57.5	18.1	0.05	43.3	79	43	43.3	36.9	33.7

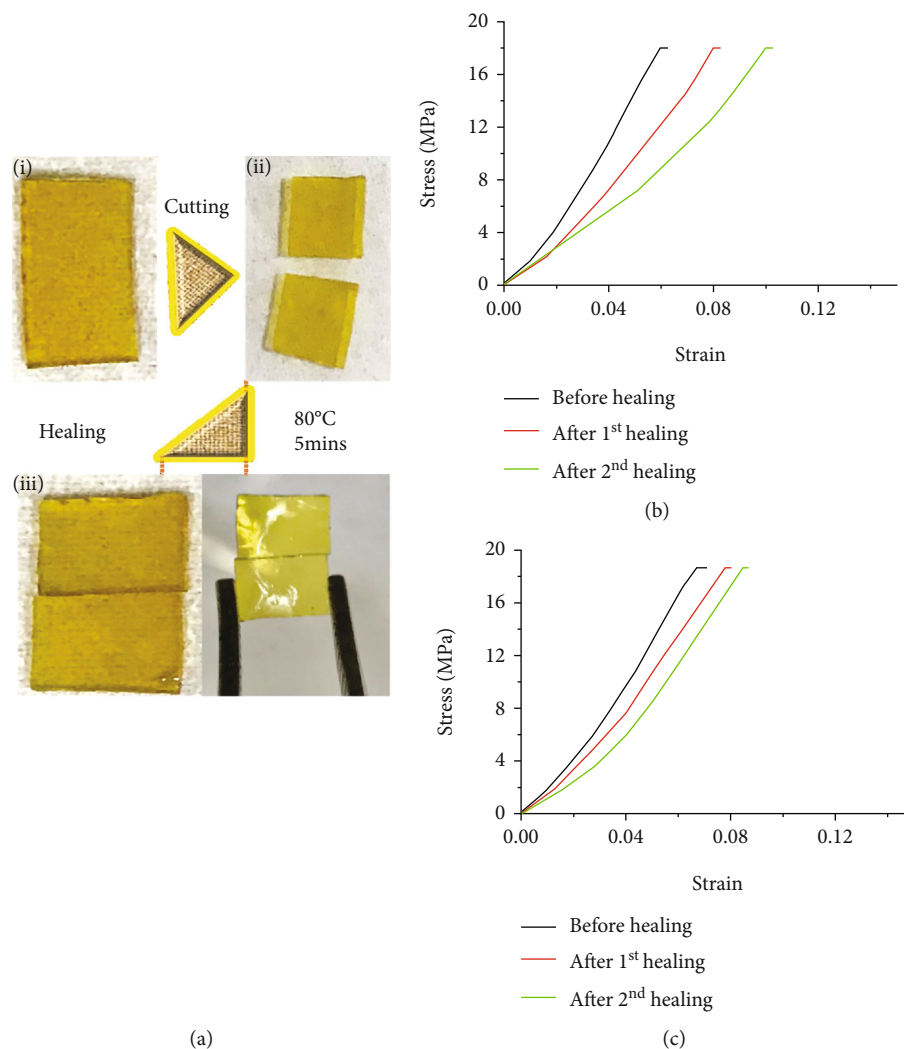


FIGURE 3: (a) Self-healing of epoxy vitrimer: (i) pristine EP-p, (ii) cut into two pieces, and (iii) rejoined. Healing efficiency of vitrimer was calculated via the stress-strain relationship for (b) EP-p and (c) EP-1.

(Figures 2(a) and 2(b)). It was found that biocomposites with 1 wt% of AC exhibit a higher storage modulus than pristine epoxy vitrimer. EP-1 demonstrates a higher storage modulus (Table 2) due to the presence of activated carbon, where matrix-interlocked fillers have restricted the chain exchanges at lower temperature. However, EP-1 has availed a faster reduction in viscoelasticity (above  $T_g$ ), where the addition of AC has reduced the glass transition temperature ( $T_g$ ) of the epoxy vitrimer (due to filler and matrix free volume space). During the period of temperature change, the performed material EP-1 has exhibited a faster decline in storage and loss

modulus than the pristine sample; thus, performed material viscoelasticity (storage and loss modulus) was entirely dependent with the solid to rubbery phase change temperature (i.e., glass transition temperature) [4].

A three-point bending test using the force ramp mode at 40°C was performed for calculating the stress-strain behavior of the prepared samples. The pristine and biocomposite vitrimers demonstrate a similar stress-strain behavior (Figure 2(c)). However, a lower strain at break and a higher flexural modulus of AC-epoxy vitrimer biocomposite compared to pristine epoxy vitrimer denote

the stiffness increment as well as high bending resistance upon the incorporation of AC. A 35% increment in flexural modulus was obtained with the addition of AC nanofillers (Table 2).

**3.3. Stress Relaxation.** Stress relaxation experiments were performed for the EP-p and EP-1 at different temperatures (60°C, 70°C, and 80°C); hence, resulted relaxation times (EP-p = 112.8 s, 40.8 s, and 34 s; EP-1 = 234 s, 129.6 s, and 46.2 s, respectively) were plotted in the Arrhenius equation (Figures S3 and S4) to identify the activation energy of the pristine epoxy vitrimer and its biocomposites (SI) [38].

The investigated relaxation time for EP-p was comparatively lower than that for EP-1. This could be due to the presence of activated carbon, which has restricted the chain mobility and increased the viscosity of epoxy vitrimer. However, free volume formation between filler and matrix was found to be the key component for reduction in glass transition temperature [35].

**3.4. Self-Healing Mechanical Properties.** The performed first healing of pristine epoxy vitrimer and AC-epoxy vitrimer biocomposite demonstrates efficient self-healing via disulfide exchanges (Figure 3(a); video 1). The specimen was cut into two pieces with the help of a razor blade, both the pieces were immediately kept together at 70°C for 5 min, and the healing efficiency was evaluated from the stress-strain relationship. The activated carbon involving EP-1 demonstrates self-healing at 70°C, owing to their low glass transition temperature, as low  $T_g$  is helpful to achieve the S-S bond rearrangement. Disulfide-mediated radical exchange is the key factor for achieving the self-healing behavior for vitrimeric materials [39], as conventional epoxy specimen devoid of 2-aminophenyl disulfide does not show the healing properties. The healing efficiency was evaluated through flexural studies (Figures 3(b) and 3(c)), where healed pristine epoxy vitrimer and AC-epoxy vitrimer biocomposites demonstrate healing efficiency 73% and 85%, respectively, after the first cycle. Then, the performed second healing demonstrates a 60% and 78% healing; a reduction in healing efficiency after each cycle was observed. However, material flexural strength was quite similar after every healing cycle, whereas a change in flexural modulus and strain at break was observed.

## 4. Conclusion

It is concluded that activated carbon from sugarcane bagasse could be an effective filler to achieve sustainable epoxy vitrimer biocomposite. The high surface area of activated carbon in AC-epoxy vitrimer biocomposite makes them capable to achieve progressive chain exchanges. The disulfide exchanges promoted temperature-dependent self-healing observed at 80°C for 5 min in pristine epoxy vitrimer, and the material had demonstrated a lower temperature self-healing at 70°C for 5 min upon the addition of activated carbon. Healing efficiency evaluated via flexural studies highlighted a prominent recovery in vitrimer biocomposites with 1 wt % of AC (EP-1), where 85% and 70% efficiency was exhibited after two consecutive healings. In the future, biocomposite vitri-

mer study would be helpful to envisage efficient vitrimer composite materials for real-time applications with sustainable properties.

## Data Availability

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

## Conflicts of Interest

The authors declare no conflict of interest.

## Acknowledgments

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## Supplementary Materials

Table 1: different nanocomposite percentage prepared with an addition of dispersed solutions (AC). Figure S1: FT-IR results for EP-pristine curing with respect to time. Figure S2: glass transition temperatures for different percentages of activated carbon involved epoxy vitrimer (1-5). Figure S3: Arrhenius equation plotted graph ( $\ln(\tau^*)$  vs.  $1/T$ ) for EP-p. Figure S4: Arrhenius equation plotted graph ( $\ln(\tau^*)$  vs.  $1/T$ ) for EP-1. (*Supplementary Materials*)

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