Comparing Life Cycle Energy and Global Warming Potential of Carbon Fibre Composite Recycling Technologies and Waste Management Options

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Abstract

Carbon fibre reinforced polymers (CFRP) are used in increasing quantities as they have some of the best properties in terms of specific strength and stiffness of any widely available material. By 2020, annual global CFRP production is expected to be over 140,000 tonnes. However, the resulting increased quantity of CFRP waste has highlighted the need for sustainable treatment options as carbon fibre manufacture has high-energy intensity. A life cycle methodology is used to evaluate primary energy demand (PED) and global warming potential (GWP) leveraging best available literature data, process models, and experimental work. Overall results indicate that recycling scenarios are generally the environmentally preferable options over landfill and incineration. However, the relative environmental benefits of advanced recycling processes (i.e., pyrolysis, fluidised bed, and chemical recycling process) depend on the method used to determine displacement of virgin carbon fibre by recycled carbon fibre. Totally, recycling processes can achieve a representative GWP of -19 to -27 kg CO₂eq. and PED of -395 to -520 MJ per kg CFRP, providing superior environmental performance to conventional composite waste treatment technologies.

Keywords: Carbon Fibre Recycling; Life Cycle Methodology; Primary Energy Demand, Global Warming Potential; Waste Treatment Options

Introduction

Recently, the steady increase in the use of carbon fibre reinforced polymers (CFRP) across a wide range of aerospace (e.g., Boeing 787 airplane wing structures), automotive (e.g., BMW i3 body panels), energy (e.g., wind turbine blades), and sporting applications (e.g., fishing rods, bicycles) has been seen as CFRP contributes to significant weight reduction of the product
while providing excellent performance. In the past 10 years, the annual global demand for carbon fibre (CF) has increased from approximately 16,000 to 72,000 tonnes and is forecast to rise to 140,000 tonnes by 2020.\textsuperscript{1} Global revenues of CFRP are estimated to be $16.1 billion in 2011, and are expected to increase to $28.2 billion in 2015 and $48.7 billion by 2020.\textsuperscript{2} However, the resulting CFRP wastes from manufacturing (up to 40% of the CFRP can be waste arising during manufacture)\textsuperscript{3} and end-of-life components are generated at an increasing speed in the next decades. Quantities of CF in production and end-of-life waste are estimated to be 62,000 tonnes by 2020.\textsuperscript{4} Existing automotive sector-specific EU regulations requires the recycling of at least 85% of end-of-life materials.\textsuperscript{5} Moreover, the high cost and energy intensity of virgin CF (vCF) manufacture (198–595 MJ/kg) also provide an opportunity to recover substantial value from CFRP wastes. Therefore, there is a clear and significant requirement to develop a CFRP waste management system dealing with CF recovery to comply with this legislation.

Recycling has been recognised as a desirable end-of-life option to deal with CFRP wastes as recycling has the potential to recover value from the waste materials rather than being disposed in landfill or incineration, fulfilling legislative and sustainability targets. Current recycling methods vary from conventional mechanical recycling to thermal recycling (e.g., pyrolysis and fluidised bed process) and chemical recycling.\textsuperscript{6,7} Mechanical processes reduce the size of the composites to micron level to be incorporated into the manufacture of new CFRP as fillers. Secondly, pyrolysis is a widely used method, being established in commercial operations. Pyrolysis thermally decomposes the resin into an oil phase of liquid hydrocarbons, a gas fraction (primarily CO, H\textsubscript{2} and methane) and char covered fibres. Another thermal process is the fluidised bed process, which has been developed for the recycling of glass and carbon fibres at the University of Nottingham for over 15 years.\textsuperscript{6} In the fluidised bed process, the polymer matrix is oxidised to enable fibre recovery\textsuperscript{6}, which can accommodate contamination in end-
of-life CFRP waste. Recovered CF (rCF) shows almost no reduction in modulus and 18-50% reduction in tensile strength relative to vCF.\textsuperscript{3,6} A chemical recycling process uses a solvent to degrade the resin and recover the carbon fibres. Recycling processes are now transitioning from lab scale to commercial facilities (e.g., ELG Carbon Fibre Ltd. in UK using a pyrolysis recycling process with an annual capacity of 2,000 t/yr). Table 1 summarizes the details of existing CFRP recycling plants. Recovered CF could reduce environmental and financial impacts relative to vCF production, while the potentially lower cost of rCF can create new markets for lightweight materials.\textsuperscript{8,9} However, to date there is no systematic study comparing the environmental performance of different recycling technologies.

Table 1 CFRP recycling technologies: geographical location, key processing parameters and capacity

Life Cycle Assessment (LCA)\textsuperscript{10,11} has been widely recognized as a valuable tool to aid decision making for waste management systems, or strategic decisions concerning resource use priority. LCA can provide an overview of the environmental aspects of different waste management strategies, and enable efficient comparison of the potential environmental impacts of these options. Prior studies\textsuperscript{12-16} have estimated energy requirements of various CFRP recycling technologies and found substantially lower energy requirements compared to vCF manufacture. However, very few studies have been undertaken to quantify environmental impacts of CFRP recycling processes. Li et al.\textsuperscript{17} found that mechanical recycling reduces GHG emissions, primary energy demand, and landfill waste generation compared to landfilling, however, it is not economically competitive. Meng et al.\textsuperscript{8,9} evaluated the energy and environmental impacts of CF recycling by a fluidised bed process and reuse of rCF to manufacture a CFRP material for the first time. A few additional studies have conducted LCA
for some CFRP recycling technologies; however, each study is based on the different system boundaries and assumption and limited data estimates for recycling process energy and material inputs. Thus, it is difficult to compare the environmental performance of different recycling technologies and determine which one offers the most benefits. While potential overall environmental benefits are claimed in technical studies of CFRP recycling processes and fibre reuse opportunities to replace vCF, these benefits have yet to be demonstrated in a comprehensive life cycle study that compares different recycling techniques.

Overall, prior analyses indicate reduced energy consumption for rCF compared to vCF. Due to the lack of inventory data on recycling processes in any life cycle inventory (LCI) databases and the markets for recycled materials, comparative LCA studies on CFRP recycling are not well established. In this study, a detailed life cycle analysis of waste treatment options for CFRP materials has been developed in order to quantify the environmental impacts (primary energy consumption, global warming potential in 100 years) in a life cycle perspective. Comparisons of conventional landfill, incineration and the most commercially/near-commercially viable recovery technologies (mechanical recycling; fluidised bed recycling; pyrolysis recycling; chemical recycling) are undertaken to identify relative environmental performance of alternative waste treatment routes on the UK basis.

**Methodology**

The life cycle model built following the ISO standards begins where the waste CFRP has been collected. System expansion has been applied in the model that includes credits from avoiding production of energy and virgin materials as potential outputs of waste treatments. “Gate-to-grave” models are developed for CFRP waste treatment by landfilling and
incineration including waste processing (disassembly, shredding), transport (100 km) (between recycling facilities only), and waste treatment (landfill, incineration) (see Figure 1). For recycling, a “gate-to-gate” approach is taken to include the production of composite materials from rCF. The production and use of CFRP prior to end–of-life is identical regardless of the waste treatment route selected, thus is not included in the model; i.e., the boundary of our study assumes availability of CFRP waste as a feedstock without any prior associated/allocated environmental burdens. Maintenance and facility construction are also excluded as the allocated emissions and energy use per functional unit from these long-lived assets are likely to be small. The functional unit is one tonne of CFRP waste treated, primary from manufacturing scrap or end of life waste from aerospace industry, which is assumed to consist of 55wt% fibre content and 45wt% matrix (epoxy resin) content, to compare waste management options (i.e., landfill, incineration, mechanical recycling, pyrolysis recycling, fluidised bed recycling, and chemical recycling) for CFRP waste. In order to complete the full life cycle model, data collected from process model based on the pilot plant, experimental data, best available literature data and LCI databases (e.g., Eco-invent\textsuperscript{19}) are the major sources to provide process-specific data. Primary energy demand (PED) and global warming potential (GWP) are the two main metrics examined from the life cycle models. Figure 1 shows a schematic representation for waste treatment routes considered in this study and final products of each process.

**Waste treatment routes on CFRP**

This study considers six potential waste treatment routes for CFRP: landfill, incineration and mechanical, pyrolysis, fluidised bed, and chemical recycling processes. Each waste flow for recycling processes can be seen in the following sections, and shown in Figure 2. For all
recycling processes, collected CFRP waste materials are transported to materials disposition facilities by 32 tonne standard dump truck for shredding and separation before waste treatment. Transport distances of 100 km to landfill and 200 km to incineration or recycling facilities were assumed. Waste residues arising from the recycling process, which is either landfilled or incinerated, is assumed to be transported an additional 100 km distance from the recycling site to the landfill site or an additional 200 km to incineration site. Combustion ash generated from incineration processes is assumed to be transported 100 km to landfill. Recycling credits either from electricity production or CF replacement are allocated to the waste treatment of CFRP waste as the methods specified in Section 2.3. The inventory data for landfill and incineration is obtained from LCI databases – Gabi and EcoInvent database. The inventory data for rCF is based on data collection of all the activities including recycling processes and transport followed by documentation of the data obtained covering inputs and outputs (see Table A1).

Figure 1 CFRP Waste treatment routes.

Landfilling

The disposition of waste CFRP in landfill is assumed to occur in a sanitary landfilleing site which is built for the final disposal of solid waste and isolates the waste from environment. Before waste CFRP burying in landfill site, shredding pre-treatment is needed to reduce the size of CFRP waste into scraps. No prior dismantling work is needed to isolate the CFRP waste as it is disposed with other landfill fractions. Transportation distance is assumed as 100 km from scrap yard to landfill. Impacts associated with CFRP landfilling are assessed based the Ecoinvent sanitary landfill dataset. After burying waste CFRP scrap is assumed to emit no
further greenhouse gas (GHG) or consume any energy due to the largely inert property of CFRP waste.\textsuperscript{17}

3 **Incineration**

Incineration of CFRP provides an alternative way to treat the CFRP waste and recovers the embodied energy. Prior results\textsuperscript{4} suggest that CFRP waste can be co-fired with municipal waste and used as an energy source.

The potential for energy production from CFRP waste is dependent on its energy content and the incinerator efficiency. Typically, CFRP energy contents is about 30 MJ/kg\textsuperscript{6,20} but varies depending on the specific CFRP composition. Based on the fibre/matrix ratio, an energy content of 32 MJ/kg is used in this study.\textsuperscript{21} Compared to advanced power stations using conventional fuels, waste incineration typically has a lower efficiency than conventional fuels; a 13\% electricity generation efficiency with a combined heat & electricity cogeneration efficiency of 38\% is assumed.\textsuperscript{19} We assume a “high-efficiency” incineration scenario with a 25\% electricity generation efficiency with a combined heat & electricity cogeneration efficiency of 80\%. It is assumed that electricity generation displaces the average UK grid mix and heat generation displaces heat produced from natural gas which is the major source for heat production in the UK. Residue material after combustion is collected and transported to disposal in landfill as described in Section 2.1.1. Emissions from waste incineration are modelled based on stoichiometric balances, assuming all the carbon content of CFRP is oxidised and emitted as CO\textsubscript{2}. Net GHG emissions are dependent on the direct emissions from combustion and avoided emissions from electricity and heat displacement.
Mechanical recycling

Among various recycling methods, the most mature technology is mechanical recycling. It is currently used on a limited industrial scale to recycle waste composites, especially glass fibre reinforced plastic. After initial size reduction, the material is ground in a hammer mill and graded into different lengths through sieving. Using mechanical recycling, CFRP wastes can be reduced to two useful fractions: fine powder and fine rCF fraction. Mechanical recycling products are commonly used as fillers in lower value materials, such as bulk or sheet moulding compounds to replace glass fibre (see Figure 2). The resulting materials have poor mechanical properties compared to virgin CFRP (vCFRP) materials and so are not suitable for lightweighting or high modulus/strength applications. Coarse fraction waste residues are assumed to be transported 100 km from the recycling site to landfill site or 200 km to incineration site.

Figure 2 System diagram showing material and energy flows for a) mechanical recycling with landfilling and mechanical recycling with incineration, b) pyrolysis recycling, c) fluidised bed recycling and d) chemical recycling processes. The arrow colour indicates a type of each flow (red – energy flow, green – credits, and black – material flow).

Pyrolysis recycling

Pyrolysis is a thermal decomposition of polymers without oxygen or in a controlled flow of oxygen at high temperatures between 300 °C and 800 °C, enabling the recovery of long fibres with high modulus. An elevated temperature of 1000 °C can be applied but it will result in a significant degradation of mechanical properties of the fibre products. Due to the significant
impact of temperature and residence time on the final quality of the rCF, the two factors must be controlled strictly in the pyrolysis reactor.

As a thermal method, a shredding preparation of CFRP wastes before fed into the pyrolysis recycling plant is required. The pyrolysis process uses external heat to allow for recovering fibres with minimum properties reduction, which can then be used in the composite manufacture industries as rCF. Studies have shown that rCF from pyrolysis could maintain 90% or more of the original mechanical performances. In addition, the polymeric matrix can potentially be recycled as an oil phase of liquid hydrocarbons. Pyrolysis has now reached early stages of commercialisation, e.g., ELG Recycled Carbon Fibre Ltd has 2,000 t of CFRP waste/yr recycling capacity with an estimated process energy of 30 MJ/kg. For this process, we assume substitution for rCF for the recovered material. There is a data limitation in that we are not able to include separation processes and so may be overestimating the environmental benefit of recovering pyrolysis products to displace primary production of these materials.

An experiment of pyrolysis recycling of CFRP is conducted in laboratory-scale to obtain the mass data. The process of the experiment setup is as follows. The CFRP waste used in this experiment comprised of 55wt% CF and 45wt% epoxy resin same as specified in Nahil et al. Five sheets of CFRP (total volume CFRP: 12”x12”x0.5”) with described composition were fed into a large laboratory furnace with the size of chamber 16”x16”x16”. Pyrolysis recycling to recover rCF from CFRP waste involves two stages of heating process. First, the pyrolysis reaction was performed at 500°C which was chosen to achieve a reasonable yield of rCF while retaining mechanical properties of CF. During the pyrolysis reaction, nitrogen gas was fed into the furnace chamber with the flow rate of 2L/min to prevent significant property degradation of the CF. After the pyrolysis reaction, CF was mostly covered with residual chars. The second
heating process was performed at 450°C to remove char and separate CF in an oxidizing atmosphere for a short time period. During the char removal step, it is assumed that residual char is completely oxidized into CO₂. Based on the specifications of the furnace, energy consumption of two heating process is calculated as follow. The required heating power is 600W and 500W at steady state to maintain pyrolysis temperature and char-removing temperature, respectively. The required power to achieve desired temperature with heating rate 5-10°C is 2 kW. Energy related inventory data are obtained from commercial operation from ELG (2000t CFRP waste/yr). 28

Fluidised bed recycling

The fluidised bed (FB) recycling process requires a shredding process before feeding CFRP wastes into the FB 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 for about 20 minutes, but degraded material remains in the bed. The operating temperature of 450 °C to 550 °C of the FB reaction is chosen to be sufficient to cause the polymer to decompose, leaving clean fibres, but not too high to degrade the fibre properties substantially. The fibres can then be removed from the gas stream by a cyclone or other gas-solid separation device and collected. 3 However, organic materials from the gas stream will be a mixture of different by-products and it may be more cost effective to oxidise them fully to recover the energy value in a high temperature chamber. These out gases can be fed to power a combined heat and power unit. Therefore, in the current pilot plant, the gas stream after fibre separation is directed to a combustion chamber to fully oxidise the polymer by-products from FB process. Heat is recovered from the gas stream to pre-heat fresh air input before being
exhausted through the stack. The mass and energy flow of fluidised bed recycling pathway can be found in Figure 2.

Inventory data is extracted from the process model, considering likely operating conditions: 500 t rCF/yr annual capacity; 9 kg CF/hr-m² 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).\(^9\) Epoxy resin matrix is assumed to be made of Diglycidyl ester of bisphenol A (DGEBA) in 87wt% and Isophorone Dianmine (IPD) in 13wt%. \(^9\)

CO\(_2\) emissions resulting from the oxidation of the epoxy matrix material are calculated on a stoichiometric basis assuming all carbon is fully oxidized to CO\(_2\).

**Chemical recycling**

Chemical recycling process utilises a liquid solvent (such as water, acid, and alcohol) to break down polymer resin and separate them from CF. The recycling process is able to recover high quality CF with only about 1.1% tensile strength loss and recover polymeric matrix as an organic compound with a solvent method in nitric acid solution as reported in\(^29\) (it is assumed to replace virgin epoxy resin). The rCF is normally semi-long or long rCF with low contamination. However, the decomposition temperature and nitric acid concentration have an impact on the mechanical properties.

Depending on the temperature and pressure process, the chemical process can be categorised into supercritical, subcritical, and near-critical chemical. Schneller et al.\(^30\) investigated the fibre-matrix separation via chemical using sub- and supercritical fluids (pure water and a water/ethanol-mixture). After chemical separation, the majority of resin content could be removed at high temperature and at long processing time (see Figure 2). Due to this, there may
be no requirement for an additional oxidising surface treatment which is used to remove the char resulted from oxidation of resin. Chemical recycling technique is feasible but processing temperature, time, solvents and equipment have negative effects on the environment.

Primary data were obtained at laboratory scale for the chemical recycling process as in\textsuperscript{31}. However, process energy source types (electricity/gas), process efficiency and recovery of non-energy inputs due to scales of plant operation can have impacts on the environmental impacts and thus bring uncertainty of results, which has been addressed in Discussion section.

**Carbon fibre replacement**

The quality of life cycle inventory data for vCF manufacture is poor: publicly available data is limited. vCF production energy requirement and sources vary significantly (198 to 595 MJ/kg from a mix of electricity, natural gas, and steam) and GHG emissions associated with vCF production have been estimated at 30-80 kg CO$_2$eq.\textsuperscript{9} Further, studies have not linked production data to CF properties despite different processing conditions required to achieve high modulus and high strength CF (between 1000–1400°C for high modulus fibers, or 1800–2000°C for high strength fibers). Therefore, there is inadequate information to match energy intensity to fiber properties. In this study, the manufacture of vCF is modelled based on existing data from literature and life cycle databases, with parameters (e.g., mass yield in acrylonitrile, polyacrylonitrile and CF conversion steps) selected based on literature consensus, expert opinion, and results from a confidential industrial dataset. All of these data have been recalculated relative to 1 kg of CF, giving an energy consumption of 149.4 MJ electricity, 177.8 MJ natural gas, and 31.4 kg steam.\textsuperscript{9} Utilising CFRP recyclate to displace vCF in composite production is desirable for maximising revenue from waste streams and minimising energy use and GHG emissions.
In order to determine the quantity of rCF that replaces vCF, it is required to have an equivalent material function. In this paper, two different CF replacement methods are considered either based on variable fibre content or variable material thickness. These methods will be described in detail in the next section.

**Replacement - Variable fibre content**

The first method considers CF replacement under equivalent specific stiffness \((E/\rho)\) can be achieved by changing the fibre volume fraction of rCFRP to mitigate the mechanical properties reduction of rCF relative to vCF. Generalised rule of mixtures for tensile modulus can be described by:

\[
E_c = \eta_\theta E_f V_f + E_m V_m
\]  

Where \(\eta_\theta\) is composite efficiency factor (Krenchel), \(\eta_\theta =1\) for unidirectional, \(\eta_\theta =0.5\) for biaxial, \(\eta_\theta =0.375\) for random (in-plane), \(E_f\) is the modulus of the fibres, \(E_m\) is the modulus of the matrix, \(V_f\) is the volume fraction of the fibres and \(V_m\) is the volume fraction of the matrix, \(V_f+ V_m=1\). As shown in Figure 3, using generalised rule of mixture the tensile modulus can be predicted based on the fibre volume fraction. Therefore, in this study, we use the rule of mixtures to predict the modulus of vCF and rCF products for comparison.

Figure 3 Tensile properties of an epoxy recycled carbon fibre composite experimentally measured. The solid and dotted lines represent the theoretical stiffness calculated using the generalized rule for randomly distributed and perfectly aligned fibres. 

Using the method based on eq (1), the fibre volume fractions of rCFRP using rCF recovered using different recycling techniques are calculated as listed in Table 2. To meet the same mechanical properties, a higher fibre fraction is required for rCF and thus more rCF to replace...
vCF. Meanwhile, the difference of polymer matrix content due to varying fibre volume fraction can be determined as well: higher fibre fraction giving corresponding less polymer matrix content for the same total thickness.

Table 2 Recycled CFRP design properties for CF replacement method based on variable fibre content.

6 Replacement - Variable material thickness

In comparison, the second method to determine the replacement of vCF is selected based on variable material thickness but constant fibre fraction. When evaluating alternative materials, functional equivalence can be maintained by considering the design material index ($\lambda$) and varying component thickness to account for differences in each material’s mechanical properties.\textsuperscript{34-36}

$$R_t = \frac{t_{rCF}}{t_{vCF}} = \left( \frac{E_{vCF}}{E_{rCF}} \right)^{\frac{1}{\lambda}}$$

$$R_m = \frac{m_{rCF}}{m_{vCF}} = \frac{\rho_{rCF}}{\rho_{vCF}} \left( \frac{E_{vCF}}{E_{rCF}} \right)^{\frac{1}{\lambda}}$$

Where $R_t$ is the ratio of component thicknesses between rCF material ($t_{rCF}$) and the reference vCF material ($t_{vCF}$), $E$ is the modulus of the two materials (GPa), and $\lambda$ is the component-specific design material index. The expected CFRP component mass can be calculated based on the relative thickness and density of alternative materials to calculate the equivalent CF replacement (i.e., method 2). The relative thickness of the components affects both the life cycle inventory, as thicker components require greater quantities of fibre and matrix materials.
Depending on design purposes, the parameter $\lambda$ value may vary between 1 and 3. $\lambda=1$ is appropriate for components under tension loading (e.g., window frame), $\lambda=2$ is for columns and beams under bending and compression conditions in one plane (e.g., vertical pillar) and $\lambda=3$ is suitable for plates and flat panels when loaded in bending and buckling conditions in two planes (e.g., car bonnet) and is selected as base case. (Actual component designs require a finite element analysis to identify the material design index that would ensure design constraints are met.) The sensitivity of environmental impacts will be assessed for $\lambda$ values ranging from 1 to 3. The mass ratios and corresponding fibre and resin mass calculated are shown in Table 2: fibre fraction is constant, so larger components require more fibre and resin.

Results and discussion

Section 3.1 describes the environmental impacts of the various waste treatment options covering gate-to-gate impacts of recycling processes while Section 3.2 shows the total life cycle environmental impacts including the use of waste treatment products (recovered energy, rCF, chemicals) to displace conventional products/production.

Gate-to-gate life cycle primary energy demand and global warming potential

The PED and GWP results, excluding recycling credits, are compared among selected CFRP waste treatment methods as shown in Table 3. Landfill emits only minor GHG emissions of 0.13 kg CO$_2$eq./kg CFRP waste (0.03 kg CO$_2$eq. from shredding and 0.10 kg CO$_2$eq. from sanitary landfill operation) plus 0.01 kg CO$_2$eq./kg from transport of the waste materials; GHG generation in landfill is not of concern due to inert nature of CFRP waste. Incineration produces larger amount of GHG emissions of 3.12 kg CO$_2$eq./kg CFRP waste. The large amount of GHG
emissions are mainly from the combustion process as the carbon content of CFRP is released to the environment as CO₂.

Excluding the benefits of energy recovery or the use of rCF materials, recycling processes require energy inputs to treat the CFRP wastes. Mechanical recycling with landfilling of the remaining coarse fraction gives GHG emissions of 0.11 kg CO₂eq./kg CFRP waste and the mechanical recycling process accounts for 0.03 kg CO₂eq. In comparison, mechanical recycling with incineration produces larger GHG emissions of 1.80 kg CO₂eq./kg CFRP waste due to combustion of coarse fraction but this option will have GHG deduction from energy outputs to displace grid electricity and heat.

Thermal recycling processes - pyrolysis and chemical recycling - require greater energy inputs and are associated with higher PED and GWP than the fluidised bed, primarily due to the recovery of energy from oxidised matrix material in the fluidised bed process. Pyrolysis is associated with 2.9 kg CO₂eq./kg CFRP waste, primarily from electricity and natural gas consumption for recycling process. Transport accounts for only 1% of the total energy consumption. Fluidised bed and chemical recycling processes have similar amount of GHG emissions arising from recycling process, producing 1.56 kg CO₂eq. and 1.53 kg CO₂eq. per kg CFRP waste, respectively.

Table 3 Primary energy demand and Global Warming Potential comparison of the carbon fibre recovery and conventional landfill and incineration without credits from the use of products of recycling processes.
Total life cycle primary energy demand and global warming potential

Including the recycling credits from CF replacement or electricity production, the PED and GWP of different CFRP waste treatment methods based on method 1 and method 2 are shown in Figure 4. Overall the LCA results indicate that recycling scenarios are generally the environmentally preferable options for PED and GWP considered in this analysis, though the exact net environmental impacts vary depending on the CF replacement method selected.

Conventional waste treatment processes (landfill, incineration) perform worst in terms of life cycle PED and GWP. Even including energy credits, GHG emissions from incineration exceed the emission saving from electricity and heat power generation giving a net GHG emission of 2.14 kg CO$_2$eq. while landfilling consumes more than 1.11 MJ energy due to no environmental credit from displacement. High-efficiency incineration scenario has the potential to further reduce the emission due to avoiding more raw energy production. Assuming higher conversion efficiencies of 25% electricity generation with a combined heat & electricity cogeneration efficiency of 80%, net GHG emissions can be reduced by half to 1.08 kg CO$_2$eq./kg CFRP. However, even in this high efficiency scenario where useful energy outputs are approximately doubled, avoided emissions associated with displacing UK grid electricity and natural gas-fired heat generation are insufficient to compensate for CO$_2$ emissions arising from CFRP combustion.

Mechanical recycling with landfilling of the coarse recyclate fraction exhibits a modest global warming potential reduction relative to mechanical recycling with incineration. The displacement of glass fibre production results in a GHG emissions credit of 0.48 kg CO$_2$eq./kg CFRP, giving a net global warming potential reduction of 0.37 kg CO$_2$eq./kg CFRP. However, if the coarse recyclate fraction is incinerated, mechanical recycling produces a net GHG
emission increase of 0.76 kg CO$_2$eq./kg CFRP. Compared to the advanced thermal recycling process, mechanical recycling was not favourable primarily due to the low mechanical performance obtained in the recovery.$^{17}$

**Carbon fibre replacement – variable fibre content**

Due to different reductions of mechanical properties of recycling processes, rCF materials must contain a higher fibre volume fraction than vCF materials of the same thickness in order to achieve the same stiffness. Results show GHG emissions from fluidised bed recycling process are mainly from the oxidation of the resin content in CFRP. However, rCF displacing vCF provides environmental benefits by reducing raw material production, resulting in a net reduction in GHG emissions.

As in Figure 4, fluidised bed recycling process shows the relative lower net primary energy demand and GHG emissions and the main credits are from CF replacement. Considering primary energy demand, the fluidised bed recycling method gives about -494.5 MJ net PED per kg CFRP waste (Figure 4) by replacing the energy intensive vCF production (834.5 MJ PED/kg$^9$). Net primary energy demand equals to total energy input to recycling minus energy reduction from displacement of vCF. In replacement method based on variable fibre content, 1 kg fluidised bed rCF can replace 0.95 kg vCF compared to 0.88 kg vCF per kg pyrolysis rCF and 0.89 kg vCF per kg chemical rCF (see Table 2). Meanwhile, pyrolysis and chemical recycling processes result in a 12% and 11% resin savings per kg rCF compared to 5% for fluidised bed process. Primarily due to the difference from fibre replacement, pyrolysis and chemical recycling processes show higher net PED (-452.9 MJ and -431.8 MJ, respectively), in spite of additional benefits of by product, such as oil/wax from pyrolysis recycling process and epoxy resin from chemical recycling process. Similarly, GWP of fluidised bed recycling
process is the lowest amongst the selected recycling options, -25.9 kg CO₂eq./kg CFRP compared to 23.6 kg CO₂eq./kg CFRP for pyrolysis process and 23.7 kg CO₂eq./kg CFRP for chemical recycling process.

Sensitivity analysis of CF replacement ratio has been performed to analyse its impact on PED and GWP for CF replacement method based on variable fibre content. If fibre recovery can be undertaken with no degradation of fibre properties, then 1 kg rCF could displace 1 kg vCF (fibre volume fraction is 40% for both rCFRP and vCFRP). Compared to the base case, pyrolysis, fluidised bed and chemical recycling process could achieve further reductions in life cycle PED (12%, 5% and 10% further avoided energy use, respectively) and GWP (13%, 5% and 11% further reductions in GWP, respectively). For 20% mechanical property reduction, 1 kg rCF can replace 0.8 kg vCF (fibre volume fraction is 50% for rCFRP compared to 40% for vCFRP) and correspondingly there is 20% reduction in epoxy resin usage per kg rCF. As a consequence, recycling processes have less PED and GWP benefits relative to base case: PED is 92%, 86% and 92% of that of base case for pyrolysis, fluidised bed and chemical recycling process, respectively and GWP has a similar trend.

Figure 4 a) Primary energy demand and b) Global warming Potential of different CFRP waste treatment options including credits (CF replacement based on variable fibre content and material thickness)

**Carbon fibre replacement – variable material thickness**

Similar with results based on variable fibre content, overall results based on variable material thickness indicate the great environmental PED and GWP benefits obtained from recovery process where benefits are primarily from replacing vCF with rCF.
Environmental benefits of recycling processes achieved by avoiding intensive energy consumption associated with vCF production can be analysed by another replacement method based on variable material thickness assuming a constant fibre volume fraction. Pyrolysis recycling can achieve a net PED of -451.4 MJ/kg CFRP by accounting for the 490.26 MJ PED reduction from avoiding 0.6 kg vCF production. Chemical recycling process can only achieve a net PED of -431.8 MJ/kg CFRP while 90% benefits from vCF replacement and 10% from avoiding of virgin epoxy resin production as epoxy resin content can be recovered using chemical recycling technique. Fluidised bed recycling activities can achieve slightly larger net PED reduction of 497.6 MJ/kg CFRP than any other thermal recycling processes. The main difference is primarily due to the fibre replacement ratios of 0.94 kg vCF/kg pyrolysis rCF, 0.98 kg vCF/kg fluidised bed rCF and 0.95 kg vCF/kg chemical rCF, respectively. GWP results show similar trend that fluidised bed rCF to replace vCF leads the best GHG emission performance, -26.2 kg CO$_2$eq./kg CFRP.

Error bars shown in Figure 4 present PED and GWP of different thermal recycling processes when $\lambda$ value ranges in 1–3 relative to base case $\lambda$ value of 2 for replacement method based on variable material thickness. Results indicate that the relative performance remains similar regardless of the rCF application. At higher $\lambda$ values, greater weight reductions are achieved in material substitution, resulting in less rCF and polymer matrix required and as such relatively greater PED and GHG emission reduction. When $\lambda =3$, recycling processes are able to provide larger PED reductions relative to base case: 2.9%, 0.3%, and 2.6% reduction for pyrolysis, fluidised bed, and chemical recycling processes, respectively. In contrast, when $\lambda =1$, net PED has 7.2%, 3.6% and 6.4% increase for pyrolysis, fluidised bed and chemical recycling processes, respectively. GWP results show similar trends that different rCF applications based on $\lambda$ values result in up to 7% difference for these recycling processes.
Sensitivity analysis of variations of rCF mechanical property reductions of 20% and 0% are performed similar with that for replacement method based on variable fibre content. If there is no reduction in mechanical property of rCF relative to vCF, 100% vCF replacement can be made and the largest PED and GWP reduction can thus be achieved. Minimum net PED is -488.2 MJ, -513.6 MJ, and -461.0 MJ per kg CFRP waste for pyrolysis, fluidised bed and chemical recycling process, respectively, corresponding to 8%, 3% and 7% less than the base case assumed; similar trends for GWP results can be seen as well. These further reductions demonstrate the need for technology improvements in minimising mechanical properties reduction in the recovery process. Expected increase of PED and GWP can be seen when the rCF mechanical property reductions go up to 20%. Especially for fluidised bed process, there are 14% less PED and 15% less GWP reductions if rCF is 20% poorer than vCF in light of mechanical property. Previously, potential trade-offs have been identified between increasing feed rate and resulting rCF properties. In recycling process, in order to avoid agglomeration at high feed rates as in fluidised bed process, fibre length must be reduced. However, fibre length may also affect the downstream CFRP manufacturing process and resulting CFRP product properties. Ongoing and future research has to balance recycling performance and rCF properties for optimising environmental impacts as discussed above.

Discussion

Weight reduction achieved with CF composites can, in some applications such as aerospace, achieve substantial reductions in life cycle energy use and GHG emissions; however, the large energy requirement for vCF production can, in some uses, outweigh weight reduction and associated environmental benefits due to reduced fuel use during the vehicle life. Delivering more environmentally-beneficial CF materials to a wider range of opportunities depends on 1)
the development of energy efficient manufacturing processes for CF and CFRP (e.g., plasma
based processing or use of renewable energy feedstocks such as lignin), and 2) the recovery
of high quality rCF so that rCF can meet partial demands for vCF materials. In the current
dpaper, environmental impacts of current advanced CF recycling technologies (i.e., pyrolysis,
fluidised bed and chemical recycling process) have been assessed and our results show that
they can achieve similar and significant environmental (PED, GWP) benefits. This is because
rCF from recovery processes only experiences minor degradation of mechanical properties and
can be reused in place of vCF. The environmental benefits via replacement are achieved
primarily by avoiding the production of energy-intensive CF and preserving primary material.

It should be noted that for pyrolysis process, we assume all pyrolysis products can be
recovered and displace primary production, but have not included any recovery activities nor
considered the financial viability of recovering these materials, especially if in small quantities.
Similarly, for chemical recycling, we assume 1 kg resin recovered can replace 1 kg new epoxy
resin without considering any additional processes nor cost aspects. Therefore, their potential
environmental benefits can only be true provided that assumptions made are met. In addition,
as we are not accounting for use phase, and how component weight would impact use fuel
consumption, future work is suggested to analyse the environmental performances among these
advanced recycling technologies in the full life cycle. A final point around the pyrolysis data
used in this study is that they were primarily based on laboratory experiments and at larger
scale they may have a lower energy footprint.

Maintaining the mechanical properties of CF through the recycling processes is a key
challenge to overcome in developing a commercial CF recovery process and trade-offs between
increasing the throughput and maintaining mechanical properties clearly exist between the
competing recycling technologies. Advanced recycling processes such as fluidised bed can recover CF showing almost no reduction in modulus and 18-50% reduction in tensile strength relative to vCF. Retention of tensile modulus and tensile strength is also achievable in the studied recycling processes. In this paper, environmental performance of recovery processes with varied rCF mechanical properties have been investigated and the most favourable 100% retain of mechanical properties in rCF still depends on technical improvement. Current commercial recycling processes (e.g., pyrolysis) cannot recover rCF without mechanical properties reduction and will have to be selective in types of waste materials. Although fluidised bed recycling process has high tolerance to contamination and varied polymer types, it is still in transition to commercial scales.

As more CF is being produced, there are more opportunities for CF recovery from waste from manufacturing and, in due course, more end-of-life CF waste will also be available for recycling. Impacts of geographical location of waste could have an impact on environmental and cost impacts of recovery processes. Therefore, an analysis can be done in what future recycling systems would look like in terms of waste CFRP availability and location, which would impact plant capacity, waste collection, transportation, location, and regional factors.

The advanced CF recycling technologies investigated in this study currently exist at varying levels of technological maturity: pyrolysis is operated at commercial scale; fluidised bed recycling has been proven at demonstration scale; and the chemical recycling process modelled here is still on a laboratory scale. There is greater uncertainty in estimating the life cycle impacts of emerging technologies; while data available for pyrolysis and fluidised bed systems are comparatively robust, present estimates for chemical recycling do not consider how the process could be developed for deployment at commercial scale (energy sources; minimisation
of heat losses; process yield and efficiency; recovery of non-consumed process inputs). As such, the environmental impacts of the chemical recycling process could, in practice, differ significantly from the results presented here and in previous publication\textsuperscript{16} and warrant further investigation through process simulation/optimisation and, ultimately, validation by operating commercial facilities. However, we note that the net energy and GHG impacts of rCF are dominated by the displacement of vCF production, which provides a credit to the system approximately 10 to 20 times greater than the estimated impacts of the recycling process. Process efficiency improvements, therefore, would have a marginal impact on net life cycle metrics evaluated in the present study, so long as the mechanical properties of rCF are maintained to enable use in place of vCF.

While CFRP recycling has a range of potential advantages, challenges and opportunities co-exist. Replacing vCF with rCF in CFRP manufacturing may be limited for applications requiring the highest integrity materials (e.g., aircraft primary structure), but is viable for non-structural lightweighting applications with less extreme strength requirement. Achieving the high level of environmental benefits via recycling also depends on potential market. The potential applications for replacing vCF with rCF are some transportation industries (e.g., automotive, or infrastructure (replace vCF in bridges), wind energy and sporting industries (e.g., races boats using rCF reported by Land Rover)).\textsuperscript{8,40} But there is of concern about the mismatch between supply of rCF (estimated at about 50,000 t/yr in 2017) and potential demands. The source of rCF is currently only industrial waste and end-of-life CFRP wastes are not available in significant quantities yet. The automotive industry, for instance, needs high volumes of material that are generally larger than industrial waste from aerospace and other sectors currently using CF (in excess of 95 million vehicles globally in 2015\textsuperscript{41}). It is therefore significant to design the best recycling capacity, combination of advanced recycling technology,
and find the best target market to obtain the maximum potential environmental benefits.

Furthermore, full consideration of their environmental and financial impacts\textsuperscript{40} would enable inclusion of trade-off between CFRP waste management strategies and help to ensure opportunities are pursued that provide the greatest overall net benefit.

Appendix A

Table A1 Inventory data of different CFRP recycling technologies

Supporting information

There is no SI file for publication.

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Notes

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Table 1 CFRP recycling technologies: geographical location, key processing parameters and capacity

| Recycling method       | Recycling plant                                      | Temperature, °C | Pressure       | Processing capacity | Source |
|-----------------------|------------------------------------------------------|-----------------|----------------|--------------------|--------|
| Mechanical recycling  | University of Manchester, UK                         |                 | Ordinary       | ~20 t/yr (scaled from lab plant) | 14     |
|                       | Toray Industries, Teijin, Mitsubishi Rayon           | 500-700         | Ordinary       | 1000 t/yr           | 16     |
|                       | Takayasu, Japan                                      | N/A             | Ordinary       | 60 t/yr             | 16     |
|                       | ELG, UK                                              | 350-800         | N/A            | 2000 t/yr           | 42     |
| Pyrolysis             | CFK Valley Stade Recycling GmbH & Co KG, Germany     | 350-800         | N/A            | 1000 t/yr           | 43     |
|                       | Carbon Conversions, USA                              | 350-800         | N/A            | 2000 t/yr           | 44     |
|                       | KARBOREK RCF, Italy                                  | 350-800         | N/A            | 1000 t/yr           | 45     |
| Fluidised bed         | Pilot plant, University of Nottingham               | 400-600         | Below atmospheric | 50 t/yr           | 9      |
|                       | Shizuoka University, Japan                           | 250-350         | 5-10 MPa       | 5 L                | 16     |
| Supercritical fluid   | Hitachi Chemical, Japan                              | 200             | Ordinary       | 12 t/yr             | 16     |
|                       | University of Birmingham, UK                         | 320             | 17 MPa         | 5 L (lab batch)     | 15     |
| Subcritical fluid     | Kumamoto University                                  | 300-400         | 1-4 MPa        | 0.5 L (lab batch)   | 16     |
Table 2 Recycled CFRP design properties for CF replacement method based on variable fibre content and variable material thickness.

|                | Fibre Tensile modulus, GPa | Epoxy resin Tensile modulus, GPa | Modulus Reduction | Density, kg/m³ | Variable fibre content | Variable material thickness |
|----------------|----------------------------|----------------------------------|-------------------|----------------|------------------------|-----------------------------|
|                |                            |                                  |                   |                | Fibre volume fraction, Vf | Fibre ratio, vCF/rCF | (Incremental) resin use, kg/kg rCF | Mass ratio, rCF/vCF | Fibre volume fraction, Vf | Fibre ratio, vCF/rCF | (Incremental) resin use, kg/kg rCF | Mass ratio, rCF/vCF |
| vCF            | 230.00<sup>46</sup>       | 5.00                             | -                 | 1.79           | 40%        | -                        | -                            | 40% | -                        | -                            | -                        |
| Pyrolysis rCF  | 202.40<sup>31</sup>       | 5.00                             | 12.0%             | 1.79           | 46%        | 0.88                      | -0.12                       | 114% | 40%                      | 0.94                        | +0.09                      | 106%                        |
| Fluidised bed rCF | 218.04<sup>31</sup>  | 5.00                             | 5.2%              | 1.79           | 42%        | 0.95                      | -0.05                       | 106% | 40%                      | 0.98                        | +0.04                      | 102%                        |
| Chemical rCF   | 204.70<sup>31</sup>       | 5.00                             | 11.0%             | 1.79           | 45%        | 0.89                      | -0.11                       | 113% | 40%                      | 0.95                        | +0.08                      | 105%                        |
Table 3 Primary energy demand and Global Warming Potential comparison of the carbon fibre recovery and conventional landfill and incineration without credits from the use of products of recycling processes.

|                  | PED, MJ/kg CFRP | GWP, kg CO$_2$eq./kg CFRP |
|------------------|-----------------|---------------------------|
|                  | Landfill | Incineration | Recycling | Transport | Total  | Landfill | Incineration | Recycling | Transport | Total  |
| Landfill         | 0.97     | -           | -         | 0.14      | 1.11    | 0.13     | -           | -         | 0.01      | 0.14   |
| Incineration     | -        | 1.17        | -         | 0.54      | 1.71    | -        | 3.09        | -         | 0.03      | 3.12   |
| Mechanical+      | 0.16     | -           | 0.69      | 0.35      | 1.20    | 0.05     | -           | 0.03      | 0.02      | 0.11   |
| Mechanical+      |          |             |           |           |         |          |             |           |           |        |
| landfill         |          |             |           |           |         |          |             |           |           |        |
| Mechanical+      | -        | -           | 0.69      | 0.43      | 1.12    | -        | 1.74        | 0.03      | 0.03      | 1.80   |
| Mechanical+      |          |             |           |           |         |          |             |           |           |        |
| incineration     |          |             |           |           |         |          |             |           |           |        |
| Pyrolysis        | -        | -           | 37.09     | 0.27      | 37.36   | -        | -           | 2.88      | 0.02      | 2.90   |
| Fluidised bed    | -        | -           | 9.98      | 0.27      | 10.25   | -        | -           | 1.54      | 0.02      | 1.56   |
| Chemical         | -        | -           | 38.12     | 0.27      | 38.39   | -        | -           | 1.51      | 0.02      | 1.53   |
Figure 1 CFRP Waste treatment routes.
Figure 2 System diagram showing material and energy flows for a) mechanical recycling with landfilling and mechanical recycling with incineration, b) pyrolysis recycling, c) fluidised bed recycling and d) chemical recycling processes. The arrow colour indicates a type of each flow (red – energy flow, green – credits, and black – material flow).
Figure 3 Tensile properties of an epoxy recycled carbon fibre composite experimentally measured. The solid and dotted lines represent the theoretical stiffness calculated using the generalized rule for randomly distributed and perfectly aligned fibres.\textsuperscript{33}
Figure 4 a) Primary energy demand and b) Global warming Potential of different CFRP waste treatment options including credits (CF replacement based on variable fibre content and material thickness)

Table A1 Inventory data of different CFRP recycling technologies

| Input                                      | Mechanical+ landfill | Mechanical+ Incineration | Pyrolysis | Fluidised bed | Chemical |
|--------------------------------------------|----------------------|--------------------------|-----------|--------------|----------|
| CFRP waste, kg                            | 1.00                 | 1.00                     | 1.00      | 1.00         | 1.00     |
| Acetic acid, kg                           |                      |                          |           |              | 0.83     |
| Deionised water, kg                       |                      |                          |           |              | 2.50     |
| Sodium hydroxide, kg                      |                      |                          |           |              | 0.07     |
| Transport, km                             |                      |                          |           |              |          |
| Cutting & shredding                       |                      |                          |           |              |          |
| Recycyling                                |                      |                          |           |              |          |
| Electricy, MJ                             |                      |                          | 7.6       | 3.4          | 6.5      |
| Natural gas, MJ                           |                      |                          |           |              |          |
| rCF, kg                                   | 0.24                 | 0.24                     | 0.62      | 0.62         | 0.62     |
| Fine powder, kg                           | 0.19                 | 0.19                     |           |              |          |
| Coarse fraction, kg                       | 0.57                 | 0.57                     |           |              |          |
| Epoxy resin, kg                           |                      |                          |           |              | 0.67     |
| Char, kg                                  |                      |                          |           |              | 0.14     |
| H₂O, kg                                   |                      |                          |           |              | 0.27     |
| CO₂, kg                                   |                      |                          |           |              | 1.02     |
| Incinerating CO₂ from natural gas, kg     |                      |                          |           |              | 0.06     |
| NO₂, kg                                   |                      |                          |           |              | 0.03     |
| Liquid                                    |                      |                          |           |              |          |
| Water                                     |                      |                          |           |              |          |
| Oil/wax                                   |                      |                          |           |              |          |
| Pentane, kg                               |                      |                          |           |              | 0.03     |
| Benzene, kg                               |                      |                          |           |              | 0.03     |
| Ethylacetate, kg                          |                      |                          |           |              | 0.10     |
| Methanol, kg                              |                      |                          |           |              | 0.04     |
| Gas                                       |                      |                          |           |              |          |
| CO, kg                                    |                      |                          |           |              | 0.001    |
| CO₂, kg                                   |                      |                          |           |              | 0.003    |
| H₂, kg                                    |                      |                          |           |              | 0.000    |
| Compound          | Mass (kg) |
|-------------------|-----------|
| CH₄               | 0.003     |
| C₂H₆             | 0.002     |
| C₃H₈             | 0.001     |
| Propene/propylene| 0.002     |
Figure shows gate-to-gate PED and GWP of waste management options for CFRP waste.

Sources from author Fanran Meng, Microsoft PowerPoint and author’s publication³

The paper is of particular relevance for Sustainability given its focus on the life cycle environmental assessment of recovery of CFRP waste materials compared with landfill and incineration.