Review Article

University of Washington URMSE Volume 1 (2020)

A timely review of recent developments in recycling of carbon-fiber-reinforced plastics

Renjie Song

Department of Materials Science and Engineering, University of Washington, Seattle, WA 98195, USA

Article Info

Submitted 25 July 2020

Keywords:
Composites
Carbon-fiber-reinforced plastics
Recycling
Processing and post-processing
Mechanical performance

Abstract

Recycling and reuse of the carbon-fiber-reinforced plastics are of great interest due to their wide adoption, the costly production of the virgin carbon fibers, and the environmental impact of traditional waste treatment approaches. In this review, several selected novel methods for recycling and post-processing of these composites are briefly evaluated for their effectiveness and process parameters.

Corresponding author: Renjie Song (rs30@uw.edu)

1. Introduction

Carbon fiber reinforced plastics (CFRPs) are determined to be able to offer great potential for lightweight structural components due to its high strength to weight ratio and corrosion resistance, as well as high thermal and electrical conductivity. However, the prevalent adoption of CFRPs is hindered by the cost, environmental impact, and lack of reusability [1]-[14]. The production of CFRPs requires a vast amount of energy and non-renewable resources while being a great contributor to CO_2 emissions, thus treating the CFRP waste by traditional incineration, marine dumping, or landfill methods is not satisfactory, not to mention the problems such as the environmental impact, legal issues, and economic losses. To maximize the reuse of the CFRPs, various methods have been investigated and adopted at different degrees since 2019: chemical recycling, thermal recycling, mechanical recycling, microwave decomposition, electrical recycling, and biotechnical recycling, and the major ones being chemical and thermal recycling of the CFRPs [10]. Other than the recycling process parameters, post-treatments were also shown to be effective in improving the performance of the composites made with recycled carbon fibers (rCFs). Such post-treatments usually alter the surface characteristics of the fibers to achieve better adhesion to the polymer matrix and increase the alignment of the rCFs to increase the modulus of the composites [1]-[14].

2. Selected novel processing methods

CFRPs can be recycled chemically, which involves a multi-step process of dissolution, fiber/matrix polymer extraction, and solvent recycling. It was proven by Knappich et al. that using different CreaSolv® CFRP formulations, some CFRP waste can be successfully recycled and reused [10]. The authors' experiment showed that the fiber recycling process with CreaSolv® formulations took roughly 3 hours at temperatures ranging from 140°C to 280°C, depending on the type of the CreaSolv® formulation and the matrix polymer. Additionally, it was predicted that the rate could be accelerated with the production scale[10]. The length of the retrieved fibers was not affected; the matrix polymer, polyamide 6 (PA6), was recovered with high efficiency by vacuum drying at 140°C and 160°C after 4 hours, with minor loss in average molecular weight. All solvents used could be separated from the mixture with vacuum distillation and reused without loss in their dissolving performance [10].

If the CFRP of recycling interest is PA6, other than the respective CreaSolv® Formulation, benzyl alcohol can also be used as the solvent with acetone being the non-solvent for filtrating the rCFs [12]. This process could take 15 hours for dissolution and fiber/matrix polymer reclamation. Despite the long dissolution time, its low processing temperatures, around 160°C for dissolution and 80°C for drying, makes it more energetically preferred and less instrument-demanding. However, a significant amount of the mechanical performance was lost: the recycled CFRPs showed a 39% decrease in the modulus and a 40.4% decrease in the tensile strength, which stayed constant as more rounds of recycling were done. The loss in the mechanical performance was attributed to the misalignment of the fibers, or fiber agglomeration, resulted from the process [12].

According to Kim et al., CFRPs can also be recycled chemically with supercritical fluid water (SCF-W) [8]. Its unique advantages include nontoxicity and economic feasibility compared to other chemical recycling methods of CFRPs. With SCF-W at around 405℃ and 280 bars, all sizing and matrix polymers can be removed from the carbon fibers, and little degradation of the expensive carbon fibers occurs. The epoxy resin examined was recovered with ethyl acetate. It was noticed that the longer the treatment time, the more effective the removal of the sizing and the matrix polymers on the carbon fibers' surfaces, and the higher the carbon crystallinity, accompanied by more degradation of their properties [8].

The parallel thermal method for recycling the CFRPs could be realized with pyrolysis at more elevated temperatures in a furnace [5]. A study showed that the carbon fibers could be retrieved from CFRPs in a furnace with high surface roughness, which could positively affect the coherence between the matrix and rCFs and thus enhance the performance of the remanufactured composites. The minimum temperature required for effectively cleaning the surface of the carbon fibers was found to be 650℃ under a steam environment, any temperature beyond this could lead to the excessive wear of the carbon fibers, lowering the average fiber diameter and impact the performance in a negative fashion. Furthermore, the treatment at 650℃ could leave some residue chemical groups at the surface of the rCFs, which improved the implementation of the rCFs into the new polymer matrix [5]. The optimum process based on this principle took 140 minutes for a batch or 60 minutes for continuous processes excluding the cooling, and retained 65% of the original fiber tensile strength, and 100% of the modulus [5].

The pyrolysis process for decomposing the polymer matrix can also be divided into two stages with different temperatures and gas contents to minimize the energy cost [7]. In the report, Kim et al. declared that the two stages can be optimized to be composed of: (1) using carbon dioxide at 400℃ without holding, and (2) using steam at 700℃ and hold. The tensile strength of the rCFs reached the maximum at 40 minutes of holding at the second stage, with 90% being retained, and the modulus could be kept to the maximum at 102.5% when holding for 100 minutes in the second stage. In the first stage, the epoxy resin was quickly decomposed because of the carbon dioxide; in the second stage, the carbon char was removed by steam and yielded clean rCFs [7].

3. Selected novel post-processing methods

From the study regarding the processing methods for recycling CFRPs, it was found that several parameters of the rCFs, especially the surface morphology, which can be altered by thermal or atmospheric plasma post-treatments, significantly affects the performance of the rCF-based composites [1, 11]. Thermally treating the rCFs at elevated temperatures around 400℃ in nitrogen was proven to increase the performance of the composites made with rCFs due to the formation of aromatic rings at the surface of the rCFs which made them more graphitic and thus increased their modulus. Plus, the presence of more acidic groups due to the thermal

Table 1. Brief conclusion of all the mentioned CFRP recycling methods

Reference	Method Category	Processing Conditions	Substances involved	Notes
[10]	Chemical	140℃ to 280℃, 3 hours	CreaSolv® CFRP Formulations	No loss in fiber length, solvent reusable, PA6 matrix recoverable with little degradation
[12]	Chemical	15 hours, 160℃ for dissolution and 80℃ for drying	Benzyl alcohol, acetone	Significant mechanical performance loss, economic feasibility
[8]	Chemical	405℃ and 280 bars, less than 2 hours	Supercritical fluid water (SCF-W), ethyl acetate	Nontoxicity, economic feasibility
[5]	Thermal	650℃, 140 minutes	Steam	65% of the original fiber tensile strength and 100% of the modulus retained
[7]	Thermal	400℃ without holding, 700℃ hold for 40 minutes	Carbon dioxide, steam	90% of the tensile strength and 102.5% of the modulus retained

treatment decreased the work adhesion of the fibers but improved the adhesion between the fiber and the polar matrix [11].

It was also proven that atmospheric plasma can be used to alter the surface characteristics of rCFs to some degree to reach the optimized adhesion between the rCFs and the matrix polymer, such as polypropylene [1]. By treating the rCFs with 100 W plasma, oxygen-containing groups could be introduced to the surface of the rCFs, that enhance the adhesion and improve the tensile strength and flexural strength by 17% and 11%, respectively. However, more energetic plasma (200 W and 300 W) was shown to be able to over etch the surface of the rCFs, which deteriorated the performance of the rCF-based composites [1].

Other than the surface morphology of the fibers, their alignment is also a critical factor contributing to the mechanical performance of CFRP [12, 13]. To improve the alignment of the fibers, a centrifugal alignment rig, coupled with a dispersion process, can be used to produce aligned rCF mats. Compared to the composites made with randomly aligned rCFs, those made with aligned fibers maintained the tensile strength and had a 37% advantage in the modulus. However, it was noticed that the processing of the sized fibers to align them in glycerol and water had a negative impact on the surface and weakened the bonding between the rCFs and the polymer matrix [13].

4. Conclusions

Several novel methods for recycling the carbon-fiber-reinforced plastics as well as the effects for post-processing of the retrieved high-value carbon fibers were introduced and reviewed, with a brief summary shown in **Table 1**. The importance and the driving force, such as the environmental concern, for recycling these composites were explained. Parameters for characterizing and evaluating the processes, especially the mechanical performance retained, were shown for different recycling approaches as well as post-processing techniques. In conclusion, various novel practices for increasing the reusability of carbon-fiber-reinforced plastics were proven to be environmentally and economically feasible, and further research on fast, high-yielding, high-property-retaining and low-cost approaches are certainly valuable for the wide adoption of the carbon-fiber-reinforced plastics as well as the recycled carbon fibers.

Acknowledgements

The author would like to thank the Society for the Advancement of Materials & Process Engineering UW Chapter for its great composites activities.

Conflict of Interest

The author declares no conflict of interest.

References

[1] L. Altay *et al.*, “The effect of atmospheric plasma treatment of recycled carbon fiber at different plasma powers on recycled carbon fiber and its polypropylene composites,” *J. Appl. Polym. Sci.*, vol. 136, no. 9, p. 47131, Mar. 2019, doi: 10.1002/app.47131.

[2] G. Fortunato, L. Anghileri, G. Griffini, and S. Turri, “Simultaneous Recovery of Matrix and Fiber in Carbon Reinforced Composites through a Diels–Alder Solvolysis Process,” *Polymers (Basel).*, vol. 11, no. 6, p. 1007, Jun. 2019, doi: 10.3390/polym11061007.

[3] W. Guo, S. Bai, Y. Ye, and L. Zhu, “Recycling carbon fiber-reinforced polymers by pyrolysis and reused to prepare short-cut fiber C/C composite,” *J. Reinf. Plast. Compos.*, vol. 38, no. 7, pp. 340–348, Apr. 2019, doi: 10.1177/0731684418822144.

[4] W. Guo, S. Bai, Y. Ye, L. Zhu, and S. Li, “A new strategy for high-value reutilization of recycled carbon fiber: Preparation and friction performance of recycled carbon fiber felt-based C/C-SiC brake pads,” *Ceram. Int.*, vol. 45, no. 13, pp. 16545–16553, Sep. 2019, doi: 10.1016/j.ceramint.2019.05.191.

[5] J. S. Jeong, K. W. Kim, K. H. An, and B. J. Kim, “Fast recovery process of carbon fibers from waste carbon fibers-reinforced thermoset plastics,” *J. Environ. Manage.*, vol. 247, no. July, pp. 816–821, 2019, doi: 10.1016/j.jenvman.2019.07.002.

[6] S. Karuppannan Gopalraj and T. Kärki, “A review on the recycling of waste carbon fibre/glass fibre-reinforced composites: fibre recovery, properties and life-cycle analysis,” *SN Appl. Sci.*, vol. 2, no. 3, 2020, doi: 10.1007/s42452-020-2195-4.

[7] K.-W. Kim, J.-S. Jeong, K.-H. An, and B.-J. Kim, “A Low Energy Recycling Technique of Carbon Fibers-Reinforced Epoxy Matrix Composites,” *Ind. Eng. Chem. Res.*, vol. 58, no. 2, pp. 618–624, Jan. 2019, doi: 10.1021/acs.iecr.8b02554.

[8] Y. N. Kim *et al.*, “Application of supercritical water for green recycling of epoxy-based carbon fiber reinforced plastic,” *Compos. Sci. Technol.*, vol. 173, no. January, pp. 66–72, 2019, doi: 10.1016/j.compscitech.2019.01.026.

[9] M. Kimm, D. Pico, and T. Gries, “Investigation of surface modification and volume content of glass and carbon fibres from fibre reinforced polymer waste for reinforcing concrete,” *J. Hazard. Mater.*, vol. 390, no. November 2019, p. 121797, 2020, doi: 10.1016/j.jhazmat.2019.121797.

[10] F. Knappich, M. Klotz, M. Schlummer, J. Wölling, and A. Mäurer, “Recycling process for carbon fiber reinforced plastics with polyamide 6, polyurethane and epoxy matrix by gentle solvent treatment,” *Waste Manag.*, vol. 85, pp. 73–81, Feb. 2019, doi: 10.1016/j.wasman.2018.12.016.

[11] B. S. Maia, J. Tjong, and M. Sain, “Material characterization of recycled and virgin carbon fibers for transportation composites lightweighting,” *Mater. Today Sustain.*, vol. 5, p. 100011, Sep. 2019, doi: 10.1016/j.mtsust.2019.100011.

[12] R. J. Tapper, M. L. Longana, I. Hamerton, and K. D. Potter, “A closed-loop recycling process for discontinuous carbon fibre polyamide 6 composites,” *Compos. Part B Eng.*, vol. 179, no. April, p. 107418, 2019, doi: 10.1016/j.compositesb.2019.107418.

[13] N. van de Werken, M. S. Reese, M. R. Taha, and M. Tehrani, “Investigating the effects of fiber surface treatment and alignment on mechanical properties of recycled carbon fiber composites,” *Compos. Part A Appl. Sci. Manuf.*, vol. 119, pp. 38–47, Apr. 2019, doi: 10.1016/j.compositesa.2019.01.012.

[14] J. Zhang, V. S. Chevali, H. Wang, and C. Wang, “Current status of carbon fibre and carbon fibre composites recycling,” *Compos. Part B*, p. 108053, 2020, doi: 10.1016/j.compositesb.2020.108053