

Article

Recycling of Carbon Fibers from CFRP Waste by Microwave Thermolysis

Jianning Deng ¹, Lei Xu ^{1,2,*}, Libo Zhang ¹, Jinhui Peng ¹, Shenghui Guo ^{1,2,*}, Jianhua Liu ² and Sivasankar Koppala ¹

¹ Faculty of Metallurgical and Energy Engineering, Kunming University of Science and Technology, Kunming 650093, China; jydengkust@163.com (J.D.); zhanglibopaper@126.com (L.Z.); jhpeng@kmust.edu.cn (J.P.); pepsiva9@gmail.com (S.K.)

² State Key Laboratory of Complex Nonferrous Metal Resources Clean Utilization, Kunming University of Science and Technology, Kunming 650093, China; liujianhua501050@163.com

* Correspondence: xulei_kmust@aliyun.com (L.X.); shguo78@hotmail.com (S.G.)

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Abstract: With the growth of the use of carbon fiber-reinforced polymer (CFRP) in various fields, the recovery of carbon fibers from CFRP waste is becoming a significant research direction. In the present work, degrading epoxy resin and recycling carbon fibers from CFRP waste by microwave thermolysis and traditional thermolysis were studied. The carbon fibers were successfully recovered by thermolysis under an oxygen atmosphere in this study. The properties of the recovered carbon fibers were characterized by field emission scanning electron microscopy (FESEM), Fourier transform infrared spectroscopy (FT-IR), X-ray diffraction (XRD), and Raman spectroscopy. The result shows that using microwave thermolysis to recover carbon fibers from CFRP waste is an attractive prospect. Compared to the traditional method, the reaction time was reduced by 56.67%, and the recovery ratio was increased by 15%. Microwave thermolysis is faster, more efficient, requires less energy, and obtains cleaner recovered carbon fibers than those recovered using traditional thermolysis.

Keywords: CFRP; recycling; carbon fibers; thermolysis; microwave

1. Introduction

Carbon fiber-reinforced polymer (CFRP) has been widely used in the aerospace, automobile, transportation, architecture, and sport industries, as well as the medical sector, due to their outstanding properties including low density, high strength and elastic modulus, and excellent resistance to corrosion and fatigue [1–5]. Carbon fibers were initially produced commercially starting in the late 1960s, and by 2006 approximately 27,000 tons of carbon fibers were being manufactured in industries around the world, an amount that will rise to 140,000 tonnes by 2020 [6,7]. CFRP is expected to be valued at over 25 billion dollars per year by 2025 [7]. However, the increasing use of CFRP has led to an increasing amount of waste being generated, consisting primarily of scraps and end-of-life components [1,8]. For instance, increasingly more plants using CFRPs will end their service life, generating massive amounts of CFRP waste [1,9–11]. Thus, the treatment of CFRP waste is becoming a more urgent problem.

Traditionally, CFRP waste has been managed by landfill disposal and incineration. However, it is predicted that traditional disposal approaches will need to be phased out due to pollution concerns, lack of economic viability, and new legal standards imposed in countries around the world [4,12,13]. Therefore, the recovery of carbon fibers from CFRP waste has become a research field that is attracting increasingly more attention. At present, various technologies have been proposed for recovering carbon fibers from carbon fiber-reinforced epoxy resin composites with excellent characterization,

including mechanical processes [14,15], chemical methods [4,16–20], and thermal technology [3,21–23]. However, long carbon fibers are difficult to obtain using a mechanical process, and their mechanical properties are seriously damaged with this technique [24,25]. Liu et al. [13], Yildirim et al. [19], and Hyde et al. [20] have investigated recycling carbon fibers from CFRP using different solvents under subcritical or supercritical conditions. These methods are expensive, difficult to industrialize, and produce large amounts of liquid waste and hazardous gases [1]. Yang et al. [3], López et al. [22], and Ye et al. [23] have studied thermolysis methods of recovering carbon fibers from CFRP. The pyrolysis process is not very economical, producing multiple hazardous gases and depositing char on the carbon fiber surfaces [1,19,25]. Therefore, research to develop new methods of recovering carbon fibers from CFRP has begun.

Microwave heating is gradually being used more often in the area of material processing due to its advantages of rapid, uniform, and selective heating [26]. Microwave heating is the process of coupling materials with microwaves, absorbing electromagnetic energy and transforming it into heat within the material volume, in which the heat is generated from the inside to the entire volume [27–29]. Therefore, the microwave method can treat uniform samples due to its features of volumetric and internal heating [30,31]. Yingguang Li et al. [32,33] have reported curing of CFRP by microwave energy, and CFRP can absorb microwave energy and be heated effectively. This study provides a great reference for microwave applications in the preparation and recovery of CFRP. Long Jiang et al. [34] reported that microwave irradiation is a flexible, easy-to-control, efficient method to recover high-value carbon fibers, and recovered carbon fibers could be directly used as reinforcement in new polymers (Polypropylene and nylon). This shows the great potential of effectively recycling carbon fiber by the microwave technique. In addition, Lester et al. [35] have reported carbon fiber recovery from polymer composites by microwave heating. Their results indicated that clean carbon fibers can be recovered using microwave heating, and that this method may be useful for recovering long carbon fibers [35]. However, these works have not undertaken enough research on the effects of different heating methods (microwave and traditional technique) on the recovery of carbon fibers. The research regarding the weight loss rate of CFRP, the recovery rate, and the chemical structure of recovered carbon fiber was not clear. In our present work, degradation of epoxy resin and recovery of carbon fibers from CFRP waste using microwave heating was studied. The influence of the reaction temperature and reaction time on carbon fiber recovery was investigated, and the surface of the carbon fibers was characterized.

2. Materials and Methods

2.1. Materials and Experimental Procedure

In this work, rectangular-shaped CFRP waste sheets with a thickness of 3 mm were provided by the Gongyi Fanruiyihui Composite Material Co., Ltd (Henan, China). The carbon fiber was Formosa Plastics TC33-3K, and the epoxy matrix was E51. The CFRP waste experiments were carried out in a high-temperature microwave furnace with a frequency of 2.45 GHz, under an oxygen atmosphere. In general, industrial microwave frequency is 2.45 GHz or 915 MHz [27,36]. However, most of the experiments regarding materials [27,28,37,38], metallurgical properties [27,30], or food [36,39] processing by microwaves are based on microwave magnetrons with 2.45 GHz frequency [28]. Therefore, the frequency of the microwave was selected as 2.45 GHz in this work. A K-type thermocouple was used to measure the temperature. The CFRP waste sheets were placed into a mullite crucible, which was placed within a polycrystalline mullite fiber cotton-based insulation chamber inside the microwave reactor. Oxygen was pumped continuously into the furnace cavity after the microwave furnace door was closed. Various microwave powers were applied to heat the CFRP waste, ultimately heating the samples from room temperature up to the desired reaction temperature, which was maintained for several minutes. In addition, the treatment of CFRP waste in a muffle

furnace under an oxygen atmosphere was also examined. The microwave experimental process is shown in Figure 1.

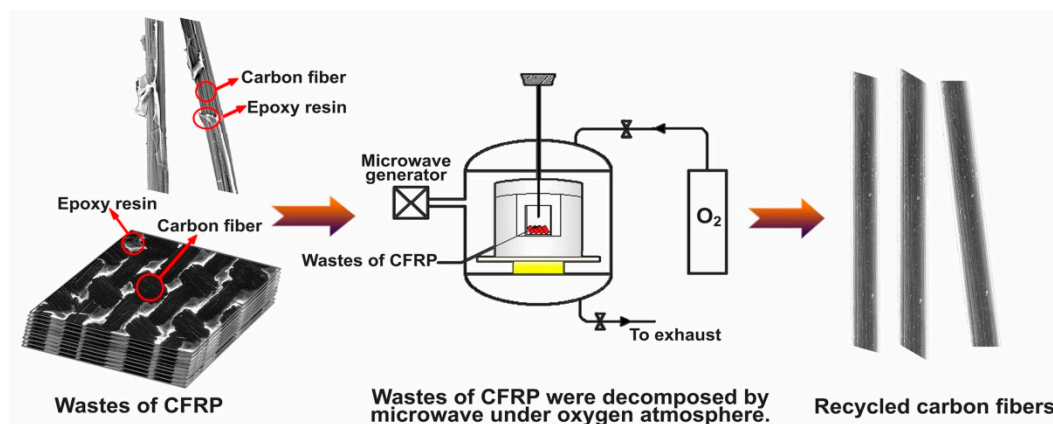


Figure 1. Schematic of carbon fiber-reinforced polymer (CFRP) waste treatment by microwave thermolysis.

2.2. Characterization

A Netzsch DSC/DTA-TG STA 449 F3 Jupiter® (Selb, Bavaria, Germany) was used to collect the thermogravimetric analysis (TGA) data. CFRP waste samples (10 mg) and virgin carbon fibers (10 mg) were heated individually from 30 °C to 1000 °C at 10 °C/min under an O₂ atmosphere (20 mL/min). The micro-morphologies and microstructures of the CFRP waste were examined using a metallographic microscope (Changfang CMM-10E, Shanghai, China) and scanning electron microscopy (SEM, Tescan VEGA3 SBH, Brno, Czech Republic). The surface images of recovered carbon fibers were examined using a field emission scanning electron microscope (FESEM, FEI Nova NanoSEM 450, Hillsboro, OR, USA). The functional groups of recovered carbon fibers were characterized by Fourier transform infrared spectroscopy (FT-IR, Thermo Scientific Nicolet iS50, Waltham, MA, USA), and the absorbance spectra were measured between 400 cm⁻¹ and 4000 cm⁻¹ using a Nicolet iS50 FT-IR spectrophotometer with a spectral resolution of 4 cm⁻¹. The crystalline phases of the recovered carbon fibers were observed by X-ray diffraction (XRD, PANalytical X' Pert3 Powder, Almelo, Overijssel, Holland). The XRD results were obtained using Cu K α radiation with a scan rate of 8°/min in a 2 θ of 5–70° at 40 kV and 40 mA. Raman spectra were obtained using a Renishaw plc. inVia Raman microscope (Gloucestershire, London, UK), with the laser power below 10.0 mW. The scanning wavenumber range was 800 cm⁻¹ to 2300 cm⁻¹.

3. Results and Discussion

3.1. Characterization of Carbon Fiber-Reinforced Polymer (CFRP) by Metallographic Microscopy and Scanning Electron Microscopy (SEM)

The structure of the CFRP waste is shown in Figure 2. The CFRP waste sheets were rectangular in shape, measuring 18 × 18 × 3 mm. Figure 2a'' shows the weave structure of carbon fibers, which indicates that the warp and weft were interwoven vertically. In the interwoven boundary of the carbon fiber bundles, it can clearly be seen that the carbon fibers are attached to each other by the epoxy resin.

The cross-sectional morphologies of CFRP waste are shown in Figure 2b–b''. As shown in Figure 2b, approximately five layers of carbon fiber fabric were laminated, subsequently impregnated by epoxy resin (Figure 2b'), and then cured for a period of time [40]. The vertical and horizontal directionality of carbon fiber bundles alternated in the cross-section.

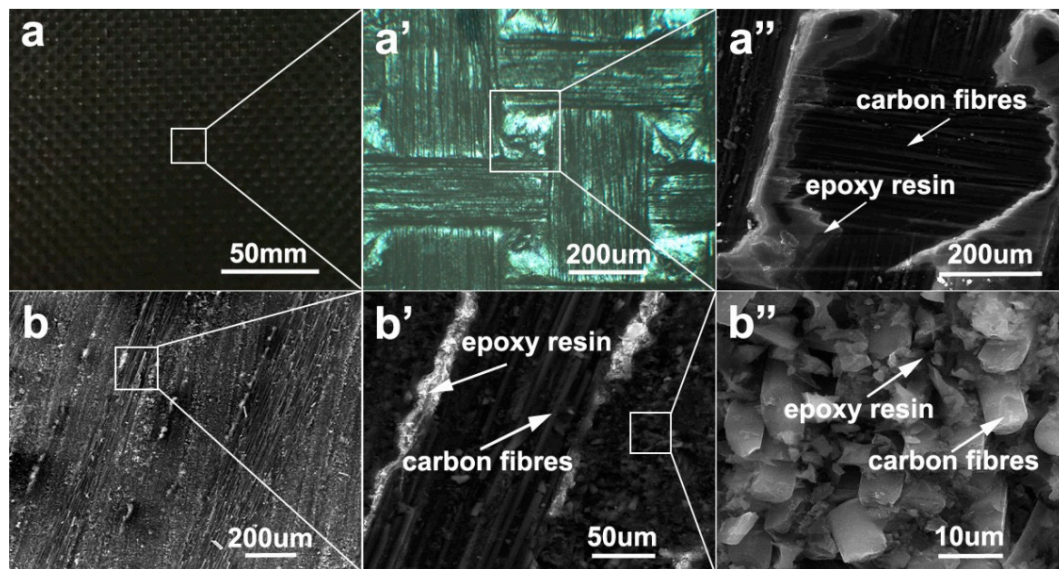


Figure 2. The structures of CFRP waste: (a–a'') surface structures, (b–b'') cross-sectional structures.

3.2. Thermal Decomposition Behavior of the CFRP Waste

The thermolytic behaviors of the CFRP waste and virgin carbon fibers under an oxygen atmosphere are shown in Figure 3. The thermolytic behavior of CFRP waste (Figure 3a) exhibited three oxidation peaks in the derivative thermogravimetric analysis (DTG) curve. The first peak corresponded to the degradation of the epoxy resin, the second peak corresponded to the oxidation of the pyrolytic carbon, and the third peak corresponded to the oxidation of the carbon fibers [3,41], indicating three reaction stages present in the TGA curve. The epoxy resin of the carbon fiber surface began to devolatilize at 230 °C with a mass loss of 11% at the end of first stage, the second devolatilization stage of the epoxy resin occurred between 350 °C and 550 °C with a mass loss of 38%, and the carbon fibers began to oxidize at 550 °C with 1.1% of residual mass. The differential scanning calorimetry (DSC) curve indicated that the reactions were exothermic. Figure 3b shows only one oxidation peak in the thermolysis behavior of virgin carbon fibers. Carbon fiber oxidation began at temperatures ranging from 500 °C to 600 °C and ended at 750 °C, with 1.3% of residual mass. This result was consistent with the result shown in Figure 3a. As shown in Figure 3c, the epoxy resin of the carbon fiber surface began to decompose at approximately 230 °C and stabilized at 450 °C, and this result was consistent with the results shown in Figure 3a,b. The temperature of the CFRP was 450 °C at 42 min. After 30 min duration, the residual mass of the CFRP was 66%, which was close to the 62% carbon fiber mass in the CFRP. Therefore, the durations were determined as 30 min in the muffle furnace. According to the TGA results, the reaction temperature (set between 400 °C and 500 °C in this study) was the critical factor for removing the epoxy resin of the carbon fiber surface and recovering carbon fibers. Some studies, such as Ye et al. [23] and Yang et al. [3], investigating the thermolytic process of recovering carbon fibers, have reported similar TGA results, so it was believed that the reaction temperature of the CFRP was relatively stable.

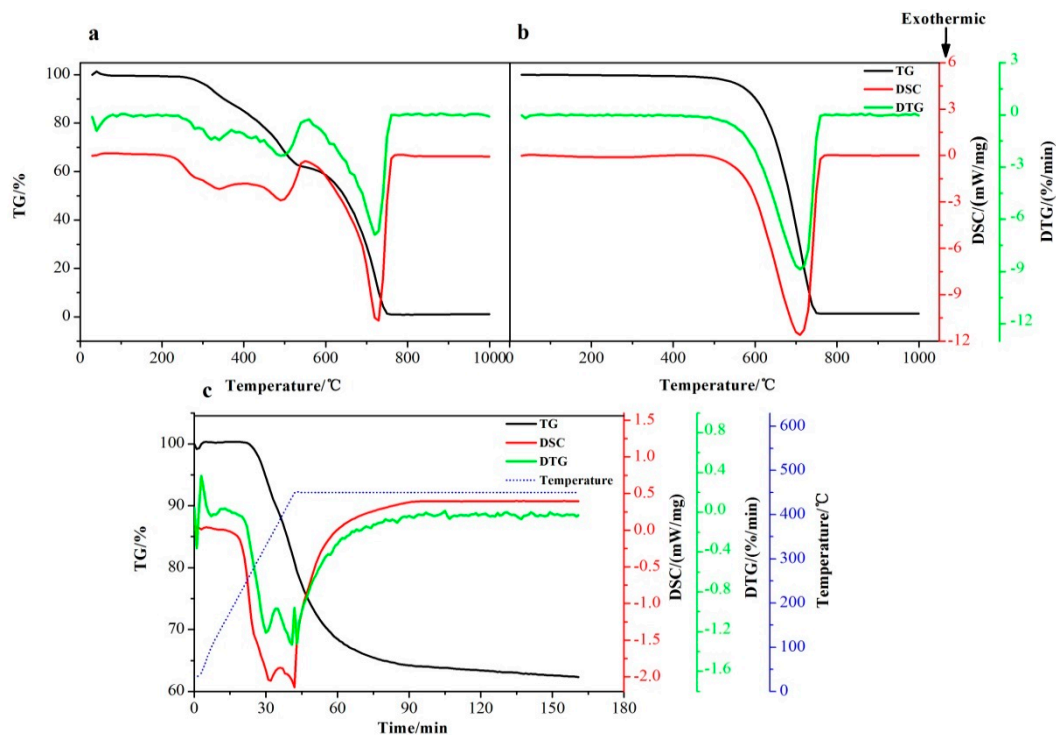


Figure 3. The thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) results under an oxygen atmosphere: (a) CFRP waste, (b) virgin carbon fibers, (c) CFRP waste maintained for 2 h at 450 °C.

The epoxy resin content in the CFRP waste was approximately 38 wt %. Table 1 shows the experimental details of the samples after treatment. The W_r , which was defined as the weight loss ratio of the CFRP waste, was calculated according to the following formula [3,19]:

$$W_r = \left(\frac{W_1 - W_2}{W_1} \right) \times 100\% \quad (1)$$

where W_1 and W_2 represent the weight of CFRP waste samples before and after experimentation, respectively.

Table 1. Details of samples under different reaction conditions.

Sample	Temperature/(°C)	Time/min	Average Power/W	Heating Method	Weight-Loss Ratio/%
1	400	30	850	Traditional	21.89
2	450	30	1060	Traditional	56.50
3	500	30	1300	Traditional	96.12
4	450	13	500	Microwave	47.03
5	450	30	500	Microwave	63.76

The R_{CF} , which was defined as the recovery ratio of carbon fibers, was calculated according to the following formula:

$$R_{CF} = \left(\frac{1 - W_r}{0.62} \right) \times 100\% \quad (2)$$

where W_r represents the weight loss ratio of the CFRP waste, and 0.62 is the mass of carbon fibers in the CFRP waste.

From Table 1, the best temperature should be between 400 °C and 450 °C, due to the 38 wt % weight-loss ratio being located between 400 °C and 450 °C. The weight-loss ratio of carbon fibers which were recovered by the microwave method at 450 °C was closer to 38 wt %, and required a shorter

reaction time than that of carbon fibers which were recovered by the muffle furnace at the same temperature. Compared to the traditional method, the reaction time of the microwave method was reduced by 56.67% at 450 °C. The recovery ratios of the microwave and traditional method were approximately 85% and 70%, respectively. Thus, the microwave heating method is faster, has a better recovery ratio, and requires less energy than the traditional heating method.

3.3. Morphology of the Recovered Carbon Fibers

Figure 4 shows the recovered carbon fibers after traditional themolysis (Figure 4a–c) and microwave themolysis (Figure 4d). The epoxy resin could not completely decompose, with the sheet CFRP retained at 400 °C. However, it could be completely decomposed when the temperature reached 450 °C, and the carbon fibers could then be obtained. On the other hand, some of carbon fibers oxidized (the grey section in Figure 4c) when the temperature was above 500 °C. This result was consistent with the TGA results.

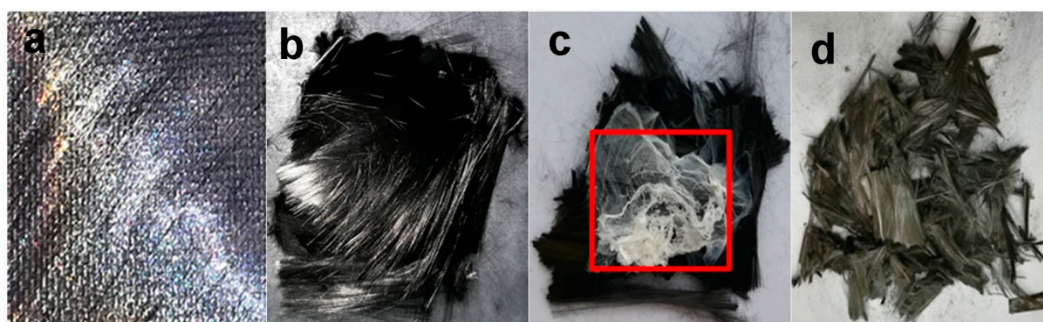


Figure 4. The macroscopic appearances of the CFRP waste samples after experiments: (a) 30 min of traditional heating at 400 °C, (b) 30 min of traditional heating at 450 °C, (c) 30 min of traditional heating at 500 °C, (d) 13 min of microwave heating at 450 °C.

The micromorphology of carbon fibers recovered by the traditional method is shown in Figure 5a–c. A small amount of epoxy resin is visible on the surface of the carbon fibers obtained at 400 °C. When the temperature was 450 °C, the carbon fibers' surfaces were relatively clean. However, when the temperature reached 500 °C, the surface of the carbon fibers were seriously damaged, exhibiting a large number of grooves indicative of partial oxidation on the carbon fiber surface. Thus, relatively clean carbon fibers could be recycled at 450 °C. From Figure 5d, we can see that the surface of the carbon fibers which underwent microwave heating at 450 °C were cleaner than those which were treated by traditional heating at the same temperature, and the reaction time was reduced by 56.67%. This conclusion was consistent with results of Table 1 and Figure 4.

Figure 5a'–d' shows the average diameter of the recovered carbon fibers. The average diameter of the virgin carbon fiber was around 7 µm. For the traditional method, the average diameter of the carbon fiber which was recovered at 400 °C for 30 min was 8.47 µm (Figure 5a') and at 450 °C for 30 min was 7.77 µm (Figure 5b'), which were both larger than the average diameter of virgin carbon fibers, possibly because the epoxy resin of the carbon fiber surface was not removed completely. However, when the temperature was 500 °C for 30 min, the average diameter of the recovered carbon fibers was 5.82 µm (Figure 5c') for the carbon fiber surface when oxidized. The average diameter of the carbon fiber which was recovered using the microwave method at 450 °C for 13 min was 7.02 µm (Figure 5d'), close to the virgin carbon fiber, which was probably due to the structure of the recovered carbon fiber not being damaged too much because of the microwave's rapid heating feature. However, the carbon fiber recovered by microwave at 450 °C for 30 min was damaged with many holes, as shown in Figure 5e,e'. Therefore, 13 min duration was chosen in the microwave furnace.

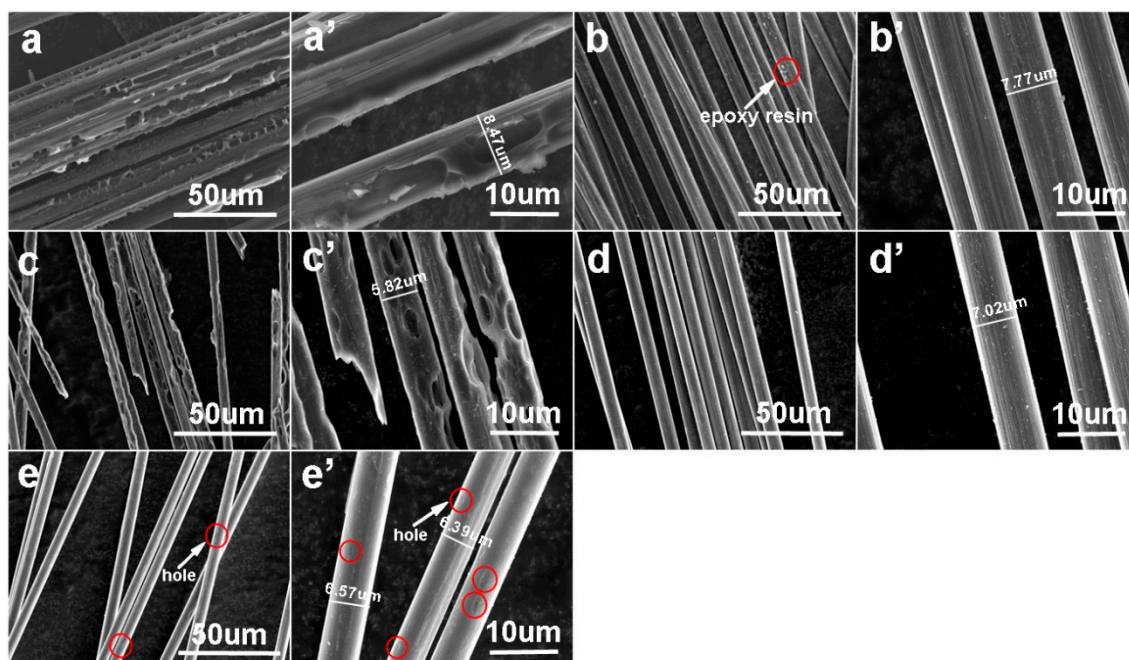


Figure 5. Field emission scanning electron microscope (FESEM) images of recovered carbon fibers: (a,a') 30 min of traditional heating at 400 °C, (b,b') 30 min of traditional heating at 450 °C, (c,c') 30 min of traditional heating at 500 °C, (d,d') 13 min of microwave heating at 450 °C, (e,e') 30 min of microwave heating at 450 °C.

3.4. Fourier Transform Infrared (FT-IR) Analysis of the Recovered Carbon Fibers

Functional groups found on the recovered carbon fibers under different experimental conditions were observed in the FT-IR spectra, as shown in Figure 6, and alterations of major chemical bonds were also studied. Functional groups of carbon fibers usually include -CO , -OH , -COH and -COOH . The broad peak at 3420 cm^{-1} corresponded to O-H stretching vibrations due to water on the surface of the recovered carbon fibers. The small broad peak at 3260 cm^{-1} corresponded to O-H stretching vibrations related to hydroxyl groups. The peaks at 2960 cm^{-1} and 1380 cm^{-1} were attributed to the C-H stretching vibrations of a methyl group, while the peaks at 2930 cm^{-1} and 2850 cm^{-1} were attributed to the C-H stretching vibrations of a methylene group. The peaks at 2360 cm^{-1} and 2340 cm^{-1} were attributed to $\text{C}\equiv\text{N}$ stretching vibrations. The peaks at 1630 cm^{-1} and 1084 cm^{-1} were attributed to the C=C and C-C stretching vibrations, respectively, of the recovered carbon fiber backbone. The peaks at 1260 cm^{-1} and 1050 cm^{-1} were C-O asymmetric and symmetric stretching vibrations, respectively.

From Figure 6, the FT-IR spectra of recovered carbon fibers were similar to those of virgin carbon fibers. However, the $\text{C}\equiv\text{N}$ bands were produced on recycled carbon fibers, possibly due to curing during the preparation of CFRP. The C=C bands decreased, while the C-H bands and C-O bands increased, which was potentially because the reaction between the carbon atom and the oxygen atom broke down the C=C bonds and produced C-H bands and C-O bands. The FT-IR spectra of carbon fibers which were recovered using the traditional method from 400 °C to 500 °C for 30 min were similar, suggesting that an increase in the reaction temperature did not significantly impact the types of chemical bonds. However, the methyl peak became stronger and the C-O band increased while the C=C band decreased with a corresponding temperature increase, possibly due to the oxidation of carbon atoms and formation of C-O bonds. Compared with the traditional method, the types of chemical bonds of the recovered carbon fibers which were obtained using the microwave method did not noticeably change, but the intensity of its absorption peaks was weakened. This observation might be explained by the oxidation of more organic molecules to smaller molecules, which were released in the form of gas by microwave heating under an oxygen atmosphere. The FESEM results indicated that the recovered carbon fibers which were obtained at 450 °C by both the microwave and traditional

methods were clean, while Figure 6 reveals that their $C\equiv N$ bonds were small, possibly due to $C\equiv N$ bond breakage related to epoxy resin of the carbon fiber surface removal.

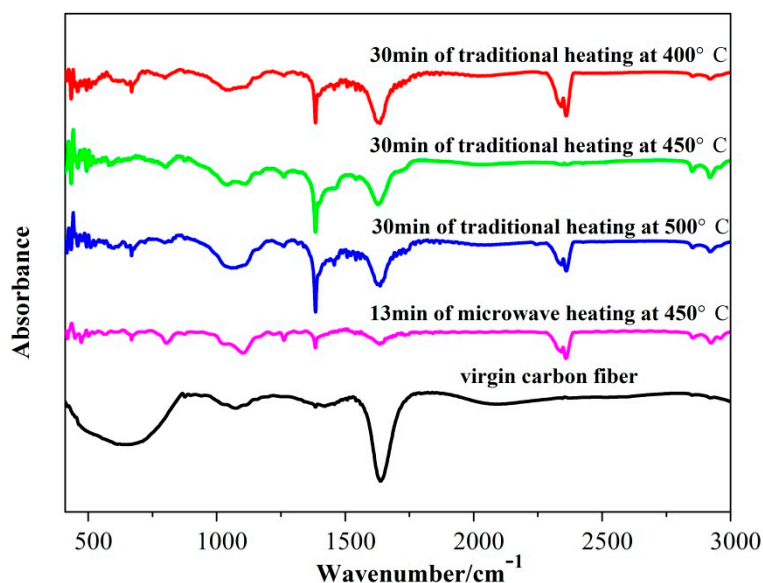


Figure 6. Fourier transform infrared (FT-IR) spectra of carbon fibers.

3.5. X-ray Diffraction (XRD) and Raman Analyses of the Recovered Carbon Fibers

The XRD results of the carbon fibers recovered under different conditions are shown in Figure 7a and 7b. A scattering peak is apparent in Figure 7a, indicating the (002) plane at approximately $2\theta = 25.5^\circ$, reflecting the carbon layer stack thickness along the “c” axis (L_c). As illustrated in Figure 7b, another scattering peak was observed at the (101) plane at approximately $2\theta = 43.5^\circ$, correlating to the graphite crystallite size along the “a” axis (L_a). The Raman spectra of the recovered carbon fibers are shown in Figure 8. The peak near 1370 cm^{-1} is called the D band, reflecting a graphitic lattice vibration mode with A_{1g} symmetry [4]. The other peak near 1600 cm^{-1} is the G band, reflecting an ideal graphitic lattice vibration mode with E_{2g} symmetry [4]. The β is the full width at the half-maximum of the peak with a unit of radian. W_D and W_G are the Raman shift of the D band and G band, respectively. A smaller integral intensity ratio R (I_D/I_G) indicates a higher degree of graphitization, or smaller proportions of the ordered structure of recovered carbon fibers [42].

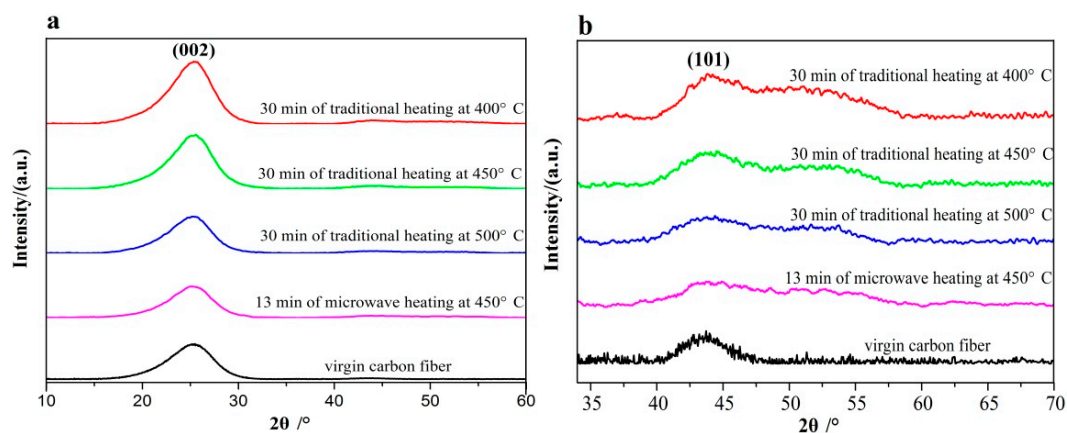


Figure 7. (a) XRD spectra of the (002) plane of carbon fibers, (b) XRD spectra of the (101) plane of carbon fibers.

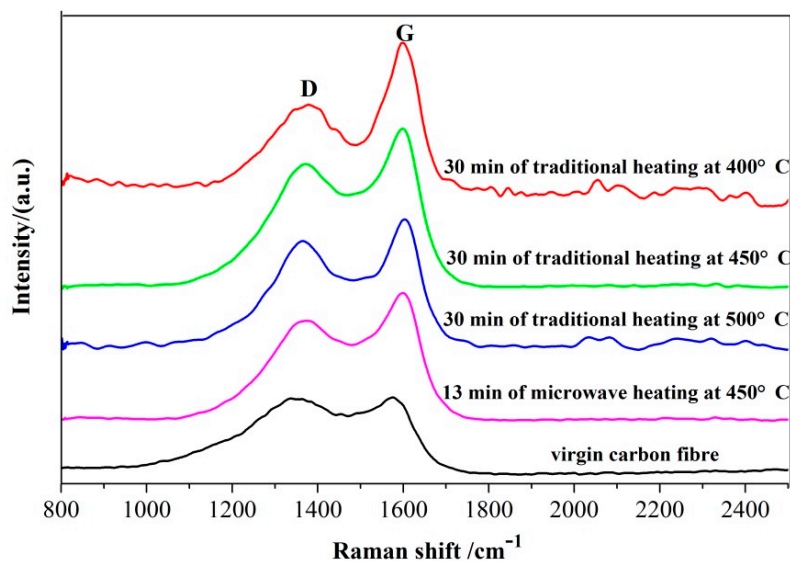


Figure 8. Raman spectra of carbon fibers.

Table 2 contains the crystallite parameters of the (002) plane and the (101) plane of the recovered carbon fibers in the XRD analysis. Upon comparison of carbon fibers which were recovered using the traditional method from 400 °C to 500 °C for 30 min, the value of β increased with increasing reaction temperature, which means the width of the scattering peaks increased, possibly due to the decrease in the crystallite size of the carbon fibers [43]. The intensity of the scattering peaks weakened significantly with increasing reaction temperature, indicating a reduction in the degree of graphitization. Comparison of the crystallite sizes (L_c and L_a) of the carbon fibers which were recovered using the traditional method from 400 °C to 500 °C showed that the size decreased obviously with an increase in temperature, revealing that the amount of amorphous carbon increased, demonstrating a lesser degree of graphitization and lower carbon fiber strength. This result was consistent with the conclusion obtained from Figure 7. Upon comparison of Samples 2 and 4, microwave heating had less influence on the degree of graphitization of recovered carbon fibers at 450 °C, due to the very similar values of L_c , and L_a . Compared with virgin carbon fibers, the properties of recycled carbon fibers decreased slightly. We believe that the reduction of graphitization is due to the increase of amorphous carbon, and the increase of amorphous carbon is due to temperature, and may also be the result of the intensified carbon combustion. In this study, we found this phenomenon, but the relationship between the decrease of graphitization degree, increase of amorphous carbon, temperature factor and carbon combustion still need to be further studied.

Table 2. X-ray diffraction (XRD) results of the (002) plane and the (101) plane of the recycled carbon fibers.

Sample	2θ (002)/°	D (002)/nm	2θ (101)/°	D (101)/nm	β (rad)	L_a /nm	L_c /nm
1	25.66	0.34684	43.52	0.20778	0.0898	3.06	1.57
2	25.60	0.34763	43.46	0.20804	0.0922	2.99	1.52
3	25.52	0.34879	43.42	0.20822	0.0963	2.73	1.46
4	25.54	0.34853	43.50	0.20789	0.0918	2.99	1.53
Virgin carbon fiber	25.36	0.35098	43.85	0.20631	0.0599	2.89	2.345

The Lorentz fitting parameters are listed in Table 3. Tuinstra et al. [44] studied the relationship between the value of R and L_a , concluding that $L_a = 4.4/R$. The L_a value of Raman was similar to the XRD results. At different temperatures with the traditional method from 400 °C to 500 °C, the D peak wave number values decreased and the R value increased with increasing reaction temperature, suggesting a reduced degree of graphitization of the recovered carbon fibers. This result indicated that increasing the reaction temperature caused greater damage to the structure of the recovered carbon

fibers. Microwave thermolysis had little influence on the degree of graphitization of the recovered carbon fibers. The conclusions obtained from the Raman analysis were in very good agreement with the results of the XRD experiments.

Table 3. Raman results for the recycled carbon fibers.

Sample	$W_D/(\text{cm}^{-1})$	$W_G/(\text{cm}^{-1})$	$R(I_D/I_G)$	L_a/nm
1	1378.24	1598.61	1.19	3.70
2	1371.17	1598.61	1.55	2.84
3	1365.52	1602.85	1.64	2.68
4	1372.59	1598.61	1.59	2.76
Virgin carbon fiber	1363.6	1576.39	1.61	2.73

4. Conclusions

Using microwave and traditional thermolysis to degrade epoxy resin and recover carbon fibers from CFRP waste has been studied in this work. The result shows that carbon fibers can be recovered from CFRP waste using thermolysis at 450 °C under an oxygen atmosphere, and the best temperature for this is 450 °C in this study. Compared to traditional thermolysis, microwave thermolysis is a faster and more efficient method that requires less energy, achieving a reduced reaction time by 56.67% and an increased recovery ratio by 15%. Additionally, the surface of the carbon fibers recovered using microwave heating was cleaner, smoother, and contained less epoxy resin. Furthermore, the increase in amorphous carbon and a lessened degree of graphitization occurred with increasing temperature, corresponding to decreased carbon fiber strength. However, microwave heating has minimal influence on the graphitization degree when used at the same reaction temperature as that of the traditional heating method, and does not alter the chemical structure of the recovered carbon fibers.

It can be predicted that CFRP waste will be increasingly generated, and its disposal will become more and more urgent. At present, many methods have been researched to recycle CFRP waste. However, microwave energy has been shown to have unique advantages, and great application prospects for recycling CFRP due to being faster, more efficient, requiring less energy, and achieving a higher recovery ratio of carbon fibers than traditional heating. In the future, continuous microwave reactors and production lines should be investigated to recover CFRP industrially. In addition, although the reuse of carbon fibers has not been studied much, it should attract more and more attention. Recycled carbon fibers can maintain reasonable properties and meet the requirements of some fields, such as bearing, filling, and more. The reuse of carbon fibers requires low cost, expands the application field, and protects the environment, which are significant positive outcomes.

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