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Application of supercritical water for green recycling of epoxy-based carbon fiber reinforced plastic

Young Nam Kim[a,b], Young-O. Kim[a], Seong Yun Kim[c], Min Park[a,d], Beomjoo Yang[f], Jaewoo Kim[a], Yong Chae Jung[a,e,*]

[a] Institute of Advanced Composite Materials, Korea Institute of Science and Technology (KIST), 92 Chudong-ro, Bongdong-eup, Wanju-gun, Jeonbuk 55324, Republic of Korea
[b] Department of Chemical and Biomolecular Engineering, Yonsei University, 262 Seongsanno, Seodaemun-gu, Seoul 120-749, South Korea
[c] Department of Organic Materials & Fiber Engineering, Chonbuk National University, 567 Baekje-daero, Deokjin-gu, Jeonju-si, Jeonbuk, 54896, Republic of Korea
[d] KHU-KIST Department of Converging Science and Technology, Kyung Hee University, Seoul 02447, Republic of Korea
[e] Department of Nanomaterials and Nano Science, Korea University of Science and Technology (UST), 217 Gajeong-ro, Yuseong-gu, Daejeon 34113, Republic of Korea
[f] School of Civil Engineering, Chungbuk National University, Chungdae-ro 1, Seowon-Gu, Cheongju, Chungbuk 28644, Republic of Korea

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ABSTRACT

The objective of this study is to fabricate conductive carbon fiber composites with thermal and electrical properties by degradation carbon-fiber-reinforced plastics (CFRPs) and recycled carbon fibers using only supercritical water without any catalyst or oxidant. We focused on a recycling method that is harmless to the human body and environment-friendly, by using supercritical fluid water rather than recycling CFRP by physical or pyrolysis methods. In particular, we recycled carbon fibers (R-CFs) in which up to 99.5% of epoxy resin was removed, by optimizing the conditions of supercritical fluid water (SCF-W) treatment, and we fabricated conductive R-CFs composites with thermal and electrical properties by combining the R-CFs with cyclic butylene terephthalate (CBT), which is a polymerizable low-viscosity thermoplastic resin. The fabricated composites had a thermal conductivity of 1.35 ± 0.05 (W/mK) and an electrical conductivity of 11.23×10^{-6} (S/cm) when the added amount of recycled carbon fibers was 5 wt%.

1. Introduction

Carbon-fiber-reinforced plastics (CFRPs) are composite materials in which carbon fibers are impregnated with thermosetting or thermoplastic polymer resins and have properties such as high ratio of mechanical strength to weight, chemical resistance, and corrosion resistance [1,2]. These properties allow them to be widely applied in various industries such as automotive, aerospace, and equipment for sports and leisure activities [3]. In particular, in the field of aircraft manufacture, CFRPs are increasingly used for maintenance cost reduction and high fuel efficiency through weight reduction of materials [4]. Currently, the usage of CFRPs is increasing yearly, and it is expected that the demand will increase sharply from $16.1 billion in 2011 to approximately $48.7 billion in 2020 [5].

Since used CFRP is very difficult to recycle due to its complex composition, the cross-linking properties of thermosetting resins, and their fusion with other materials, most CFRP waste is currently landfilled or incinerated. Although some waste carbon fibers or carbon powders are recovered and used as low-cost battery additives or rubber fillers, the recycling rate is low due to inconsistent physical properties compared to existing materials. In addition, research on recycling technologies for CFRPs has been gaining attention in Europe as well as in Asia, because landfilling and incineration of expensive carbon fibers is regarded as an unsatisfactory solution due to environmental impact, legal issues, economic losses, and resource management issues. In 2009, the UK government selected “Increasing Sustainability and Recycling” as one of its three major goals of a strategy for the composites industry and has regulated CFRP [6,7]. The Japanese government has also recently regulated the landfilling of waste CFRP and encouraged its recycling [8].

As methods of recycling waste CFRPs, there exist physical [9,10], pyrolysis [11–14], and chemical methods [15–20]. The physical method destroys composites by shredding, crushing, grinding or other similar mechanical processes, and After that, powders and fibrous

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[*] Corresponding author. Institute of Advanced Composite Materials, Korea Institute of Science and Technology (KIST), 92 Chudong-ro, Bongdong-eup, Wanju-gun, Jeonbuk 55324, Republic of Korea.

E-mail address: ycjung@kist.re.kr (Y.C. Jung).

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materials from the generated scraps are separated by sieving, and recycled as new composites or used as construction materials (e.g. artificial wood, asp halt or mineral resources for cement). However, since these are used as low-cost materials, quality improvement methods need to be applied to maintain the quality level of composites. The pyrolysis method, the most widely used recycling process, recovers carbon fibers by treating CFRPs waste at high temperature under anaerobic conditions. However, there are limitations to this method due to toxic gas generated during treatment, the introduction of defects into the carbon fibers.

A decomposition method using supercritical fluids is a typical chemical method. A supercritical fluid displays strong solvent properties by accelerating mass transfer rate with low viscosity and high diffusion coefficient under conditions above a certain critical point (critical temperature and critical pressure) for each material [21]. In particular, among the fluids to be used, water and carbon dioxide have the advantages that they are harmless to the human body, inexpensive, and can be easily recovered and reused.

Among previous studies on recycling using supercritical fluids, the Lester group of Nottingham University in UK conducted a study on the recovery/restoration of the surface properties of carbon fiber composites using supercritical n-propanol [15], and the Pickering group reported a study on the separation and recycling of carbon fiber/epoxy resin composites according to fluid treatment conditions using the same method [22]. The Goto group at Kumamoto University in Japan conducted a study on the development of synthetic/separation based technologies according to supercritical or subcritical conditions for recycling polyethylene terephthalate (PET) infusible/insoluble plastic from fiber-reinforced plastic (FRP) [23]. The Y. Liu group showed that the decomposition efficiency of bisphenol-A-type epoxy resin hardened with diaminodiphenylmethane was higher than 95% by adding phenol and potassium hydroxide to catalyze the supercritical fluid water (SCF-W) process, proving this method to be effective [24]. R. Piñero-Hernanz reported the decomposition of epoxy resin in CFRP using supercritical methanol (above its critical temperature of 239 °C and critical pressure of 8.1 MPa), ethanol (above its critical temperature of 243 °C and critical pressure of 6.1 MPa), *n*-propanol, and acetone (above its critical temperature of 236 °C and critical pressure of 4.7 MPa) [25]. As shown in the above examples, most methods of recycling CFRP using supercritical fluids also use organic solvents or catalysts, therefore introducing issues such as solvent recovery, recycling, and secondary pollution.

Therefore, in this study, we aim to recycle CFRPs by decomposing it with only supercritical fluid water (SCF-W), without using a separate catalyst, to recycled carbon fibers (R-CFs). In particular, we investigated the decomposition behaviors of CFRPs and structural changes of the R-CFs depending on supercritical treatment conditions. In addition, we fabricated conductive R-CFs composites with thermal and electrical properties by using the R-CFs and compounding them with cyclic butylene terephthalate (CBT), and evaluated their properties.

2. Experimental

2.1. Supercritical water treatment of epoxy-based carbon fiber reinforced plastics

The CFRPs was supplied by Decc Carbon, Prepregs were prepared using TORAYCA® T700S with epoxy KBR-1729 (Cycloaliphatic Glycidyl ether modified type) and KBH-1088 (Acid anhydride hardener type) as curing agents. The manufactured prepreg was laminated 50 pieces and CFRPs was produced through an autoclave process. In order to carry out the experiment, we made the dimensions of 1x1x1 cm cubic structure sample (width x length x thickness) by using a laser cutter (ILS 12.75, University Laser System). We used equipment built by ourselves for the treatment of SCF-W. This equipment is a batch type reactor of total 150 ml capacity equipped with 4 blades. The laser cut sample was put into 80 ml of water in the reactor. The sample was heated to 405 ± 2 °C, which made the internal pressure of the reactor reach to 280 ± 10 bar. The CFRP sample was degraded in supercritical water with different reaction times of 10, 30, 60, and 120 min. After the reaction finished, the reactor was cooled down at room temperature.

2.2. Manufacture of conducting carbon fiber composite using recycled carbon fibers (R-CFs)

In order to fabricate conductive R-CF composites with thermal and electrical properties by using R-CFs, cyclic butylene terephthalate (CBT, CBT160, Cyclics Schenectady NY, USA) and R-CFs were mixed using a planetary centrifugal mixer “THINKY MIXER” (Are-310, Thinky Cor., Japan) and hot-pressed at 15 MPa and 250 °C for 2 min using a hot-melt press (Model-1006C, Ocean Science, Korea).

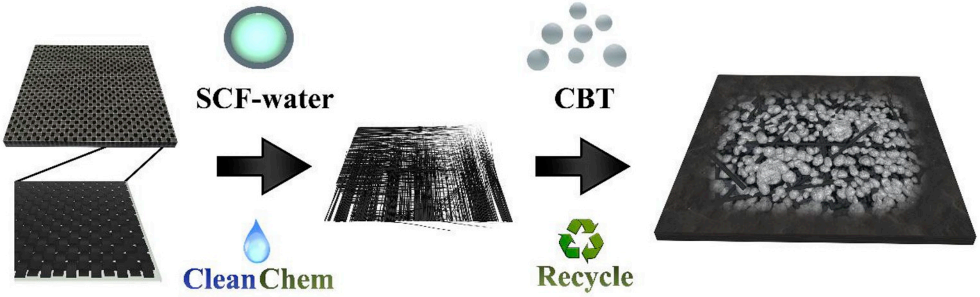
2.3. Characterizations

To observe the structure and surface properties of the R-CFs that were degraded from raw samples (CFRPs), we used Renishaw inVia Raman microscope (514 nm laser lines, Renishaw Corporate, UK), FT-IR (Nicolet iS10, ThermoFisher Scientific, USA), X-ray photoelectron spectroscopy (XPS, K-Alpha, ThermoFisher Scientific, USA), and filed-emission scanning electron microscopy (FE-SEM, Nova NanoSEM 450, FEI, USA). Thermogravimetric analysis (TGA, Q50, TA Instruments, USA) was used to analyze the thermal degradation of R-CFs; a 10 mg sample was heated from 40 °C to 900 °C at a rate of 10 °C/min under a N_2 atmosphere. The mechanical properties of the Pristine carbon fiber (Pristine CF) and R-CFs were examined by tensile tester for a single fiber (FAVIMAT+, Textechno, Germany. We used the following measurement conditions: gauge length = 25 mm; crosshead speed = 2 mm/min. At least 10 samples were tested, and the average value was reported. In addition, we used dimensional (3D) non-destructive X-ray micro computed tomography (micro-CT) (Sky Scan1172, BRUKER, Belgium), and a thermal conductivity meter (TPS 2500 Hot Disk, Sweden) to investigate the structural, thermal and electrical properties of fabricated conductive R-CFs composites. The electrical conductivity of the obtained R-CFs composites was measured using a LORESTA-GP (Mitsubishi Chemical, MCP-T600) electric analyzer. A standard four probe (TFP type probe) method was used to measure the electrical conductivity of all the R-CFs composites at ambient conditions of 25 °C. The thermal conductivity of a square-(25 mm × 25 mm x 2 mm) R-CFs composites was measured using a steady-state heat flow measurement apparatus (LFA447-Nanoflash, NETZSCH). The residues obtained after SCF-W treatment were extracted by ethyl acetate and analyzed by GC-MS equipment (GCMS-QP2010-Plu, Shimadzu, Scientific Instruments).

3. Results and discussion

Schematic 1 is a diagram illustrating the recycling of CFRPs in this study. Samples that were cut to a certain size were treated with SCF-W and degraded, then conductive carbon fiber composites were fabricated by compounding the R-CFs with polymerizable low-viscosity thermoplastic resins (CBT) using a hot press (see Scheme 1).

Fig. 1 shows photos and FE-SEM images of the R-CFs before and after SCF-W treatment. Fig. 1(a) shows photos of R-CFs carbon fibers from which epoxy is removed by treating CFRP samples cut to a certain size with SCF-water. Fig. 1 (b) and (c) are images of CFRP observed before and after SCF-W treatment using FE-SEM. Fig. 1(b) shows cross-section and surface images of the samples, showing that epoxy resin surrounds the carbon fibers. However, it is clear that the epoxy resin is completely degraded and removed after treatment with SCF-W as shown in Fig. 1(c). Fig. S1 show the IR spectrum according to the reaction conditions, CFRP before the reaction shows a C-O-C peak (1033 cm^{-1}). As the reaction proceeds, the C-O-C peak is reduced,



Scheme 1. Schematic diagram of the green recycling process of CFRP by SCF-W.

while peaks that are notable features of carbon fiber, such as the CH_3 bend peak (1388 cm^{-1}), C=C aromatic peak (1635 cm^{-1}), and C-H stretch peak (2915 cm^{-1}) remain constant. In other words, as the SCF-W reaction proceeds, the epoxy resins are removed from the carbon fibers of CFRP.

To find out the optimum degradation conditions for CFRP using SCF-W treatment, we analyzed the structure and physical properties of carbon fibers after recovering them at different critical pressures and temperatures, and various treatment times. Fig. 2(a–d) show images showing the extent of degradation of the epoxy resin and surface structural changes of the recovered carbon fibers observed by FE-SEM while increasing the treatment times from 10 to 30, 60, and 120 min, respectively, at the critical pressure of 280 ± 10 bar and the critical temperature of 405 ± 2 °C. They show that not only is the epoxy around the carbon fibers removed but also the morphology of the R-CFs surface is partially clean as SCF-W treatment time increases. We can confirm from Fig. 2(a and b) that a large amount of epoxy resin particles remains on the R-CFs surface without being decomposed after SCF-W treatment for approximately 10–30 min. However, as treatment time is gradually increased, the epoxy resin observed around the R-CFs gradually disappears, and we obtain only R-CFs with clean surfaces. We confirmed that the diameter of the obtained carbon fibers was

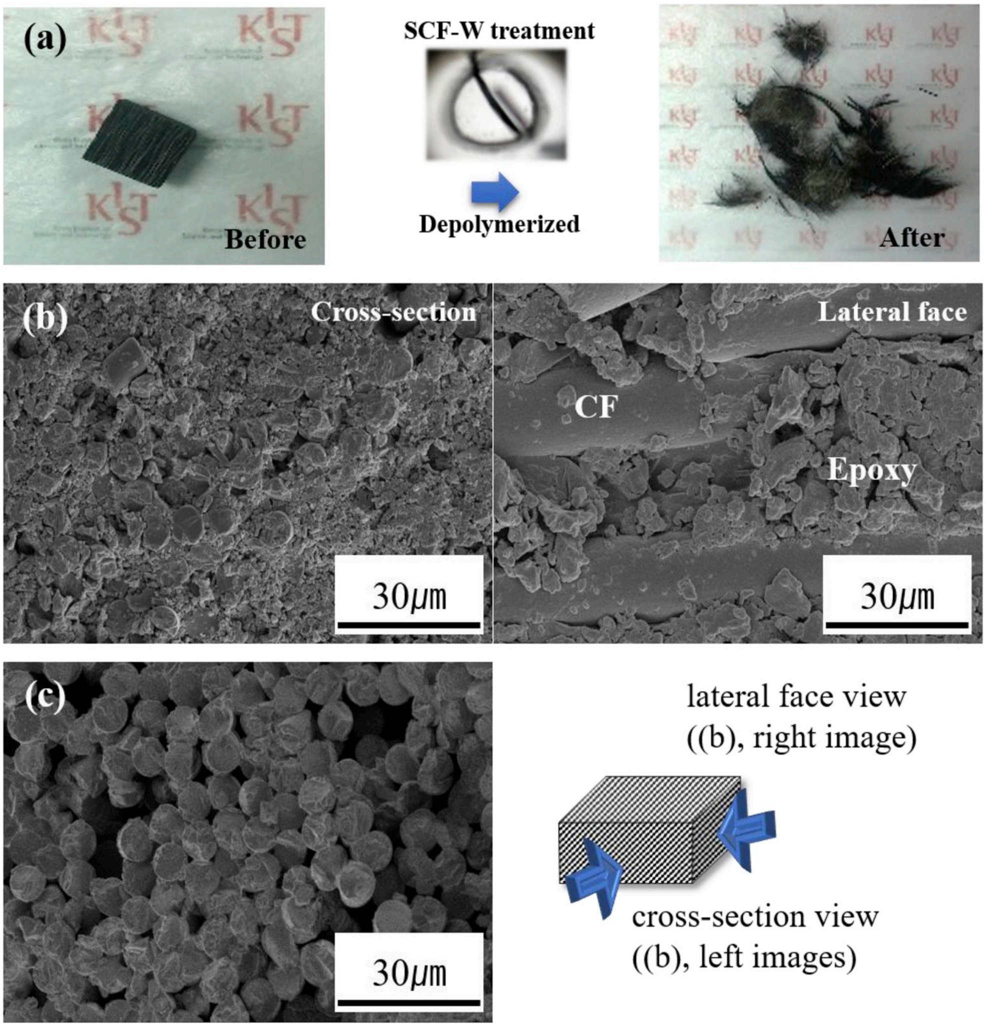


Fig. 1. (a) Photo images of untreated CFRP sample cubic (left) and RCFs (right); SEM photos (b) before SCF-W treatment (cross-section and lateral face), and (c) after SCF-W treatment.

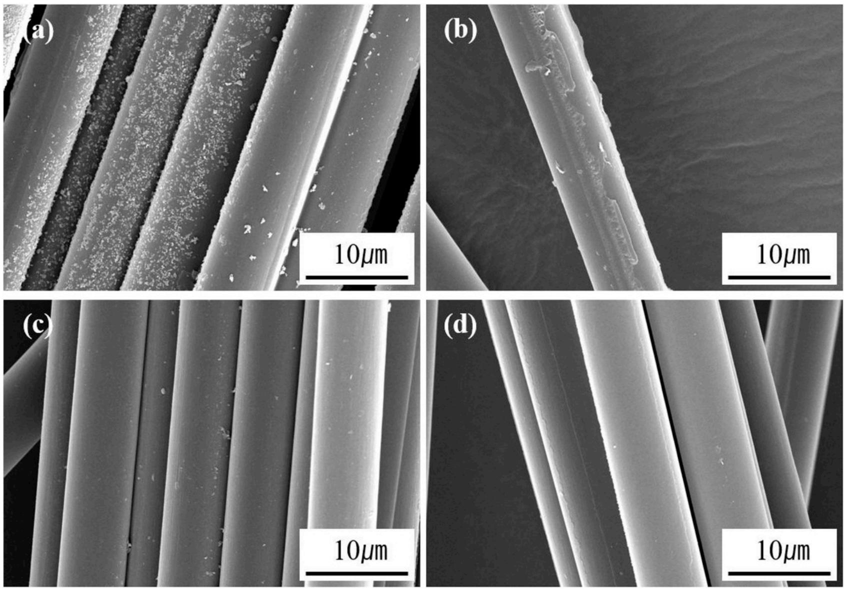


Fig. 2. SEM images of R-CFs of after SCF-W treatment: (a) 10 min, (b) 30 min, (c) 60 min, and (d) 120 min treatment, respectively.

7 ± 1 μm with a relatively uniform distribution (see Fig. S3, Supporting Information).

On the other hand, we can confirm that the epoxy is partially decomposed when treating at subcritical water conditions (critical pressure 150 ± 10 bar and critical temperature 300 ± 2 °C) for 30 and 120 minutes, but a large residual amount of epoxy resin that was not degraded from R-CFs was observed on the surface of the R-CFs (Fig. S2). It is considered that, compared to the subcritical state, the supercritical state helps rapid penetration of fluid into the microcavities of the CFRP due to rapid heat and mass transfer, low viscosity, and high diffusion coefficient of the fluid, thus leading to more effective degradation of the epoxy resin. In particular, the SCF-W used in this study differs in its density, dielectric constant, and electrical conductivity, and has a high solubility for various solutes when it is above a certain critical value (temperature 374 °C, pressure 22.1 MPa), therefore it shows totally different characteristics from water or water vapor below the critical point [26].

One of the important properties in describing the structure of water in the supercritical region is the static dielectric constant. The dielectric constant is a numerical value indicating the degree of hydrogen bonding and polarity of water. The dielectric constant of water is 80 at room temperature and atmospheric pressure, and a large dielectric constant means that hydrogen bonding between water molecules is strong. According to the results of Marcus et al., the dielectric constant above 25 MPa has a value of 10 at 420 °C, 5 at 420 °C, and 2 at 490 °C [27]. This means that the hydrogen bonding between water molecules is weakened and the degree of hydrogen bonding is also weakened as the density of water in the critical region is reduced. The dielectric constant of SCF-W has a value similar to that of non-polar solvents (e.g., benzene, ethyl ether, and nucleic acids), meaning that SCF-W acts as a weakly polar or non-polar solvent. In the supercritical region, water can act as a non-polar solvent, and is completely mixed with organic compounds that are insoluble at room temperature and atmospheric pressure; all components are present in a single phase when the mixture reaches a supercritical state. Therefore, SCF-W can be considered an excellent solvent for organic compounds that are insoluble in normal water.

Fig. 3 and Table 1 shows the results of Raman, thermogravimetric analysis TGA and XPS, in order to identify any structural changes of the R-CFs obtained from the degradation treatment conditions of SCF-W, and the residual amount of epoxy. Fig. 3(a) shows the results of Raman analysis ($\lambda = 514$ nm) to identify the structure and defect level of carbon fibers. A D-band (disorder band) appears in the defect structure of carbon at around 1300 cm^{-1} and the G-band of graphite arising from the E_{2g} mode is confirmed at 1580 cm^{-1}. The G-band peak, which has a relatively large peak size compared to the D-band, is generally found in isotropic pitch-based carbon fibers and represents a typical carbon fiber structure. In Raman spectra, R value is used to investigate the crystallinity of carbon materials. The R value is the D peak area divided by the G peak area, so that the degree of increase and decrease of the non-sp2 bond can be understood [28]. In Fig. 3 (a), R value of Pristine CF is 1.81, which is attributed to the sizing process on the CF surface. However, with increasing the time of the supercritical treatment, R value of the CF surface decreased, which is considered that as the supercritical treatment time increases, both the remaining sizing and epoxy materials on the surface of the CF disappears. This is similar to the TGA data in Fig. 3 (b). As a result, as SCF-W reaction time increased, the degree of carbon crystallinity of the R-CFs surface increased and the R value tended to decrease.

Fig. 3(b) shown that the decomposition efficiency of epoxy is approximately 90% when the SCF-W time is around 10 min, and the decomposition efficiency is over 99% when the time is 120 min. In addition, it can be confirmed that the thermal degradation curve for cured epoxy resin, which disappears at around 400 °C after degradation, is in stark contrast to the thermal degradation curve of the samples before treatment. These results are supported by XPS analysis. As shown in Table 1, for Pristine CF, binding energy peaks corresponding to C1s, O1s, and Si are confirmed at around 282 eV, 532 eV, and 101 eV, respectively. Here, pristine CF has carbon accounting for 70.84% of the total composition, oxygen for 17.79%, and Si2p for 11.37%. The Si2p peak is derived from sizing treatment that forms a more stable physical interface by inducing chemical bonding between the carbon fiber surface and resin after coating the carbon fibers with interface bonding agent [29–31].

On the other hand, for the R-CFs after SCF-W treatment time increased, the Si2p peak (sizing effect) gradually decreased about 72.47% from 11.37% to 3.13% was pristine observed. As the treatment time was increased, the composition of C1s increased from 76% to 90.5% and that of O1s from 16.04% to 6.37%, showing that the ratio of oxygen decreases as the ratio of carbon increases. In particular, the O1s/C1s atomic ratio was 0.251 for untreated carbon fibers whereas it was 0.07 after of 120 min of treatment. The reason for this is thought to be the gradual elimination of the sizing agent and the cured epoxy resins present on the surface of the R-CFs.

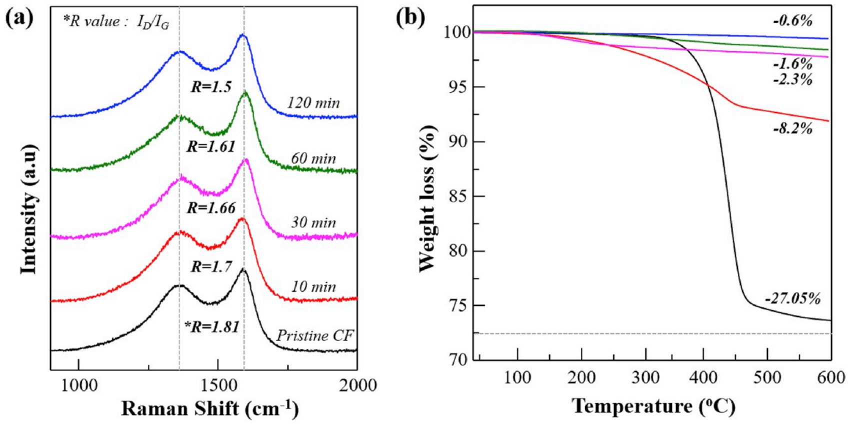


Fig. 3. (a) Raman spectra results for *Pristine CF and R-CF at various supercritical fluid conditions (reaction time), and (b) TGA results for R-CF at various supercritical fluid conditions (reaction time); (red) 10 min, (pink) 30 min, (green) 60 min, (blue) 120 min treatment, and black line is untreated CFRPs. *Pristine CF: Commercial Toray T700 grade carbon fibers treated sizing agent.

Table 1
Atomic composition of pristine and SCF-W treated CF samples, respectively.

Sample code		Carbon (%)	Oxygen (%)	Si2P (%)	*O1s/C1s*
[a]Pristine CF		70.84	17.79	11.37	0.251
SCF-W reaction time	10 min	76	16.04	7.96	0.211
	30 min	81.79	12.39	5.82	0.151
	60 min	81.27	13.83	4.9	0.170
	120 min	90.5	6.37	3.13	0.070

[a] Pristine CF: Commercial Toray T700 grade carbon fibers treated sizing agent.

In summary, we obtained R-CFs with high isotropic structure and high crystallinity by effectively degradation of the cured epoxy resins through SCF-W treatment of CFRPs, and simultaneously eliminating the sizing treatment used to increase interfacial adhesion with the resin on the surface of the carbon fibers. In addition, it is an important issue that degraded polymers produced by recycling process of CFRPs are recovered and reused. Therefore, the residues after SCF-W treatment for 120 minutes were extracted by ethyl acetated and analyzed using GC-MS chromatography. They contained phenol-like chemicals such as o-Cresol, p-Cresol, m-isopropyl phenols, m-xylene and m-ethyl phenol (see Fig. S4, Supporting Information). As a result, it was confirmed that not only R-CFs but also petrochemical products can be partially recycled in CFRP recycling process by SCF-W treatment [20,32].

Fig. 4 shows the Strain-Stress curve measured by FAVIMAT to investigate the mechanical properties of the R-CFs after degradation. While the tensile strength and modulus of elasticity of pristine CFs were 4.5 ± 0.68 GPa and 277 ± 20.3 GPa, respectively, the tensile strength of the R-CFs obtained by SCF-W was 2.9–3.7 GPa, a reduction of 18–36%. The modulus of elasticity was 221.3–257.7 GPa, a reduction of 7.2–20.2%. These results are expected from the analysis results of Raman, TGA and XPS. That is, is the degradation of strength is due to the elimination of cured epoxy resin and sizing agent on the surface of carbon fibers through SCF-W treatment, and the mechanical properties of the treated material are similar to those of T300 carbon fiber [33].

Fig. 5 shows a schematic diagram of the solvent free process, micro-CT image and CBT/R-CFs composite sample of FE-SEM cross-section images in which CBT and R-CFs are compounded to fabricate conductive composites. We checked the distribution of R-CFs and CBT inside the composites before hot-press molding, and Fig. 5 ((a), left SEM image) shows that R-CFs with a length of approximately 100–150 μm are 3-dimensionally and randomly mixed with CBT. The thermal and electrical properties of R-CFs composite samples are known to depend largely on their pore content, the amount of R-CFs and the interface properties between R-CFs and resins, as well as the physical properties of the raw materials. In order to evaluate the pores in the carbon fiber composite samples, and therefore the quality of impregnation of R-CFs with CBT resins, we examined the surface and the cross-section of the samples by FE-SEM and confirmed that R-CFs were uniformly distributed in the CBT matrix as shown in Fig. 5(b). In addition, we analyzed the internal structure of the samples using 3D C-Scan (Micro-CT), a nondestructive analysis technique, to evaluate the porosity of the area for the entire composite sample, and the result shows that there were no pores larger than 1 μm (Fig. 5(c) and (d)). We added R-CFs at 1, 3, and 5 wt% of CBT weight, manufactured samples of this composite, and then investigated their thermal and electrical properties. As the amount of added R-CFs increased from 1 wt% to 5 wt%, the thermal conductivity increased by 189% from 0.45 ± 0.01 to 1.35 ± 0.05 W/mK, and increased by approximately 1250% compared to CBT (0.1 ± 0.05 W/mK). And the electrical conductivity also increased from 3.52×10^{-7} to 1.23×10^{-6} S/cm (see Table 2).

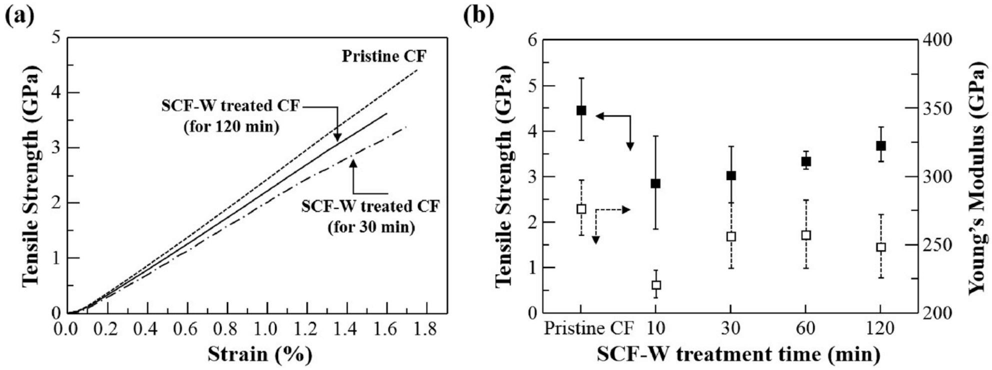


Fig. 4. Mechanical properties of single fiber of Pristine CF and R-CF at various supercritical fluid conditions: (a) strain-stress curves, and (b) tensile strength and modulus specimens, respectively.

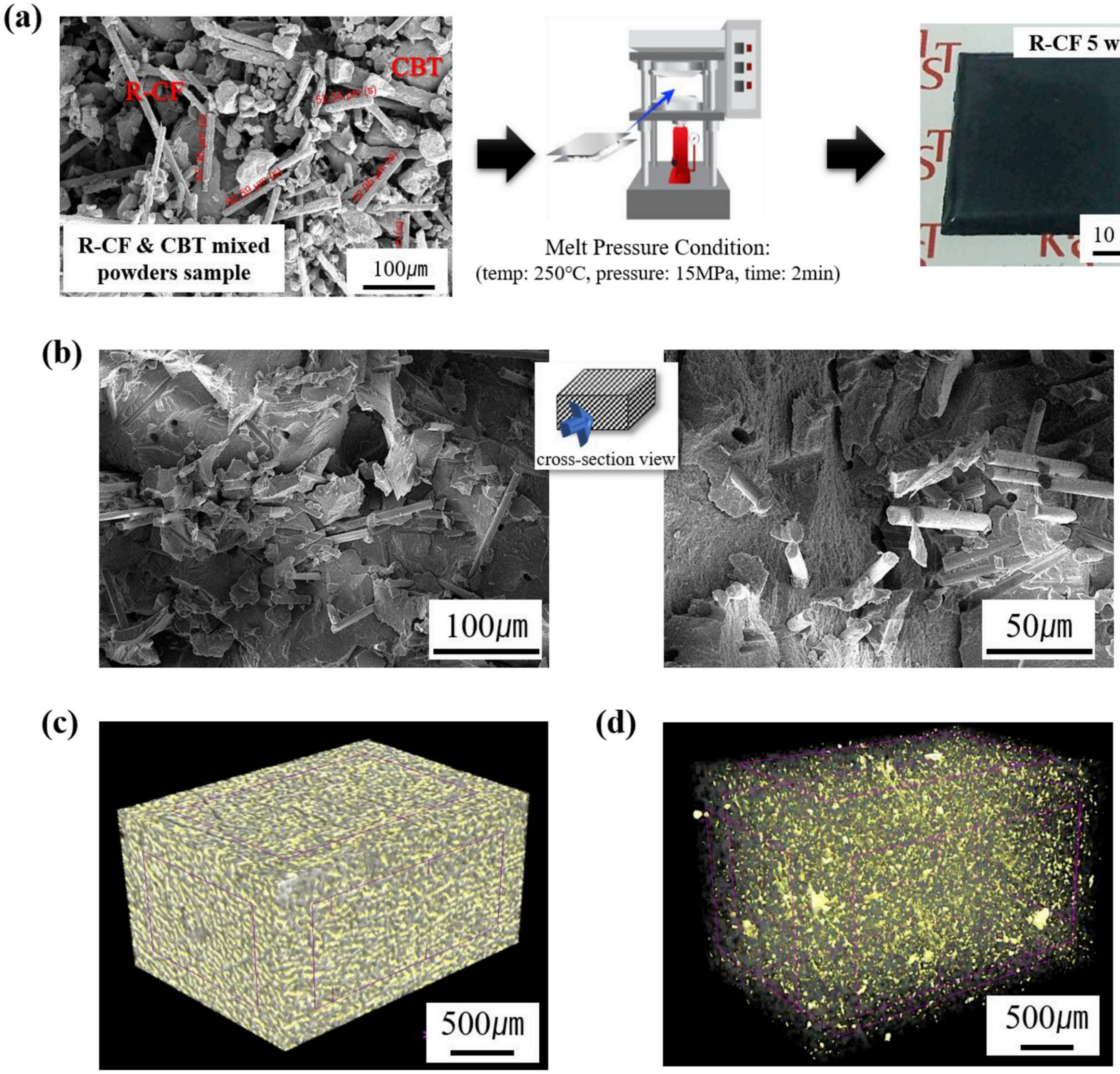


Fig. 5. (a) Schematic diagram of the solvent free process based on simple powder mixing and in-situ polymerization of cyclic butylene terephthalate (CBT) oligomers for preparation of R-CFs -polymer composites, (b) cross-section SEM images, and (c–d) micro-CT scan image; (c) original images, and (d) image before CBT removal of CBT/R-CF composites, respectively.

Table 2
Thermal conductivity and electrical conductivity of CBT/R-CFs composite at different R-CFs concentrations.

	CBT/R-CF 1 wt%	CBT/R-CF 3 wt%	CBT/R-CF 5 wt%
Thermal conductivity (W/mK)	0.45 ± 0.007	0.98 ± 0.003	1.35 ± 0.05
Electrical conductivity (S/cm)	3.52×10^{-7}	8.51×10^{-7}	1.23×10^{-6}

4. Conclusion

We recycled CFRPs with a decomposition efficiency of 99% or more using only water without any catalyst or oxidant after degradation of CFRP under supercritical condition. We then compounded the R-CFs with CBT to fabricate conductive R-CFs composites, with thermal (1.35 ± 0.05 W/mK) and electrical (1.23×10^{-6} S/cm) properties through a relatively simple process using a hot press. Through this study, we propose an eco-friendly recycling method for reusing carbon composites that are currently mostly landfilled or incinerated, and in the future this process could be applied for zero-waste low-carbon resource recycling that reduces pollution.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.compscitech.2019.01.026.

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