FRPRCS-14, Belfast - June 2019

Characterisation of carbon fibre recovered by pyrolysis using thermal gravimetric analysis (TGA)

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Abstract

Fibre reinforced polymer (FRP) composites are finding increasing number of applications in lightweight structures due to their unique combination of high strength and low weight. Due to the inherent cross-linked nature of matrix in thermoset polymers, they cannot be melted down and remoulded like thermoplastics, makes it hard to recover carbon fibre from the composite. This paper presents the results of a pyrolysis process on fibre reinforced polymer composites for recovery of carbon fibre. The pyrolysis is carried out in a thermogravimetric analyser (TGA) and the effect of different process parameters during pyrolysis on the properties of reclaimed carbon fibres is investigated. The parameter include pyrolysis temperature, heating rate and isothermal dwell time. The characterisation is performed with the focus on the decomposition of the bond between carbon fibres and epoxy adhesive.

Keywords: FRP composite; Carbon fibre; Pyrolysis; Epoxy; Pyrolysis temperature; Thermal gravimetric analysis; Carbon fibre recovery

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Introduction

It has been more than two decades since the application of FRP composites has attracted the attention of civil engineers for construction of new structures or the rehabilitation of existing structural elements. FRP composites have been used in the civil engineering industry since the late 1980s [1,2]. FRP composites get their strength from long, precisely aligned carbon fibres (CFs), fixed within a polymer matrix (e.g. epoxy) that is cured at ambient or high temperatures with or without presence of pressure. Once cured, due to the mixed nature of the composition, CFRP composite cannot simply be melt down and reformed. As a result, most of the waste produced is disposed of in landfill or incinerated [3].
Mechanical recycling is currently the most common method of CFRP recovery. It involves the composite break-down by shredding, crushing or milling, resulting in a mix of powdered and fibrous material. Due to the harming of fibre surface during crushing or grinding, the mechanical properties of recycled fibres are considerably lower than that of virgin carbon fibre (VCF). Thermo-chemical decomposition processes have gained popularity over mechanical recovery methods since they allow the recovery of higher quality fibres. Pyrolysis is one of the most common methods of thermo-chemical processes [4]. In pyrolysis, CFRP material is heated in the absence of oxygen and is broken down into lower molecular weight organic liquid/oils, gases and solids. Carbon fibre recycled with pyrolysis shows strength retention up to 90% of that of VCF which makes it suitable for applications in construction such as facades, noise absorption walls and pipes or even structural elements for the retrofitting of existing structures [5, 6]. However, to produce rCFRP with high grade properties, an optimisation of pyrolysis with regards to its key process parameters (e.g. temperature and dwelling time) is required.
This research investigates the influence of different parameters such as temperature, heating rate and isothermal dwell time on the pyrolysis of CFRP plates by thermogravimetric analysis (TGA). Results of this research can be used for optimisation of the pyrolysis parameters and recovery of CFRP plates which are usually used in construction industry.

Experimental Set-up

Carbon fibre reinforced polymer (CFRP) composites were fabricated from 2-ply, 500 × 500 mm^2 unidirectional carbon fibre sheets via wet lay-up processing technique (Figure 1a). Epoxy resins were mixed according to manufacturer recommendations. A layer of epoxy was applied to a non-stick flat surface and then, carbon fibre sheets were layered over the gel coat. Rollers used to impregnate the fibres and remove excess resin and air pockets. After curing, CFRP composites were cut into coupons and prepared according to specifications of ASTM D3039/D3039M-17 [7]. Epoxy was a two-part resin used to impregnate carbon fibre sheets. The material properties for the carbon fibre and epoxy resin used in this study are summarised in Table 1.

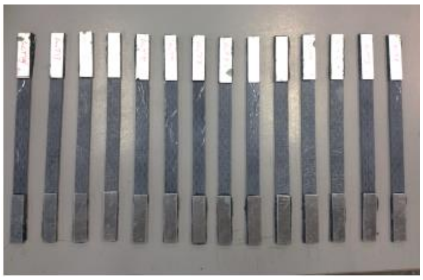
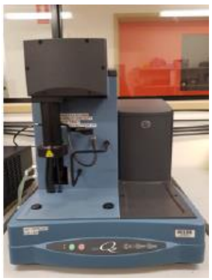
 (a)  (b)

Figure 1: (a) CFRP coupon specimens, (b) TGA Q50 machine

The experiments were carried out in a TGA Q50 (Figure 1b). CFRP specimens were heated until 800 ºC in order to separate the fibre from surrounding epoxy matrix. The inert atmosphere during the pyrolysis was created by nitrogen with a gas flow rate of 60 mL/min. To investigate the effect of temperature rates on the pyrolysis process, samples were tested with three different heating rates: 10 ºC / minute, 20 ºC / minute, and 30 ºC / minute. The initial weight of samples for each heating rate was 11.38 mg, 10.34 mg, and 9.82 mg, respectively. Different isothermal dwell times (10, 20, 30, 40, 50, and 60 minutes) were considered for these tests. Figures 2 illustrates the pyrolysis process that is used in this research.

Table 1: Material properties

	Material Property	**Value**
	Areal weight (g/m^2)	300
	Tensile modulus (GPa)	210
	Tensile strength (MPa)	3,000
	Nominal thickness (mm/ply)	0.12
Carbon fibre samples	Density (g/cm^3)	1.79
	Viscosity - Shear rate (/s)	50
	Tensile modulus (GPa)	4.5
	Flexural modulus (GPa)	3.8
	Tensile strength (MPa)	30
	Glass transition temperature, T_g	+58°C

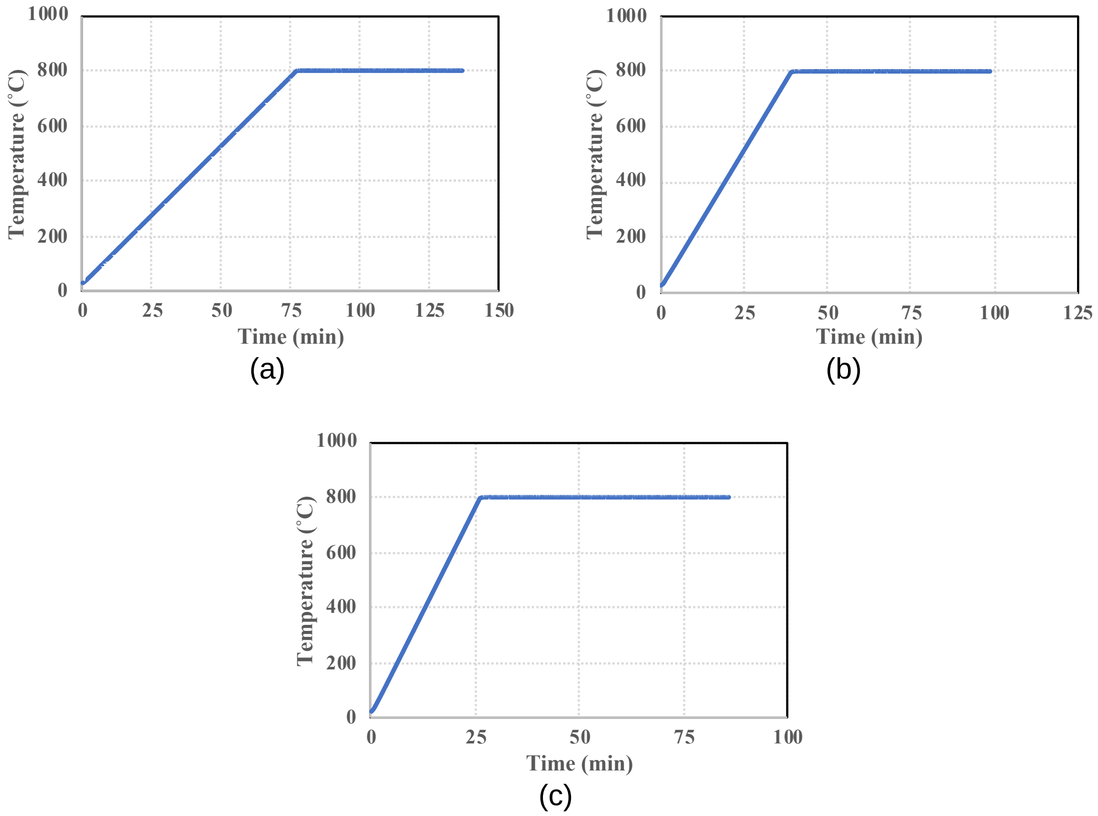
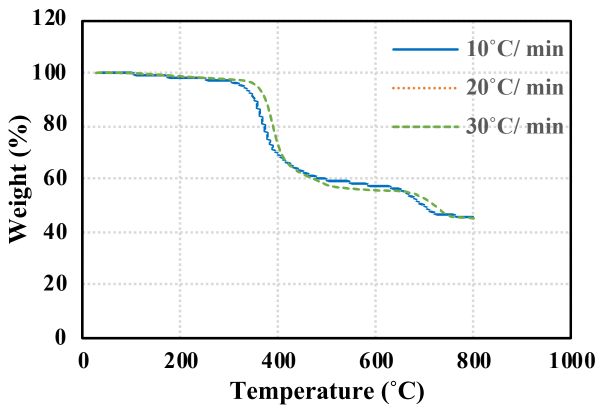


Figure 2: Temperature versus time profile for tests with (a) 10 ºC / min, (b) 20 ºC / min, and (c) 30 ºC / min temperature increase rate

Results of Pyrolysis on CFRP Plates

This section provides the outcomes of the pyrolysis of CFRP plates which are carried out by thermogravimetric analysis. Figure 3 shows the weight changes of specimens for various temperatures during pyrolysis. The decomposition reaction between carbon fibres and epoxy resin is similar for all heating rates. The release of molecules occur at around 350 ºC (370 ºC for the specimen with 30 ºC / min heating rate) which is followed by a rapid weight reduction until 400 ºC. This can be due to the decomposition of the organic epoxy matrix [6]. After 400 ºC, the decomposition slows down until around 650 ºC which another sudden reduction is observed. Above 750 ºC, this decomposition is completed and negligible changes in the sample weight can be observed.



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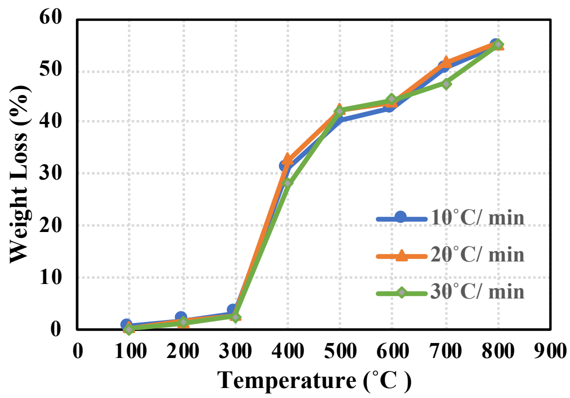

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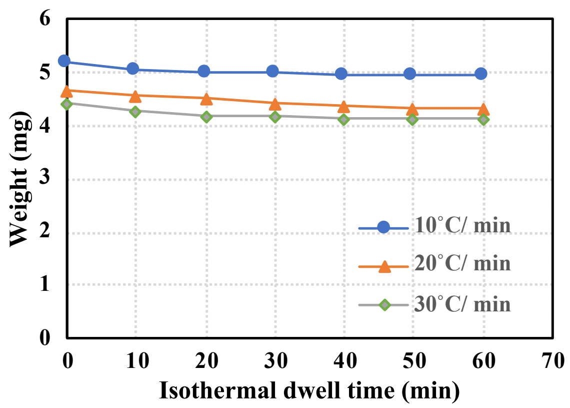
Figure 3: Specimen (a) weight and (b) weight loss versus pyrolysis temperature



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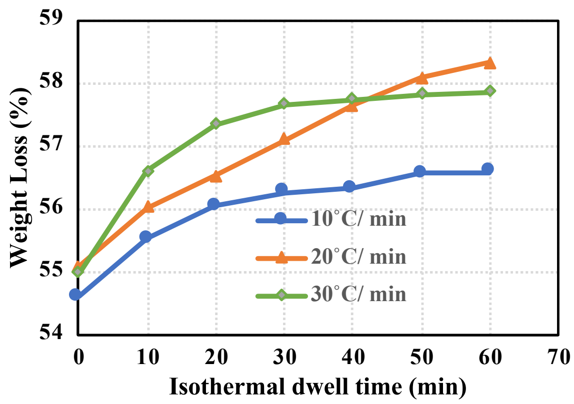

(b)

Figure 4: (a) Weigh and (b) weight loss of specimens over isothermal dwell time

Figure 4 illustrates the effect of isothermal dwell time on the samples weight. It can be seen that after 30 minutes dwell time, there is not significant weight changes for both specimens under 10 ºC / min and 30 ºC / min heating rate. However, the weight reduction is stopped after 50 minutes dwell time for the specimens with heating rate of 20 ºC / min. Further investigations on maximum temperature and isothermal dwell times (Figures 3 and 4) reveal that pyrolysis of the CFRP in inert atmosphere such as nitrogen leads to pyrolytic carbon residues even at high temperatures, i.e. 800 ºC. This indicates that the pyrolysis process leaves char on the fibre which prevents the fibres to develop a good bond with resin matrix. An extra oxidation step at high temperatures is required to post-treat this char and recover clean fibres with a highly activated surface. This post-treating condition may significantly impact physical properties of the fibres. However, it should be also considered while high temperatures (> 700 ºC) can lead to production of a clean fibre, they can severely diminish the tensile properties of recycled carbon fibre, such as tensile strength.

Conclusions

This research presented the outcomes of TGA analyses on CFRP plates processed with wet lay-up manufacturing technique. Results can provide information on optimising pyrolysis method for recovery and recycling of manufactured CFRP products. It was shown that heating rates during pyrolysis do not have significant effect on the weight loss of the CFRP. However, the partial oxidation of pyrolytic carbon from the carbon fibre surface after pyrolysis has to be performed in order to achieve recycled fibres with minimum residue. Clear recyclate carbon fibre with excellent surface characteristics can enhance the bond between recycled fibre with epoxy matrix after remanufacturing which leads to higher mechanical properties.

Acknowledgement

Authors would like to thank the School of Civil Engineering at the University of Sydney for supporting this project. First author would also like to acknowledge the support provided by the Association of Commonwealth Universities (ACU) and the CASS Foundation.

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