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Fabrication and properties of composites utilizing reclaimed woven carbon fiber by sub-critical and supercritical water recycling

Chase C. Knight, Changchun Zeng*, Chuck Zhang [1], Richard Liang

High-Performance Materials Institute, Florida State University, Department of Industrial & Manufacturing Engineering, FAMU – FSU College of Engineering, Tallahassee, FL 32310, USA

HIGHLIGHTS

- Sub-critical and supercritical water is highly efficient for composite recycling.
- 12-layer aerospace grade carbon fiber composites were recycled in woven form.
- Multi-layer composites were fabricated using the recycled woven fiber.
- Recycled-fiber composites maintained 80–95% of original flexural strength.
- Reuse of the reclaimed matrix polymer is feasible with this recycling technology.

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ABSTRACT

Supercritical fluid recycling has emerged as an appealing method for recycling carbon fiber reinforced plastics (CFRP). Under supercritical conditions, the high reactivity, low viscosity and high diffusivity of water greatly facilitate the efficient degradation of the polymer matrix to allow the harvesting of clean and mostly undamaged fibers. We previously reported the successful use of supercritical water recycling to recover carbon fibers from high-performance single-layer composites and possibly multi-layered composites. The fibers are reclaimed in the original woven architecture, which is beneficial for direct use for reclaimed-fiber composites. In this study, the fabrication of reclaimed-fiber composites (RFC) was investigated using fibers recycled from aerospace-grade IM7/8552 (Hexcel) 12-layer composites. Two fabrication methods – hand lay-up and vacuum infusion – were attempted. The recycled matrix materials were also combined with fresh resin and cured. The reclaimed-fiber composites exhibited 80–95% of flexural strength of virgin carbon fiber composites. This paper also discusses the manufacturing issues associated with the reuse of reclaimed materials.

1. Introduction

The carbon fiber market is forecast to grow at an annual rate of 17% over the next five years, reaching an estimated 118,600 tons (value of $7.3 billion) by 2017. By extension, the carbon fiber reinforced plastics (CFRP) market is predicted to grow at a rate of 16% annually from 2012 to 2020 [1]. The utilization of CFRP continues to increase rapidly in several industries. As such, there is an increased concern regarding the disposal of these materials at the end of their life cycle. As these waste disposal problems arise, increasing emphasis is being placed on the necessity to recycle CFRP.

Landfill disposal is currently the main option for waste CFRP. However, the same superior properties of chemical and environmental resistance that make the CFRPs appealing for many applications also make these materials nearly impossible to be broken down by natural means. To address environmental concerns and to possibly reclaim expensive CFRP materials, researchers are seeking to develop suitable CFRP recycling technologies [2]. For example, Adherent Technologies, Inc. (USA) developed a catalytic pyrolysis process that is capable of producing epoxy free carbon fibers with a tensile strength 83–99% of the virgin materials [3–6]. Researchers at the University of Nottingham [7–9] and Harbin Institute of Technology [10,11] have investigated supercritical fluid recycling of unidirectional composites. However, the recycling of higher-valued multi-layered, woven carbon fiber composites has not been reported.

* Corresponding author.
E-mail address: zeng@eng.fsu.edu (C. Zeng).
[1] Current address: H. Milton Stewart School of Industrial & Systems Engineering, Georgia Institute of Technology, Atlanta, GA 30332, USA.

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Under supercritical conditions, water has low viscosity, high mass transfer coefficients and high diffusivity. The dielectric constant is significantly reduced and the hydrogen bonding essentially disappears. The supercritical water behaves similar to organic solvents and has a high solvating power for organic compounds [12]. These properties enable supercritical water to efficiently degrade the polymer matrix, while essentially causing no damage to fibers [7].

Our previous work suggests the feasibility of recycling woven fiber based carbon fiber composites using supercritical water [13–15]. Relatively clean fibers were recovered directly from the multi-layer woven fiber composites, and the woven architecture of the fibers was well preserved, indicating the advantage of direct reuse in this valuable form. Our current study represents a further investigation of the supercritical water (SCW) recycling process and explores the manufacturing issues associated with the reuse of the reclaimed materials. The recovered fibers were directly reused to manufacture composites. A direct comparison was also made between the strength of virgin fibers and the reclaimed fibers in composites. In addition, the reclaimed resin matrix materials were combined with fresh resin then cured, and their mechanical properties were evaluated. To our knowledge, this investigation is the first of its kind and provides critical information in advancing the supercritical recycling technology to a relevant scale suitable for industry.

2. Experimental

2.1. Materials and sample preparation

The composite materials for the first set of experiments were a high-performance, aerospace grade prepreg system comprising Hexcel 8552 resin (38% by weight) and eight-harness satin weave IM7 fabric, supplied by Hexcel Corporation (Stamford, CT, USA). The resin was a toughened, multi-component system. The complete composition is proprietary, but the base components are known to be tetraglycidyl-4,4′-diaminodiphenylmethane (TGDDM) with 4,4′-diaminodiphenylsulfone (DDS) as the curing agent. Twelve layers of prepreg were vacuum bagged and cured in accordance with the manufacturer's specifications: 1 h at 107 °C, followed by 2 h at 177 °C. The cured panels, which were an average of 4.3 mm thick, were cut into 50 × 60 mm samples, as shown in Fig. 1.

A second set of composite samples was fabricated and used for direct comparison of the flexural properties of original fiber composites with those of the RFC. Three layers of Toray T-300 3K plain weave fabrics were vacuum infused with Epon 862 and curing agent W, both provided by Miller-Stephenson. A large panel was cured at 120 °C for 2 h, followed by an additional 2 h at 180 °C, and then cut to samples approximately 60 mm × 50 mm × 0.8 mm.

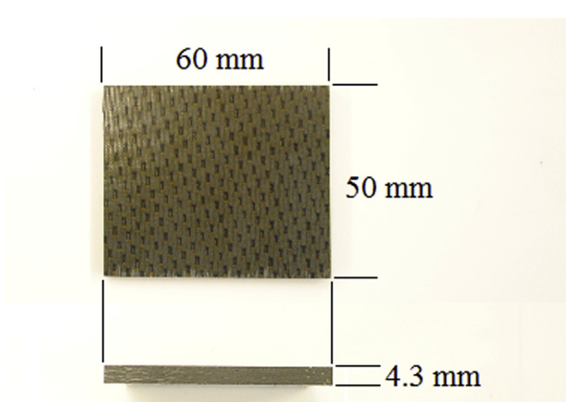

Fig. 1. A typical virgin sample for recycling study.

2.2. Recycling of CFRP and reuse in composite fabrication

2.2.1. Supercritical water recycling

Recycling experiments were conducted using a previously described high-temperature, high-pressure stirred reactor [15]. Three experimental runs were executed using 90 ml of 0.05 M KOH solution at a target temperature of 395 °C. After reaching the target temperature, the reaction was allowed to run at intervals of 15, 30 and 60 min, with the samples and products corresponding to each run were labeled as D1, D2, D3, respectively. The heating up time for each run was approximately 50 min. The process temperatures and pressures were monitored and recorded using a computer, which was connected to the thermocouple and pressure transducer. For each run, the pressure at 395 °C was 27.0 MPa, which is above the critical point of water. At the end of each run, the vessel was rapidly cooled using a series of ice baths. The recovered carbon fiber fabric, which was held together by a wire cage, were thoroughly washed with acetone and then dried.

The second set of virgin composites (Epon 862/T-300) was subjected to the same supercritical water process conditions of 395 °C and 27.0 MPa. A reaction time of 15 min was applied to all three samples to obtain the results in triplicate.

2.2.2. Fabrication of reclaimed-fiber composites

Before being used to fabricate composites, the reclaimed fabric layers were dried for 12 h at 70 °C, weighed, dried for an additional 12 h, then weighed again to ensure thorough drying. Two methods were evaluated for fabricating multi-layer composites by directly reusing the reclaimed fabrics: hand-layup and vacuum infusion. The resin system used was Epon 862 resin – curing agent W (100:26.4), which was cured at 120 °C for 2 h followed by 180 °C for 2 h.

2.2.3. Reuse of reclaimed degradation product

The degraded product from the recycling process was recovered and placed in an oven at 50 °C until the water was completely evaporated of from the mixture. The resulting solids were ground into a powder form and mixed (5% by total weight) with Epon 862 – curing agent W (100:26.4). The mixture was cured at 120 °C for 2 h followed by 180 °C for 2 h.

2.3. Characterization

2.3.1. Resin elimination efficiency and reclaimed-fiber morphology

The resin elimination efficiency is defined as the ratio of resin removed from the fibers during the recycling process to the initial mass of resin in the virgin composite sample. This ratio was estimated by:

$$\mathrm{REE} = \frac{M_i - M_f}{M_r} \tag{1}$$

where $M_r = x_r M_i$, x_r is the prepreg resin weight fraction, M_i is the initial mass of composite before recycling, M_f is the mass of recovered fibers after complete drying.

The samples were taken from the middle and the two outermost layers of the reclaimed fabrics to observe their surface for residual resin or visible damage using a JOEL (JSM-7401 F) scanning electron microscope (SEM). The specimens were mounted on the SEM sample stubs and sputter coated with Au/Pd. Images were acquired at an accelerating voltage of 10 kV.