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# Influences of processing conditions on mechanical properties of recycled epoxy-anhydride vitrimers

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## Abstract

The chemically crosslinked network structures make epoxies, the most common thermosets, unable or hard to be recycled, causing environmental problems and economic losses. Epoxy-based vitrimers, polymer networks deriving from epoxy resins, can be thermally malleable according to bond exchange reactions (BERs), opening the door to recycle epoxy thermosets. Here a series of experiments were carried out to study the effects of processing conditions (such as particle size distributions, temperature, time, and pressure) on recycling of an epoxy-anhydride vitrimer. Polymer powders from the epoxy-anhydride vitrimer with different size distributions were prepared and characterized, and the influence of particle size on the mechanical performance of recycled epoxy-anhydride vitrimers was investigated by dynamic mechanical analysis and uniaxial tensile test. Experimental results demonstrated that finer polymer powders can increase the contacting surfaces of recycled materials and thus result in high quality of recycled materials. In addition, the influences of other treating parameters, such as temperature, time, and pressure, were also discussed in this study. Adjusting these treating parameters can help the design of an optimized reprocessing procedure to meet practical engineering applications.

Biao Zhang and Honggeng Li contributed equally to this work.

## 1 | INTRODUCTION

Epoxyes are the most common thermosets that have been widely used in coating,<sup>[1]</sup> encapsulating for electrical components,<sup>[2]</sup> laminates,<sup>[3]</sup> adhesives,<sup>[4]</sup> and many others.<sup>[5]</sup> The market of epoxyes has been growing rapidly and is anticipated to be \$27.5 billion in 2020.<sup>[6]</sup> However, the chemically crosslinked network structures make epoxyes unable or hard to be recycled, causing environmental problems and economic losses. This view is being changed by a revolutionary group of new polymers—vitrimers, which was firstly named by Leibler and coworkers who developed the epoxy-based covalent adaptable networks (CANs) enabled by the transesterification reaction.<sup>[7]</sup> These vitrimers behave like traditional thermosets at room temperature but are malleable like thermoplastics at high temperatures due to the thermally induced bond exchange reactions (BERs) between hydroxyl and ester groups.<sup>[7,8]</sup> The simultaneous breaking and reconnecting between hydroxyl and ester groups reshuffles the macromolecular chains and essentially allows the polymer network to switch between multiple equilibrium states. Such behavior makes epoxy vitrimers not only maintain the properties of traditional thermosetting polymers, such as excellent mechanical performance, chemical resistance, and thermal stability, but also thermally malleable like thermoplastics. This unique nature of vitrimers opens the door to the exciting possibility of new manufacturing and remanufacturing processes. To date, epoxy vitrimers have been widely used as shape memory polymers,<sup>[9]</sup> matrix materials for composites,<sup>[10]</sup> 3D printing materials,<sup>[11]</sup> and so on.<sup>[8c, 12]</sup>

To well understand the reprocessing and recycling process of epoxy vitrimers, the influences of processing conditions (e.g., time, temperature, and pressure) on the mechanical performance of the recycled vitrimers have been systematically investigated.<sup>[7b, 13]</sup> Yu et al.<sup>[13b]</sup> experimentally studied the reprocessing and recycling ability of an epoxy-acid vitrimer in a pulverous state, and the experimental results indicated that the increase in either processing time or temperature improves the mechanical performance of the recycled samples because of the stress relaxation property of vitrimers caused by the time-temperature dependent BERs.<sup>[13b]</sup> Qi and coworkers<sup>[13]</sup> also found that the increase in pressing pressure results in a larger contact area between powder particles, thus shortens the healing time of the epoxy-acid vitrimer.<sup>[13b]</sup> Besides processing time, temperature, and pressing pressure, the size of particles is also an important parameter that affects the contacting surfaces between polymer particles in pulverous states, and thus determines the mechanical properties of recycled materials. However, to our best

knowledge, there are no systematic studies to investigate the influence of particle sizes on the mechanical performance of recycled vitrimers. Moreover, most studied epoxy-based vitrimers are elastic epoxy-acid vitrimers that prepared by mixing fatty acids and diglycidyl ether of bisphenol A (DGEBA). The relatively low glass transition temperatures ( $T_g$ ) ( $\sim 15^\circ\text{C}$ ) and modulus ( $\sim 3$  MPa) limit their applications as load-bearing materials.

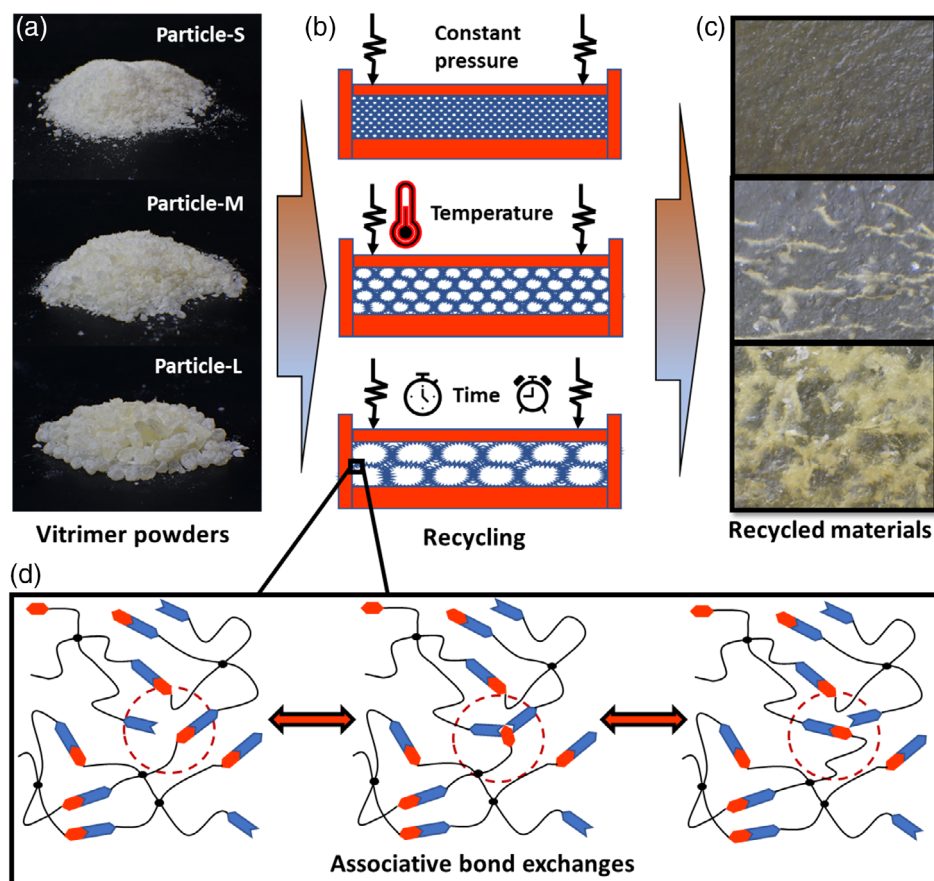
In contrast, Leibler and co-workers developed a hard epoxy-anhydride vitrimer that can be synthesized by crosslinking diglycidyl ether of bisphenol A (DGEBA) and glutaric anhydride. This epoxy-anhydride vitrimer exhibits high  $T_g$  of  $\sim 80^\circ\text{C}$  and room-temperature modulus of  $\sim 1.8$  GPa where the mechanical performance is close to that of conventional commercial epoxy thermosets.<sup>[7b]</sup> However, compared with elastomeric epoxy-acid vitrimers, the investigations about the influences of processing conditions on the mechanical performance of the recycled hard epoxy-anhydride vitrimer are limited due to the difficulty of applying controllable high pressing pressure in a heated environment, and grounding the hard epoxy-anhydride vitrimer into powders with controllable size. The reported work from Li and coworkers just indicated that long grinding time of the hard vitrimer with a ball mill results in finer particles and high healing efficiency.<sup>[13e]</sup> In this work, an epoxy-anhydride vitrimer was synthesized by mixing diglycidyl ether of bisphenol A (DGEBA) and glutaric anhydride with zinc acetylacetonate as the BER catalyst.<sup>[7b]</sup> Powders with different sizes resulting from the synthesized vitrimer (Figure 1a) were treated under different conditions by varying treating pressure, temperature, and time (Figure 1b). The recycled vitrimers with different morphologies (Figure 1c) were prepared due to the triggered associative bond exchanges between ester and hydroxyl groups under different processing conditions (Figure 1d). The particle size distributions of prepared vitrimer powders were analyzed by ImageJ software. The effects of the particle size, treating time, temperature, and pressure on the mechanical performance of recycled epoxy-anhydride vitrimers were systematically studied by dynamic mechanical analysis (DMA) and uniaxial tensile test.

## 2 | EXPERIMENTAL

### 2.1 | Material synthesis

The epoxy-anhydride vitrimer was synthesized according to the method reported by Leibler and coworkers.<sup>[7]</sup> All the chemical products are commercially available and were provided by Sigma-Aldrich in Singapore (Details

**FIGURE 1** (a) The resulting polymer powders (Particle-S, Particle-M, and Particle-L) from epoxy-anhydride vitrimers by using 0.5, 1.0, and 2.0 mm trapezoidal perforation sieve rings, respectively. (b) The treating parameters for recycling materials and (c) the resulting new shapes from epoxy-anhydride vitrimers powders with different particle sizes. (d) The associative bond exchanges between carbonyl and hydroxyl groups [Color figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]



**TABLE 1** Reagents for synthesis of epoxy-anhydride vitrimers

Chemical	Structure	MW (g/mol)	Provider
DGEBA		340.41	Sigma Aldrich
GA		114.10	Sigma Aldrich
Zn(acac) <sub>2</sub>		263.61	Sigma Aldrich

Note: DGEBA, diglycidyl ether of bisphenol A; GA, glutaric anhydride; Zn(acac)<sub>2</sub>, zinc acetylacetonate.

can be seen from Table 1). 5% epoxy equivalents zinc acetylacetonate was first dissolved in diglycidyl ether of bisphenol A (DGEBA) by heating with a heat gun (~130°C) for ~10 min, and then glutaric anhydride was

added. After stirring, a miscible mixture was obtained and was poured into a customized Teflon mold curing for 12 hr at 140°C. The stoichiometry between epoxy and anhydride is 1:0.5.

## 2.2 | Pulverous sample preparation

To obtain polymer particles with controllable size distribution, bulk samples of epoxy-anhydride vitrimer were ground using the Variable Speed Rotor Mill PULVERISETTE 14 (Fritsch, Germany) with different types of trapezoidal perforation sieve rings.

## 2.3 | Preparation of recycled samples

The PRESS (Model 4386, Carver, Inc.) was used to prepare the recycled samples. The machine was first preheated to a target temperature. The polymer powders with different particle size distributions were poured into a customized aluminum mold. Then the mold was transferred into the chamber and heated for a set time under an applied constant pressure. Figure 1c shows healing extent of the recycled vitrimers under different processing conditions. Obviously, temperature, time and pressure promoted material healing extent. The surface morphology of the samples was affected by the surface quality of the molds.

## 2.4 | Dynamic mechanical analysis

Dynamic mechanical analysis (DMA, Q800 DMA, TA Instruments) was used to characterize the thermomechanical properties of recycled materials in the tension film mode. Samples with the dimension of 15 mm × 5 mm × 0.5 mm were tested at a frequency of 1 Hz and an amplitude of 5  $\mu$ m. The temperature was first equilibrated at −50°C for 5 min, and then gradually increased to 120°C at a heating rate of 2°C/min. The glass transition temperatures ( $T_g$ ) were determined from the peak of the  $\tan \delta$ . The value of storage modulus at 30°C was taken as the glass modulus, and the value of storage modulus at 120°C was taken as the rubber modulus. For each test, at least three samples were tested, and the average values were reported.

## 2.5 | Uniaxial tensional tests

To perform the Young's modulus and ultimate tensile strength of recycled epoxy-anhydride vitrimers, Instron machine (Model 5943, Germany) was used to carry out tension tests at room temperature. The loading rate was chosen to be a small value (5% per min for all tests) to minimize viscoelastic effects. The samples were trimmed uniformly into the size of 15 mm × 5 mm × 0.5 mm for

testing. For each tension test, at least three samples were tested, and the average values were reported.

## 3 | RESULTS AND DISCUSSIONS

### 3.1 | Characterization of polymer particles

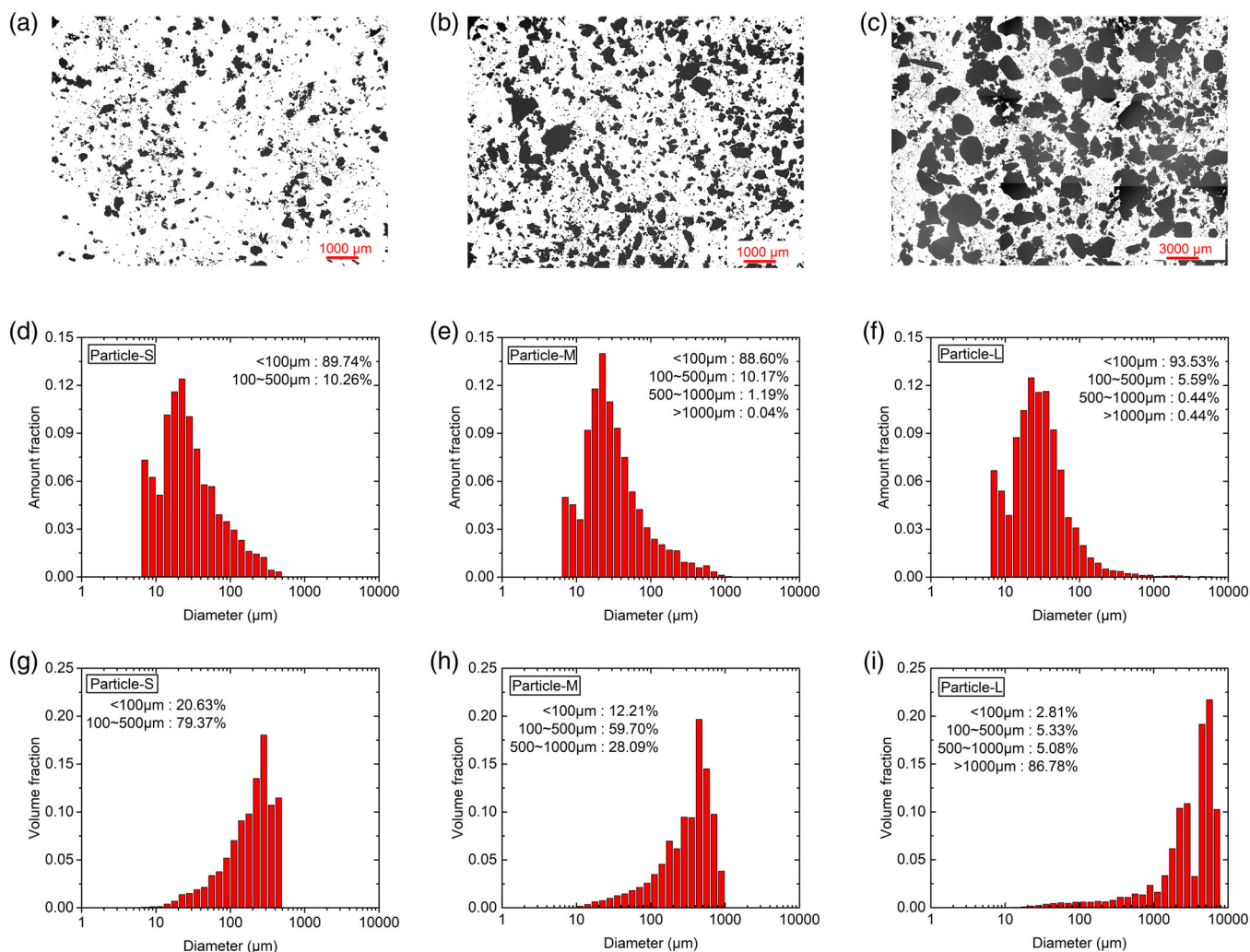
The synthesized epoxy-anhydride vitrimer was ground into powders by using 0.5, 1.0, and 2.0 mm trapezoidal perforation sieve rings, respectively. Figure 2a–c show the microscopic observation of the resulting isolated polymer particles which are all in irregularly shapes.

To quantify the particle size distribution of resulting polymer powders, the commercial image processing software ImageJ, which is a Java-based program developed at the National Institutes of Health (NIH), was used to analyze the resulting microscope images. The outlines of polymer particles were extracted, and the particle diameters were taken to be the diameters of the circle with equal area of each corresponding particle profiles. The analyzed particle size distributions of the polymer powders are shown in Figure 2d–i. In general, the particle sizes become larger with the dimension of used trapezoidal perforation sieve rings. According to the content fraction, almost 90% of the polymer particles obtained by three different trapezoidal perforated sieve rings are less than 100  $\mu$ m in diameter. According to volume fractions, almost 80% of polymer particles with diameters range in 2,000–8,000, 200–1,000, 100–500  $\mu$ m with the size of used trapezoidal perforation sieve rings at 2.0, 1.0, and 0.5 mm, respectively. This result is consistent with the observation from microscopic images that the diameter of the polymer particles increases with the size of used trapezoidal perforation sieve rings. In the following, Particle-L, Particle-M, and Particle-S are used to represent the resulting polymer particles from large to middle and small, respectively.

### 3.2 | Effect of the particle size on recycled thermomechanical properties

As interface contacting is a prerequisite that allows the transesterification exchange reactions happening during the recycling, the size of polymer particles, which can affect the contacting area, is an important factor that decides the mechanical performance of recycled vitrimers. To well understand the effect of particle sizes, the vitrimer powders with different size distributions (i.e., Particle-S, Particle-M, and Particle-L) were thermally treated under a controlled pressure of 5 MPa for





**FIGURE 2** Images of polymer particles (a–c) and the corresponding distributions (d–i) from epoxy-anhydride vitrimers by using 0.5, 1.0, and 2.0 mm trapezoidal perforation sieve rings, respectively [Color figure can be viewed at [wileyonlinelibrary.com](https://onlinelibrary.wiley.com)]

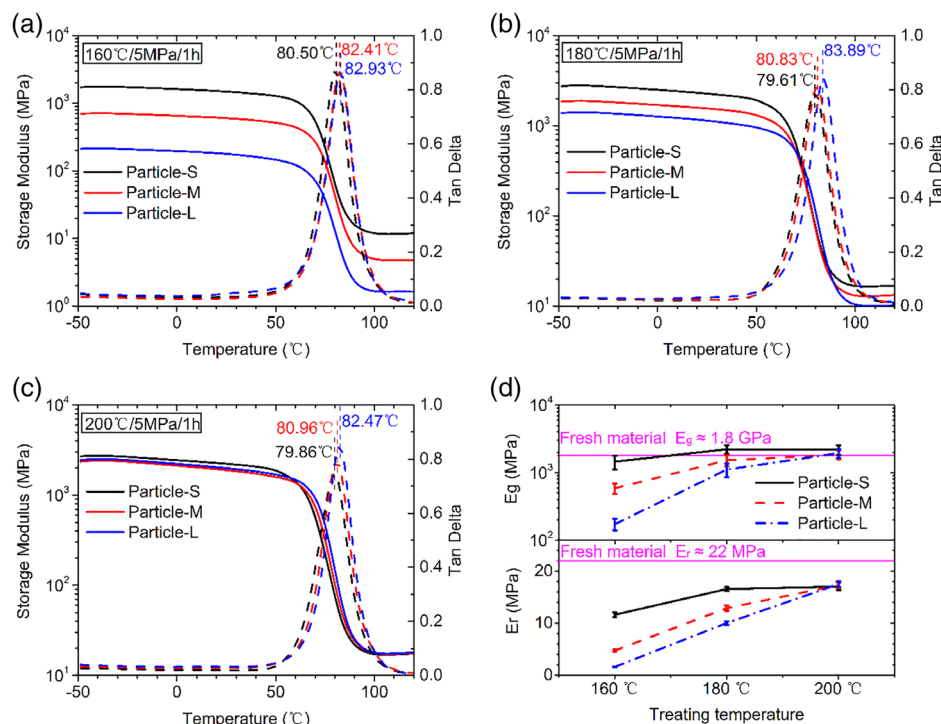
1 hr, and the recycling samples were characterized by dynamic mechanical analysis and uniaxial tensile test. Figure 3a–c show the influence of particle size distributions on the dynamic mechanical behaviors of recycled samples treated at 160, 180, and 200°C, respectively. When the treating temperature is at 160°C, the increase of particle size from small to large results in a gradual decrease of glassy modulus ( $E_g$ ) which is measured at the high modulus plateau where the temperature is far lower than  $T_g$  and the material is at its glassy state.<sup>[14]</sup> (Figure 3a). When raising the treating temperature to 180°C, the difference of glassy moduli from recycled samples with different particle sizes still exists but becomes smaller (Figure 3b). Further increasing the treating temperature to 200°C leads to close DMA curve performance, showing the reach of equilibrium state of all treated samples (Figure 3c), and indicating that increasing the treating temperature to 200°C can help reduce the effect

of the polymer particle size distributions on the thermomechanical properties of the final materials. This is because that the BERs are time–temperature dependent, and the high treating temperature at 200°C results in faster diffusion of reactive polymer chains and BERs between particles, which makes the effect of particle size negligible at the time scale of DMA tests. Overall, these results demonstrate that finer powders are easier to be recycled with the mechanical properties reaching the original materials at a relatively lower temperature. Figure 3d summarizes the effect of particle sizes on the glassy modulus and rubbery modulus. Here, the glassy modulus ( $E_g$ ) is measured from the modulus plateau where the temperature is far below  $T_g$  and the material is at its glassy state; while the rubbery modulus ( $E_r$ ) is measured at the low modulus plateau where the temperature is above  $T_g$  and the material is at its rubbery state. It can be seen that the  $E_r$  of recycled materials also changes

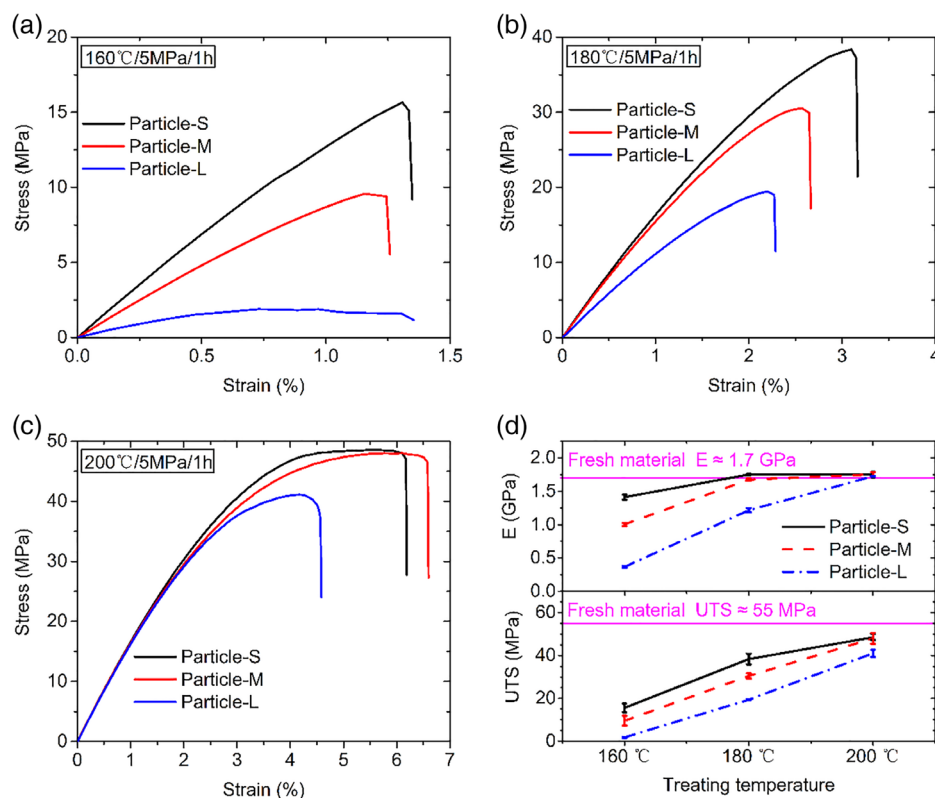
with particle size distributions, showing the similar behavior with  $E_g$ .

Figure 4 shows the uniaxial tensile test results (at room temperature) of recycled materials treated with different heating temperatures. An improved elongation

strength can be obtained with decreasing polymer particle sizes where the ultimate tensile strength (UTS) increases from 2.5 to 13.4 MPa, 19.1 to 38.2 MPa, 41.3 to 48.2 MPa when the fixed processing temperature is 160, 180, and 200°C, respectively. Young's modulus ( $E$ )



**FIGURE 3** Effect of the particle size on thermomechanical performance of recycled epoxy-anhydride vitrimers. (a–c) DMA curves of recycled epoxy-anhydride vitrimers from polymer powders with different size distributions (i.e., Particle-S, Particle-M, and Particle-L) at 160, 180, and 200°C, respectively. (d) The effect of particle sizes on glassy modulus ( $E_g$ ) and rubbery modulus ( $E_r$ ). Solid line, storage modulus; dash line: rubbery modulus. All the samples were prepared under the same controlled pressure for 1 hr [Color figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]



**FIGURE 4** Effect of the particle size on mechanical performance of recycled epoxy-anhydride vitrimers. (a–c) Uniaxial tensile test results of recycled epoxy-anhydride vitrimers from polymer powders with different size distributions (i.e., Particle-S, Particle-M, and Particle-L) at 160, 180, and 200°C, respectively. (d) The effect of particle sizes on Young's modulus and ultimate tensile strength.  $E$ , Young's modulus; UTS, ultimate tensile strength. All the samples were prepared under the same controlled pressure for 1 hr [Color figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]

decreases with polymer particle size distributions when the treating temperature is 160 or 180°C. When the treating temperature is elevated to 200°C, the values of  $E$  are not affected by the particle sizes and maintain at around 1.7 GPa. Figure 4d summarizes the tendencies of  $E$  and  $UTS$  of recycled vitrimers with particles size distributions, showing that the behaviors of  $E$  and  $UTS$  are consistent with the behaviors of  $E_g$  and  $E_r$  (shown in Figure 3).

In short, two common trends can be concluded: (a) the decrease of particle sizes leads to the increase of the modulus of the recycled sample, and the  $E_g$  of the samples recycled with Particle-S could almost recover to the one of a fresh sample. (b) The increase of treating temperature can reduce the effect of particle size on the mechanical properties of recycled materials. The reasons leading to the two trends are (a) the smaller particle size leads to more interfacial bonds which facilitate the welding between particles and therefore result in high modulus of recycled samples. (b) The BER is time–temperature dependent. When the treating temperature increases, the reaction proceeds faster and more particles can bond together.

### 3.3 | Effect of heating temperature

The influence of heating temperature on the recycling of epoxy-anhydride vitrimers was also studied. The polymer powders were thermally treated for 1 hr under a controlled pressure of 5 MPa at 160, 180, and 200°C, respectively. Figure 5a–c shows the influence of treating

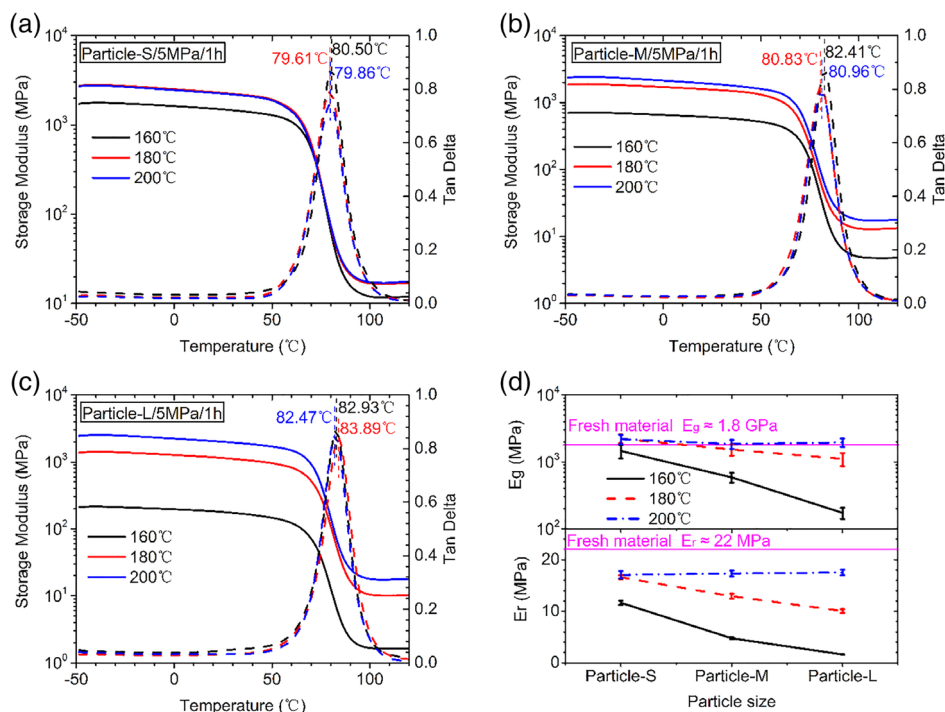
temperatures on the DMA curves of recycled samples from polymer powders with different size distributions. When the recycled samples are from Particle-S, elevating treating temperature from 160 to 180°C results in the increase of  $E_g$  and  $E_r$  from 1.46 GPa and 11.7 MPa to 2.19 GPa and 17.1 MPa, respectively (Figure 5a). After 1 hr of treatment at 5 MPa, the recovery efficiency of Particle-S at 200°C is similar to the recovery efficiency at 180°C. For the recycled samples prepared with Particle-M and Particle-L, both  $E_g$  and  $E_r$  are increased with the treating temperature (Figure 5b,c). Figure 5d summarizes the effect of treating temperature on the  $E_g$  and  $E_r$  of recycled vitrimers. 1 hr heating at 200°C can recover  $E_g$  of recycled samples close to the fresh material, even the particle size is large. Meanwhile, the  $E_r$  of the recycled sample can recover ~80% of its fresh material when Particle-S was treated under 200°C.

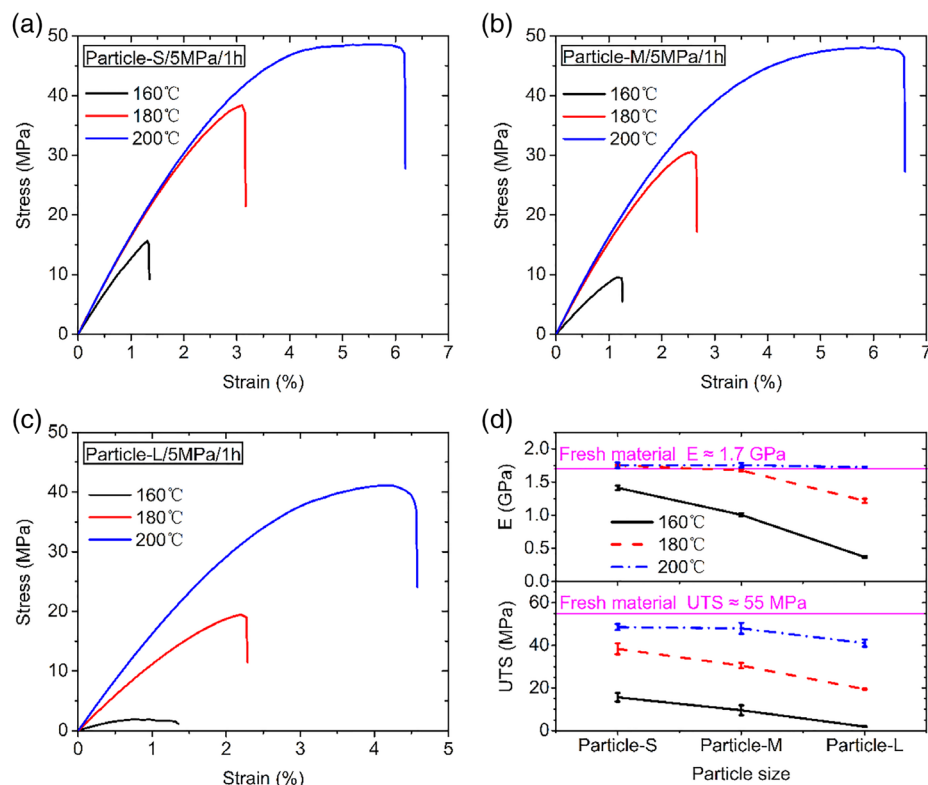
Besides, uniaxial tensile tests were performed to study the influence of the treating temperature on the recycled materials (Figure 6).  $E$  and  $UTS$  of recycled vitrimers increase with heating temperature, showing the similar behavior with  $E_g$  and  $E_r$  (shown in Figure 5). Furthermore, Figure 6 also indicates that the finer particles are helpful for recycling process when other conditions are the same.

### 3.4 | Effect of heating time

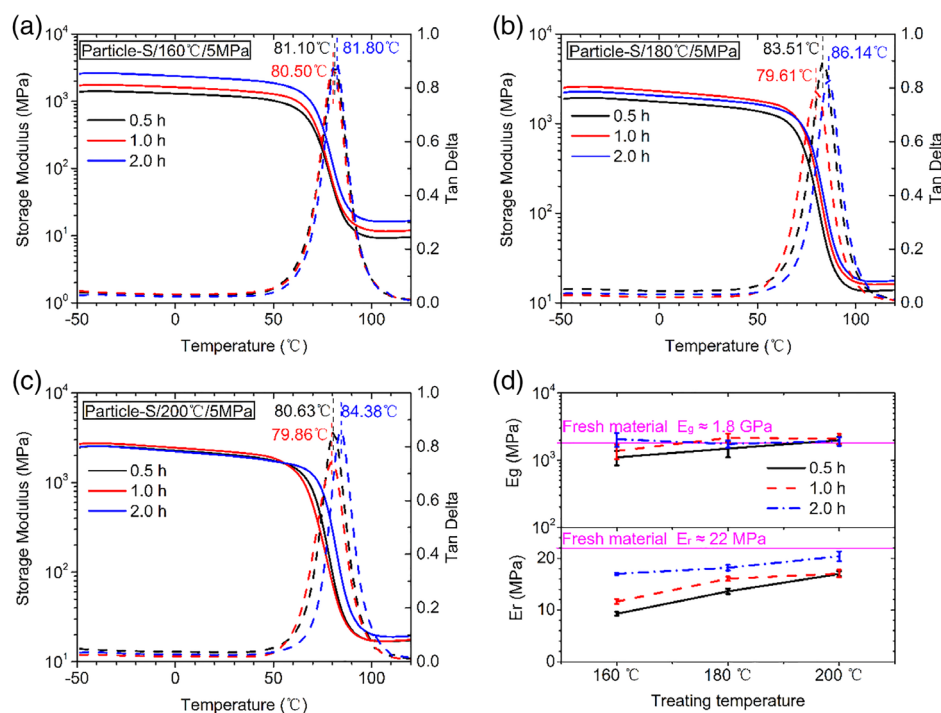
The effect of heating time on thermomechanical performance of recycled materials is demonstrated in Figures 7

**FIGURE 5** Effect of heating temperature on thermomechanical performance of recycled epoxy-anhydride vitrimers. (a–c) DMA curves of recycled epoxy-anhydride vitrimers treated at 160, 180, and 200°C from polymer powders with different size distributions (i.e., Particle-S, Particle-M, and Particle-L). (d) The effect of treating temperature on glassy modulus ( $E_g$ ) and rubbery modulus ( $E_r$ ). Solid line, storage modulus; dash line, rubbery modulus. All the samples were prepared under the same controlled pressure for 1 hr [Color figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]





**FIGURE 6** Effect of heating temperature on mechanical performance of recycled epoxy-anhydride vitrimers. (a–c) Uniaxial tensile test results of recycled epoxy-anhydride vitrimers treated at 160, 180, and 200°C from polymer powders with different size distributions (i.e., Particle-S, Particle-M, and Particle-L). (d) The effect of treating temperature on Young's modulus and ultimate tensile strength.  $E$ , Young's modulus; UTS, ultimate tensile strength. All the samples were prepared under the same controlled pressure for 1 h [Color figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]



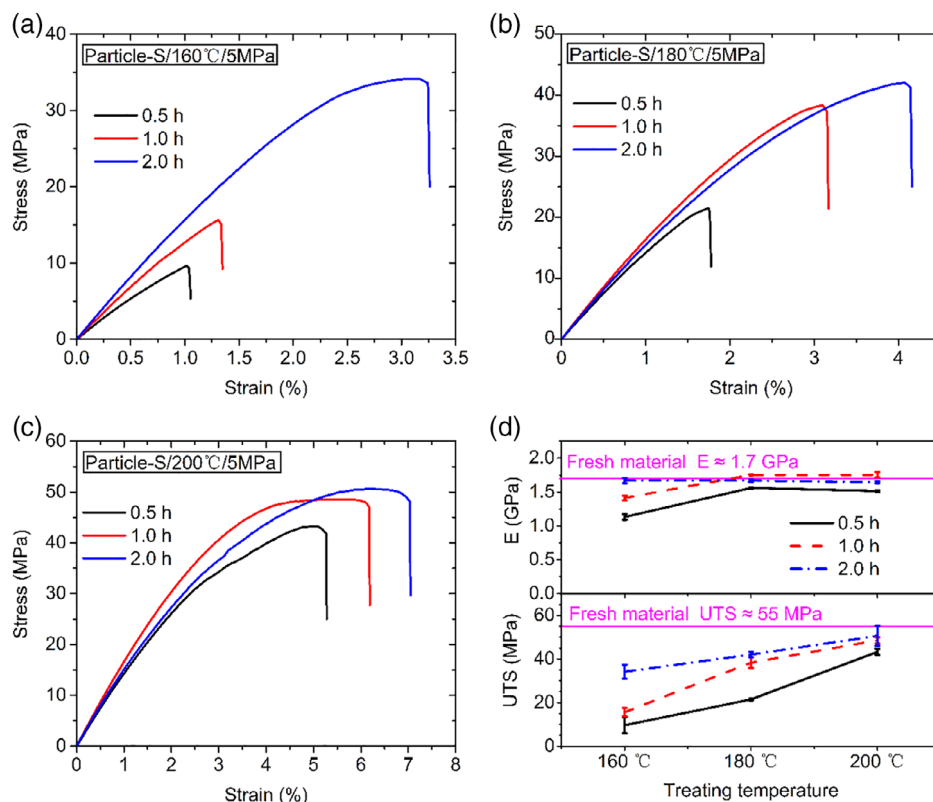
**FIGURE 7** Effect of heating time on thermomechanical performance of recycled epoxy-anhydride vitrimers. (a–c) DMA curves of recycled epoxy-anhydride vitrimers treated at 160, 180, and 200°C from Particle-S. (d) The effect of treating time on glassy modulus ( $E_g$ ) and rubbery modulus ( $E_r$ ). Solid line, storage modulus; dash line, rubbery modulus. All the samples were prepared under the same controlled pressure [Color figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]

and 8. Here, the recycled samples were thermally treated by using Particle-S under a controlled pressure of 5 MPa for 0.5, 1, and 2 hr, respectively. Figure 7a–c show the storage and loss modulus of recycled samples with time, where the samples are treated at 160, 180, and 200°C,

respectively. When the treating temperature is at 160°C, the increase of the heating time from 0.5 to 2 hr results in a gradual increase of  $E_g$  and  $E_r$  from 1.10 GPa and 9.3 MPa to 2.05 GPa and 17.0 MPa, respectively. When the treating temperature was increased to 180°C or



**FIGURE 8** Effect of heating time on mechanical performance of recycled epoxy-anhydride vitrimers. (a–c) Uniaxial tensile test results of recycled epoxy-anhydride vitrimers treated at 160, 180, and 200°C from Particle-S. (d) The effect of treating time on Young's modulus and ultimate tensile strength.  $E$ , Young's modulus;  $UTS$ , ultimate tensile strength. All the samples were prepared under the same controlled pressure [Color figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]



200°C, the  $E_g$  is no longer subjected to the treating time, and all the values maintain at around 1.8 GPa, closing to that of original sample, while  $E_r$  still demonstrates a growing trend with heating time. When the recycled samples were treated at 180°C, the  $E_r$  increased from 13.6 to 18.2 MPa with the heating time extending from 0.5 to 2 hr. Figure 7d summarizes the effect of heating time on  $E_g$  and  $E_r$  of recycled epoxy-anhydride vitrimers.

To further study the influence of heating time on the mechanical performance of recycled epoxy-anhydride vitrimers, uniaxial tension tests were carried out and the results are shown in Figure 8. The elongation strength improves with increasing heating time where the  $UTS$  increases from 10.2 to 34 MPa, 22 to 38.2 MPa, and 44.6 to 50.2 MPa when the fixed processing temperature is 160, 180, and 200°C, respectively. When the treating temperature keeps at 160°C,  $E$  increases with heating time from 1.17 to 1.64 GPa. However, when the treating temperature elevated to 180 or 200°C, the value of  $E$  reaches to around 1.7 GPa, which is not affected by heating time, showing the similar behavior with  $E_g$  shown in Figure 7.

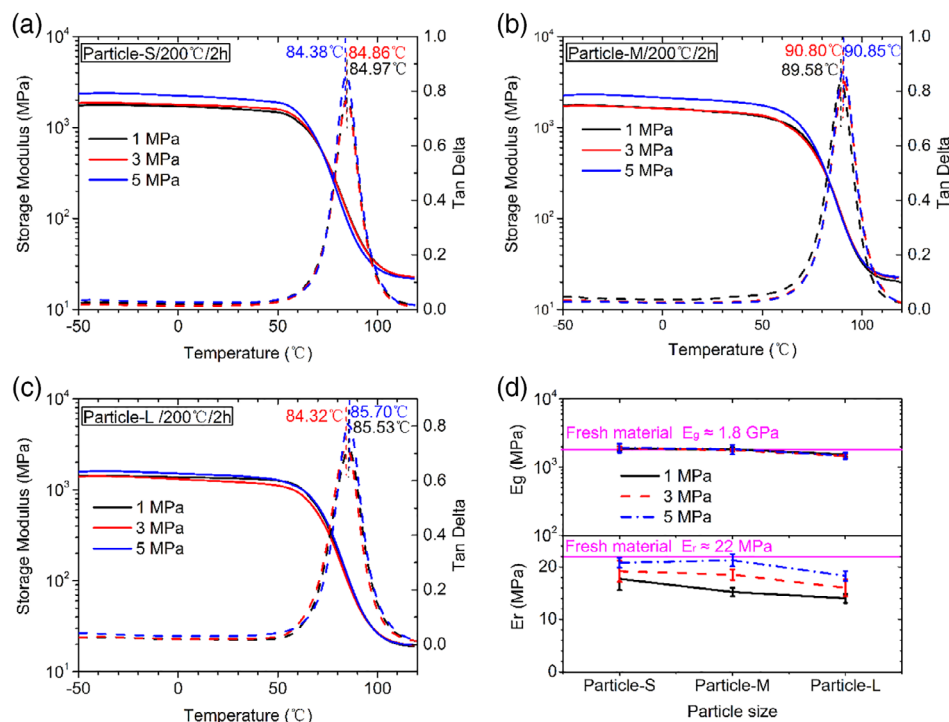
The similar tendency of mechanical performance with increasing heating time and temperature can be explained by that the interfacial welding of vitrimers follows the Arrhenius-type time and temperature superimpositions (TTSP).<sup>[7]</sup> Both time and temperature can affect the extent of BERs on the interfaces of vitrimer particles. The interfacial chain density increases with time or

temperature, and thus leads to the increase of modulus and strength of recycled samples.

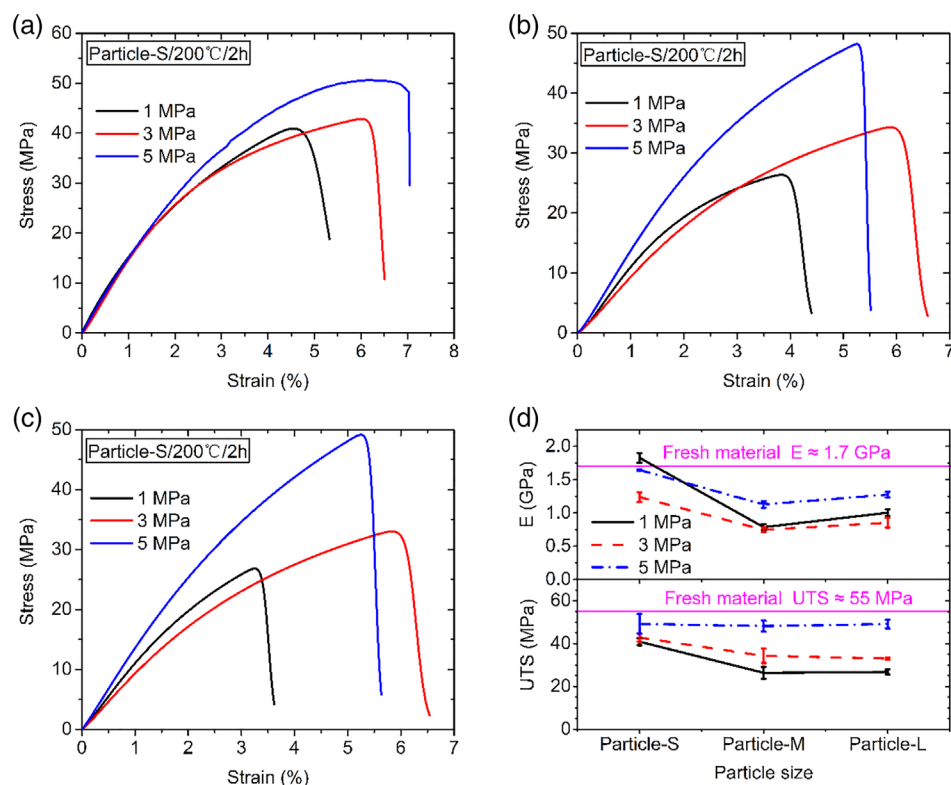
### 3.5 | Effect of pressure

Previous work by Yu et al. demonstrated that pressure can affect the welding efficiency of epoxy-acid vitrimers.<sup>[13b]</sup> To well know the effect of pressure on the recycling process of epoxy-anhydride vitrimers, the vitrimer powders with different particles sizes were thermally treated for 2 hr under different pressures at 200°C, respectively. Figure 9a–c demonstrates the storage and loss modulus of recycled samples with time, where the samples are treated under 1, 3, and 5 MPa, respectively. It is seen from figures that  $E_g$  is around 1.8 GPa, which is almost unaffected by the treating pressures and particle sizes, while  $E_r$  of recycled materials gradually increases from 14.3 to 20.4 MPa when the treating pressure arises from 1 to 5 MPa with the used polymer particles changing from Particle-L to Particle-S. The tendencies of  $E_g$  and  $E_r$  with pressures and particle sizes are summarized in Figure 9d.

The influence of pressure on the mechanical performance of recycled epoxy-anhydride vitrimers was further studied by uniaxial tensile test. As shown in Figure 10, an improved elongation strength can be obtained with the decrease of polymer particle sizes.  $UTS$  increases



**FIGURE 9** Effect of the pressure on thermomechanical performance of recycled epoxy-anhydride vitrimers. (a–c) DMA curves of recycled epoxy-anhydride vitrimers from polymer powders with different size distributions (i.e., Particle-S, Particle-M, and Particle-L) treated under 1, 3, and 5 MPa, respectively. (d) The effect of treating pressure and particle sizes on glassy modulus ( $E_g$ ) and rubbery modulus ( $E_r$ ). Solid line, storage modulus; dash line, rubbery modulus. All the samples were treated for 2 hr at 200°C [Color figure can be viewed at [wileyonlinelibrary.com](https://onlinelibrary.wiley.com)]



**FIGURE 10** Effect of the particle size on mechanical performance of recycled epoxy-anhydride vitrimers. (a–c) Uniaxial tensile test results of recycled epoxy-anhydride vitrimers from polymer powders with different size distributions (i.e., Particle-S, Particle-M, and Particle-L) treated under 1, 3, and 5 MPa, respectively. (d) The effect of pressure and particle sizes on Young's modulus and ultimate tensile strength.  $E$ , Young's modulus; UTS, ultimate tensile strength. All the samples were treated for 2 hr at 200°C [Color figure can be viewed at [wileyonlinelibrary.com](https://onlinelibrary.wiley.com)]

from 26 to 42 MPa, 34 to 43 MPa, 51 to 54 MPa when the fixed processing pressure is 1, 3, and 5 MPa, respectively.  $E$  of recycled samples from Particle-S shows higher values compared with that of samples from Particle-M and Particle-L. Figure 10d summarizes the tendencies of  $E$  and UTS with polymer particles sizes and treating

pressures. Generally, the higher treating pressures and smaller polymer particle sizes contribute to higher Young's modulus of recycled vitrimers. Such phenomenon can be explained by the good interfacial contact caused by smaller polymer particles and higher treating pressure during the welding.<sup>[13a]</sup>

## 4 | CONCLUSIONS

In summary, we systematically investigated the effects of treating parameters, such as particle size distributions, treating time, temperature, and pressure, on the mechanical properties of recycled epoxy-anhydride vitrimers. The recovery of  $E_g$ ,  $E_r$ ,  $E$ , and  $UTS$  was investigated to assess the efficiency of the recycling process. Experimental results indicated that an acceptable recovery of mechanical properties requires the lowest particle size coupled to the highest values of temperature, time, and pressure. When the recycled sample from Particle-S was treated at 200°C under 5 MPa for 2 hr,  $E_g$  and  $E$  of recycled sample is close to the fresh material. Meanwhile,  $UTS$  and  $E_r$  can recover ~95 and ~80% of its fresh material, respectively. Especially, the effect of the particle size distribution is remarkable. Finer polymer powders can increase the contacting areas of recycled materials and thus reduce the requirements of other processing conditions. When the particle size keeps the same, enough thermal processing pressures with long treating time and high treating temperatures are needed to induce higher modulus and strength. Adjusting these treating parameters can help the design of an optimized reprocessing procedure to meet the practical engineering applications.

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## CONFLICT OF INTEREST

The authors declare no competing financial interest.

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