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# Recycling of carbon fiber-reinforced epoxy resin composite via a novel acetic acid swelling technology

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#### ABSTRACT

Compared with thermal or chemical recycling, the mechanical recycling of the carbon fiber-reinforced polymer (CFRP) is simpler, more economical, and produces less environmental pollution. However, the recycled products of mechanical recycling are only fillers or short fibers with poor reinforcing properties. In this study, acetic acid was used to achieve the full swelling and slight degradation of cured epoxy resin (CEP) in CFRP. The stiff CFRP became soft in the acetic acid medium at 140 °C, and it delaminated into soft single CFRP layers at 160 °C-220 °C within 1 h. Thus, the swollen CFRP can be easily cut into long strips, large thin slices, or other customized shapes. The length and strength of the carbon fibers were preserved well during the reusing process. The shear and delamination products exhibited excellent reinforcing properties, and they can be prepared into new CFRP products through hot pressing. The flexural strength of a newly prepared CFRP board can reach 47%–89% that of the original CFRP board. The swelling and degradation characteristics and mechanism of CEP in acetic acid were also investigated in this study. Thus, this work provides a novel method for recycling thermosetting composites.

#### 1. Introduction

Given its light weight, high strength, and good corrosion resistance [1,2], carbon fiber-reinforced polymer (CFRP) has been widely used in aircraft, wind turbines, aerospace products, automotive vehicles, and sporting goods [3–5]. The extensive application of CFRP will lead to a high amount of waste CFRP products, expired prepregs, and leftover materials. Global CFRP waste is projected to increase to 20 kt annually by 2025 [6]. Improperly treated CFRP waste may occupy large areas of land and cause environmental pollution. Moreover, the production of CFRP waste can considerably reduce production cost and save energy [7]. Thus, the recycling of CFRP waste has become an important challenge toward attaining environmental protection and social sustainable development [8].

Thermosetting polymers (e.g., epoxy and unsaturated polyester) are mostly used as matrices for CFRP structures. They provide CFRP with excellent mechanical strength and chemical stability. Emerging technologies have focused on recovering long, high-modulus carbon fibers because this form of CFRP is the most valuable. However, the irreversible cross-linked structure of thermosetting polymers cannot be melted, remolded, or reprocessed, making the recovery of CFRP extremely difficult [2]. In general, three CFRP recovery approaches are used at present: thermal processing [9–11], chemical recycling [12–14], and mechanical recycling [15–18].

Long valuable carbon fibers can be recovered from waste CFRP via thermal processing or chemical recycling. However, a major proportion of the cured epoxy resin (CEP) content of CFRP is degraded into lowerweight molecules. The components of the degradation products of epoxy resin exhibit a complex structure and are difficult to recover. These degradation products contain a variety of toxic aromatic hydrocarbons and phenols. Thus, they should be treated or recycled properly to avoid environmental pollution. Mechanical recycling mostly involves grinding CFRP into small pieces or powder through shredding, grinding, or milling. Thus, the mechanical method is simple, inexpensive, and can avoid environmental pollution caused by waste chemical reagents or the degradation products of thermosetting polymers. However, the length and tensile strength of carbon fibers cannot be retained well during mechanical recycling [2]. The products of mechanical recycling are fillers or short fibers with poor reinforcing properties. Thus, the value of mechanically recycled products can be significantly improved if the length and strength of the original carbon fibers can be retained well

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#### during a mechanical process.

Although CEP is difficult to dissolve using common chemical reagents, it can be swelled to a certain extent by some organic solvents. The weight increase of CEP in acetic acid (180 °C, 6 h) is more than 56%, which is considerably higher than those of methanol (18.79%), ethanol (20.85%), n-propanol (16.16%), isopropanol (11.06%), and water (3.80%) [19]. The swelling phenomenon of CEP enables the free migration of the catalyst in the epoxy network, significantly increasing resin degradation rate. In our recent studies, we found that the swelling phenomenon is not only involved in the catalytic degradation of CEP, but it also significantly changes the physical properties of CFRP. After acid swelling treatment, the softening phenomenon acetic (100 °C-140 °C) and automatic delamination (160 °C-220 °C) occurred successively as swelling temperature increased. Swelling products were easy to cut into long strips or other shapes. Acetic acid swelling treatment exerts insignificant effects on the tensile strength and tensile modulus of recycled carbon fibers [19]. Moreover, new CFRP products can be obtained via hot pressing when a new resin matrix is added to the sheared or layered products. All the carbon fibers and most of the CEP (undegraded) content of CFRP can be synchronously recovered using this method.

In the current study, a novel practical and environmentally friendly process for recycling CFRP was developed on the basis of the acetic acid swelling method. The objectives of this study were as follows (1) to evaluate the swelling and degradation characteristics of CEP in acetic acid, (2) to disscuss the possible swelling and degradation mechanisms of epoxy resin in acetic acid, (3) to analyze the mechanical characteristic of the swelling residues, and (4) to examine the flexural strength of the newly prepared CFRP boards.

#### 2. Experimental section

#### 2.1. Materials

CFRP prepreg (WP-3011, 0.2 mm thick) that contained 60 wt% carbon fiber woven cloth (weave angle:  $0/90^{\circ}$ ) and 40 wt% bisphenol A type epoxy resin (6508 epoxy resin, dicyandiamide curing agent) was purchased from Shandong Weihai Guangwei Carbon Fiber Co. Ltd. (China). The same uncured 6508 epoxy resin (already mixed with dicyandiamide curing agent) used in the CFRP prepreg was also purchased from Shandong Weihai Guangwei Carbon Fiber Co. Ltd. (China). The chemical reagents used in the experiments were of analytical grade unless otherwise stated.

#### 2.2. Preparation process of CFRP boards

CFRP prepreg was cut into small pieces (80 mm  $\times$  10 mm  $\times$  0.2 mm). Eight layers of prepregs were bonded together to prepare a CFRP board (~2 g, 80 mm  $\times$  10 mm  $\times$  2 mm) via hot pressing (cured at 140 °C for 180 min). Molding pressure was ~0.10 MPa.

#### 2.3. Acetic acid swelling recycling process

The detailed procedure of acetic acid swelling recycling is illustrated in Fig. 1. Swelling treatments were performed using a 100 mL highpressure reactor. In a typical swelling treatment, one CFRP board and 45 mL of acetic acid were placed into the reactor and then the reactor was sealed. The CFRP board was completely immersed in acetic acid. The examined temperatures were as follows: 100 °C, 120 °C, 140 °C, 160 °C, 180 °C, 200 °C, and 220 °C. The corresponding pressure ranged from 0.07 MPa to 1.03 MPa. Approximately 60 min was consumed to achieve the set temperature and maintained it for 0.5, 1, 2, and 4 h. After the swelling treatments, the reactor was allowed to naturally to room temperature. The swelling products were then removed from the autoclave.

After the swelling treatment, the original stiff CFRP board became soft (140 °C) and automatically delaminated (160 °C-220 °C) within 1 h. The utilization methods of the softening and delaminated products differed. The softening products obtained at 140 °C were first cut into long strips (80 mm  $\times$  2 mm  $\times$  2 mm) and then dried at 105 °C for 24 h. A certain amount of epoxy resin (~0.50 g) was added to the long strips, and a new CFRP board (80 mm  $\times$  10 mm  $\times$  2 mm) was prepared via hot pressing (cured at 140 °C for 180 min, and molding pressure was 0.10 MPa). The delaminated products obtained at 160 °C-220 °C were first dried at 105 °C for 24 h and then mixed with epoxy resin (~1.50 g) to prepare a new CFRP board (80 mm  $\times$  10 mm  $\times$  2 mm) via hot pressing (cured at 140 °C for 180 min, and molding pressure was 0.10 MPa). The newly added epoxy resin was used to fill the cracks of the swelling products during hot pressing.

#### 2.4. Analysis

The swelling ratio (S) and mass loss rate (D) of CEP in acetic acid were calculated using the following equations:

$$S(\%) = \frac{M_1 - M_2}{M_0 * M_r - M_3} \times 100\%$$

$$M_3 = M_0 - M_2$$
(1)



Fig. 1. Detailed procedure of acetic acid swelling recycling.

$$D(\%) = \frac{M_3}{M_0 * M_r} \times 100\%$$
 (2)

where  $M_0$  is the initial mass of the CFRP board before swelling treatment,  $M_1$  is the mass of the CFRP board after swelling treatment (wet weight, including the sweller),  $M_2$  is the remaining mass of the dried CFRP board after swelling treatment,  $M_r$  is the mass fraction of the epoxy resin in the initial CFRP (40%), and  $M_3$  is the mass reduction of the CFRP board before and after swelling treatment. After swelling treatment, the ratio of the acetic acid mass absorbed by the CFRP board to the residual resin mass was used to reflect the approximate swelling degree of CEP. However, the excessive swelling and slight degradation of CEP led to the formation of cracks in the CFRP board, and many carbon fibers were exposed. The cracks and exposed carbon fibers may adsorb a certain amount of acetic acid, and thus, the measured swelling ratio was higher than the actual swelling degree of CEP, particularly at high temperatures (160 °C-220 °C).

The flexural strength of the CFRP board was calculated via a threepoint bending test, and loading speed was 2 mm/min. Flexural strength was calculated using the following equation:

$$\sigma = \frac{3PL}{2bh^2} \tag{3}$$

where P is the measured load; L is the span; and b and h are the width and thickness of the CFRP board, respectively. The surface microstructure of the swelling product was examined via scanning electron microscopy (SEM) using a Zeiss Supra 40 scanning electron microscope (Germany). The primary chemical structure of the resin products was characterized via Fourier transform infrared spectroscopy (FTIR) using Shimadzu AIM-9000 FTIR microscope system (Japan).

#### 3. Results and discussion

#### 3.1. Swelling and degradation characteristics of CEP in acetic acid

Fig. 2a shows the effects of temperature on swelling ratio and mass loss rate when the added amount of acetic acid and swelling time were fixed at 45 mL and 1 h, respectively. Swelling ratio and mass loss rate increased rapidly with an increase in temperature. Swelling ratio was only 30.74%–60.85%, and no evident degradation phenomenon occurred at  $\leq$  120 °C. The CFRP board remained stiff at 120 °C. Swelling ratio increased to 106.2% at 140 °C, and CEP began to degrade (mass loss rate: 5%). The stiff CFRP board became soft at 140 °C. Swelling ratio and mass loss rate increased to 145.88% and 13.11% at 160 °C, respectively. CFRP was dissociated into eight layers under the combination of the swelling action and the degradation effect. Swelling ratio and mass loss rate reached the maximum values of 239.21% and 23.33% at 220 °C, respectively, with an increase in temperature.

Temperature exerted an important influence on swelling ratio and mass loss rate. High temperatures can increase the diffusion rate of acetic acid in CEP. When the swelling temperature was raised, swelling ratio and amount of solvent absorbed at equilibrium increased. In accordance with bond–energy theory, when temperature is sufficiently high, the acetic acid swelling environment can provide sufficient energy to break polymer bonds [20]. Therefore, raising the temperature can increase swelling ratio and mass loss rate.

Fig. 2b depicts the effects of swelling time on swelling ratio and mass loss rate when the added amount of acetic acid was fixed at 45 mL. Swelling ratio and mass loss rate increased with the extension of swelling time at different treatment temperatures. Swelling ratio was low at 120 °C. When swelling time was extended to 4 h, swelling ratio was only 94.13%. Meanwhile, mass loss rate can be disregarded in the first 2 h, and it increased to 2.54% at 4 h. The CFRP board remained hard after 2 h of swelling treatment, and it began to soften at 4 h. Swelling ratio ranged from 100.93% (0.5 h) to 144.62% (4 h) at 140 °C, while mass loss rate ranged from 0.93% (0.5 h) to 10.59% (4 h). The CFRP board treated at 140 °C for 1 h became soft, and partial dissociation of the CFRP board was observed when swelling time was extended to 4 h. The CFRP board quickly became soft after 0.5 h of swelling treatment at 160 °C, and four CFRP layers fell off from the board. Complete dissociation of the CFRP board was achieved within 1 h at a swelling ratio and mass loss rate of 146.21% and 13.26%, respectively. In addition, CEP is a typical thermosetting resin that can only swell to a certain extent. Apart from the swelling effect, acetic acid also exhibits a certain catalytic degradation effect on CEP [21]. The degradation of CEP can further destroy the internal network structure of the resin and accelerate the diffusion rate of acetic acid in the resin. Thus, the degradation of CEP can enhance the swelling effect.

#### 3.2. Possible swelling and degradation mechanism of CEP in acetic acid

Solvent molecules can weaken and replace polymer–polymer interactions with polymer–solvent interactions [22,23]. Moreover, the degree of swelling is maximized for a polymer network in solvents with similar solubility parameters [22,24]. The Hansen solubility parameter ( $\delta$ ) is a 3D solubility parameter that depends on dispersion force, polar (dipole–dipole) bonding, and hydrogen bonding. The solubility parameters of epoxy resin, acetic acid, methanol, ethanol, *n*-propanol, and water were 19.85–20.46 MPa<sup>1/2</sup>, 21.4 MPa<sup>1/2</sup>, 29.6 MPa<sup>1/2</sup>, 26.5 MPa<sup>1/2</sup>, 24.35 MPa<sup>1/2</sup>, and 47.8MPa<sup>1/2</sup>, respectively [25]. Among these common solvents, the solubility parameter of acetic acid (21.4 MPa<sup>1/2</sup>) was closest to that of epoxy resin (19.85–20.46 MPa<sup>1/2</sup>). Acetic acid is a polar solvent with carbonyl (C=O) and hydroxy (C–OH) moieties. CEP is a cross-linked polymer that is largely composed of C–O–C, C–C, and C–N bonds. Hydrogen bond formation between CEP and acetic acid is one of the important causes of resin swelling. The availability of four



Fig. 2. Effects of different factors on swelling ratio and mass loss rate: (a) temperature and (b) swelling time.

hydrogen donor sites makes acetic acid a good sweller due to the formation of a hydrogen bond between CEP and acetic acid (Fig. 3) [26,27]. The simple fundamentals of chemistry may help explain the swelling mechanism, but the exact mechanism of interaction must be explored further.

Fig. 4 shows the FTIR results of the virgin CEP and the swelling products (220 °C, 1 h). For the swelling products, the remarkable reduction in peak intensity for the C–N bond at 1420 cm<sup>-1</sup> and the C–O–C bond at 1031 cm<sup>-1</sup> indicated the cleavage of the C–N bond and C–O–C bond during swelling treatment. The chemical bond energy of the C–N bond (305 kJ/mol) and C–O–C bond (326 kJ/mol) was lower than those of the C–C bond (332 kJ/mol), Ph–O bond (358 kJ/mol), Ph–CH<sub>2</sub>Ph bond (378.2 kJ/mol), and Ph–CH(CH<sub>3</sub>)<sub>2</sub> bond (413.8 kJ/mol) [28]. Thus, the cleavage of the C–N bond and C–O–C bond was prior to those of the other chemical bonds and resulted in the destruction of the 3D network structure of the resin. In addition, acetic acid can provide an external source of hydronium to reinforce the acid–base catalysis degradation effect on CEP [21,28].

#### 3.3. Mechanical characteristic analysis of swelling products

Fig. 5a presents a comparison of the thicknesses of the swelling products obtained at different temperatures when holding time was fixed at 1 h. The swelling phenomenon of CEP caused the gap between each carbon fiber layer to become larger at high temperatures. Thus, the thickness of the swelling products increased with an increase in swelling temperature. The thickness of the original CFRP board was  $\sim$ 2 mm. When temperature increased from 120 °C to 140 °C, thickness rapidly increased from 0.23 mm to 0.31 mm. The gap between each carbon fiber layer was evident, and the swelling products can be cut into long strips. The thickness of the CFRP board increased from 0.38 mm at 160 °C to 0.42 mm at 220 °C. When temperature was higher than 160 °C, the CFRP board can be dissociated into eight layers. The swollen single CFRP layer was quite soft. A single CFRP layer still exhibited certain elasticity even after drying, and it can be easily bent or cut.

Fig. 5b shows the load–deformation comparison images of the swelling products (without drying) obtained at different temperatures. One end of the carbon fiber plate was fixed, while a 200 g weight was suspended at the other end. The original CFRP board was sufficiently stiff, and no bending occurred under the action of an external force. The CFRP board could be slightly bent at 120 °C, and the bending angle was  $\sim$ 5°. When temperature increased to 140 °C, the bending deformation degree of the CFRP board increased and the bending angle was  $\sim$ 50°. This swelling phenomenon indicated the rapid penetration of acetic acid



Fig. 4. FTIR spectra of (a) virgin CEP and (b) swelling products obtained at 220  $^\circ\text{C}.$ 

into CEP. When temperature was higher than 160  $^{\circ}$ C, the CFRP board was dissociated into eight single layers, and the bending deformation degree of the CFRP board reached its maximum value.

Fig. 6 shows the SEM images of the original CFRP board and the swelling products obtained at different temperatures. Fig. 6a presents the SEM image of the original CFRP board. All the carbon fibers were covered by CEP, and the surface of the composite material was smooth and complete. At 140 °C, part of CEP was peeled off from the surface of the carbon fibers. Long strips of resin fell off from the gap between adjacent carbon fibers. This phenomenon indicated that the excessive expansion of CEP during the swelling process led to a decrease in adhesion between the resin and carbon fibers. Meanwhile, the excessive swelling and slight degradation of CEP may cause its fragmentation [26]. Therefore, the stiff CFRP became soft and freely bent at 140 °C. Swelling ratio and mass loss rate gradually increased with an increase in temperature. More resin was peeled off from the surface of the carbon fibers, leading to the delamination of the CFRP board into eight layers at 160 °C. The mass loss rate of CEP in the current study was attributed to two reasons: (1) mass loss was caused by the falling-off phenomenon of CEP from the CFRP board, and (2) mass loss was caused by the slight degradation of CEP. The minute CEP particles were precipitated out of the CFRP board into the acetic acid solution. These particles can be recovered through filtration and recycled as fillers or fuels. The resin degradation products included aromatic hydrocarbons, such as bisphenol A, phenol, or isopropyl phenol. These resin degradation products can be removed from an acetic acid solution via an organic solvent



Fig. 3. Hydrogen bond interactions resulting in the swelling of CEP in acetic acid.



(a)



(b)

Fig. 5. Pictures of the swelling products obtained at different temperatures: (a) thickness comparison pictures and (b) deformation comparison pictures under a constant load.

extraction method.

## 3.4. Flexural strength of the swelling products and the re-prepared CFRP board $\$

The soft CFRP board treated at 100 °C-140 °C (without delamination) possibly became hard again when acetic acid gradually evaporated from the swollen resin. The flexural strength of the treated CFRP board (after drying) is shown in Fig. 7a. The flexural strength of the original CFRP board was 604.61 MPa. When temperature increased from 100 °C to 140 °C, the flexural strength decreased from 484.22 MPa to 205.57 MPa. Research has shown that an acetic acid swelling system (180 °C, 6h) exerts an insignificant effect on the tensile strength of carbon fibers [19]. Possible reasons for the decrease in flexural strength were as follows. (1) The excessive expansion and shrinkage of CEP during the swelling and drying processes led to the formation of cracks in CEP. (2) The overexpansion of CEP caused the reduction in adhesion between the resin and carbon fibers. (3) A larger amount of CEP acting as matrices was degraded at high temperatures. Thus, the flexural strength of the swollen CFRP board decreased rapidly with an increase in temperature.



Fig. 6. SEM images of the original CFRP and the swelling products obtained at different temperatures: (a) original CFRP, (b) 140 °C, (c) 160 °C, and (d) 220 °C.



Fig. 7. Flexural strength of (a) swelling products after drying and (b) re-prepared CFRP board.

When the temperature was  $\geq$ 160 °C, the CFRP board automatically delaminated into eight layers. A single CFRP layer exhibited certain elasticity and can be bent freely. Thus, flexural strength was extremely low and can be disregarded.

After 1 h of swelling treatment, the CFRP board remained stiff and hard to cut at  $\leq 120$  °C. A longer time (more than 4 h) was necessary to acquire a soft CFRP at low temperatures. Considering energy consumption, the swelling products obtained at 140 °C-220 °C were used to prepare a new CFRP board via hot pressing. Many cracks were formed during the swelling process due to the excessive expansion and slight degradation of CEP. Uncured epoxy resin in liquid form was mixed with the swelling products. The epoxy resin in liquid form can permeate into the swelling products through these cracks under the action of hot pressing. Meanwhile, the liquid epoxy resin was gradually cured. Thus, the shear products or delaminated products can be bonded together by the newly added epoxy resin after hot pressing. Fig. 7b illustrates the flexural strength of the newly prepared CFRP board. The flexural strength of this board (made from 140 °C swelling products) was only

282.28 MPa, which was 47% that of the original CFRP board (604.61 MPa). At a low temperature (140 °C), the excessive expansion and shrinkage of CEP during the swelling and drying processes caused an evident reduction in adhesion between the resin and carbon fibers. Meanwhile, the excessive expansion of CEP also resulted in the formation of cracks in CEP. The two reasons led to the low flexural strength of the swelling product (after drying), i.e., only 205.57 MPa. In addition, the mass loss rate of CEP was only 5% at 140  $^\circ\text{C},$  indicating that the cracks between the carbon fibers were small. The uncured epoxy resin in liquid form cannot penetrate into the interior of the shear products through these small cracks. Instead, a large proportion of the newly added epoxy resin only adhered onto the surface of these shear products. Thus, the flexural strength of the newly prepared CFRP boards was not high at lower temperatures. At high temperature (160 °C-220 °C), the swelling ratio and mass loss rate of CEP increased rapidly. A higher amount of CEP was peeled off from the CFRP board or degraded by acetic acid. Consequently, more cracks were formed in the CFRP and the flexural strength of the swelling products was extremely low. However,

the newly added epoxy resin can easily penetrate into the interior of the delamination products to compensate for the flexural strength loss caused by the swelling or degradation of CEP. The flexural strength of the newly prepared CFRP board quickly increased to 388.89 MPa at 160 °C, reaching the maximum value at 220 °C (537.94 MPa), which was 89% that of the original CFRP board. Carbon fibers exhibited high thermal and chemical stability. The experimental results indicated that the length and strength of the carbon fibers can be preserved well during the mild reusing process.

#### 3.5. Energy consumption of the swelling treatment

In the current study, waste CFRP boards were immersed in acetic acid medium at 140 °C-220 °C for 1 h. The energy consumption of the swelling treatment was used to heat the acetic acid, carbon fibers and epoxy resin. The corresponding specific heat capacity was  $2.08 \times 10^3$  J/(kg.°C),  $0.7-0.9 \times 10^3$  J/(kg.°C) and  $1.0-2.0 \times 10^3$  J/(kg.°C), respectively. The solid-to-liquid ratio was 1:25, and 1 kg of CFRP contained 0.4 kg of epoxy resin and 0.6 kg of carbon fibers. The energy required to treat 1 kg of CFRP via the acetic acid swelling method was as follows:

 $\begin{array}{l} E_a \,=\, C_a \,\times\, M_a \,\times\, (T{-}T_0) \,=\, 2.08 \,\times\, 10^{3} \,\, J/(kg.^\circ C) \,\times\, 25 \,\, kg \,\times\, (120{-}200 \,\,^\circ C) \,=\, 6.2 \,\times\, 10^{3}{-}10.4 \,\times\, 10^{3} \,\, \text{KJ}, \end{array}$ 

 $E_c=C_c\times M_c\times (T-T_0)=0.8\times 10^3$  J/(kg.°C)  $\times$  0.6 kg  $\times$  (120–200 °C) = 57.6–96.0 KJ,

 $E_e = C_e \times M_e \times (T-T_0) = 1.5 \times 10^3$  J/(kg.°C)  $\times$  0.4 kg  $\times$  (120–200 °C) = 72.0–120.0 KJ,

Energy consumption per 1 kg of CFRP =  $E_a + E_c + E_e = 6.3 \times 10^3 {-} 10.6 \times 10^3$  KJ.

In the preceding expressions,  $E_a$ ,  $E_c$ , and  $E_e$  respectively denote the energy consumption of acetic acid, carbon fibers, and epoxy resin;  $C_a$ ,  $C_c$ , and  $C_e$  indicate the specific heat capacity of acetic acid, carbon fibers, and epoxy resin, respectively;  $M_a$ ,  $M_c$ , and  $M_e$  represent the mass of acetic acid, carbon fibers, and epoxy resin, respectively; T is the swelling

temperature (140 °C-220 °C); and  $T_0$  is the initial temperature (20 °C).

The calculation results of energy consumption indicated that energy consumption was largely attributed to the heating of acetic acid while the energy consumption of carbon fibers and epoxy resin can be disregarded. Continuous swelling treatment can be applied to the actual process to reduce energy consumption. Once a batch of swelling treatment was completed, the swelling CFRP was removed from the reactor and another batch of CFRP was placed in the swelling reactor. Thus, the swelling treatment system was only required to supply a small amount of energy to heat epoxy resin and carbon fibers. In addition, the softening and delaminated products contained a certain amount of acetic acid that should be recovered using evaporation and condensing methods. The recovery of acetic acid was helpful in reducing treatment cost and inhibiting the effect of acetic acid on the strength of the newly prepared CFRP board.

#### 3.6. Different reuse methods of CFRP swelling products

Different reuse methods of CFRP swelling products are presented in Fig. 8. The reuse method primarily depended on the size of CFRP waste. For small CFRP waste, such as carbon fiber fishing rods, bicycles, and tennis racquets, the swelling products can be cut into long strips. Then, the shear products were used to make new CFRP boards or other structural components via hot pressing, as shown in Fig. 8a. Such reuse method is simple, but it may consume more epoxy resin to fill the gaps in the disordered shear products. A high amount of epoxy resin in CFRP materials may decrease the flexural strength of the new CFRP product. For larger CFRP waste, such as carbon fiber wind blades, aircraft wings, and racing boats, the swelling product was suitable for delaminating into large single CFRP layers. Then, these layers were cut into rectangular or round sheets or ribbons in accordance with the shape of the final product. Many single CFRP sheets can be bonded together via hot pressing, and a new CFRP board was obtained as shown in Fig. 8b. A Carbon fiber tube can be also prepared from a single CFRP sheets as shown in Fig. 8c. Carbon fibers are still arrayed regularly and compactly



Fig. 8. Different reuse methods of CFRP swelling products.

in new CFRP products. This reuse method can reduce the usage of epoxy resin and restore the original strength of CFRP materials. In actual applications, the selection of swelling treatment conditions primarily depends on the flexural strength requirements of the final product.

#### 4. Conclusion

Acetic acid swelling was utilized to develop a practical and environmentally friendly approach for recycling CFRP waste. Apart from its excellent swelling effect, acetic acid also exerts a certain catalytic degradation effect on CEP. The slight degradation phenomenon of CEP can promote the softening and delamination of CFRP. After 1 h of swelling treatment, the stiff CFRP became soft at 140 °C and was automatically delaminated at 160 °C-220 °C. The swelling ratio and mass loss rate of CEP increased with an increase in temperature and swelling time. The swelling products can be easily cut into long strips or other shapes. The length and strength of carbon fibers were retained well during reuse. The overexpansion and slight degradation of CEP caused the flexural strength of the dried swelling products to decrease with an increase in temperature. Epoxy resin was used to bond the shear and delaminated products together via hot pressing. The newly added epoxy resin can easily penetrate into the interior of the delaminated products to compensate for the loss in flexural strength caused by the swelling or degradation of CEP. The flexural strength of the new CFRP board can reach 47%-89% that of the original CFRP. Apart from carbon fibers, the CEP content of waste CFRP can be utilized to reduce environmental pollution caused by resin degradation.

#### Author statement

Mingfei Xing: Conceptualization, Methodology. Zixin Li: Writing-Original draft, Investigation, Visualization. Guohang Zheng: Resources, Validation. Yajie Du: Date Curation. Chun Chen: Software. Yaping Wang: Writing- Reviewing and Editing, Formal analysis.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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