# Energy and environmental assessment and reuse of fluidised bed recycled carbon fibres

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## 1 ABSTRACT

2	Carbon fibre reinforced plastic (CFRP) recycling and the reutilisation of the recovered carbon fibre (rCF)
3	can compensate for the high impacts of virgin carbon fibre (vCF) production. In this paper, we evaluate the
4	energy and environmental impacts of CF recycling by a fluidised bed process and reuse to manufacture a
5	CFRP material. A 'gate-to gate' life cycle model of the CFRP recycling route using papermaking and
6	compression moulding methods is developed based on energy analysis of the fluidised bed recycling process
7	and processing of rCF. Key recycling plant operating parameters, including plant capacity, feed rate, and air

8 in-leakage are investigated. Life cycle impact assessments demonstrate the environmental benefits of recycled

- 9 CFRP against end of life treatments-landfilling, incineration. The use of rCF to displace vCF based on
- 10 material indices (equivalent stiffness and equivalent strength) therefore proves to be a competitive alternative
- 11 for composite manufacture in terms of environmental impact.

#### 12 KEY WORDS

13 Fluidised bed CFRP recycling, Compression moulding, Energy analysis, Life cycle assessment

## 14 1 INTRODUCTION

15 Growing demand for carbon fibre reinforced polymers (CFRP) for lightweighting in aerospace applications 16 and, to a lesser extent, automotive applications contributes to fuel efficiency objectives in the transportation 17 sector. In the past 10 years, the annual global demand for carbon fibre (CF) has increased from approximately 18 16,000 to 72,000 tonnes and is forecast to rise to 140,000 tonnes by 2020 [1]. The generation of CFRP-based 19 wastes is correspondingly increasing, arising from manufacturing (up to 40% of the CFRP can be waste 20 arising during manufacture [2-4]) and end of life products/components. CF recovery from wastes is a priority 21 due to the energy intensity and high financial cost of virgin CF (vCF) production. Boeing aims to recycle at 22 least 90% of retired airplane materials [5], which will increasingly require CF recovery in the future. Existing 23 EU regulations aim to reduce the quantities of all wastes sent to landfill [6], while automotive sector-specific 24 policy requires the recycling of at least 85% of end-of-life materials from 2015 [7]. In contrast to industry and 25 policy goals, the vast majority of CFRP waste at present is not recovered: in the UK, for example, up to 98% 26 of composite waste is disposed of in landfill or incinerated [8, 9]. Recovery of metals from end-of-life aircraft 27 has proven to be beneficial in terms of cost and energy intensity relative to virgin material production [10], 28 however, there is no detailed energy and cost information of the CFRP recycling processes. Thus, there is a 29 need to identify an environmentally beneficial recycling technique to address these issues.

30	Recycling techniques take different approaches to recovering fibres from the cross-linked thermoset matrix
31	material, including mechanical size reduction [11] and thermal processes to partially or fully decompose
32	matrix [3]. Pyrolysis is a widely used thermal method, being established in commercial operations, e.g., ELG
33	Carbon Fibre Ltd [12, 13]. A related thermal process is the fluidised bed process, being the subject of this
34	paper, which has been developed for the recycling of glass fibre and CF at the University of Nottingham for
35	over 15 years [3]. Although it has shown a strength reduction of up to 50% [3, 14, 15], this continuous
36	process has been shown to be particularly robust in dealing with varied polymer types containing mixtures of
37	different materials and other contaminants. Very low residual char remains on the fibre surface as organic
38	material is oxidized and any metallic material, such as aluminium honeycomb, rivets etc. remains in the
39	fluidised bed and can be removed by regrading the bed.
40	Prior studies have estimated energy requirements of various CFRP recycling technologies, finding
41	substantially lower energy requirements compared to vCF manufacture. For instance, industry reports claim
42	that recovered CF (rCF) achieves about 95% energy reduction to manufacture compared to vCF while the
43	mechanical performance is comparable [5, 10]. To be specific, recycling energy consumption has been
44	reported to be 0.17-1.93 MJ/kg for mechanical recycling of GFRP [16], 0.27-2.03 MJ/kg for mechanical
45	recycling of CFRP [11], 3-30 MJ/kg for pyrolysis recycling of CFRP [16], 19.2 MJ/kg for solvolysis
46	recycling of CFRP [17] and 60-90 MJ/kg for chemical recycling of CFRP in Japan [18] compared to 198-595
47	MJ/kg for vCF production. However, no recycling capacity or other processing details were specified in most
48	literature, nor the modelling methodology for the energy intensity. Little work was focused on energy demand
49	and environment burden particularly for fluidised bed recycling of CFRP, which needs to be addressed.
50	In addition, to comprehensively assess the environmental performance of CF recycling, however,
51	evaluations should extend beyond the recycling process and account for the reutilisation of rCF in place of
52	current materials. Life cycle assessment (LCA) is an internationally accepted environmental assessing method
53	to quantify and evaluate the environmental impacts such as energy use and greenhouse gas emissions as
	3

54 described by [19]. A number LCA studies evaluating the use of CFRP in lightweighting applications have 55 been conducted, generally finding that weight reductions owing to the use of CFRP to replace conventional 56 materials such as steel and aluminium potentially leads to both energy and greenhouse gas (GHG) emission 57 reduction in either aerospace or automotive industries [2, 20-25]. However, these studies have not considered 58 the end-of-life of CFRP components and therefore do not completely assess environmental impacts. Very 59 few studies have estimated the environmental impacts of a CFRP recycling technology [21, 26], however 60 these have relied on hypothetical data regarding the energy intensity of the recycling process. The lack of data regarding CFRP recycling process inputs and impacts is a barrier to developing informative LCA models. 61 While potential environmental benefits are claimed in technical studies of CFRP recycling processes and fibre 62 63 reuse opportunities, these benefits have yet to be demonstrated in a comprehensive life cycle study. 64 Environmentally-beneficial recycling strategies are essential to support the role of CF-based materials in 65 lightweighting applications to reduce transportation energy consumption. In this study, comprehensive life 66 cycle models are developed to consider the fluidised bed CFRP recycling process and subsequent reuse of 67 rCF in composite materials. Process models of the recycling process and composite manufacture with rCF are 68 developed and validated against pilot plant data. Inventory data (material and energy inputs; direct emissions) 69 are derived from the process models and input to the LCA models. Key environmental impacts (primary 70 energy consumption, global warming potential) are assessed. Environmental impacts of composite production 71 from rCF are estimated and compared with vCF-based composite material.

## 72 **2 METHODS**

This study evaluates the life cycle environmental impacts of CFRP recycling by the fluidised bed process and subsequent manufacturing of composite material from rCF. Results are compared with vCF materials to determine the environmental impacts of using rCF in place of vCF in composite materials and the following composites are considered:

77	1) Recycled CFRP (rCFRP): rCF recycled from fluidised bed process is processed by a wet						
78	papermaking process co-mingled with polyamide (PA) fibre to make a non-woven fabric,						
79	followed by compression moulding with a fibre volume fraction of 25%.						
80	2) Virgin CFRP (vCFRP) 1: vCF co-mingled with PA fibre is manufactured into an intermediate						
81	non-woven mat via wet-papermaking to manufacture CFRP products by compression moulding						
82	with a fibre volume fraction of 25%.						
83	3) vCFRP 2: a prepreg comprising bi-directionally woven vCF and epoxy resin is moulded in an						
84	autoclave to give a fibre volume fraction of 50%.						
85	Key activities included in the study are shown in Fig. 1. Waste CFRP (including scrap and end-of-life						
86	CFRPs), comprised Toray T600SC CF in MTM28-2 epoxy resin, is first shredded and input to the fluidised						
87	bed process, which yields a fluffy fibre product. A co-mingled mixture of rCF and PA fibre is then						
88	manufactured into a non-woven mat via a wet-papermaking process before compression moulding into rCFRP						
89	products. For the vCF materials, we consider two separate manufacturing pathways (see Fig. 1). The first						
90	considers a similar process wherein chopped vCF (Tenax-A HTC124) co-mingled with PA fibre is						
91	manufactured into an intermediate non-woven mat via wet-papermaking to manufacture CFRP products by						
92	compression moulding (vCFRP1). As rCF recovered via fluidised bed exhibit no appreciable degradation of						
93	modulus but strength is reduced by 25-50%, the vCFRP1 material has better mechanical properties than						
94	rCFRP materials as experimentally measured[27]. A second, vCF material typical of high-performance						
95	applications is considered wherein woven vCF (Toray T600SC) is impregnated with epoxy resin and						
96	components are subsequently manufactured via autoclave moulding (vCFRP2).						
97	Process models of the fluidised bed recycling and CFRP manufacturing processes are developed to estimate						
98	the energy and material requirements of commercially operating facilities and are validated using pilot plant						
99	operating data for the fluidised bed recycling process. Direct process inputs and emissions (derived from						
100	recycling and CFRP manufacturing process models) are input to the life cycle model and supplemented with						

101 LCA databases to estimate the impacts of producing and using material and energy inputs (e.g., Gabi

102 Database [28], Eco-invent [29]). The LCA evaluates CF recycling and CFRP manufacture on a "gate-to-gate"

103 basis, corresponding to the activities shown in Fig. 1. Composite use and end-of-life activities are excluded

104 from this study; potential implications of these activities are discussed in Section 3.3. Infrastructure and

105 labour are not included within the study, as is common practice where impacts are expected to be small

relative to the operation of the facilities (e.g., Zhang et al., 2010 [30]).

107 Two environmental impacts are quantified: primary energy demand (PED) in terms of MJ and global

108 warming potential (GWP), based on the most recent IPCC 100-year global warming potential factors to

109 quantify GWP in terms of CO<sub>2</sub> equivalents (CO<sub>2</sub> eq.) [31]. The functional unit for the analysis is one generic

110 CFRP panel which could find use in a range of applications, including automotive and aircraft interior

111 components. For this analysis, a panel size of 300mm x 190mm is selected to correspond to prior

experimental analyses [27]. The component thickness is variable and is adjusted so that equivalent mechanical

113 properties (equivalent bending stiffness and equivalent bending strength) can be met by CFRP produced from

both rCF and vCF. Further details regarding the mechanical properties of the CFRP products are provided in

115 Section 2.5.

116

## 6 2.1 Fluidised bed recycling process

A schematic diagram of the fluidised bed recycling process is shown in Fig. 2. CFRP wastes are shredded to typically between 6-20mm [32] before entering the fluidised bed reactor. The silica sand bed is used to volatilise the shredded scrap material and thus to decompose the epoxy resin and release the fibres. The fluidising air is able to elutriate the released fibres, while non-organic contaminants (e.g., metal) remain in the bed. The operating temperature of the fluidised bed is chosen to be sufficient to cause the polymer to decompose, leaving clean fibres, but not too high to degrade the fibre properties substantially. At the operating temperatures of 450°C to 550°C, resin decomposition products are oxidised to recover energy

124 content. The fibres are then removed from the gas stream by a cyclone or other gas-solid separation device
125 and collected. In the current pilot plant, the gas stream after fibre separation is directed to an oxidiser
126 (combustion chamber) to fully oxidise the polymer decomposition products. Heat is recovered from the gas

stream to pre-heat fresh air input before being exhausted through the stack.

127

128 Mass and energy models of the fluidised bed process are developed to evaluate the impact of operating 129 parameters (CF feed rate per unit of fluidised bed area (kg/hr-m<sup>2</sup>); annual plant capacity (t/yr)) on energy 130 consumption and associated environmental impacts. Key details of the recycling process model are presented 131 here subsequently; additional information can be found in the Supplementary Data (Section 1.1). Waste CFRP 132 is assumed to be from manufacturing scrap or end-of-life prepreg, typically composed of high strength Toray T600SC CF (53% vf; 62% wt) and MTM28-2 epoxy resin. Parameters for the fluidised bed model are based 133 134 on experience from operation of a 50 t/yr pilot scale facility but are selected to best represent expected 135 conditions of a commercial operating facility. For all model variations, equipment and piping are sized 136 assuming a representative fluidising velocity of 1m/s, pipework air velocity of 20 m/s, and minimal pipe 137 length to accommodate equipment size for practical operation and maintenance. Thermal energy balances, 138 including heat losses from equipment and pipework are calculated assuming a representative fluidised bed 139 temperature of 550°C and oxidiser temperature of 750°C.

140 Inefficiencies arise in the process from heat loss to the surroundings and in-leakage of air due to the

141 operation of the system slightly below atmospheric pressure. Energy inputs to the system are quantified by

estimating process energy requirements and heat losses for each section within the fluidised bed system. Heat

143 losses in the exhaust are mitigated by high efficiency heat recovery from the oxidiser outlet prior to

144 exhausting. Pipework and equipment insulation are determined by economic optimisation of insulation costs

145 and potential energy savings with minimum net present value. Fan power requirements are calculated to

- achieve airflow through the system and to maintain fluidised bed operating pressure at 500 Pa below
- 147 atmospheric pressure to ensure that there is no leakage of gases from the system into the air [33]. The energy

148	model is verified by comparing with experimental results from the pilot plant. The energy consumption
149	predicted using the model agrees to within 1% of the pilot plant data, suggesting the model is reliable to
150	develop life cycle inventory data.
151	Although the full chemical formulation of the epoxy resin is not available, for the purposes of
152	stoichiometry calculations, it is assumed to be made of Diglycidyl ester of bisphenol A (DGEBA) in 87 % wt
153	and Isophorone Dianmine (IPD) in 13 % wt. CO <sub>2</sub> emissions resulting from the oxidation of the epoxy matrix
154	material are calculated on a stoichiometric basis assuming all carbon is fully oxidised to CO <sub>2</sub> . Data on other
155	potential GHG emissions (methane, nitrous oxide) are not available and are assumed to be negligible.

## 157 **2.2 Wet papermaking process**

158 The wet papermaking process has been successfully demonstrated to be an effective way to produce non-159 woven mats from rCF [27]. The process can also use chopped vCF as a reinforcing fibre. CF with matrix 160 polymer fibres (PA) is dispersed in a viscous aqueous solution, laid into a mat in random orientation, and 161 dried via vacuum drying (with recovery of aqueous dispersion media for reuse) and a final thermal drying 162 stage. The resulting mat consists of 25% CF (by volume) and an areal density of 100 gsm. Co-mingling of the 163 CF and PA fibres during dispersion has the advantage of bringing reinforcement and polymer fibres close 164 together, thereby reducing the melt flow distance in subsequent manufacturing stages and promoting more 165 complete resin impregnation with minimal void formation.

Energy and material requirements of the papermaking process are estimated based on experimental data and, where possible, energy efficiency data of standard equipment. Process parameters are selected to achieve fibre dispersion and drying with minimised energy input, based on experimental evidence and model outputs. A critical parameter is the total fibre volume content (CF and PA fibres) of the dispersed slurry, which is

assumed here to be 0.1% to avoid agglomeration of fibres during processing [34]. Increasing the fibre content

171 could substantially reduce the energy requirements for papermaking and is the subject of ongoing research.

172 Further details regarding the wet-papermaking process model can be found in the Supplementary Data,

173 Section 1.3.

174

## 4 2.3 Manufacture of composite components via compression moulding

175 Components produced from co-mingled mats (rCFRP, vCFRP1) are manufactured via a compression 176 moulding process. Mats are cut to size (300 mm x 190 mm) and stacked to achieve the required component 177 thickness, typically requiring 15 layers of 100gsm mats to fill up the mould cavity. The moulding is 178 subsequently compressed between two steel tools and heated to form the component. Energy analysis of each 179 step (i.e., heating stage, curing stage, pressure build-up stage and finishing stage) based on heat transfer 180 theory has been carried out based on the processing parameters for CF-PA composite (See Supplementary 181 Data, Section 1.3). Differences between vCF and rCF component thickness to compensate for differences in 182 material properties (see Section 2.5) are accounted for in our analysis of CFRP manufacturing energy use.

## 183 **2.4** Virgin carbon fibre composite manufacture

184 The manufacture of vCF is modelled based on existing literature data. The life cycle inventory data input to

185 our LCA models information is described in the Supplementary Data Table S2 and comprises data from

186 literature and life cycle databases, with parameters selected based on literature consensus, expert opinion and

187 results from a confidential industrial dataset. Publicly available data on vCF manufacture is limited and, in

188 many cases, is lacking in key details that should be incorporated into LCA studies, in particular variations in

189 CF mechanical properties (high strength vs high modulus) and corresponding production energy

190 requirements.

- 191 Two vCF materials are considered: vCFRP1, which is produced via paper-making/ compression moulding
- as described above (Sections 2.2 and 2.3); and vCFRP2, a woven CF/ epoxy composite material manufactured
- 193 via autoclave moulding of prepreg (Toray T600s and epoxy resin) with 50% fibre volume fraction. The

194 composite has fibres oriented at 0° and 90° and exhibits a Young's modulus of 70 GPa in both the

195 longitudinal and transverse directions [35]. Energy requirements of the autoclave process are obtained from

196 literature, including prepreg production (4 MJ/kg) and autoclave moulding (average of reported values 29

197 MJ/kg) [21, 23, 36].

## 198 **2.5** Mechanical properties of composite materials

199 To properly compare CFRP production routes from different CF sources (rCF, vCF), it is essential that the

200 CFRP products exhibit identical mechanical properties. Two metrics are considered to determine functional

201 equivalence of CFRP materials: bending stiffness and bending strength. To compensate for variations in

202 material properties between the three materials (rCFRP, vCFRP1, and vCFRP2), thickness of the panel is

203 varied in order to achieve the required bending stiffness or strength for a composite beam. The relative

204 thickness of the components impacts both the life cycle inventory, as thicker components require greater

205 quantities of fibre and matrix materials. Mechanical properties of the rCFRP and vCFRP1 materials are

206 measured experimentally from samples prepared with 10 to 15 layers as required to fill the mould cavity [27].

207 Material properties for vCFRP2 are from the manufacturer's data [35] (see Table 1) while mechanical

208 properties of rCF and vCF are measured experimentally as shown in Table S2.

209 The thickness ratio  $(R_t)$  comparing a CFRP components with a reference material (subscript *ref*) for

210 constant bending stiffness and strength can be determined by:

$$R_{t-stiffness} = \frac{t}{t_{ref}} = \left(\frac{E_{ref}}{E}\right)^{\frac{1}{3}} \tag{1}$$

$$R_{t-strength} = \frac{t}{t_{ref}} = \left(\frac{\sigma_{ref}}{\sigma}\right)^{\frac{1}{2}}$$
(2)

211 Where *t* is the material thickness (mm), *E* is the material tensile modulus, and  $\sigma$  is the tensile strength of the 212 materials. Further details can be found in Supplementary Data, Section 1.5.

213 In assessing the relative thickness and corresponding relative mass of CFRP materials, the vCFRP1

214 material is selected as the reference. Relative to this material, rCFRP requires 5% and 7% greater thickness to

achieve equivalent bending stiffness and strength, respectively (see Table 1). The woven vCFRP2 material

216 exhibits superior mechanical properties and as such has low thickness ratios: 0.66 and 0.55 for equivalent

217 stiffness and strength, respectively.

- 218 **3 RESULTS AND DISCUSSION**
- 219 **3.1** Fluidised bed recycling process

220 CF can be recovered from CFRP with energy expenditure as little as 6 MJ/kg CF for the fluidised bed 221 operating parameters considered. Fig. 3 shows the energy balance of the recycling process, including energy 222 inputs (natural gas, electricity), energy release from resin oxidation, and heat losses, for a fluidised bed plant 223 with 100 t/yr of annual throughput of rCF. The energy requirements of the fluidised bed recycling process are 224 primarily dependent on two factors: the feed rate of CFRP processed per unit bed area (kg CF/hr-m<sup>2</sup>), and the 225 in-leakage of ambient air. At lower feed rates, relatively more air needs to be heated and transferred through 226 the system per kg of CF recovered, leading to greater natural gas demand for thermal energy and electricity 227 for the fans. At higher feed rates, thermal energy requirements are significantly reduced to the extent that 228 most process heat can be provided by resin oxidation. Beyond a feed rate of 5 kg/hr-m<sup>2</sup>, energy efficiency gains are minor as the resin energy input is fully exploited in heating the fluidised bed to 550 °C and there is 229 230 minimum quantity required to raise the oxidiser temperature to 750 °C. Gas exhaust from the system 231 following the oxidation and heat recovery stage is the primary mode of heat loss from the fluidised bed 232 system. The heat recovery system is arranged to give the maximum practical heat recovery but that 233 nevertheless the exhaust gases from the stack where exhaust temperatures range from 98°C to 208°C across parameters calculated in this study. Heat recovery from the stack for other process uses could therefore 234

235 improve overall efficiency. Fluidised bed plants with annual throughputs of 50 to 1000 t/yr are analysed, 236 finding that plant capacity has minor impacts on energy use, as shown in Supplementary Data, Fig. S4. 237 While energy efficiency gains are identified to be achievable by increasing feed rate, there are potential 238 trade-offs in terms of resulting rCF properties. To avoid agglomeration in the recycling process at high feed 239 rates, fibre length must be reduced [37]. However, fibre length may also affect the downstream CFRP 240 manufacturing process and resulting CFRP product properties. It is expected that fibre lengths in the range of 241 1-10mm will be preferred for balancing fluidised bed performance and rCF properties for CFRP manufacture, 242 however; this is a topic of ongoing research.

243 As described before, as the fluidised bed system operates below atmospheric pressure there is the potential 244 for air in-leakage at pipework joints and in particular at shaft seals on the high temperature fans in the system. 245 The air in-leakage rate impacts the thermal energy requirements as this introduces a mismatch in mass flow 246 rate: additional air must be heated to 750 °C at the oxidiser, thereby resulting in greater exhaust heat losses. 247 Air leakage also places an impact on fan power consumption by changing the mass flow rate. Air in-leakage 248 rates up to 10% are evaluated, showing that natural gas and electricity requirements increase by up to 340% 249 and 1% respectively (see Supplementary Data, Fig. S5). Air in-leakage could be minimised in a commercial 250 plant design, but some would be unavoidable given the need to operate below atmospheric pressure to prevent 251 emissions of untreated gases.

252 In comparison to rCF from composite recycling, vCF production is energy intensive, with reported energy

requirement ranging from 198 to 595 MJ/kg [10, 20, 24, 38]. Across the range of operating parameters

considered in the study, energy required to recover CF is generally less than 10% of that required to produce

vCF, while operation of the fluidised bed process at higher feed rates with well controlled air in-leakage could

reduce this figure to 3%.

- 257 Data is extracted from the fluidised bed recycling process model to input to the life cycle analysis,
- considering likely operating conditions: 500 t CF/yr annual capacity; 9 kg CF/hr-m<sup>2</sup> fluidised bed feed rate;

and 5% air in-leakage. These parameters correspond to an energy requirement of 7.7 MJ/kg rCF, comprised of
1.9 MJ/kg (natural gas) and 5.8 MJ/kg (electricity).

#### 261 3.2 CFRP manufacturing

Direct energy requirements for the CFRP manufacturing processes (papermaking and compression
moulding; prepreg and autoclave) are shown in Table 2. These results are presented on a mass basis and a part
basis accounting for the relative stiffness/strength of the CFRP materials. Direct energy results presented in
Table 2 assume a component thickness of 2 mm; implications of component thickness on manufacturing
energy use are discussed below.

267 Papermaking – including fibre dispersion, vacuum drying and thermal drying steps – accounts for 268 approximately 15% of the energy consumed during the manufacturing process for the rCFRP and vCFRP 1 269 materials. Based on expected process parameters, total energy requirement is estimated as 14 MJ/kg CF mat 270 with approximately half from fibre dispersion and half from drying. Model parameters for fibre dispersion 271 and drying affect energy requirements of the papermaking process; an assessment of the sensitivity of results 272 to variations in these parameters and insights are presented in the Supplementary data (Section 2.2). As results 273 are based on expected process parameters, it is noted that these could be varied in actual processes which 274 could impact results presented here.

As shown in Table 2, final component manufacturing by compression moulding is much more energy intensive than the papermaking process. The heating and pressure build-up stages of compression moulding accounting for 60% and 16% of total manufacturing energy requirements, respectively. Energy requirements for compression moulding of a component are not strongly related to the component thickness for thicknesses in the range of the current study (1 to 10 mm). Heat losses and pressure build up energy consumption are largely dependent on the cross-sectional area of the panel and independent of component thickness. When considering components of different thickness, energy consumption in terms of MJ/kg can vary between

282 materials as energy consumption is very similar despite differences in component mass (see Supplementary

283 Data, Fig. S7). As a result, compression moulding energy requirements are similar for the rCFRP and

vCFRP1 materials even when relative thicknesses required to achieve equivalent stiffness and strength are

285 considered. Compression moulding energy consumption results presented here are high relative to literature

sources (approximately 9 MJ/kg) [22] due to the higher processing temperature of PA fibre (270°C) in this

study relative to unsaturated polyester resins used in the literature (30-80°C cure temperature).

288 For the autoclave component (vCFRP2), total manufacturing energy associated with prepreg production

and autoclave moulding energy consumption (4 MJ/kg and 29 MJ/kg, respectively) is similar to values

290 reported for the papermaking-compression moulding route on a mass basis. However, the vCFRP2 component

291 can achieve equivalent strength and stiffness with a lower mass than with the rCFRP and vCFRP1 materials.

292 Accounting for the relative mass of components results in lower manufacturing energy requirements for the

vCFRP2, approximately 50% of energy consumption for rCFRP and vCFRP1.

## 294 **3.3** Life cycle energy use and greenhouse gas emissions

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295 The LCA results are presented by considering two environment metrics- primary energy demand (PED) in 296 and global warming potential (GWP). All LCA results consider the relative thickness and mass of 297 components required to achieve equivalent stiffness and strength. CFRP production from rCF results in 298 substantially lower life cycle PED and greenhouse gas (GHG) emissions than vCF components of equivalent 299 mechanical properties (see Fig. 4). The total PED for rCFRP component (51.1 MJ/part under equivalent 300 stiffness; 51.8 MJ/part under equivalent strength) is 50%-51% of the vCFRP 1 component and 56%-68% of 301 the vCFRP 2 component when considered under both equivalent stiffness and equivalent strength bases. This 302 is primarily due to high PED associated with vCF manufacture, which represents 53% of total PED for the 303 vCFRP1 component and 80% for the vCFRP2 component. In contrast, the energy consumption of the

fluidised bed recycling process is relatively small, representing only 2% of the total energy use in rCFRP

305	production. Manufacture of the matrix material (PA fibre) for the compression moulding pathways for either
306	vCFRP1 manufacture or rCFRP is the next largest contributor to PED (21% of vCFRP1 manufacture; 44% of
307	rCFRP manufacture). To achieve equivalent stiffness and strength, the rCFRP component requires greater
308	relative thickness than the vCFRP materials (8% and 5% thicker than vCFRP1 and 96% and 60% thicker than
309	vCFRP2 on equivalent strength and stiffness bases, respectively). Correspondingly rCFRP has larger
310	requirements for fibre and matrix materials than the vCF alternatives. Production of the epoxy resin matrix
311	material for the vCFRP2 material is associated with relatively small PED relative to matrix materials for the
312	rCFRP and vCFRP1 components, due primarily to lower material requirements arising from the higher fibre
313	volume fraction and lower relative mass of this material. Compression moulding and wet papermaking
314	processes make up 20% and 7% of PED for the vCFRP1, respectively compared to 40% and 14% for the
315	rCFRP under equivalent stiffness. The autoclave moulding and prepreg production are relatively less energy
316	intensive, constituting 10% and 2% of the total CFRP manufacture, respectively.
317	The GWP results follow similar trends as the PED results. Total GWP is 5.8 kg CO <sub>2</sub> eq./part for vCFRP1
318	component, 5.1 kg CO <sub>2</sub> eq./part for vCFRP2 component and 2.8 kg CO <sub>2</sub> eq./part for rCFRP component,
319	respectively under equivalent stiffness (Fig. 4b)) while total GWP is 5.8 kg CO <sub>2</sub> eq./part for vCFRP1
320	component, 4.3 kg CO <sub>2</sub> eq./part for vCFRP2 component and 2.9 kg CO <sub>2</sub> eq./part for rCFRP component,
321	respectively under equivalent strength (Fig. 4d)). For the vCFRP1 and vCFRP2 components, the manufacture
322	of vCF is the main GHG emissions source. Emissions associated with papermaking, compression moulding,
323	and PA fibre manufacture are significant for both vCFRP1 and rCFRP, whereas component manufacture by
324	autoclave, matrix materials represent relatively smaller contributions to life cycle GWP for vCFRP2. Avoided
325	GWP associated with the rCFRP is dependent on the basis for comparison. Recycled CFRP can reduce GHG
326	emissions by approximately 45% relative to vCFRP1 and vCFRP2 on an equivalent stiffness basis; however,
327	as the autoclave produced vCFRP2 component exhibits a higher specific strength than the comparator
328	materials, displacing this material with rCFRP can reduce GHG emissions by only 33%.

329 The life cycle PED and GWP results are robust across the range of fluidised bed operating parameters and

330 air in-leakage rates considered in this study. At the least favourable operating conditions (low feed rate (3

331 kg/hr-m<sup>2</sup>); high air in-leakage rate (10%)), the rCFRP component still exhibits substantial savings relative to

the vCFRP component in terms of PED (45% reduction against vCFRP1 and 38% reduction against vCFRP2)

and GWP (48% reduction against vCFRP1 and 40% reduction against vCFRP2) under equivalent stiffness.

334 While vCF production dominates the energy intensity in manufacturing vCFRP, optimization of the fluidised

bed process could further reduce the overall environmental impacts of rCFRP manufacture.

Results presented here consider a 'gate-to-gate' approach and as such do not consider the use phase of the

337 manufactured components. If used in transport applications, lighter weight components would achieve energy

338 savings by reducing mass induced fuel consumption. As such, the slightly greater mass of the rCFRP

339 component relative to the vCFRP materials (see Section 3.2) could incur greater in-use energy consumption

340 and associated GHG emissions relative to the vCFRP components. Disposal of rCFRP material is not

341 considered as there is uncertainty as to whether rCFRP could be recycled for rCF recovery at end of life, or if

a lower value recovery approach (e.g., incineration, mechanical recycling) would be required. Subsequent

343 work will extend the current analysis to consider the full life cycle of rCFRP and vCFRP components to

344 assess the net PED and GWP impacts.

345 It is found that CF recycling in the fluidised bed process can achieve substantially greater environmental

346 performance than conventional waste treatments or mechanical recycling [39]. Fluidised bed recovery of CF

347 and use of rCF to displace vCF-based composites provides substantial savings in PED (~ 65-330 MJ energy

348 savings per kg of CFRP waste), offering an order of magnitude greater net PED savings compared to waste

349 CFRP incineration. Further, avoided GHG emissions range from 3 to 19 kg CO<sub>2</sub> eq. per kg of CFRP waste

350 processed by fluidised bed, depending on the vCF material displaced (vCFRP1, vCFRP2). In contrast,

351 mechanical recycling of CFRP waste produces rCF suitable only for displacing glass fibres, achieving a far

352 smaller GHG emissions reduction of  $0.38 \text{ kg CO}_2 \text{ eq. per kg CFRP waste [39]}$ .

Results of the current study are robust, despite potential implications of model assumptions and simplifications. The main contributors to PED and GWP of rCFRP materials are the production matrix material and the compression moulding process, for which well-established data are available. Operating parameters for the fluidised bed and papermaking processes are based on operating demonstration facilities and the best available experimental data, but are less certain. However, these stages represent only 2% and 14% of GWP, respectively, and so variations are not expected to appreciably impact the overall results.

## 359 4 CONCLUSION

360 A life cycle analysis of CF recycling process has been carried out in this study, based on a novel process 361 model of the fluidised bed recycling process. Key recycling plant operating parameters, including plant 362 capacity, feed rate, and air in-leakage are investigated. The feed rate per unit bed area is identified as the most 363 important parameter for achieving energy-efficient CF recycling. The energy model shows that energy 364 requirement of rCF production is very low relative to vCF and robust across likely operating conditions. 365 Further optimisation of fluidised bed recycling process needs to balance to maximising feed rate per bed area 366 to minimise process energy use and potential implications for rCF properties. Opportunities exist for recovering stack heat loss which could further improve the energy efficiency of the fluidised bed process. 367 368 The use of rCF to displace vCF in composite applications is found to achieve significant gate-to-gate life 369 cycle energy and GHG emissions reductions. Resulting rCFRP components with identical mechanical 370 properties to those produced from vCF can reduce PED by 32% to 50 % and GWP by 33% to 51%. 371 Environmental performance far exceeds that of conventional waste treatment routes (i.e., landfilling, 372 incineration). 373 A key question for future progress is the value of the rCF. The intrinsic value of CF is due to their high cost 374 as virgin materials but excessive levels of touch labour associated with recovery and potentially high levels of

375 energy use during recovery and rCF processing to a great extent weaken the business case for recycling

- activities. As the rCFs from fluidised bed process are in a fluffy form with random discontinuous length
- 377 distribution as well as some strength reduction, it provides a number of challenges for cost-effective and high
- value re-use to manufacture CFRP composites. A series of studies [27, 40-43] at the University of
- 379 Nottingham have proposed various routes to enable the use of rCF, including wet-papermaking process to
- 380 manufacture non-woven mat analysed in this study. Furthermore, fibre alignment techniques and
- 381 thermoplastic injection moulding methods have also demonstrated alternatives to process the rCF to
- 382 manufacture high-value composite products. Progress in recycling process optimisation and manufacturing
- 383 method development are key to achieving the significant environmental benefits that CF recycling can
- 384 contribute to the aerospace and automotive industries.

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## 388 APPENDIX A. SUPPLEMENTARY DATA

389 Supplementary data related to this article is available.

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**Fig. 1.** Block flow diagram of CFRP manufacturing by waste CFRP recycling and compression moulding.



486487 Fig. 2. Main components and flow directions of the fluidised bed CFRP recycling process



**Fig. 3.** Energy flows including heat losses from each component and energy value from resin and energy





492 Fig. 4. a) Primary energy demand (PED) and b) Global warming potential (GWP) for composites produced
493 from vCF and rCF under equivalent stiffness; c) PED and d) GWP for composites produced from vCF and
494 rCF under equivalent strength

## 496 **Table 1.**

497 Mechanical properties of carbon fibre composite materials and corresponding thickness ratio and mass ratio

498 required to achieve equivalent component stiffness and strength.

	Tensile	Tensile modulus (GPa)	Equivalent stiffness		Equivalent strength		Fibre
	strength (MPa)		Thickness ratio	Mass ratio	Thickness ratio	Mass ratio	(mm)
vCFRP1	171.59±6.64	19.74±1.19	1.00	1.00	1.00	1.00	12
vCFRP2	570	70	0.66	0.75	0.55	0.62	continuous
rCFRP	148.56±9.56	16.95±0.46	1.05	1.05	1.07	1.07	1.43

# **Table 2.**

- 502 Direct energy consumption of CFRP manufacturing steps considering a nominal component thickness of
- 503 1mm.

		Energy consumption in MJ/kg (MJ/part)					
Process steps			vCFRP 1	vCFRP 2 Equiv. Bending Stiffness	vCFRP 2 Equiv. Bending Strength	rCFRP Equiv. Bending Stiffness	rCFRP Equiv. Bending Strength
		Fibre dispersion	3.45 (0.34)	-	-	3.45 (0.35)	3.45 (0.36)
	ermaking	Vacuum drying	2.44 (0.24)	-	-	2.44 (0.25)	2.44 (0.26)
ssing		Thermal drying	1.56 (0.15)	-	-	1.56 (0.16)	1.56 (0.16)
CF proces	Papo	Resetting (winding,wa shing, belt conveying)	0.41 (0.04)	-	-	0.41 (0.04)	0.41 (0.04)
	Prepreg for	autoclave	-	4.00 (0.44)	4.00 (0.44)	-	-
	Cutting		0.37 (0.06)	0.37 (0.04)	0.37 (0.04)	0.37 (0.06)	0.37 (0.06)
	Compression moulding	Heating stage	32.78 (4.84)	-	-	31.26 (4.85)	30.56 (4.85)
facture		Curing stage	2.10 (0.31)	-	-	2.01 (0.31)	1.96 (0.31)
manui		Pressure build-up	8.53 (1.26)	-	-	8.13 (1.26)	7.94 (1.26)
l part		Water cooling	0.90 (0.13)	-	-	0.90 (0.14)	0.90 (0.14)
Fina	Autoclave moulding		-	29.00 (3.19)	29.00 (2.67)	-	-
	Deflashing a	Deflashing and drilling		1.20 (0.13)	1.20 (0.11)	1.20 (0.19)	1.20 (0.19)
Total			53.75 (7.54)	34.57 (3.81)	34.57 (3.26)	51.73 (7.60)	50.79 (7.64)