Recycling of Carbon Fiber Reinforced Polymers in a Subcritical Acetic Acid Solution

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Abstract

A novel single-stage solvolysis process is demonstrated for recycling carbon fibers from an epoxy-based composite material using 50 wt% acetic acid solution under subcritical conditions. The process yields 100% recovery efficiency for fibers in less than 30 min at 300 °C. Qualitative SEM / EDS analysis of the fibers reveals that the recovered fibers are entirely free of resin, while the carbon fiber surfaces were not damaged. SEM images and gravimetric measurements of the composites treated at lower temperatures and short residence times show an initial increase in mass of the CFRP samples, suggesting a two-step process, consisting of initial composite swelling due to uptake of solvent, followed by depolymerization and chemical decomposition of the polymer. FTIR and GC-MS analyses confirm resin decomposition and production of aromatic and aliphatic compounds, which could potentially be recovered for re-use.

Keywords: Carbon Fiber, Recycling, Solvolysis, Acetic Acid, Subcritical

Introduction

The use of composite materials can be dated back to the Mesopotamian era, where people glued strips of wood at different angles and multiple layers to create plywood. Today, composite materials are used in automotive and aerospace industries, sports equipment, and other consumer products. Widely used in the aviation industry, carbon fiber reinforced polymer (CFRP) primarily consists of two components: the matrix and the reinforcement. The carbon fiber is used as reinforcement, providing strength to the composite. The matrix is generally a resin polymer or epoxy, which binds the carbon fiber. The two components are combined in a controlled proportion and process, yielding the desired material properties, *e.g.*, a high strength-to-weight ratio. CFRPs achieve tensile strength 3 to 5 times greater than stainless steel while being 30% to 50% lighter. The ubiquitous use of CFRPs raises the questions of their re-use and recycling practices from the perspective of environmental sustainability and quality of the recycled feedstock. [1-4] It is estimated that 30% of the total production is disposed of as waste, including the waste generated during the manufacturing process, constituting the End-of-Life (EoL) for the composites. Therefore, recycling most of the generated waste would significantly benefit both the environmental and economic aspects of the CFRP life-cycle. [2] Recycled fibers can be reintroduced for non-structural applications in various industries such as the automotive industry, sports, and consumer goods.[5-6]

Current CFRP recycling technologies can be broadly categorized into three groups: mechanical, thermal, and chemical processes. Mechanical processes employ a grinding step to physically break the composite material and produce grain-size particles that can be introduced in the manufacturing of CFRP composites. Thermal techniques use high temperatures to vaporize the polymer matrix and leave the fibers as a product that can be recovered. Chemical techniques utilize various solvent-water mixtures that can chemically decompose the polymers, leaving the fibers intact for subsequent collection. Thermal and chemical processes are preferred over mechanical techniques due to their ability to recover long strands of fibers. The mechanical properties of the components manufactured using mechanically recovered fibers have inferior performance due to reduced fiber length. [7]

Thermal recycling of CFRP by pyrolysis requires temperatures in the 450 to 700 °C range and an inert atmosphere to degrade and volatilize the polymer matrix, yielding clean carbon fibers.[8-10] In pyrolysis, the polymer matrix decomposes into two distinct product phases: gaseous low molecular weight (MW) compounds and high MW compounds such as oils and tars. Both product streams face subsequent discharge challenges, as the gaseous phase must be treated to meet emissions requirements, and the liquid phase is often classified as hazardous waste. Pyrolysis also may lead to deposition or condensation of tar and char on the recovered fibers, reducing carbon fiber quality. [9-10] The high MW species can be converted to low MW species via secondary pyrolysis at the higher temperatures or fully carbonized [11], exposing the composite matrix to elevated temperatures can damage the fibers, reducing the mechanical properties of the recycled CFRP composite. [12-14]

Chemical recycling of CFRP, or solvolysis, chemically de-binds the CFs from the polymer matrix by dissolution and depolymerization of the polymer. Acids, bases, alcohols, and other solvents have been explored with varied success.[15-17] Water has been used as a solvent at high-temperature, high-pressure (HTHP) conditions. CFRP recycling has been explored in both subcritical (<374 °C) and supercritical water (SCW) environments, which both can be effective at decomposing organic compounds [18-19] and recalcitrant chemicals. [20] Decomposition rates of organic compounds in SCW are typically faster than in subcritical water conditions [18, 21-22], especially when an oxidant is added to promote oxidation in supercritical water. [23-26]

Several promising chemical CFRP recycling studies used acids and solvents at subcritical conditions. Das et al. [27] used peracetic acid (acetic acid combined with H_2O_2) to decompose CFRP coupons at atmospheric pressure and moderate temperatures. Tensile strength of the fibers was reported as similar to virgin fibers, and SEM / EDS analysis confirmed little-to-no damage of the fibers. This chemical oxidation process at atmospheric pressures may allow for the continuous recycling of fibers. Still, disposal or re-use of the liquid product remains a challenge, and operating costs must be assessed against existing practices. Yu et al. [28] demonstrated that ethylene glycol (EG) could be used to decompose CFRP resin at 160 to 180 °C, and again the fibers retained >95% of the virgin fibers' tensile strength.

Chemical recycling can be used for a wide size range of composite materials. Once the resin matrix is decomposed, the RCFs can be collected and cleaned before re-use. [29-31] Fibers also tend to retain their original shape and tensile strength through chemical recycling. [15, 32-33] Chemical recycling promises the opportunity to recover and re-use the polymer decomposition products, which are captured in the liquid phase. [34-36] An improved understanding of promising CFRP chemical recycling conditions, chemistries, and reaction mechanisms is needed to advance the technology toward wider commercial adoption.

This work demonstrates a novel single-stage solvolysis process for recycling carbon fibers from an unknown epoxy-based resin composite material using 50 wt% acetic acid solution under subcritical conditions. The process yields 100% recovery efficiency for fibers in less than 30 min at 300 °C. The proposed mechanism is a two-step process: (i) initial composite swelling due to solvent uptake, (ii) depolymerization and chemical decomposition of the polymer.

Materials and Methods

Experimental Apparatus. Experiments were carried out in a high-pressure batch reactor system, comprising a 316 stainless steel vessel (High Pressure Equipment Co.; Erie, PA), a tube furnace (Barnstead Thermolyne Corp.; Dubuque, IA), and an internal K-type thermocouple to monitor the reaction temperature, as shown in Figure 1. The reactor vessel dimensions are 304.8 mm L x 25.4 mm D, with an internal volume of 50 mL. Fiberglass insulation was used to insulate the reactor vessel while it was placed inside the furnace. A thermocouple data acquisition (DAQ) module with a proportional–integral–derivative (PID) controller was used to set the temperature during experiments.

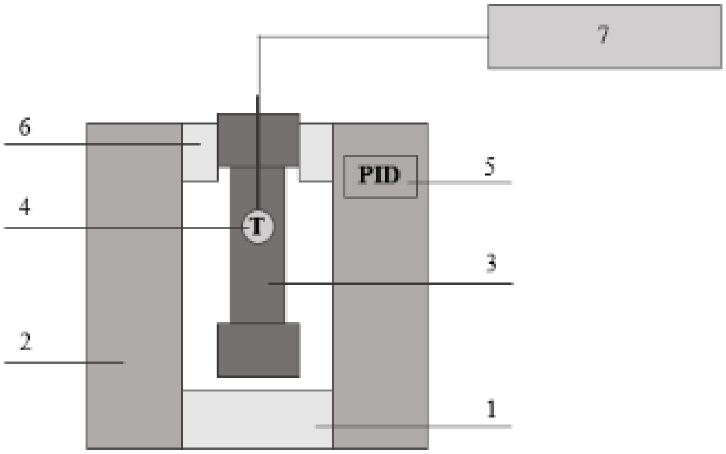

Figure 1. Schematic of a batch reactor system used for CFRP recycling consisting of (1) and (6) insulation, (2) electric tubular furnace, (3) reactor vessel (4) internal thermocouple, (5) internal PID controller, and (7) DAQ

Reagent Purity and Preparation. A thermoset CFRP composite with an unknown composition was used for all experiments. The total fraction of resin in the composite matrix was determined by thermogravimetric analysis (TGA, TA Instruments, Q50), and it was found to be in the range of 26 to 32 % of the total mass of the original CFRP. The composite sample was cut into coupons

50 mm x 10 mm x 2 mm. An aqueous solution of 50 wt% glacial acetic acid (99. %, Fischer Scientific) and 50 wt% DI water (resistivity 18.2 MΩ-cm) was used in all tests.

Data Collection and Analysis. The resin removal rate (R_R) for the composite matrix was calculated based on the mass of CFs recovered at the end of each experiment, as shown below:

$$R_R = \frac{W_{CFRP} - W_s}{W_{CFRP}} \times 100$$

where, W_{CFRP} is the mass of CFRP composite before the start of the experiment, W_s is the mass of the RCFs collected after drying the remained solids.

The chemical composition of the liquid samples was analyzed using ATR-FTIR (ThermoFisher Scientific, Nicolet iS 10 FTIR Spectrometer) and GC-MS (Agilent Technologies, MS: 5973; GC: 6890 with 7683 autosampler). The liquid samples were preprocessed before analyzing in the GC-MS. An aliquot of 5 mL was mixed with 5 mL of dichloromethane (DCM) to extract the organic solvents present in the sample. Sodium hydroxide (ACS reagent, ≥99.0%, anhydrous, granular, Sigma Aldrich) was added to the extracted liquid to remove the water from the organic extracts. The organic compounds in the solution were identified by comparing the mass spectra with the inbuilt NIST database of the instrument.

Experimental Conditions. During each experiment, 35 mL of acetic acid solution and a 5 g CFRP coupon were loaded into the batch reactor and sealed. The reactor was then placed inside the tube furnace, heated to the target temperature (T=200-300 °C), and held for the desired residence time (τ_{res}=0-120 min). The residence time datum (τ_{res}=0) was set when the target experimental temperature was reached in the reactor. On average, the heat-up time was ~80 min. Expansion of the solution during heating resulted in an autogenic pressure rise to ~10 MPa, keeping all reagents in a compressed liquid phase. After the target exposure time was reached, the reactor was cooled rapidly by immersion in cold water until it returned to room temperature. The reactor contents were then collected and filtered using Grade 1 Whatman filter paper (Millipore Sigma, Burlington, MA, US). The liquid consisting of solvent and decomposed resin was stored in a Nalgene vial for further analysis. The recycled carbon fibers (RCFs) were then rinsed with DI water and cleaned in an ultrasonic bath for 15 min in IPA (99.9 %, Sigma Aldrich) to complete the removal of liquid residues from the recovered fibers. The cleaned fibers were then dried in a desiccant dehumidifier for 24 hr. Figure 2 shows the example of the recovered sample after 30 min exposure at 300 °C.

Figure 2: Clean fibers recovered after exposing the composite to a 50 wt% acetic acid solution at 300 °C for 30 min.

Results & Discussion

Analysis of Recovered Fibers. All recovered fibers were dried and weighed to assess resin decomposition during each experiment. Figure 1 shows an image of the recovered fibers from one experiment, while Table 1 shows the resin removal percentage for all conditions in the experimental matrix.

Table 1. Resin removal rate (%) after exposure to 50 wt% acetic acid solution at various temperatures and exposure times. Positive values indicate a mass decrease, while negative values indicate a weight gain, indicating swelling of the polymer matrix.

Temp. (°C)	Resin removal rate (%)				
	0 min	30 min	60 min	90 min	120 min
200	0	-3.96	-3.61	-4.40	-5.99
225	-4.35	-4.49	-6.53	-18.6	-3.92
250	-6.82	-3.39	-2.04	-16.52	3.98
275	-6.00	1.01	15.81	20.92	25.71
300	2.08	28.49	31.92	32.97	31.71

Figure 3 shows the resin decomposition trends at various temperatures. For all residence times at 200 °C, 225 °C, and 250 °C, the overall weight of the RCF coupon increases, with a maximum weight gain after 90 minutes for exposure at 225 °C and 250 °C. This trend is followed by a loss of mass at longer residence times, indicating that the exposed resins undergo an initial weight gain phase, likely due to solvent uptake into the polymer matrix, which has been previously reported.[37-39] After the initial swelling step, we infer that the polymer begins to degrade in earnest, likely due to a combination of decomposition and depolymerization mechanisms. The temperature window from 200 °C and 250 °C show relatively to be a slow swelling process (>60 min), while higher temperatures encourage a faster swelling process, followed by more rapid decomposition and depolymerization, which can be seen in weight loss of coupons exposed to the solution at 275 °C and 300 °C. Very little resin remained after treatment at 300 °C at $\tau_{res}>30$ min. We hypothesize that the swelling process plays a crucial role in the ability of the acetic acid solution to degrade the polymer matrix efficiently, and it may be possible to leverage this effect for more rapid decomposition by soaking the CFRP at a lower temperature before thermal treatment. Interestingly, that heating process ($\tau_{res}=0$ min experiments) results in no mass gain for T=200 °C, about 5-7 % gain for T=225 °C -275 °C, and mass loss when the sample was heated up to T=300

°C. It is apparent this process is temperature-dependent, and it may be that some "activation" temperature is required for the acetic acid solution to penetrate the CFRP matrix.

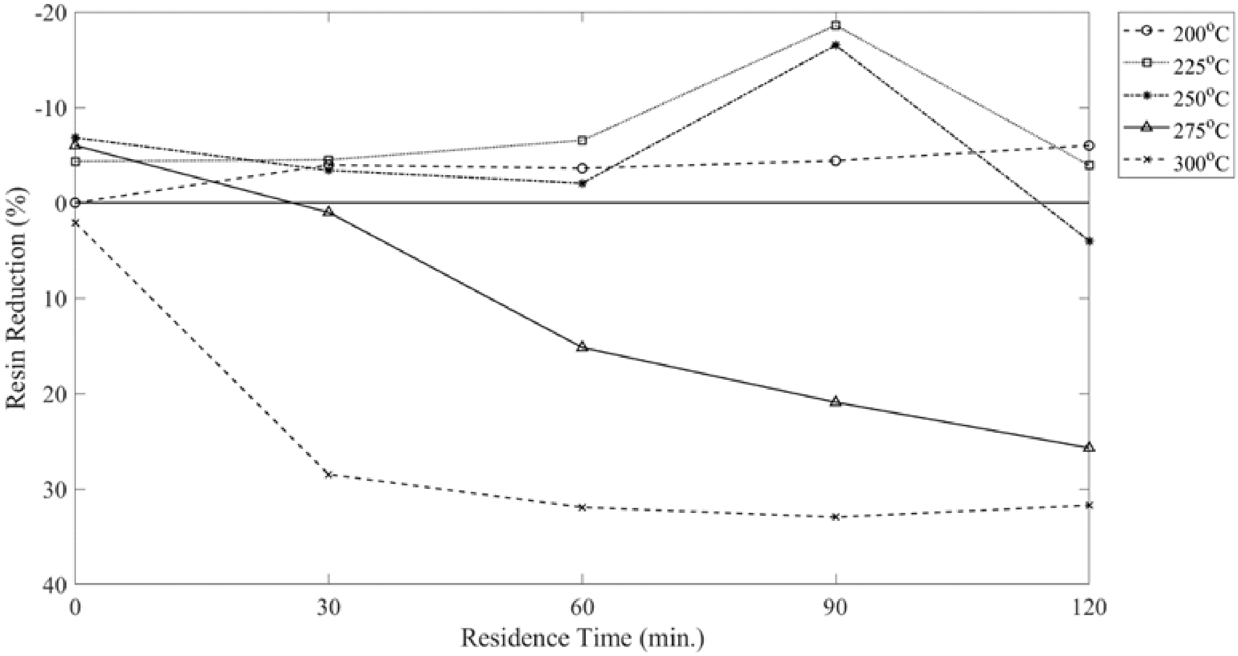

Figure 3. Resin removal rate at various experimental temperatures. The lower temperature experiments show sample mass gain before the weight reduction at the longer residence times.

SEM images of the composite matrix and recovered fibers indicate that 100% resin removal is achieved after exposure to the 50 wt% acetic acid solution at 300 °C. Figure 4 shows SEM images of the CFRP composite before the exposure, while Figure 5 shows SEM images of the fibers recovered after the 300 °C exposure experiments. EDS analysis detected no other elements present on the recovered fiber surfaces aside from carbon.

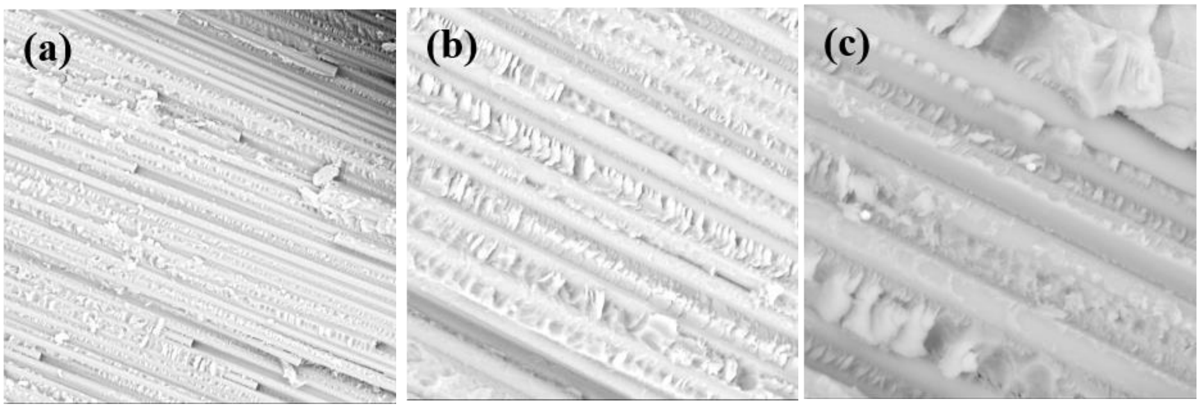

Figure 4. SEM images of CFRP composites before exposure (magnification: 850x, 2250x, and 4000x respectively)

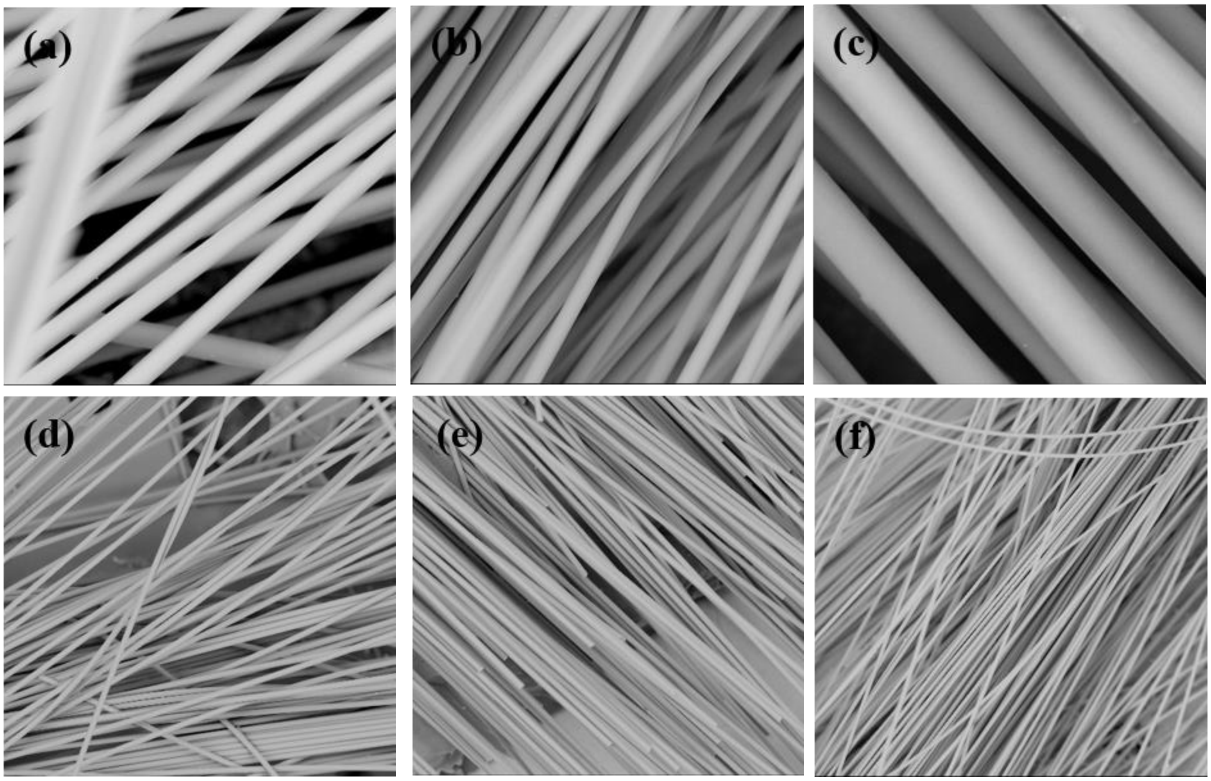

Figure 5. SEM images of recovered carbon fibers after exposure at 300 °C. (a), (b), and (c) show strands of fibers with 100% resin removal (magnification: 2500x) while (d), (e), and (f) show multiple layers of clean fibers (magnification: 590x)

Analysis of Liquid Products. The liquid products from each experiment were analyzed using ATR-FTIR to determine their composition. Figure 6 shows the ATR-FTIR spectra of the liquid products recovered after exposure at 200 °C and 300 °C for 30 min. Peaks at 1390 cm^{-1} and 1703 cm^{-1} indicate the presence of phenolic, carbonyl, and aliphatic ketones in the recovered liquids.[40-42] The notable absence of an absorption peak in the C-N bond region at 1110 cm^{-1} seemingly suggests that the polymer decomposition may initiate through C-N bond cleavage. Higher temperature treatment seems to indicate the formation of more C=O bonds via the peak at 1703 cm^{-1}. Additional peaks corresponding to alkyl aryl ethers, amines, and aliphatic nitro compounds can be observed at 1265 cm^{-1}, 1640 cm^{-1}, and 1370 cm^{-1}, respectively. The formation of species with similar functional groups has been previously reported in the degradation of DGEBA epoxies which are generally used for aerospace applications.[27-28, 40-43]

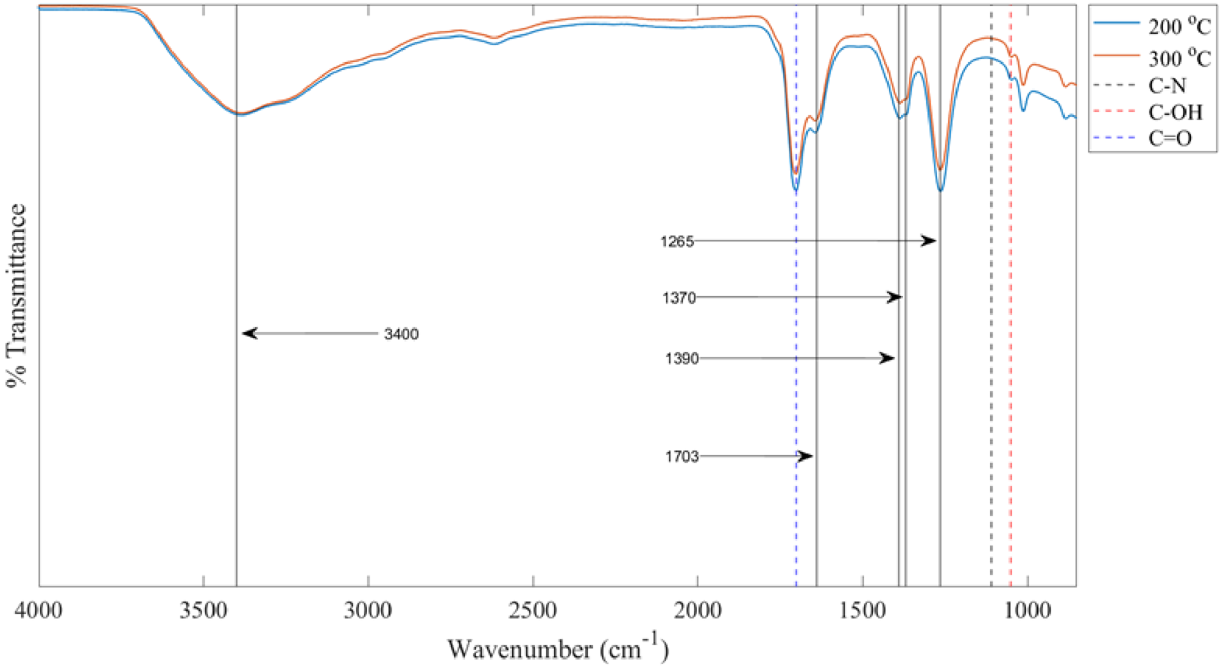

Figure 6. ATR-FTIR spectra of liquid products from CFRP decomposition experiments in 50 wt% acetic acid solution at 200 °C (blue) and 300 °C (red)

Conclusions

The recycling of aerospace-grade CFRP composite material in hot-compressed 50 wt% acetic acid solution is demonstrated. Decomposition is witnessed to be a function of exposure time and temperature, with temperatures between 200 and 250 °C promoting acetic-acid-induced swelling of the polymer and temperatures above 250 °C promoting more rapid polymer degradation and depolymerization. A 100% resin decomposition and fiber recovery are demonstrated at 275 °C (residence times ~120 min) and 300 °C (residence times ~30 min). Analysis of the liquid products reveals the presence of aromatic and aliphatic compounds, which could potentially be recovered for re-use.

ASSOCIATED CONTENT

Supporting Information

None

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Author Contributions

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ABBREVIATIONS

ATR-FTIR, attenuated total reflection Fourier transform infrared; CF, carbon fiber; CFRP, carbon fiber reinforced polymer; DAQ, data acquisition; DCM, dichloromethane; DGEBA, diglycidyl ether of bisphenol A; DI, deionized; EDS, energy dispersive X-ray spectroscopy; EoL, end-of-life; FTIR, Fourier transform infrared; GC-MS, gas chromatography - mass spectrometry; H_2O_2, hydrogen peroxide; HTHP, high-temperature, high-pressure; IPA, isopropyl alcohol; NMR, nuclear magnetic resonance; PID, proportional integral derivative; RCF, recycled carbon fiber; SCW, supercritical water; SEM, scanning electron microscope; SS, stainless steel; TOF-MS, time of flight - mass spectrometry; UD, unidirectional

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