A Sustainable Approach to the Low-Cost Recycling of Waste Glass Fibres Composites towards Circular Economy

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Abstract: For practical applications, both environmental and economic aspects are highly required to consider in the development of recycling of fibre reinforced polymers (FRPs) encountering their end-of-life. Here, a sustainable, low cost, and efficient approach for the recycling of the glass fibre (GF) from GF reinforced epoxy polymer (GFRP) waste is introduced, based on a microwave-assisted chemical oxidation method. It was found that in a one-step process using microwave irradiation, a mixture of hydrogen peroxide (H$_2$O$_2$) as a green oxidiser and tartaric acid (TA) as a natural organic acid could be used to decompose the epoxy matrix of a waste GFRP up to 90% yield. The recycled GFs with ~92.7% tensile strength, ~99.0% Young’s modulus, and ~96.2% strain-to-failure retentions were obtained when compared to virgin GFs (VGFs). This short microwave irradiation time using these green and sustainable recycling solvents makes this a significantly low energy consumption approach for the recycling of end-of-life GFRPs.

Keywords: glass fibre composites; recycling; circular economy; sustainability; microwave irradiation

1. Introduction

Due to the high strength and stiffness of fibre reinforced polymers (FRPs), they are widely used in a growing number of fields including automotive, aircraft, aerospace, wind turbines, and civil infrastructure [1–5]. The net global glass fibre (GF) market is expected to reach USD 12 billion by 2025, according to a 2019 Global Market Insights, Inc. report. Market reports have also stated that the global GF reinforced epoxy polymer (GFRP) composites market is likely to grow at a steady pace as the material finds varied end-use applications. During the forecast years of 2018 to 2025, the GFRP composites market is expected to register a compound annual growth rate of 9.18% [6]. This growth in the use of GFRP composites will lead to the production of higher amounts of waste [7–10]. Therefore, it is highly necessary to develop a GFRP waste management system dealing with GFs recovery to comply with this legislation [11,12].

Recently, in order to close the loop of GFRPs life-cycle, the complete recycling of GFRPs comprising of the recycling process of both fibres and polymer matrix has been at the centre of attention [13,14]. In contrast to thermoplastics, the recycling of FRPs fabricated by thermosetting resins is always more challenging because of highly cross-linked networks through irreversible chemical reactions, which make them insoluble and infusible in common solvents [9,15]. As yet, various approaches have been industrialized to reclaim GFs from GFRPs which are as follows [16–20]: mechanical recycling...
consists of shredding, crushing, and milling; pyrolysis and fluidized bed process; and chemical recycling. Although mechanical recycling is a cost-effective process, the mechanical performance of the fibres has greatly been ruined. Moreover, only 70–75% strength retention in reclaimed fibres is reported when using the pyrolysis and fluidized bed processes. It should be mentioned that these approaches also consume significant amounts of energy and release highly toxic gases [6]. Although these days environmentally clean chemical approaches are used to digest thermoset resins, in the past, strong oxidation mediums consisting of sulphuric and nitric acids were typically used to recycle the fibres [21,22]. Nowadays, enormous advances in chemical/electrochemical recycling methods have been made to minimize the damage to the fibres during recycling processes and to keep consistency of the fibre architecture [23–25].

Focusing on sustainable and green chemistry aspects [26–30], chemical methods which are based on supercritical solvents in which an organic solvent or water with or without catalysts are the most promising methods. These methods can retain fibre strength up to >90% and generate a limited quantity of by-products and wastes [31–35]. Nevertheless, they are still considered high energy-consuming processes because of the fact that reaching the supercritical status of solvents requires high processing temperatures and extremely high pressures. Therefore, the development of methods based on low temperatures and ambient pressure conditions are still demanding [36]. For the first time, Xu et al. [37] used H\textsubscript{2}O\textsubscript{2} in combination with an organic solvent as an efficient and green oxidiser medium, which can fulfill the supercritical state of the solvents but at lower temperatures and pressures for recycling a carbon fibre (CF) epoxy composite. They achieved recycled CFs with a strength >95% of the original strength and a degradation ratio of >90%. Peracetic acid, a mixture of H\textsubscript{2}O\textsubscript{2} and acid acetic, was introduced by Das et al. [38] to degrade a highly cross-linked amine–epoxy resin system, leaving behind CF with 94% retention of its original strength. However, the potential of these methods for recycling of GFRPs and their effects on the final properties of GFs have not been investigated yet. The concept of microwave irradiation for producing the required heat of the processes in lieu of conventional heating to increase efficiency of reactions has been researched during the past decades, and is confirmed to be more energy efficient and environmentally friendly nature [39,40]. In this regard, Zhao et al. [41] studied the effect of a microwave accelerated catalytic degradation approach in an anhydride–epoxy resin system. Their two-step process consists of diethylenetriamine as solvent to reach the highest degradation ratios in a long period of time. However, the effect of their proposed process on the mechanical properties of the recycled fibres have not been evaluated.

Here, we have developed a cost-effective, fast, efficient, and sustainable chemical method for degradation of a high-performance GF/amine–epoxy composite waste and recycling of its GFs under microwave irradiation. Tartaric acid (TA), which is a natural organic acid, in combination with H\textsubscript{2}O\textsubscript{2} was employed as a mild and green oxidiser medium. In this process, the green and sustainability aspects are reflected in using “safer solvents”, “renewable feedstocks”, and a “design for energy efficiency” [42]. The effect of various processing parameters on the degradation ratio of the GFRPs and reclaimed GF properties have also been evaluated.

2. Experimental

2.1. Materials

Tartaric acid (TA) and hydrogen peroxide (H\textsubscript{2}O\textsubscript{2}, 50 wt %) were provided by Paramount Chemicals (Melbourne, Australia). A viscose epoxy resin (RIM935 grade) and an aliphatic diamine based liquid curing agent (RIM936 grade) were supplied by Hexion Inc. (Columbus, OH, USA). Waste composite panels (WCP) with a 5 mm thickness, fabricated using vacuum resin infusion technique using woven GF fabrics (Hexcel, Washington, USA) and cured at 120 °C for 2 h, were kindly provided by Ironbark Composites, Torquay, Australia.
2.2. Recycling Process

To produce the recycling solvent, tartaric acid (TA) and hydrogen peroxide (H\textsubscript{2}O\textsubscript{2}, 50 wt %) were mixed in different ratios by a magnet stirrer at room temperature for 30 min. A waste glass fibre reinforced epoxy–amine composite panel (WCP) piece with dimensions of approximately 5 cm × 2 cm × 5 mm was immersed in the TA/H\textsubscript{2}O\textsubscript{2} solvent and irradiated using a conventional microwave (800 watts) for different time durations. After 1.5 min microwave irradiation, the hot mixture was taken out and kept idle for 30 min to simultaneously decompose the WCP and cool down the mixture. For the samples with higher microwave time durations, this process was repeated to reach time durations of 3 and 4.5 min, cooling for 30 min between each step. Two different control experiments using solutions of pure H\textsubscript{2}O\textsubscript{2} and pure TA (5 M) were also performed. Afterwards, the remaining WCP, which mainly consisted of recycled GFs, was washed with acetone in an ultrasonic cleaner for 5 min, followed by drying at 60 °C. The decomposition yield (D\textsubscript{y}) was calculated according to the following Equation (1) [38]:

\[
D_y = 100 (m_i - m_r) / w_i \times m_i
\]

where \(m_i\) is the initial weight of the WCP, \(m_r\) is the weight of remaining WCP after the recycling process, and \(w_i\) is the mass fraction of epoxy resin in the WCP which was determined by TGA. Table 1 presents the conditions of all experiments and their D\textsubscript{y} values.

| Sample | H\textsubscript{2}O\textsubscript{2}/TA (v/w) | Time (min) | D\textsubscript{y} (%) |
|--------|-----------------------------------------|------------|---------------------|
| RGFs1  | 1                                       | 1.5        | 64                  |
| RGFs2  | 1                                       | 3          | 73                  |
| RGFs3  | 1                                       | 4.5        | 81                  |
| RGFs4  | 2                                       | 1.5        | 84                  |
| RGFs5  | 2                                       | 3          | 90                  |
| RGFs6  | 2                                       | 4.5        | 92                  |
| RGFs7  | 3                                       | 1.5        | 85                  |
| RGFs8  | 3                                       | 3          | 92                  |
| RGFs9  | 3                                       | 4.5        | 93                  |
| RGFs10 | 3                                       | 6          | 93                  |
| RGFs11 | H\textsubscript{2}O\textsubscript{2}     | 4.5        | <3                  |
| RGFs12 | TA (5M)                                 | 4.5        | <3                  |
| RGFs13 | 3                                       | 360 \*     | 67                  |

\* Conventional heating at 70 °C under reflux.

2.3. Characterizations

GFs tensile properties were measured using a Favimat + Robot 2 single fibre tester (Textechno H. Stein, Bavaria, Germany). The force–elongation data for individual fibres (at least 75 single fibres) loaded into a magazine were automatically recorded. Tensile load–elongation curves were obtained at a test speed of 2.0 mm/min using a gauge length of 25 mm and a pretension of 0.5 cN/tex according to ASTM D1577 linear density and ISO 11566 for fibre tensile test. The Weibull modulus was calculated based on Weibull theory [43,44]. Thermogravimetric analysis (TGA) was performed under a nitrogen atmosphere with a heating rate of 10 °C/min from room temperature to 800 °C holding for 30 min. Fourier transform infrared (FTIR) analysis was performed by a Bruker Vertex 70 FTIR spectrometer. SEM imaging was performed by a scanning electron microscope (ZEISS Supra 55 SEM VP, Oberkochen, Germany) operated with a 5 kV accelerating voltage.
3. Results and Discussion

3.1. Recycling Process Optimization

The proposed recycling process in which H$_2$O$_2$ and TA were used as a green recycling solvent is schematically presented in Figure 1. Upon microwave irradiation, reaction temperature increased which led to the self-accelerated recycling process. During the cooling process, fibres were gradually separated into individual filaments. After 1.5 min microwave irradiation, the temperature increased to 100 °C, which provided enough energy for reaction of TA with H$_2$O$_2$. Various H$_2$O$_2$/TA ratios and microwave irradiation time durations were evaluated to optimize the process and obtain high values of $D_y$. As presented in Table 1, when the H$_2$O$_2$/TA ratio increased from 1 to 3 at a constant microwave irradiation time, $D_y$ increased gradually. For instance, for 3 min microwave irritation time, $D_y$ increased from 73% to 92% when H$_2$O$_2$/TA ratio increased from 1 to 3. More importantly, increasing microwave irradiation time up to 4.5 min increased $D_y$, reaching a $D_y$ of 93%. After 4.5 min time, a higher $D_y$ could not be achieved by increasing irradiation time, (e.g., 6 min, RGFs10 sample) and it remained as high as 93% for an H$_2$O$_2$/TA ratio of 3 and microwave irradiation time of 6 min. The pure H$_2$O$_2$ and pure TA solutions (5 M) were also applied as recycling solvents (samples RGFs11 and RGFs12, respectively), recording $D_y$ values of <3%. This implies that only the reaction of TA and H$_2$O$_2$ under microwave irradiation provides an oxidation medium suitable for epoxy matrix decomposition.

It can be discussed that the reaction of H$_2$O$_2$ and TA can produce various reactive species (e.g., organic peroxides) which can decompose epoxy resin possibly through C–N bonds cleavage [23,33,37,38]. Moreover, to investigate the role of microwave irradiation, an H$_2$O$_2$/TA solution without WCP was also treated under microwave irradiation, which can only raise the temperature up to ~70 °C within 1.5 min irradiation. This shows that WCP is a microwave absorbing material that can transfer the microwave energy to the H$_2$O$_2$/TA solution, leading to the increase of temperature over a short period of time. The effect of conventional heating on $D_y$ was also studied (RGFs13 sample). At an H$_2$O$_2$/TA ratio of 3, a $D_y$ of only 67% was obtained after 360 min, which is a very long time compared to 4.5 min microwave irradiation. Three samples of RGFs5, RGFs6, and RGFs9 which have maximum $D_y$ were chosen for further analyses.

Figure 1. Schematic presentation of the recycling process of WCP under microwave irradiation to recover its glass fibres (GFs) using a green solvent containing H$_2$O$_2$ and TA.

Figure 2a demonstrates the TGA thermogram of WCP in which a 16% weight loss at 800 °C was observed. This denotes that the WCP consisted of ~84 wt % GFs (equal to ~72% volume fraction) and ~16 wt % epoxy polymer, while the TGA thermogram of virgin GFs (VGFs) showed only a minor weight loss of ~1.5% at 800 °C. However, RGFs5, RGFs6, and RGFs9 demonstrated higher weight losses, showing higher polymer residues remaining on the fibres. A rational correlation between weight loss of the various samples and their $D_y$ can be understood according to the $D_y$ values. In other words, a lower $D_y$ leads to a higher weight loss due to higher amounts of polymer residue on the fibre surface. However, a higher $D_y$ might also result from slight etching on the surface of the GFs due to
the reaction of $\text{H}_2\text{O}_2$ with the surface, leaving behind sodium hydroxide [45]. Figure 2b shows FTIR of VGFs and RGFs5. VGFs exhibited typical peaks related to the structure of the GFs (e.g., 1020 cm$^{-1}$ (Si–O–Si) and 3550 cm$^{-1}$ (Si–OH)). Apart from the Si–O–Si structural peak, new peaks at 3400 cm$^{-1}$, 2860–2970 cm$^{-1}$, and 840 cm$^{-1}$ appeared in FTIR spectra of RGFs5. These wavelengths are respectively associated with Ph–OH/C–OH, C–H, and C–O–C [46,47], which are the main adducts of epoxy matrix which remained on the surface of the GFs.

![Figure 2](image-url)

**Figure 2.** (a) TGA thermograms of the various samples including virgin GFs (VGFs), different recycled GFs, and WCP; and (b) FTIR spectrums of VGFs and RGFs5.

Figure 3a,b depicts SEM images of WCP before and after the recycling process, respectively. Before recycling of WCP, the GFs were completely covered by epoxy polymer. After the recycling process, no resin-rich area was observed on the GFs (Figure 3b), denoting successful recycling of WCP to remove the epoxy polymer. The SEM images of the VGFs surface (Figure 3c,d) present a clean and smooth surface. However, SEM images of RGFs5 (Figure 3g,h) demonstrate a relatively rough surface consisting of some minor polymer residue particles on its surface.

![Figure 3](image-url)

**Figure 3.** SEM images of the WCP (a) before and (b) after recycling process, and SEM images of (c,d) VGFs and (e,f) reclaimed RGFs5.

3.2. Mechanical Properties of Recycled GFs

Mechanical properties of the recycled GFs were evaluated using tensile testing of the single filaments to investigate the effect of the proposed process. Typical stress–strain curves of the VGFs and various recycled GFs are presented in Figure 4a. Using these curves, different parameters such as
tensile strength, Young’s modulus, strain-to-failure, and Weibull modulus were obtained. As presented in Figure 4b–d, VGFs showed 2.87 ± 0.75 GPa tensile strength, 83.66 ± 3.61 GPa Young’s modulus, and 3.73% ± 1.07 strain-to-failure. The closest values for these properties were for the RGFs5 sample with 2.66 ± 0.82 GPa tensile strength, 82.69 ± 4.92 GPa Young’s modulus, and 3.68% ± 1.15 strain-to-failure. Compared to VGFs, RGFs5 showed only a 7.3% reduction in tensile strength, a 1.0% reduction in Young’s modulus, and a 1.3% reduction in strain-to-failure. These insignificant reductions indicate that the recycling process using the H₂O₂/TA solution under microwave irradiation did not significantly affect the mechanical performance of the GFs and the entire process only decomposed the cured amine–epoxy system. Although RGFs6 and RGFs9 samples had slightly higher Dₚ values compared to RGFs5, their mechanical performance reduction was higher than RGFs5, probably due to applying longer microwave irradiation times in their processes.

Figure 4. (a) Stress–strain curves, (b) tensile strength, (c) Young’s modulus, and (d) strain-to-failure values of the various GFs according to Table 1.

Mechanical properties of the GFs are a multiplex parameter because each GF bundle consisted of thousands of single filaments and it is not precise to describe them only by the single tensile strength values. In this regard, the Weibull model provides a useful tool to understand the intrinsic statistical nature of the fracture strength [48]. The Weibull modulus in this model is a determining parameter which can control the length dependence of the average fracture stress and bundle efficiency, reflecting the shape of the defect population in the GFs [48]. Using Weibull plots presented in Figure 5a, Weibull modulus can be calculated (Figure 5b). Despite reduction in tensile strength, Young’s modulus, and strain-to-failure of RGFs5 compared to VGFs, there was no statistical difference between Weibull modulus of RGFs5 and VGFs. However, RGFs6 and RGFs9 samples showed lower Weibull modulus compared to both RGFs5 and VGFs.
4. Conclusions

In GFRP composites waste recycling, it is crucial to develop low cost approaches for recycling the GFs, otherwise it will not be beneficial and affordable since commercial GFs are not very expensive reinforcing fibres. When it comes to the cost of recycling processes, the combination of time, temperature, and pressure always leads to a higher cost. Figure 6 compares outcomes of our presented approach with other approaches already reported in the literature. In chemical approaches, often long-term heat treatment and/or high pressure are required. On the other hand, the pyrolysis approach, which is considered an environmentally unfriendly and high energy-consuming method, is usually a distractive method for the GFs recycling.

In this work, a low cost and rapid process for the recycling of GFs from waste composites was successfully introduced. A microwave accelerated reaction between H$_2$O$_2$ and TA effectively decomposed a highly cured amine–epoxy resin in a waste GFRP. The microwave irradiation was conducted only for three minutes and the recycling process was completed over a 60 min cooling period. The yield of this process was as high as 90%, leading to obtaining individual GFs. This low energy-consuming approach resulted in the reclaimed GFs presenting only 7.3% reduction in tensile strength, 1.0% reduction in Young’s modulus, and 1.3% reduction in strain-to-failure, when compared to their original commercial GFs.
Figure 6. Proposition of this work compared to other approaches for both GFRPs and CFs reinforced polymers (CFRs) in terms of recycling time and temperature, and tensile strength retention of the GFs (a bigger circle means a higher temperature required for the process).

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