Sankar Karuppannan Gopalraj

IMPACTS OF RECYCLED CARBON FIBRE AND GLASS FIBRE AS SUSTAINABLE RAW MATERIALS FOR THERMOSETTING COMPOSITES

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Abstract

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Impacts of recycled carbon fibre and glass fibre as sustainable raw materials for thermosetting composites
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The fibre-reinforced polymer composites were implemented in various applications as a potential alternative to traditional metals. In particular, the carbon fibre-reinforced polymer (CFRP) and glass fibre-reinforced polymer (GFRP) composites were heavily utilised in high-performance applications in the past two decades. As these composites reach their end-of-life stage, the consumers and waste management industries still rely on non-sustainable waste disposal options such as landfill and incineration. The cumulating tonnes of composite wastes contain valuable carbon fibres (CFs) and glass fibres (GFs), which still possess the potential to serve in applications for multiple cycles. When these composite wastes are adequately recycled and reused to close their life-cycle loop into new recycled composites, the wastes can be reduced. At the same time, the global demand for composites can be supplied without manufacturing virgin composites. However, there are certain challenges to overcome in order to achieve such goals. Thanks to the government regulations and public awareness on the environmental impacts of producing these virgin composites instead of reusing the existing composite wastes.

This dissertation focuses on overcoming such challenges in closing the life-cycle loop of the CFRP and GFRP composite materials with fibre recovery as a primary focus. The different types of recycling processes currently available were initially studied, focusing on the mechanical properties of the recycled fibres/composites and the environmental impacts of the recycling processes. Subsequently, a thermal recycling process was developed practically with optimised conditions to successfully recycle both the CFs and GFs from their respective composite wastes and remanufactured by compression moulding employing a fresh resin system. The mechanical properties of the recycled fibres and their newly produced composites were investigated using ISO standard tensile and impact tests both experimentally in the laboratory and numerically using finite element methods. Plus, the environmental impacts of the developed process were analysed and compared with traditional CFRP and GFRP composite waste disposal methods using life cycle analysis methods.

The results show that the developed thermal recycling process is capable of recycling both CFRP and GFRP composite wastes to produce unidirectional, long and continuous CFs and GFs. The mechanical properties of the recycled fibres and composites are in an acceptable range with similar results of recycled composites from literature. The environmental impacts are favourable to recycling CFRP wastes using the developed thermal recycling process and GFRP wastes using co-incineration as feedstock during cement production. Furthermore, discussions are made to overcome the challenges observed during this developed open-loop recycling process. When these challenges are addressed while upscaling the processes into a pilot and further to an industrial scale, a closed-loop recycling process can be achieved with mechanical properties of the recycled fibres/composites similar to their virgin counter. Thus, successfully establishing a circular economy.

Keywords: life-cycle loop, circular economy, waste management, carbon fibres, glass fibres

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This research work was carried out in LUT School of Energy Systems, primarily in the LUT Fiber Composite Laboratory and partially in the Department of Sustainability Science at Lappeenranta-Lahti University of Technology LUT, Finland, between 2018 and 2022. First of all, I would like to express my immense gratitude to professor Timo Kärki for providing me with this opportunity to pursue a doctoral degree under his supervision. Next, I would like to express my deepest gratitude to Professor Andreas Krause and Dr Tommi Vuorinen for their priceless time and effort in reviewing this dissertation work. I am thankful to receive such positive feedback and constructive comments to improve the quality of this work further. Additional thanks to Dr Tommi Vuorinen for gracefully accepting the role of the opponent in my public examination.

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Sankar Karuppannan Gopalraj
August 2022
Tampere, Finland

Contents

List of publications

This dissertation is based on the four peer-reviewed articles published in internationally recognised scientific journals. All these articles were published under open-access publishing agreements, and the reprints of each article are included at the end of this dissertation. The rights have been granted by publishers to include the articles in dissertation. They are referred to in the text by their numerals.

I. **Karuppannan Gopalraj, S.**, and Kärki, T. (2020). A review on the recycling of waste carbon fibre/glass fibre-reinforced composites: fibre recovery, properties and life-cycle analysis. *SN Applied Sciences*, 2, 433, pp. 1–21

II. **Karuppannan Gopalraj, S.**, and Kärki, T. (2020). A Study to Investigate the Mechanical Properties of Recycled Carbon Fibre/Glass Fibre-Reinforced Epoxy Composites Using a Novel Thermal Recycling Process. *Processes*, 8(8), 954, pp. 1–16

III. **Karuppannan Gopalraj, S.**, and Kärki, T. (2021). A Finite Element Study to Investigate the Mechanical Behaviour of Unidirectional Recycled Carbon Fibre/Glass Fibre-Reinforced Epoxy Composites. *Polymers*, 13(8), 3192, pp. 1–24

IV. **Karuppannan Gopalraj, S.**, Deviatkin, I., Horttanainen, M., and Kärki, T. (2021). Life Cycle Assessment of a Thermal Recycling Process as an Alternative to Existing CFRP and GFRP Composite Wastes Management. *Polymers*, 13(24), 4430, pp. 1–17

Author’s contribution

Mr Sankar Karuppannan Gopalraj is the first and corresponding author and principal investigator in all the articles. The four published articles and this dissertation are documented under the supervision of Prof. Timo Kärki from LUT Fibre Composite Laboratory, Finland.

Publication I: A review article briefly describing the background literature to recycle carbon and glass fibre-based composite wastes emphasised identifying the primary research gaps in their recycling processes, recycled properties and sustainability aspects.

The study conception, material preparation, data collection, manuscript structuring and writing of the original draft was performed by the first author, Sankar Karuppannan Gopalraj. The second author, Prof. Timo Kärki, supervised the project, contributed to the manuscript structuring and commented on the manuscript drafts before submitting the final article.

Publication II: A research article investigating a laboratory-scale thermal recycling process to recycle fibre composite wastes. The study also encloses compression moulding the recycled fibres into new composites and measuring their mechanical properties utilising ISO standard tests.

The raw materials collection, sample preparation, process development, design of experiments, laboratory work, sample testing, results analysis and writing of the original manuscript were performed by the first author, Sankar Karuppannan Gopalraj. The second author, Prof. Timo Kärki, supervised the project and provided feedback to the manuscript drafts before submitting the final article.

Publication III: A research article investigating the mechanical properties and failure behaviours of unidirectional recycled fibre composite materials utilising finite element methods.

The study conception, methodology, data analysis and calibration, software, validation, modelling investigation, result analysis and writing of the original manuscript were performed by the first author, Sankar Karuppannan Gopalraj. The second author, Prof. Timo Kärki, supervised the project and provided feedback to the manuscript drafts before submitting the final article.

Publication IV: A research article to evaluate the environmental impacts of various fibre composite waste management options. The study includes a comprehensive analysis of the developed thermal recycling process compared to the traditional waste management options.

The conceptualisation was performed by the first author, Sankar Karuppannan Gopalraj, based on the discussions with Prof. Timo Kärki. The first author formulated the research plan, research aims, assisted the LCA study and wrote the original manuscript. The second author, Dr Ivan Deviatkin, selected the methodology, performed the LCA study, wrote the manuscript's inventory analysis, and provided feedback to the final manuscript draft. Prof. Timo Kärki supervised the project and provided feedback to the manuscript drafts before submitting the final article. Prof. Mika Horttanainen supervised the project.

Supplementary studies

- Karuppannan Gopalraj, S., and Kärki, T.: Fire properties of CFRP and GFRP. *FiBreMoD Conference*, KU Leuven, Belgium, December 11–12, 2019

- Karuppannan Gopalraj, S., and Kärki, T.: An investigative study on the compression moulding process to manufacture fibre-reinforced epoxy composites using recycled carbon and glass fibres. *26th International Conference on Production Research (ICPR 26)*, Taichung University, Taiwan, July 18–21, 2021

- Ullah, M., Karuppannan Gopalraj, S., Gutiérrez Rojas, D., Nardelli, P., and Kärki, T.: IoT framework and requirement for intelligent industrial pyrolysis process to recycle CFRP composite wastes: application study. *26th International Conference on Production Research (ICPR 26)*, Taichung University, Taiwan, July 18–21, 2021

- Karuppannan Gopalraj, S., and Kärki, T.: Financial and industrial-scale viability of recycling carbon and glass fibres using a novel thermal recycling process. *HyFiSyn Conference*, KU Leuven, Belgium, September 15–16, 2021

- Karuppannan Gopalraj, S., and Kärki, T.: Investigating a thermal recycling process to recover carbon and glass fibres from CFRP and GFRP composite wastes. *23rd International Conference on Composite Materials (ICCM 23)*, Belfast, UK, July 30–August 4, 2021 (Paper accepted, but conference postponed until 2023 due to COVID–19)

Nomenclature

Latin alphabet

F^{pl}	plastic deformation
F^{el}	elastic deformation
F^{T}	total deformation

Greek alphabet

$\bar{\varepsilon}_0^{pl}$	equivalent plastic strain rate
$\bar{\varepsilon}_D^{pl}$	damage onset
η	function of stress triaxiality

Abbreviations

ADP	Abiotic depletion potential
CC	Cone calorimeter
CF	Carbon fibre
CFE	Carbon fibre-reinforced epoxy
CFRP	Carbon fibre-reinforced polymer
FBP	Fluidised Bed Process
EC	European Commission
EoL	End-of-Life
EP	Epoxy resin
EU	European Union
FVF	Fibre volume fraction
FEM	Finite element methods
GF	Glass fibre
GFP	Glass fibre-reinforced polyester
GFRP	Glass fibre-reinforced polymer
GWP	Global warming potential
ISO	International Organisation for Standards
LCA	Life cycle analysis
UD	Unidirectional
rCF	Recycled carbon fibre
rCF/EP	Recycled carbon fibre-reinforced with epoxy
rCFRP	Recycled carbon fibre-reinforced polymer
rGF	Recycled glass fibre
rGF/EP	Recycled glass fibre-reinforced with epoxy
rGFRP	Recycled glass fibre-reinforced polymer
RVF	Resin volume fraction
SEM	Scanning electron microscopy
SS	Stress-strain
vCF	Virgin carbon fibre

vCFRP	Virgin carbon fibre-reinforced polymer
vGF	Virgin glass fibre
vGFRP	Virgin glass fibre-reinforced polymer
WPCB	Waste printed circuit board
wt%	Weight percentage

Units

CO_2-eq	Carbon dioxide equivalent
°C	Celsius
m^3	Cubic meter
E	Young's modulus (Elastic modulus)
GPa	Gigapascal
g	Grams
GW	Gigawatt
kg	Kilogram
kJ	Kilojoule
μm	Micrometer
m	Mass
MJ	Megajoules
MPa	Megapascal
mm	Millimeter
min	Minutes
N	Newton
Pa	Pascal
m^2	Square meter

1 Introduction

Fibre-reinforced polymer (FRP) composites have gained popularity in recent decades due to their immense development in mass manufacturing strategy, mechanical properties and value for money. These composites have gradually replaced traditional materials such as metals and alloys in various applications [1,2]. Amongst these FRP composites, carbon fibre-reinforced polymer (CFRP) and glass fibre-reinforced polymer (GFRP) composites have predominantly dominated high-performance applications—such as automobile, aeronautics, aerospace, defence, electronics, construction, sports, medicine, electronics and renewable energy—where safety is prioritised before the performance [2,3]. These composites have a higher strength-to-weight ratio, which means they are stronger than metals but half their weight.

The technical advancement in recent times has exponentially boosted the usage of CFRP and GFRP composites. The composite market analysis reports [4,5] from 2016 highlighted that the FRP composite industry was globally valued at $114.13 billion, with GFRP composites having the highest contribution of 65%, followed by CFRP composites at 11–13%. The reports also forecasted that by 2025 the global market would be valued at $282.9 billion with a steady increase primarily focusing on CFRP and GFRP composites. Furthermore, the GFRP reports [6,7] suggest that in 2020 the global market value was $46 billion and is expected to grow up to $65.9 billion by 2027. Similarly [8], in 2019, the global market value for CFRP composites was $20.55 billion and is expected to grow up to $34.69 billion by 2026. The forecast reports have spontaneously indicated steady growth in the composite market value. The advancements in modernised manufacturing processes are cable of producing these composites efficiently at a faster rate without proper awareness of their waste management options in the later phase.

The higher volume consumption of GFRP over CFRP composites can be associated primarily with their cost. The commercial selling price of 1 kg carbon fibres (CFs) varies between $20–$90 [9]. In some cases, premium aerospace-grade CFs can cost up to $200 [10]. On the other hand, the commonly used E-type glass fibres (GFs) for reinforcing with polymers as GFRP composites sell around at < $2.5 per kg. Such drastic differences in the price range reflect the production process involved in manufacturing virgin fibres. CF production is a highly energy-intensive process that consumes around 200–600 MJ/kg [11], and expensive it becomes to achieve premium grades. As a result, virgin carbon fibre (vCF) production generates higher carbon footprints compared to traditional steel production and even virgin glass fibre (vGF) production. It is to be noted that GFRP possesses considerable impacts as they are produced in a higher volume than CFRP composites contributing equal impacts altogether. As these fibres are often reinforced with thermosetting polymers, the possibilities for sustainable waste management options are even more challenging.

Despite their price tag and environmental impacts, composite industries have heavily consumed CFRP and GFRP composites in the past decades and employed them in diverse applications. One such example can be noticed in the advanced aircraft carriers. Figure 1 illustrates the aircraft's heatmap concerning the material contribution. As seen, CFRP composites dominate the material composition covering a significant portion. Modern aircraft like Airbus a350 xwb (extra wide body) possess 53% composite in their material profile by weight [12], and Boeing 787 Dreamline possess 50% composite employment compared to their predecessor 777 with only 12% composite material used in the overall profile [13]. It can be stated that the composite consumption in these aircraft will continuously increase as the manufacturers significantly progress towards lightweight aircraft to reduce fuel consumption. Furthermore, the recent EU launch of clean aviation to accomplish zero-emission by 2050 and achieve climate-neutral in aviation has estimated a target of 75% of traditional civil aviation aircraft to be replaced with hydrogen-powered electricity run aircraft [14]. Thus, these aircraft can be expected sooner or later to be in their waste management phase.

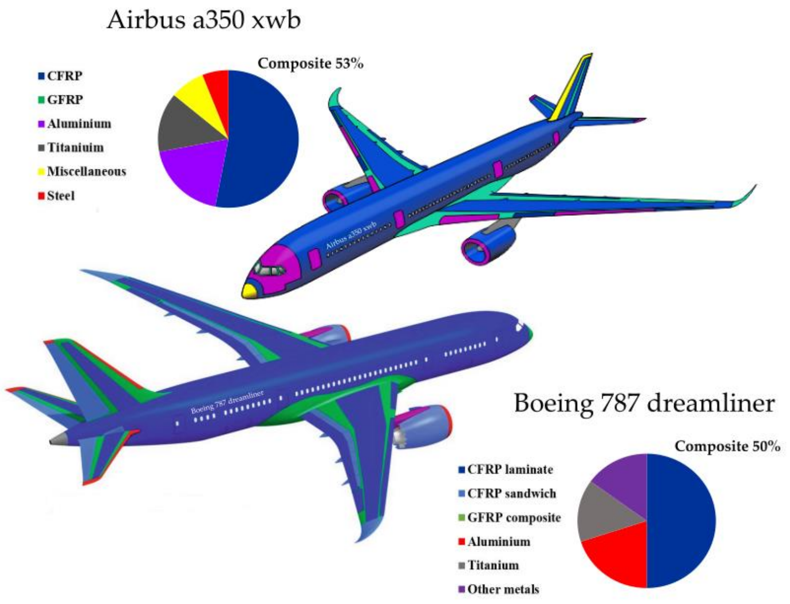


Figure 1: CFRP and GFRP composite used in modern aeroplanes [12,13]

Similarly, these composites have also been used in higher volumes in wind turbine applications. Figure 2 illustrates the materials used in wind turbine blades. GFRP composites dominate the material consumption, with GFs occupying 60–70% and resins 30–40% by weight [15]. CFRP composites are also used in a lower volume compared to GFRP composites. The blades size increases 50–80 m as the wind turbine capacity increases resulting in heavy composite consumption. According to the latest report [16], the global wind turbine installation capacity per annum in 2020 was 93 GW, and in order to achieve net-zero carbon emissions by 2050, the estimated installation rate should be three times more per annum which means high consumption of CFRP and GFRP composites.

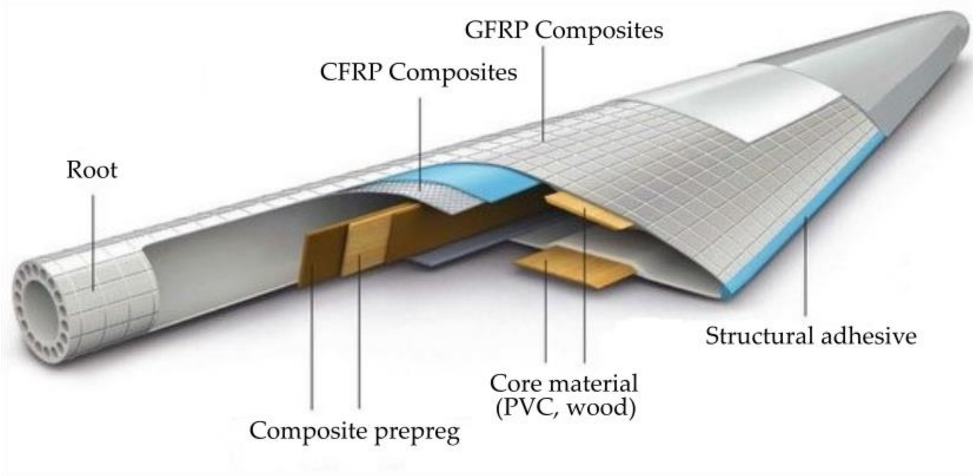


Figure 2. Cross-section of a wind turbine blade [17]

At the same time, the wastes generated by CFRP and GFRP composites are in enormous quantities. The sources of these cumulating wastes can be primarily tracked towards end-of-life waste categories such as old aeroplanes and wind turbines from the 1980s whose service period of 20 years exceeded and is available for the disposal phase. By 2050, the CFRP composite EoL wastes from the aviation industry are estimated to collect half a million tonnes [18]. A similar volume of GFRP composite EoL wastes from wind turbine applications is estimated to be available for disposal [19]. The rest of the waste source categories are automobile, electronic waste, otherwise known as e-waste, composite manufacturing waste and expired prepregs. For decades, the well-known domestic waste disposal route, landfilling, has predominantly been the favourite waste management option for these composite wastes. However, the transportation to the landfilling location typically far from the urbanised regions demanded the necessity for a cheap alternative disposal option by the waste management industries, which resulted in the non-eco-friendly route of incinerating the composite wastes with energy recovery.

The government policies [20] towards proper waste management with recycling as the highest priority was implemented in 2008, changing the attitude of the recycling companies to see waste as a valuable raw material when adequately recycled. Eventually, it paved new ways for recycling industries and composite experts to develop novel recycling methods to recover the valuable CF and GF from their CFRP and GFRP composites [2]. Additionally, heavy taxation was implemented to reduce landfilling. As the global demand for new composites increases, recycling and reusing the CFRP and GFRP composite wastes were promising to reduce the cumulating waste while satisfying the global composite demand promoting a circular economy model. As a result, emerging recycling processes reduced the practice of incineration

1.1 Background and motivation

The demand for CFRP and GFRP composites have shown a steady increase similar to their corresponding wastes. As the wastes contain CFs and GFs, properly recovering and reusing them will solve the global demand to a certain extent. Thanks to government regulations and environmental protection organisation's awareness, the possibility of waste recycling is likely to become a reality towards sustainable waste disposal route. The initial efforts of the EU directive (2000/53/EC) in EoL automobile materials, a recovery rate of 95% is required, among which 85% mass to be reused further by 2015 made new trends towards environmental protection [21]. This was followed by the EU directive (2008/53/EC) defining a framework to introduce the basic concepts towards waste management, establishing a waste management hierarchy with recycling given the high priority [20].

Additionally, heavy taxation towards landfilling has made it even more expensive. For example, it costs around € 150–170 per tonne of composite waste to be landfilled in the UK. Also, the report [22] from 2011 has stated to treat GFRP waste using co-incineration process to recover both material and energy and use as feedstocks in cement production. As landfilling and incineration are no longer reliable waste management methods, industries have seen this opportunity to adequately recycle the composite wastes in order to establish a new market which both profitable and sustainable. Despite the pandemic, the post-pandemic period is expected to be developing towards sustainable recycling options. The evidence can be observed from various Horizon 2020 [23] and Horizon Europe [23] digital twin EU funding programs ready to support research centres, universities and composite industries to come forward in topics relevant to developing CFRP and GFRP recycling processes. Thus, recycling and reusing are seen as a sustainable future, and further exploration of various recycling processes and approaches is required to make it a reality.

1.2 Aim and scope

The main aim of this thesis is to ***close the life-cycle loop of the CFRP and GFRP composite wastes***.

The study primarily focuses on developing selection criteria to identify a suitable recycling approach to recycle, remanufacture, and reuse the valuable CFs and GFs from their respective CFRP and GFRP composite wastes for numerous cycles, encouraging a circular economy. Subsequently, the study will provide insights into developing an open-loop recycling approach consisting of an optimised laboratory-scale recycling process with remanufacturing and mechanical testing of the recycled composites both experimentally and numerically. Additionally, the environmental impacts of the developed approach are investigated further to compare with the traditional CFRP and GFRP composite waste management options.

1.3 Hypothesis and research questions

The hypothesis of this thesis work is established to be a closed-loop recycling approach as a sustainable waste management option for CFRP and GFRP composite. It will focus on recycling, remanufacturing, and reusing the composite wastes into a similar application as an alternative for virgin CFRP and GFRP composites to achieve a circular economy.

The following research questions are discussed in this study:

1. What are the process parameters to thermally recycle CFRP and GFRP composite wastes?
2. What are the mechanical properties of the recycled CFRP and GFRP composites?
3. What are the challenges in performing a simulation for recycled composites?
4. What are the environmental impacts of a thermal recycling process compared to traditional waste management routes for CFRP and GFRP composites?
5. Are recycled composites a suitable replacement for virgin composites?

1.4 Framework

Figure 3 illustrates the framework of the thesis work.

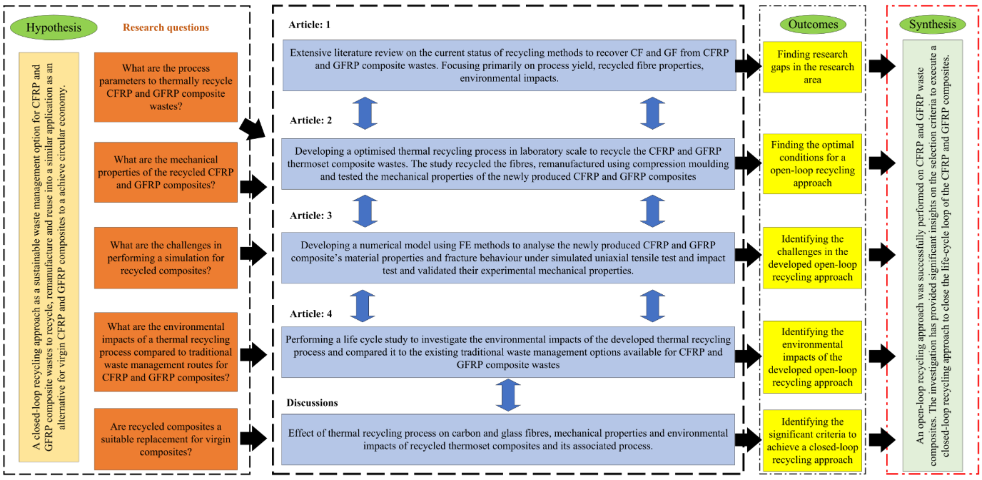


Figure 3. Proposed framework of the thesis

2 State of the art

2.1 Sustainable waste management

The focus towards sustainable waste management options has increasingly developed new government policies influencing the composite waste's fate directly and, in some cases, indirectly. The updated EU regulation in 2018 has solidly concentrated on defining the rules to classify waste categories enabling various possible options for waste management industries. At the same time, it has primarily promoted reusing, forcing it further in order to create a benchmark for all future waste management options. The regulation 2018/852 [24] is considered to be a significant update to its previous regulations such as 1994/62/EC, 1999/31/EC, 2000/53/EC, 2008/98/EC focusing on waste management and setting new goals to achieve carbon-neutral gradually in the following years. Even though these regulations have no direct influence over the CFRP and GFRP composite waste management from applications such as aeroplanes and wind turbines, researchers have adopted approaches resembling to achieve a sustainable route to prevent the cumulating composite wastes [25].

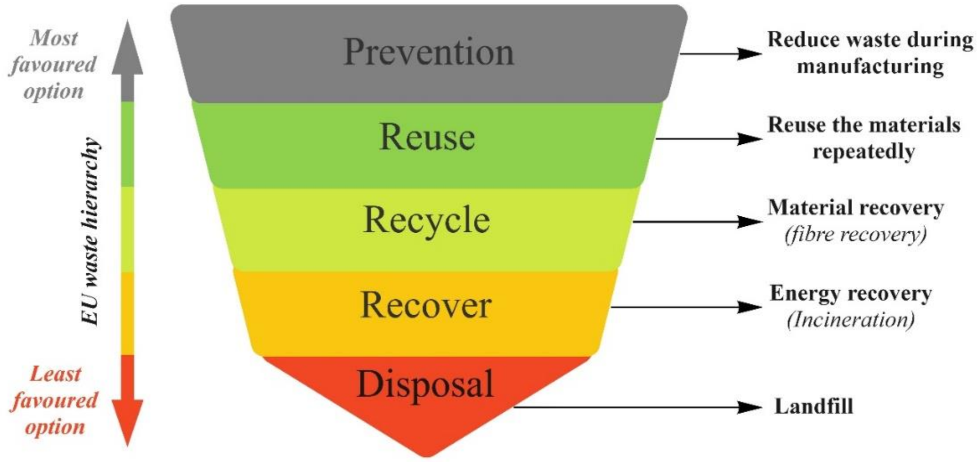


Figure 4. The waste hierarchy defined by the EU

As a result of these regulations and policies, a well-organised waste management system was established, as seen in Figure 4. It illustrates the state-of-art EU waste management hierarchy, with waste disposal being the least preferred option. Recycling and reusing the wastes with proper material recovery in a higher percentage is considered the most preferred option in the waste management phase. However, there are certain challenges to overcome in order to achieve maximum waste recycling. Typically, the composites fundamentally have two materials, the fibres and the thermoset polymers called resin, to reinforce and bind the fibres as one single composite. Various studies [2][26] have highlighted that fibre recovery is a potential starting point for CFRP and GFRP composite wastes. It contains the valuable CF and GF. However, factors like fibre arrangement, type, orientation, and length play a significant role in recycling. It is required to carefully execute the recycling approach as the goal is to close the life cycle loop of the composite waste.

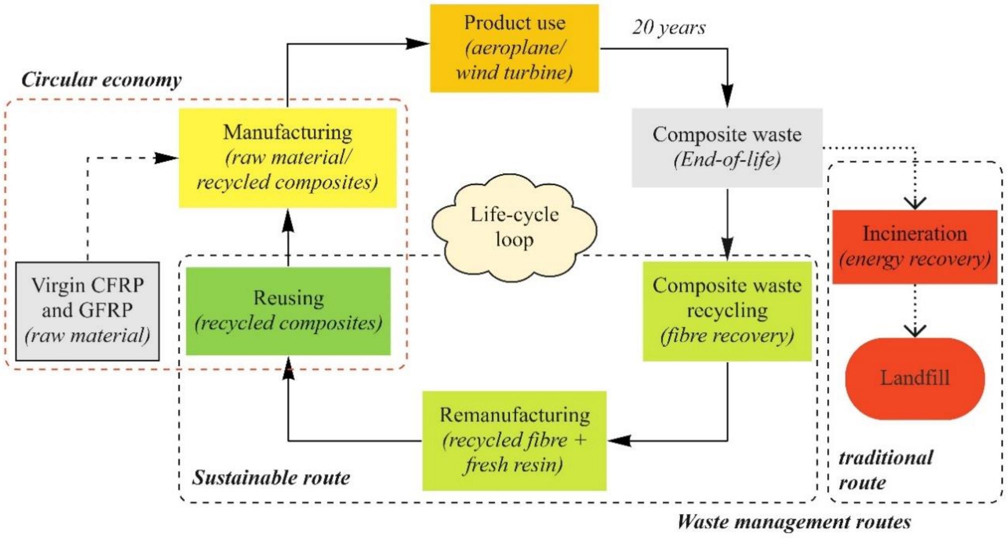


Figure 5. Life-cycle loop of CFRP and GFRP composites

Figure 5. illustrates a typical outlook of a CFRP and GFRP composite waste recycling model to close their life-cycle loop. As seen, non-material recovery processes such as landfills and incineration tend to stop the life-cycle loops with either a non-profitable energy recovery approach or holding the wastes for the future. However, with a sustainable alternative route by adequately recycling the wastes implementing fibre recovery, remanufacturing and reusing into applications replacing virgin composites can be considered closing their life-cycle loop once successfully. It is required to repeat the cycle multiple times until their potential is fully utilised. This sustainable approach not only uses recycled composites but, at the same time, reduce virgin composite production to a certain extent. Overall, a circular economy can be provoked, satisfying the EU's latest carbon-neutral demands by 2050. Still, there are certain challenges in executing such a sustainable route with maximum efficiency in fibre recovery and reclaiming their strength to match the mechanical properties of their virgin counterparts. Researchers have contributed to overcoming such challenges for the past two decades. They have developed various recycling processes capable of recovering CFs and GFs with higher mechanical properties and maximum fibre recovery rates.

2.2 Recycling to Remanufacturing: processes and parameters

To effectively close the life-cycle loop of the CFRP and GFRP composite wastes, it is required to follow the sustainable route favouring the material recovery approach. Figure 6 illustrates the various material reclaiming processes currently available to recycle the CFRP and GFRP composite wastes. These available recycling processes primary focus on separating the CFs and GFs from their resin system by predominantly utilising heat and chemicals at various favourable conditions. Additionally, the development of these processes is available in various stages such as laboratory-scale, pilot plant and fully operational industrial-scale. The mechanical recycling process was initially started as a primary recycling process. The process reduces the size of the composite wastes and can operate on both CFRP and GFRP wastes [27–29]. It involves processes like cutting, shredding, and milling the composite wastes of 50–70 m to achieve sizes in the range of 50–100 mm. The size-reduced wastes are combined with new materials into low-value applications like additives to strengthen the concretes.

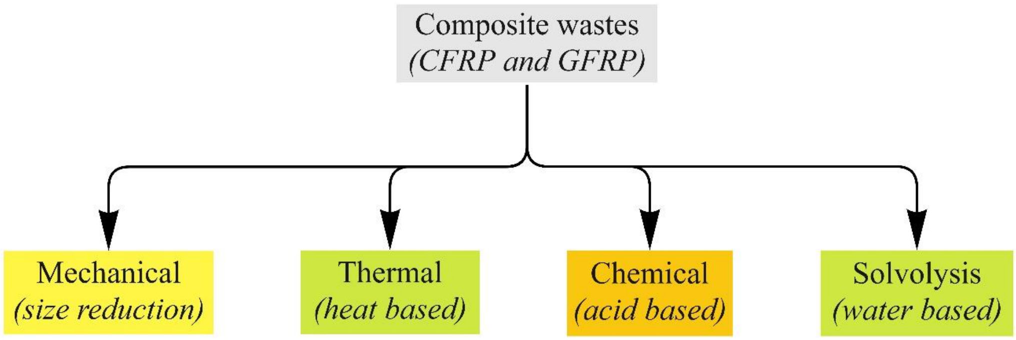


Figure 6. Available recycling processes

The chemical recycling process utilises acids at different atmospheric conditions and operating temperatures in the range of 60–360 °C. These acids enable depolymerising the resin system to separate the valuable CFs and GFs from their waste [30–34]. Both the process yield and the retained strength of the recycled fibres are >90%. However, this process was majorly studied on a laboratory scale due to the involvement of strong chemicals, upscaling into an industrial scale would reflect on the environmental aspects to the associated process outcomes. The solvolysis process follows a similar principle of resin depolymerisation using water and mild alcohols at their supercritical and subcritical conditions [35–39]. The resin removal rate is >95%, and the retained mechanical properties are >90%. The recycling process efficiency and the retained mechanical properties vary for different recycling processes. Therefore, not all processes require a follow to the secondary process, but still, it is essential to a certain extent. The recycling processes have a significant influence on the remanufacturing process. The outcome of these processes indicates a suitable manufacturing method based on the fibre length and its orientation. The process has the potential to reach an industrial scale.

The thermal recycling process utilises heat at various temperature ranges 450–700 °C to vaporise the resin system in favourable conditions inside the chamber. The process is well established and explored by various studies. The fluidised bed process (FBP) and pyrolysis are the two well-established processes capable of reclaiming the fibres with a process yield in the range of 65–75%. Their associated mechanical properties range 50–60% for FBP and 85–95% for pyrolysis. The processes are capable of recycling both CFRP and GFRP composite wastes but favourable to recycling CFs. As GFs suffers a negative effect when exposed to the thermal-based recycling process, studies have highlighted additional options for GFRP composites with secondary treatments to reclaim the mechanical properties. The FBP process has been studied in both laboratory-scale and support industrial-scale feasibility (see Figure 7). The FBP process was developed at the University of Nottingham [27,29,40–42] with intense research to recycle both CFRP and GFRP composite wastes. Additionally, FBP also has the potential to be upscaled into commercialised plants in the near future.

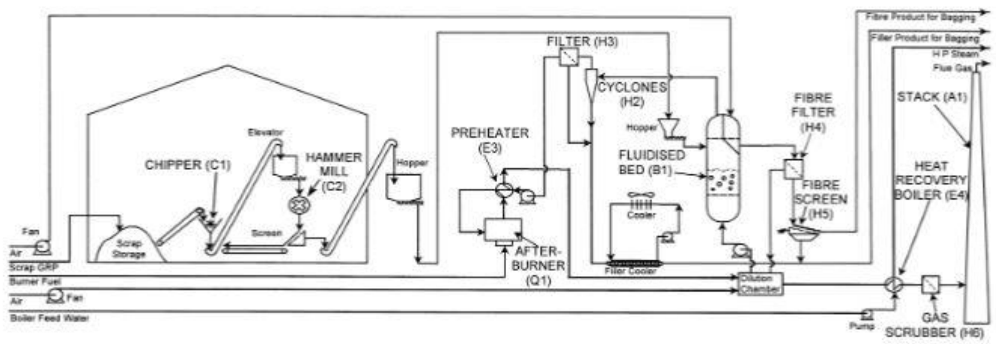


Figure 7. Commercial-scale Fluidised Bed Process [42]

Similar to FBP, the pyrolysis process has also been studied in both laboratory-scale and possess an industrial-scale plant. The ELG carbon fibre (UK), recently changed to gen2 carbon, claims to have a functional industrial-scale pyrolysis plant (see Figure 8) capable of recycling up to 5 tonnes of CFs per day, retaining 90% of their mechanical properties and sold for a 40% lower price range compared to virgin CFs in commercial markets. The rCFs from the industrial-scale processes have been widely used in various studies examining the mechanical performance of the fibres and their respective composites. Similarly, the CFK Valley Stade Recycling GmbH and Co. KG in Germany performs industrial-scale continuous recycling processes focusing on recycling CFRP composite wastes. It is to be noted that the processes majorly operate with CFRP wastes and are not favourable to recycle GFRP waste due to the loss in their mechanical properties after recycling. These upscaled recycling processes are evidence for the change in the industry's attitude towards a sustainable waste disposal approach with material recovery as their highest priority.

Figure 8. Industrial-scale pyrolysis process operational in ELG Carbon fibre, UK [43]

To close the life-cycle loop of these waste composites, remanufacturing the recycled fibres plays an important role. Figure 9 presents the different manufacturing processes based on their cycle time and part size. Amongst the processes, resin transfer moulding (RTM) related approaches have the upper hand in manufacturing FRP composites. Especially, CFRP and GFRP composites are favourable to adopt such RMT processes [44,45]. Still, the compression moulding process [30,46,47] has become popular in recent times to remanufacture recycled CFRP and GFRP composites due to its ability to maintain the composite's structural rigidity at the same time, suitable to operate various types of fibre arrangements like short, long, continuous and discontinuous. However, there is no definite remanufacturing process to utilise over the rCFs and rGFs. Thus, selecting the process based on the final product requirement is required.

After selecting the manufacturing processes, the primary task is to define the operating parameters to remanufacture the recycled fibres into new composites by employing a fresh resin system. For CFRP and GFRP based composites, studies have predominantly used epoxy-based resin with hardener at selective temperature, pressure and curing duration. Typically the parameters are in the range of 8–16 hrs curing time, post-cured at a temperature range of 60–80 °C and under atmospheric pressure conditions or 1–2 bars vacuum pressure conditions depending on the fibre arrangements. Additionally, the remanufacturing processes are also influenced by woven or non-woven based final products. Overall, it can be concluded that the final product defines the remanufacturing process and its associated stages.

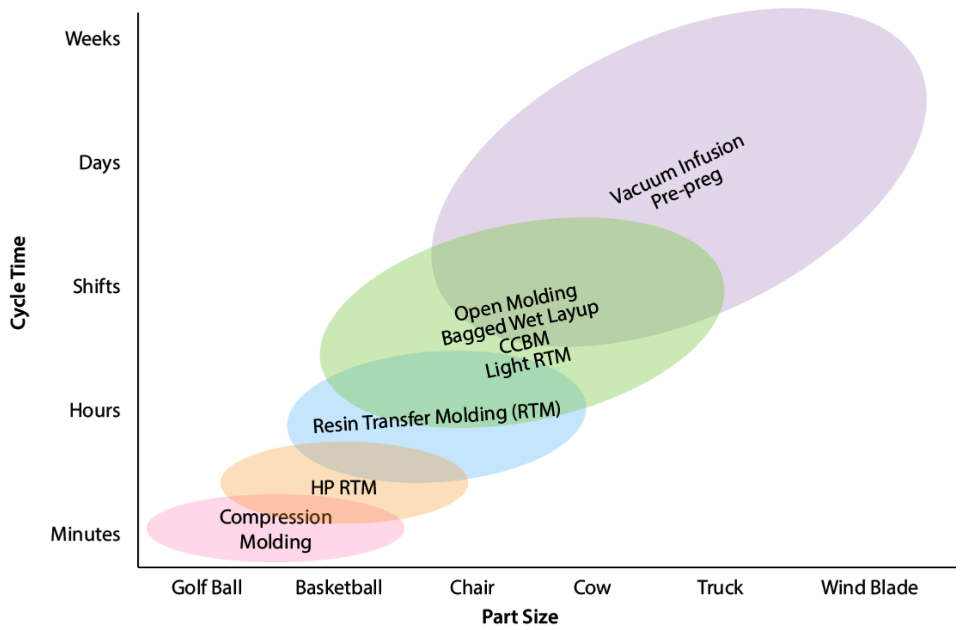


Figure 9. Remanufacturing the recycled composites [48].

2.3 Environmental impacts of the available recycling methods

The Life Cycle Analysis (LCA) studies using environmental indicators to identify the sustainability aspects of the currently available recycling processes to recycle the CFRP [11,49–53] and GFRP [54–58] composite wastes have explored various aspects of the process. The majority of the processes are capable of recycling and recovering the CFs and GFs from their respective wastes. However, the government regulations not just encourage any material recovery waste management path but focus towards sustainability should also not be compromised significantly to achieve the carbon-neutral goals. Thus, the use of LCA studies has resulted in showcasing the pros and cons of the processes to compare within the sustainable point of view. Additionally, the advantages of the environmental impacts of the material recovery process can be compared to the non-material recovery process.

As seen in the literature, most of the LCA studies [49,50,67,59–66] focus on recycling CFRP composite wastes. Such observations are expected as the price of CFRP composites is expensive. Their energy-intensive production process results in focusing on recycling these wastes properly to use as alternatives to replacing their virgin composites. The LCA outcomes of the recycling processes also highlight these negative carbon footprints with direct substitution with virgin composite production (see Figure 10). Additionally, these studies have highlighted the mandatory indicators to be used in newly developed processes, such as global warming potential (GWP), greenhouse gas emission, human toxicity and abiotic depletion potential (ADP). The outcomes of the LCA processes provide primary points to be taken under consideration during the selection criteria to adopt a recycling process to close the life-cycle loop of these CFRP and GFRP wastes. Furthermore, to optimise the process to upscale the process parameters into industrial-scale operation, the sustainable data using LCA studies has resulted in promoting the final products and altogether avoiding traditional non-renewable approaches like landfill and incineration.

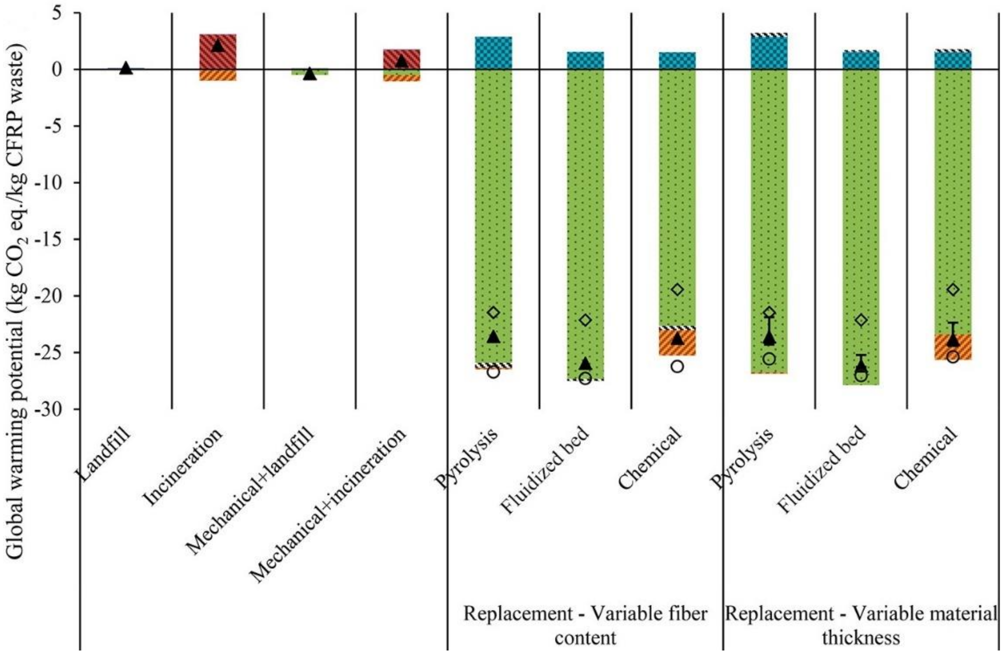


Figure 10. Environmental impacts of the available recycling process [11]

2.4 Composite wastes a valuable raw material

Based on the intense research focusing on sustainable waste disposal options related to CFRP and GFRP composite wastes present in high-performance applications such as aeroplanes [68,69] and wind turbine blades [70,71], composite industries have developed new renewable approaches towards implementing proper waste management solutions from a practical viewpoint. One such example in Finland [72] is the recent case study by Finnair partnered with Kuusakoski Oy to recycle an EoL (21-year old) Airbus A319 commercial passenger aircraft with only 0.8% of the aircraft ended up in non-sustainable disposal (see Figure 11). Furthermore, the obtained composite materials are subjected to research to minimum achieve an energy recovery approach. Compared to the past traditional landfilling and incineration waste management approaches by composite manufacturers and waste management industries, the recent attitudes towards wastes have predominantly changed for a mandatory energy recovery process. Thus, further elevated research into upscaling the material recovery processes will definitely be welcomed.

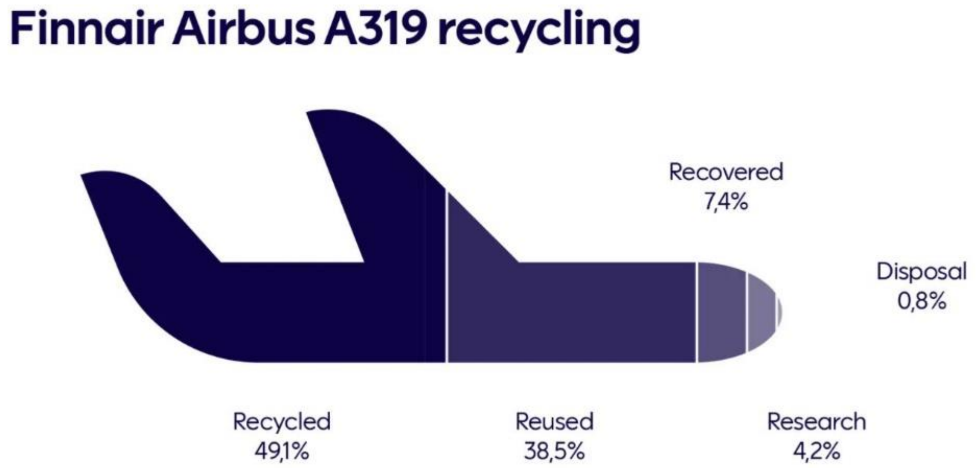


Figure 11. Finland’s first commercial aircraft recycling [72]

Recently, the practical utilisation of these composite wastes was also observed in public places (see Figure 12) by effectively utilising the large EoL wind turbine blades. The waste blades are converted into bike parking spots (Figure 12a) [73] in Denmark by the re-wind team [74] and converted into playgrounds (Figure 12b) [75] by Terneuzen in the Netherlands. Such innovative transformations utilising the cumulating tonnes of wand turbine blades possess a better alternative purpose than landfilling or just incinerating them to obtain energy after an energy-intensive size reduction process. These projects promote public awareness for waste management options at the same time, promotes renewable products with a possibility of developing a circular economy.

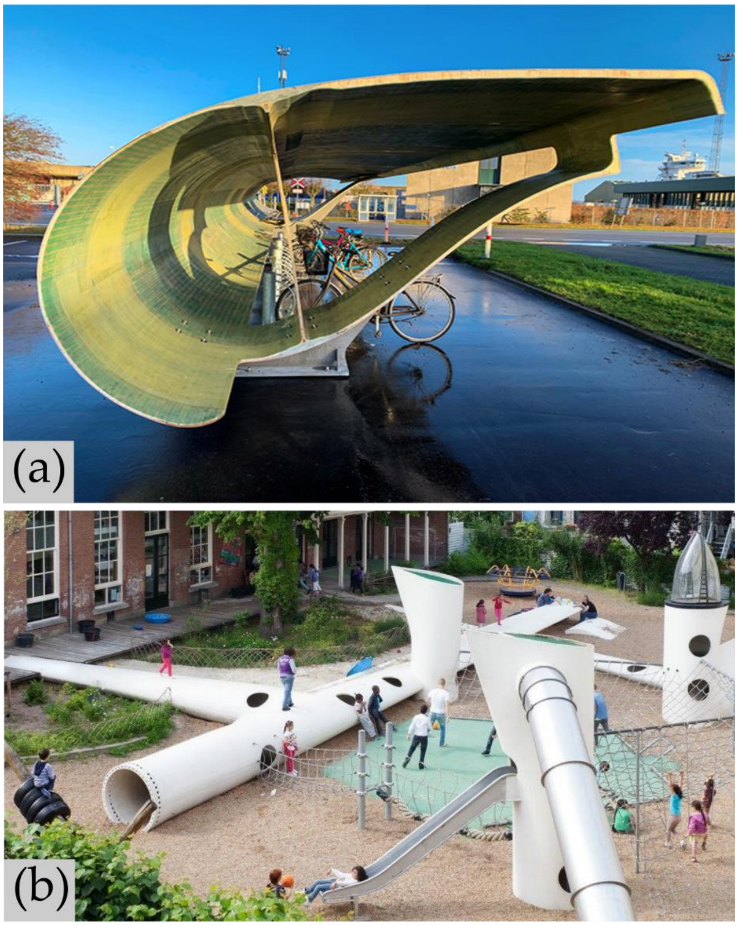


Figure 12. EoL wind turbine blades reused for domestic purposes [73,75]

Overall, composite wastes like CFRP and GFRP consist of expensive and mechanically enhanced carbon, and glass fibres are considered as raw materials then as wastes to be disposed of. Thus, properly recycling and recovering these valuable fibres have a higher potential to be reused into real-time applications. Plus, with improvised recycling approaches that are able to recover the mechanical properties of these recycled composites similar to their virgin composites, the possibility to sustainably replace the virgin composite with recycled ones are of higher chance.

3 Methodology

The methodology used in this dissertation was synthesised from four articles (I–IV). Table 1 presents the details about the adopted materials and methods.

Table 1. Materials and methods used in the articles

Article	*Materials*	*Methods*
I	Collection of previously published studies about recycling CFRP and GFRP composite wastes.	Literature review
II	Manufacturing based composite wastes such as carbon fibre-reinforced epoxy (CFE), glass fibre-reinforced polyester (GFP) and fresh laminating epoxy (EP)	Cone calorimeter equipment to recycle the composite wastes, compression moulding to remanufacture, mechanical tests: ISO 527-2 for tensile test using testXpert II software, ISO 179-1 for impact test and SEM analysis.
III	Recycled carbon fibre-epoxy (rCF/EP) and recycled glass fibre-epoxy (rGF/EP) composite's tensile test results (SS curve data) and impact test results from article II	Finite element methods (FEM) study performed using Abaqus/CAE. Elastoplastic for non-linear behaviour and ductile damage for fracture initiation and propagation.
IV	Life-cycle assessment (LCA) data from published studies, data from GaBi software (version 9.0.0.42, DP service pack 38), data from EuCIA (Eco impact calculator) and data from article II	LCA study performed using ISO 14040 and ISO 14044 with GaBi software (version 9.0.0.42, DP service pack 38)

The literature study for all the articles (I–IV) was performed primarily utilising keywords such as carbon fibre, glass fibre, recycling methods, composite waste and life cycle assessment with the help of search tools like Scopus and Web of Science. Plus, supportive tools, namely MS Excel, MS Visio, and Mendeley, were used to analyse the data further. Figure 13. illustrates the overall outlook of the processes involved in articles (II–IV) to close the life-cycle loop of the CFRP and GFRP composite wastes. As seen, it was predominantly focused on a sustainable waste management route by utilising the principles of recycling, remanufacturing and reusing the composite wastes.

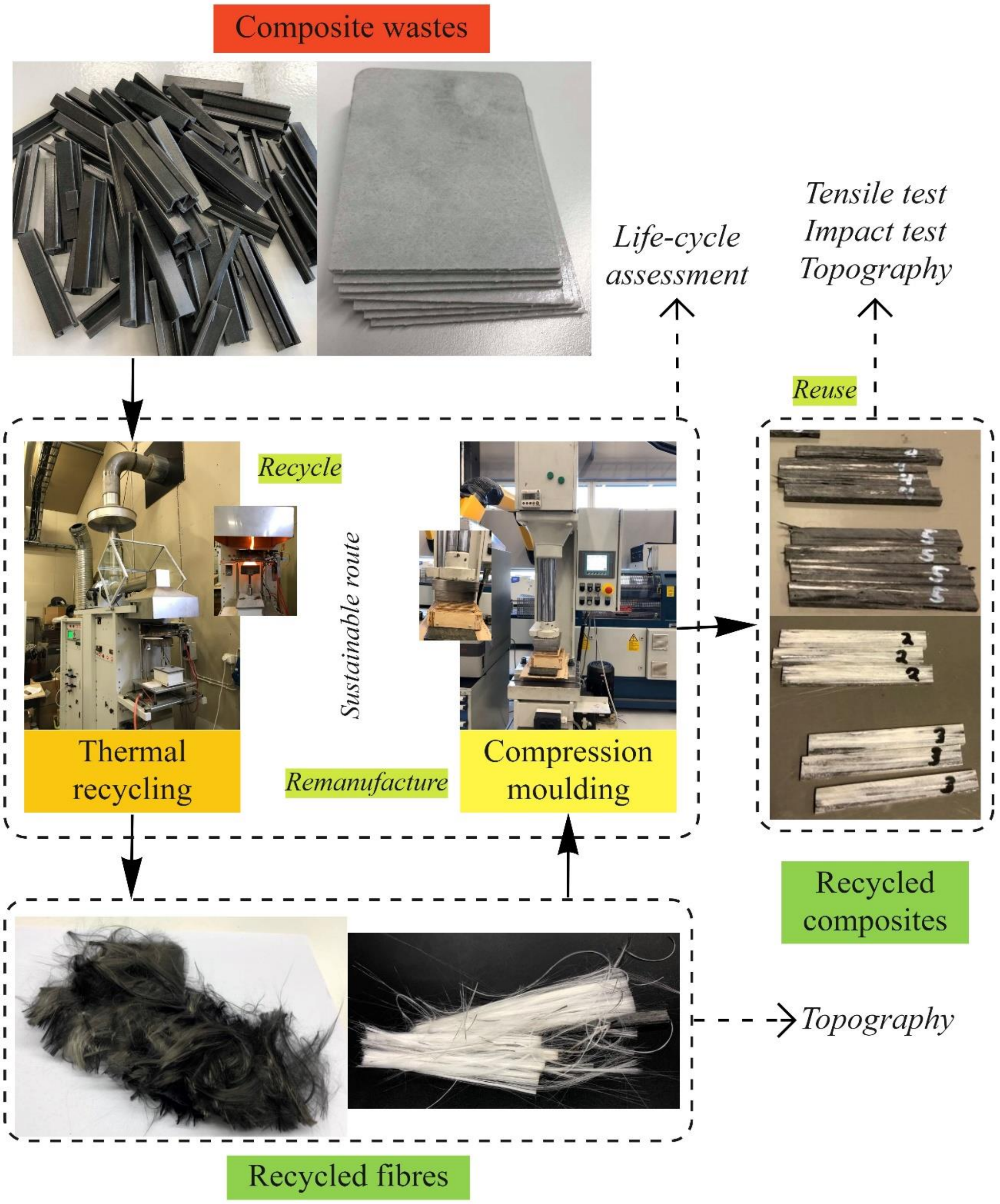


Figure 13. Life-cycle loop of the adopted sustainable route

Based on the collected literature, Article I was conducted. It focused on a brief literature review to present an overall outlook concerning the current status of recycling CFRP and GFRP composite wastes. The study analysed the recycling process types, the reclaimed fibre properties and the environmental impacts of the process. Overall, the study primarily aimed to identify existing research gaps in the various recycling processes involving CFRP and GFRP composite wastes. Additionally, a reliable recycling process was selected to practically develop on a laboratory scale to close the life-cycle loop of the composite wastes.

For laboratory experiments, a thermal-based process was adopted. The research materials were obtained from manufacturing wastes such as trimmings and off-cuts of carbon fibre-reinforced with epoxy (CFE) and glass fibre-reinforced with polyester (GFP) composites. These composite wastes were used as raw materials for Article II. Table 2. presents the raw material and composite design data. The study aims to develop a thermal recycling process to recover CFs and GFs from their composite wastes and remanufacture them into new composites to investigate their mechanical properties. A cone calorimeter equipment was employed to recycle the composite wastes, and the remanufacturing process was performed using a compression moulding technique with a customised plywood mould. The newly produced composites were subjected to uniaxial tensile loading using Zwick Roell (Z020) tester operated by testXpert II software and impact test using manually operated pendulum impact tester. The recycled fibre and the remanufactured composites were investigated microscopically in various magnifications using a Jeol JSM-5800 LV scanning electron microscope.

Table 2. Composite compositions used in this study

Raw materials					
Composite	**Fibre**	**Fibre volume (wt%)**	**Resin**	**Resin volume (wt%)**	**Composite density (g/cm^3)**
CFE	Carbon fibre	55	Epoxy	44.5	1.81
GFP	Glass fibre	44	Polyester	56	1.52
Composite design					
Composite	**Fibre**	**Fibre volume (wt%)**	**Resin**	**Resin volume (wt%)**	**Density range (g/cm^3)**
rCF/EP	Recycled carbon fibre	60	Epoxy	40	1.5–1.7
		40		60	
rGF/EP	Recycled glass fibre	60		40	1.7–1.9
		40		60	

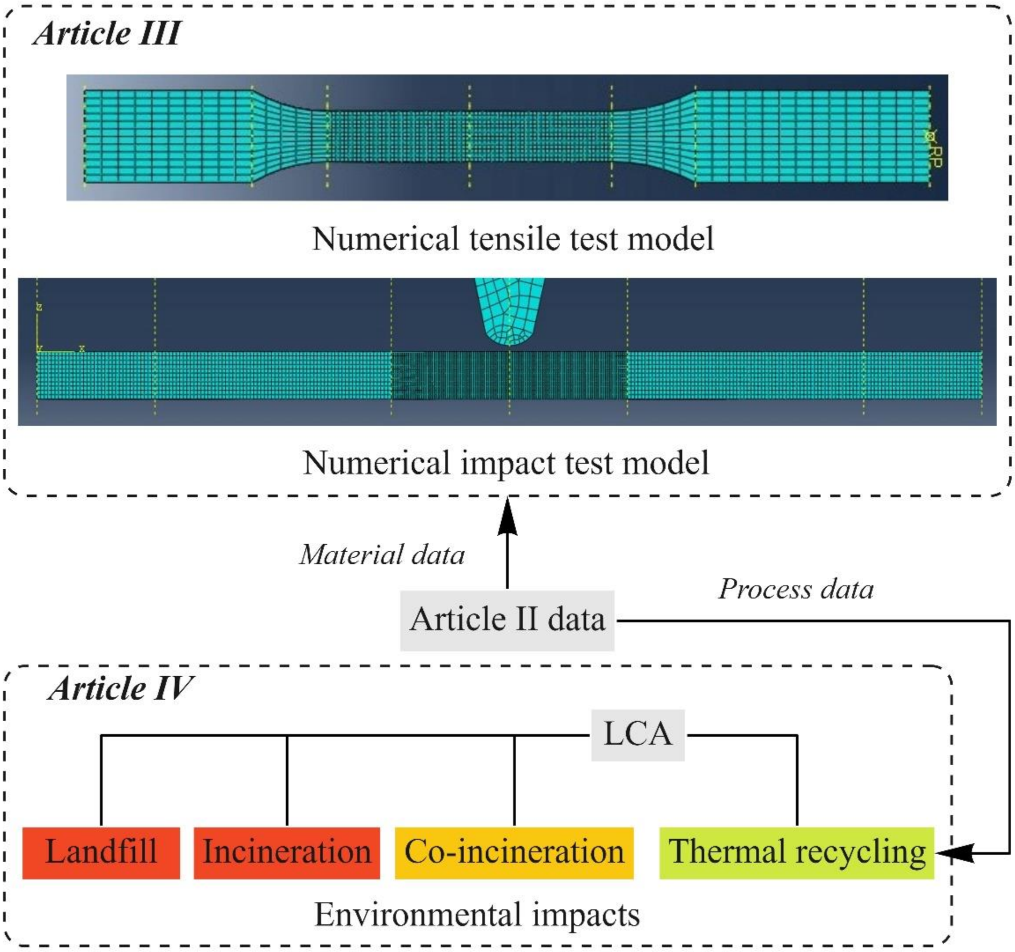


Figure 14. Article II data as primary data for Articles III and IV

These measured material data from Article II were primarily stress-strain values from the tensile test result. The data recorded the stress experienced by the recycled composites for the applied force at various strain rates. Each rCF/EP and rGF/EP was composed of two types, 40 and 60%, with a 20% increase in fibre volume. The obtained data were processed further using computing tools such as MATLAB and MS excel and fed as inputs for the material modelling to Abaqus FEM software (see Figure 14). The data were further processed using an elastoplastic isotropic model to simulate the non-linear behaviour of the recycled composites using equation (1).

$$F^{T} = F^{el} + F^{pl} \quad (1)$$

The model assumes that the plastic deformation (F^{pl}) follows after the elastic deformation (F^{el}). The total deformation (F^{T}) of the material is the sum of the deformations. Furthermore, ductile damage modelling was utilised to investigate the fracture initiation and development for the rCF/EP and rGF/EP composites. The model assumes that the damage onset ($\bar{\varepsilon}_D^{pl}$) is a function of stress triaxiality (η) and the equivalent plastic strain rate ($\bar{\varepsilon}_0^{pl}$), as seen in equation (2).

$$\bar{\varepsilon}_D^{pl}(\eta, \bar{\varepsilon}_0^{pl}) \qquad (2)$$

Furthermore, the measured data from Article II were used as raw materials for Article IV to perform a life cycle analysis (LCA). The thermal recycling and compression moulding processes data were used for inventory analysis. Additionally, literature data from Article I, Gabi and EuCIA software's database were also incorporated for the inventory analysis. The study used GaBi software (version 9.0.0.42, DP service pack 38) to investigate the environmental impacts of the thermal recycling process developed in Article II and further studied and compared the environmental impacts of various traditional waste disposal options like landfilling, incineration and co-incineration for CFRP and GFRP composite wastes (see Figure 14).

4 Results and discussion

4.1 Recycling process challenges and selection parameters

The primary findings from the literature review (Article I) concerning recycling the CFRP and GFRP composite wastes have resulted in identifying significant research gaps. The forthwith intense research focusing on the mechanical recycling process has shifted it into a pre-recycling process instead of considering it as a primary process. Despite the possibility for material recovery, it was not easy to adopt the process considering their final product as size reduced waste composite instead of proper fibre recovery. Additionally, alternative approaches [76,77] for the traditional mechanical recycling process using high voltage current were cost-effective, lower environmental impact, and time-efficient but will remain as pre-process for the available primary recycling processes like thermal, chemical and solvolysis.

The traditional chemical recycling processes to reclaim the fibres by depolymerising the resin system using strong acids have resulted in cleaner CFs and GFs retaining higher strength percentages with minimum damages on the fibre surface. The process typically consumes lower energy and results in maximum resin elimination. However, this approach has a heavy environmental impact when upscaling to an industrial scale to recycle tonnes of cumulating composite waste. The recent improvements using electrochemical as an alternative to traditional methods possess reasonable environmental impacts. Still, the processes have not fully evolved from a laboratory scale. Solvolysis following a similar depolymerising approach possesses better results compared to the chemical recycling process. The use of cheap and sustainable solvents like waster and mild solvents under their supercritical and subcritical conditions results in maximum resin elimination from the composite wastes with lower environmental impacts. The processes have a higher yield and fully retains the mechanical properties of the CFs and GFs. However, the solvolysis recycling process is still on a laboratory and semi-pilot scale as it requires further investigation to practically upscale into an operational pilot and industrial scale.

So far, the thermal recycling process like FBP and pyrolysis have dominated the CFRP and GFRP composite waste recycling. The FBP process is capable of producing clean fibres with a higher process yield. However, the fibres are short and randomly oriented, making it difficult to close their life cycle loop. It is well known that longer the recycled fibres better its ability to recycle multiple times. Plus, the rCFs encountered a 40% strength loss and rGFs a 60% strength loss. Despite these drawbacks, the process was made into the pilot scale, and a theoretical upscaling study is available to establish an industrial-scale process. Thus, when secondary treatments regain the strength of the recycled fibres. The FBP can potentially recycle the composite wastes as it also possesses reasonable environmental impacts.

Unlike FBP, pyrolysis was practised on an industrial scale. The process had a sustainable value by producing by-products such as oil and gas in addition to recycling the fibres. These by-products derived from the resin system have similar calorific values to gasoline and are capable of being used as feedstocks in other processes. The rCFs had maximum strength recovered. In contrast, the rGFs needed to be chemically treated in order to reclaim their original strength as heat affects the surface of the rGFs. The process was also capable of recycling long fibres. Overall, the process possessed lower environmental impacts compared to chemical-based processes. Thus, pyrolysis is a significant process to recycle the CFRP and GFRP composite wastes amongst the other available processes.

Based on the outcomes from the literature review (Article I), it can be highlighted that the pyrolysis process can efficiently recycle the CFRP and GFRP composite wastes. At the same time, an industrial scale exists, making it a favourable process to optimise further to extract maximum output. This process was seen as a practical solution to recycling the cumulating wastes. However, specific practical difficulties related to the pyrolysis-based process were unknown. Exploring it further requires the need to develop a thermal recycling process related to the pyrolysis principle and easy to upscale into higher capacity.

4.2 Recycling process and recycled composite’s mechanical properties

4.2.1 Developed thermal recycling process

Based on the results from Article I, a laboratory-scale thermal recycling process was developed and investigated further. The developed process utilises cone calorimeter (CC) equipment. The process operates by a conical heat radiator at 50 kW/m^2 of heat radiation equal to 750 °C coil temperature as a primary heat source to induce combustion over the waste composites. The results from the process are present in Figure 15. The measured surface temperature on the composite wastes during the process was 550 °C. The process was executed in a batch-based reactor depending on the capacity of the material holder within the CC equipment. For a better understanding, the process results were upscaled to 1 kg of CFRP and GFRP composite wastes. As seen, rapid resin removal occurs within the combustion zone contributing to the maximum percentage of the process. The recycling process duration for CFRP and GFRP composite wastes were 20–25 min and 25–30 min depending on the thickness and surface area of the wastes. As observed, the wastes with higher surface area and lower thickness possessed faster recycling. The developed process evaporated the resin system and resulted in individual rCFs and rGFs. Overall, using the developed process, the fibre recovery rate for rCFs was 95–98 wt% and for rGFs was 80–82 wt%.

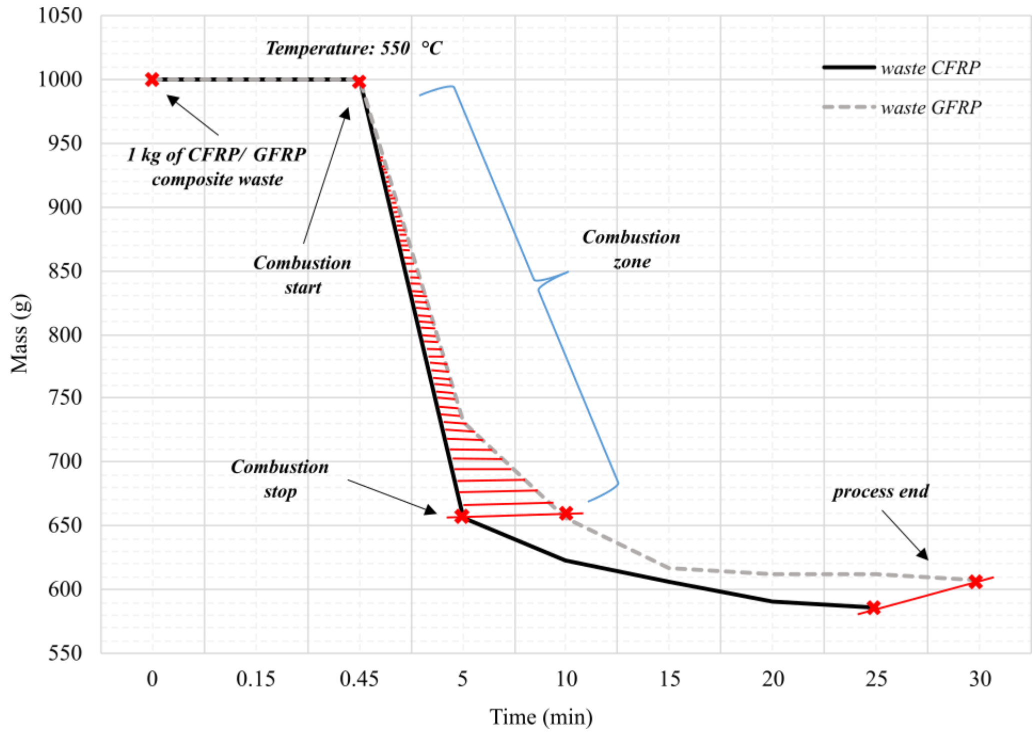


Figure 15. Thermal recycling process parameters

4.2.2 Characteristics of the recycled fibres

The recycled fibres, both rCFs and rGFs, possessed a unidirectional (0°), long (105 ± 2 mm) and continuous fibre arrangement, i.e. uniform length from start to end. Not all recycling processes are capable of producing consistent outcomes maintaining the fibre's structural integrity. As seen in research focused on thermal recycling [42,78–81], the fibres are short and randomly oriented, making it challenging to remanufacture to the desired format. Thus the developed thermal recycling process can produce longer recycled fibres. Furthermore, the feasibility of the thermal recycling process can be observed from the SEM results of both rCFs and rGFs.

Figure 16 presents the significant SEM images from rCFs. As seen, these recycled fibres have a clean and resin-free fibre surface with negligible resin residues scattered across the fibre length. Additionally, during the recycling process, increasing the reaction period damaged the fibres (overcooked) and decreasing the time left higher resin residues (undercooked). Figure 17 presents the significant SEM images from rGFs. As seen, the volume of resin residues was higher compared to rCFs. These residues were clearly noticeable across the rGFs. During the process, increasing the reaction period burnt the fibres into ashes and decreasing the time left higher resin residues with a char formation.

The observed residues in rGFs were flaky and were not completely glued to the fibre surface.

The SEM images of the recycled fibres function as proper evidence to finalise the parameters of the thermal recycling process. The process was fixed to be isothermal with constant heat radiation of 750 °C. The process duration played a significant role in resin removal. The resin can be efficiently removed from the waste composites by varying the time for the sample to be exposed to the heat radiation. The measured diameter of the rCFs was averagely 6.5 ± 0.2 µm, and for rGFs, it was 20.5 ± 0.25 µm. As noticed, the rCFs were extremely fluffy and hard to handle. On the other hand, the rGFs were fragile and broke even with gentle uneven pressure during handling. These recycled fibres were remanufactured using the compression moulding process by employing laminating epoxy resin.

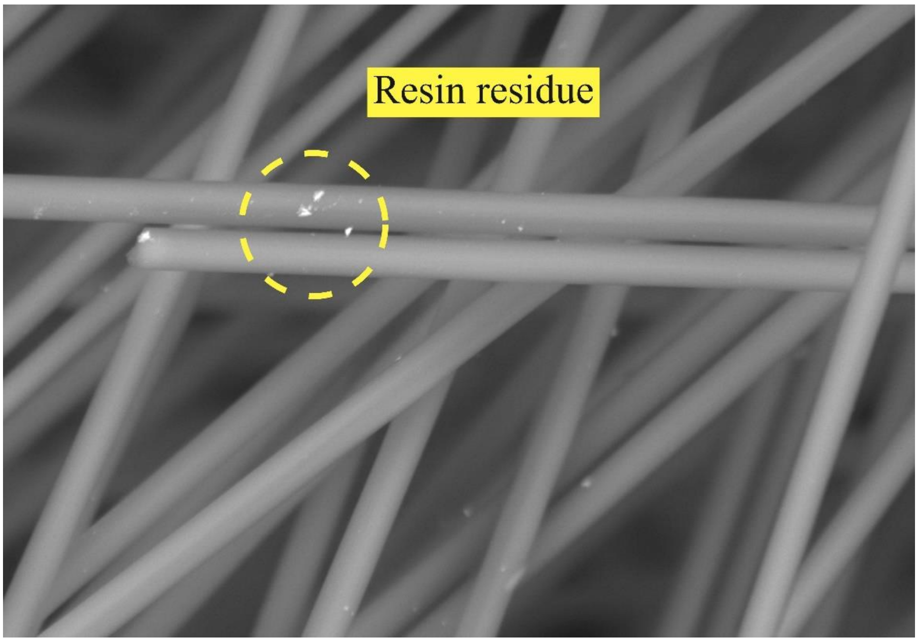


Figure 16. Recycled carbon fibres under scanning electron microscope

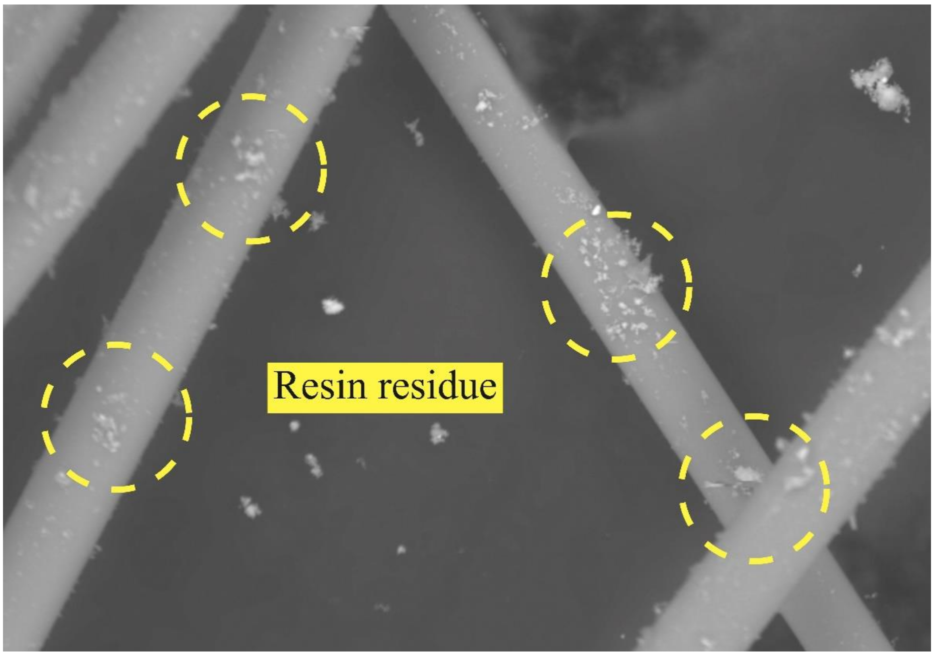


Figure 17. Recycled glass fibres under scanning electron microscope

4.2.3 Tensile and impact properties of the recycled composites

Figure 18 and Figure 19 presents the results from the uniaxial tensile testing of the rCF/EP and rGF/EP composites. The applied force was parallel to the fibre direction (0°). As seen in Figure 18, the rCFRP composites possess higher tensile strength compared to the rGFRP composites. However, it can be observed that in rGFRP composites, the increase in 20% fibre volume has resulted in extending their overall tensile strength by 75.14%. On the other hand, such a phenomenon was not noticeable in rCFRP composites. Still, the 20% increase in fibre volume has extended 12% of their overall tensile strength.

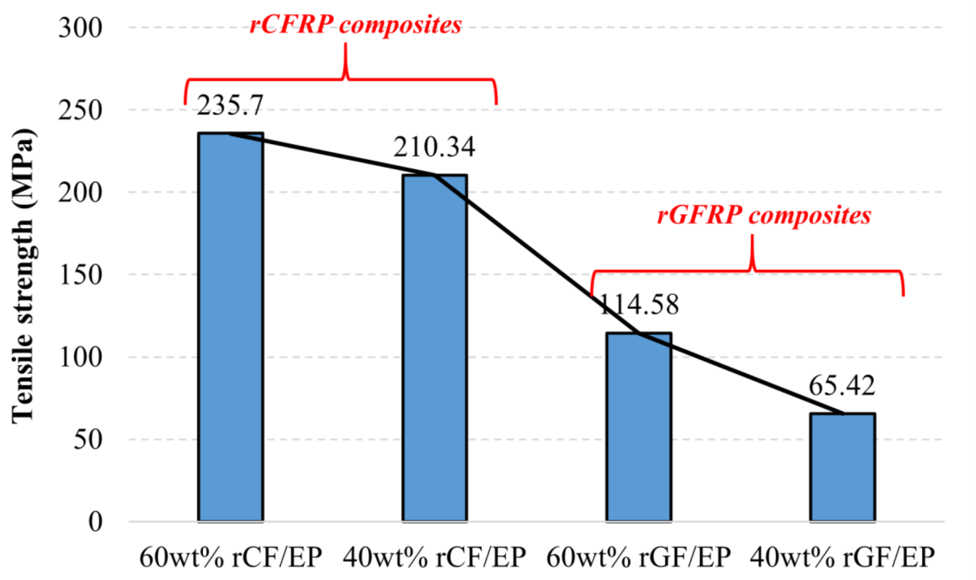


Figure 18. Tensile strength results from the experimental testing

Similar observations can be noticed in the measured Young's modulus. As seen in Figure 19, the rCFRP composites possess higher values compared to the rGFRP composites. However, it can be observed that in rGFRP composites, the increase in 20% fibre volume has resulted only in a 12.23% increase in Young's modulus and 34.27% in rCFRP composites. This is in contrast to their tensile strength behaviour. But can be explained based on the UD fibre arrangement characteristics within the recycled composites. As the applied force was in parallel to the fibre direction, adding fibres resulted in increasing the tensile strength of the composites without much effect on their elastic behaviour. The mechanical properties of their virgin composites were unknown. When compared the results to the similarly available studies like [82,83] for rCFRP and [84–86] for rGFRP composites, the values lay within the acceptable range.

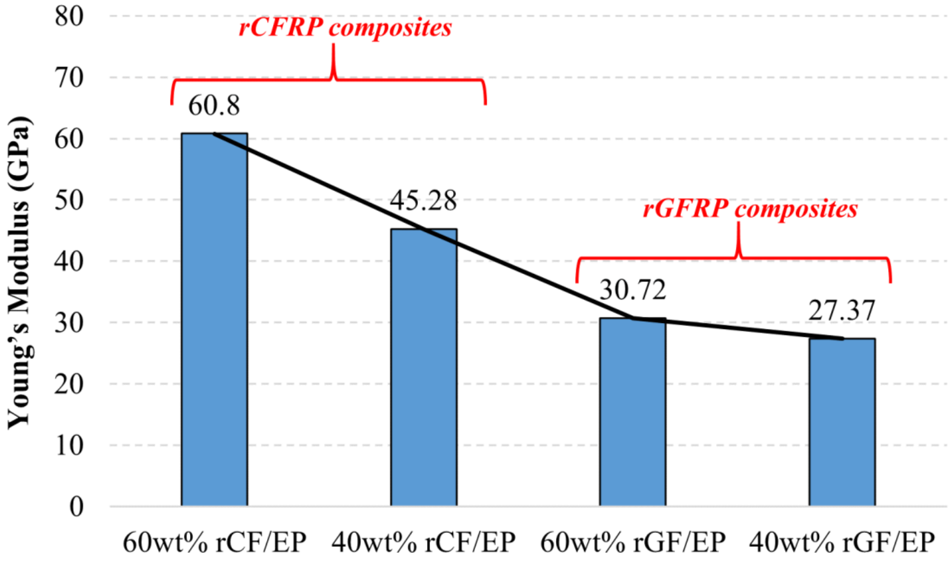


Figure 19. Young's modulus results from the experimental testing

Figure 20 presents the Weibull distribution of the individual sample results from the tensile testing of the rCF/EP and rGF/EP composites. The failure rate of the samples can be analysed using the obtained Weibull modulus. As seen, the distributions with the samples have low variation, and it represents that the tested samples have uniform values despite their defects. The external defects like poor wettability for rCF/EP composite samples and char formation for rGF/EP samples were observed during experimental testing. These defects are from the recycling and remanufacturing processes. However, based on the Weibull distribution, the values highlight that the sample's failure rate and the observed defects are evenly distributed without vast deviation within each sample population.

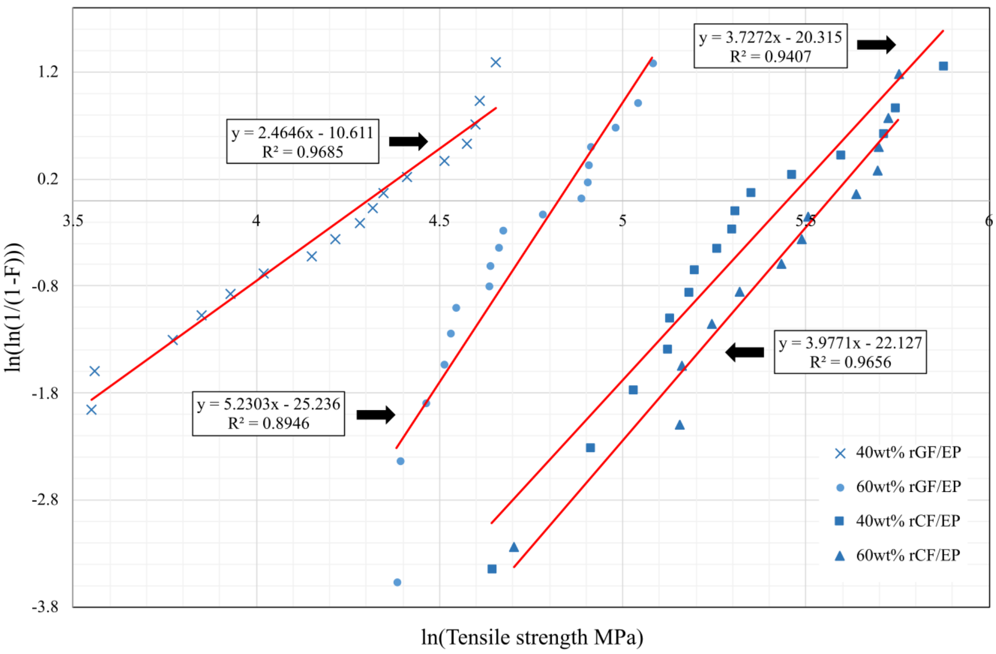


Figure 20. Weibull distribution of the recycled composites under tensile test

Figure 21 presents the results from the impact testing of the rCF/EP and rGF/EP composites. As seen, the rCFRP composites possess higher tensile strength compared to the rGFRP composites. However, it can be observed that in rGFRP composites, the increase in 20% fibre volume has resulted in extending their overall impact resistance by 116.16%. This phenomenon was not noticeable in rCFRP composites, but the 20% increase in fibre volume has extended 7.26% of their overall impact resistance. The defects observed from the recycling processes like poor wettability in rCFRP composites and char formation in rGFRP composites have a significant role during the impact testing of the samples. As the impactor (hammer) strikes the samples, the zones with defects experienced fracture compared to those with fully cured resin samples.

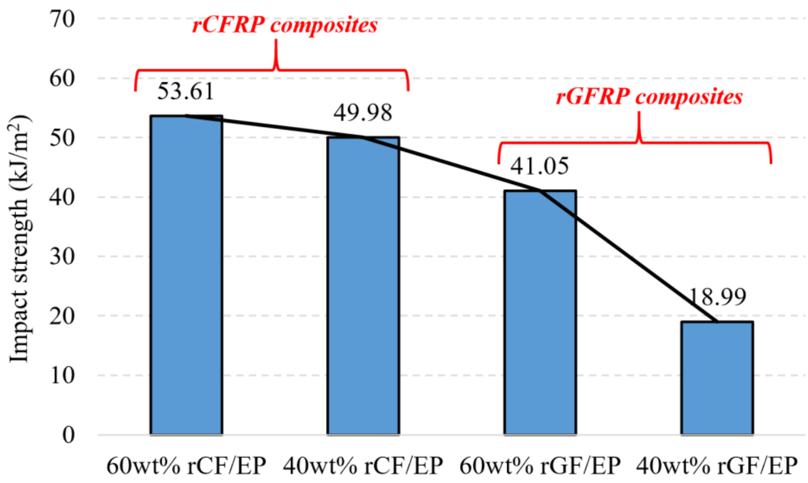
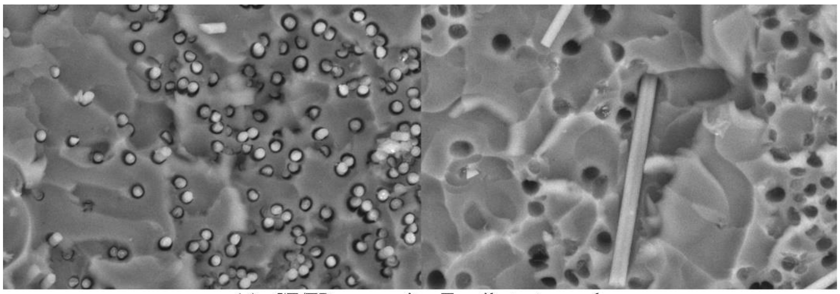


Figure 21. Impact test results from experimental testing

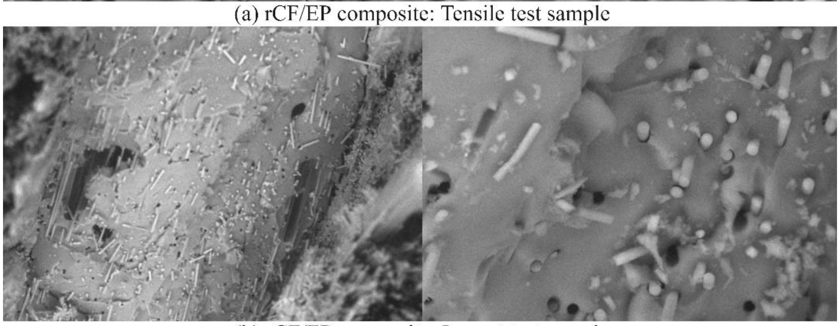
4.2.4 Characteristics of the recycled composites

The reliability of the recycled composites can be observed from the SEM images of the fractured samples. Figure 22 presents the SEM images of the rCF/EP composites after their tensile and impact test results. The cross-section of the samples was examined to investigate the fracture patterns possessed by the recycled composites. As seen, most of the samples failed under tension showcased fibre pull-outs (see Figure 22a). However, in the case of impact tested samples, as the samples experienced tension on one face and compression opposite to it, the matrix damage with evidence of shear failure was observed along with the fibre pull-outs (see Figure 22b).

Figure 23 presents the SEM images of the rGF/EP composites after their tensile and impact test results. As seen, the resin was evenly distributed between individual rGFs compared to the rCF/EP composites. A similar fibre pull-out can be noticed in the tested tensile samples (Figure 23a). Plus, multiple fibre fractures were visible, indicating the brittle nature of the rGFs. The impact tested samples possessed shear bands on the resin surface of the fractured area. Plus, such shear bands were also visible on the surface of the fractured rGFs (Figure 23b).

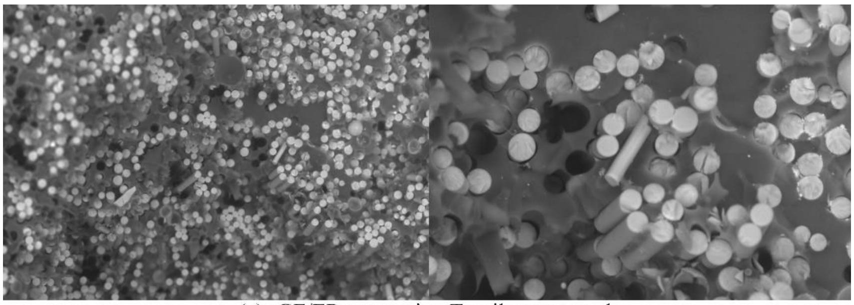


(a) rCF/EP composite: Tensile test sample

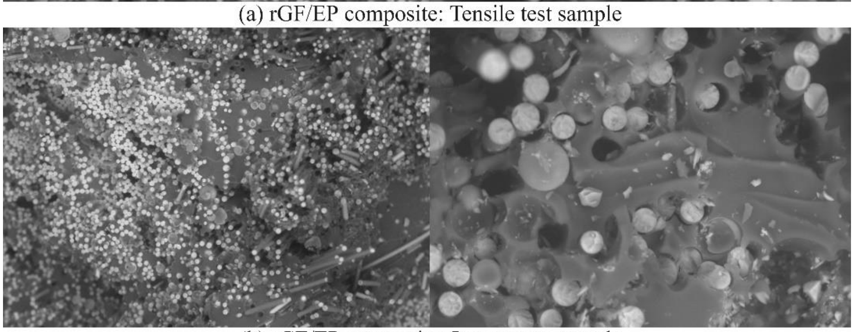


(b) rCF/EP composite: Impact test sample

Figure 22. Cross-section of the tested recycled CFRP composite



(a) rGF/EP composite: Tensile test sample



(b) rGF/EP composite: Impact test sample

Figure 23. Cross-section of the tested recycled GFRP composite

4.3 Recycling defects and their associated failures

Figure 24 presents the stress-strain curves of the numerical tensile test results from Article III compared with the experimental tensile test results from Article II. These stress-strain curves consist of the rCF/EP and rGF/EP composite types that possess a uniform non-linear curve until the damage initiation point. However, the material behaviour from the damage initiation until the final sample fracture (damage development area) has non-uniform resembles. Overall, the composites tested under tensile and impact conditions experienced failure by the general failure behaviour along with the influence of the defects present in the composite samples due to their respective recycling and remanufacturing process.

In any UD FRP composites, the primary failures involved under uniaxial tensile loading conditions are interlaminar damage, debonding in the fibre-matrix interface, composite delamination and crack development within the matrix phase near the fibre pull-outs [87–89]. Under unnotched low-velocity charpy impact test conditions, the primary failure involved are plastic deformation and cracking in the matrix, fibre breakage, and composite delamination [90,91]. All such damage behaviours were observed in the tested samples. In addition to the primary failure, the composites possessed secondary factors which can be related to the sample fracture. The observed defects present in the rCFRP composites are the poor wettability of the rCFs with the resin system. During the sample testing, the failure in the samples was noticed from the section occupied by unwet fibres. Such weak zones with defects have induced the fracture within the samples. Similarly, rGFRP composites were also possessed with a defect like the presence of char. It made the tested sample brittle at zones containing such char formation. However, the rGFs were thoroughly wet and incorporated into the resin system.

Figure 25 presents the observed impact energy values of the numerical tensile test results from Article III compared with the experimental tensile test results from Article II. The numerical results were analysed using von misses stress distribution. It was observed that the composite's fibre volume along with their strength increased, resulting in a stiffer composite, with the 60% rCF/EP sample being the stiffest. These numerically observed energies were higher than their experimental results but lower than similar virgin composites from the literature. Such results were expected, as the developed models did not incorporate any external defects from the recycling and remanufacturing processes, such as a poor resin wettability in the rCF/EP samples leading to a bundle formation in high fibre volume fraction in the 60% rCF/EP samples and a char formation in the rGF/EP samples. Alternatively, the developed models predicted the recycled composites as defect-free during damage behaviours and energy absorption.

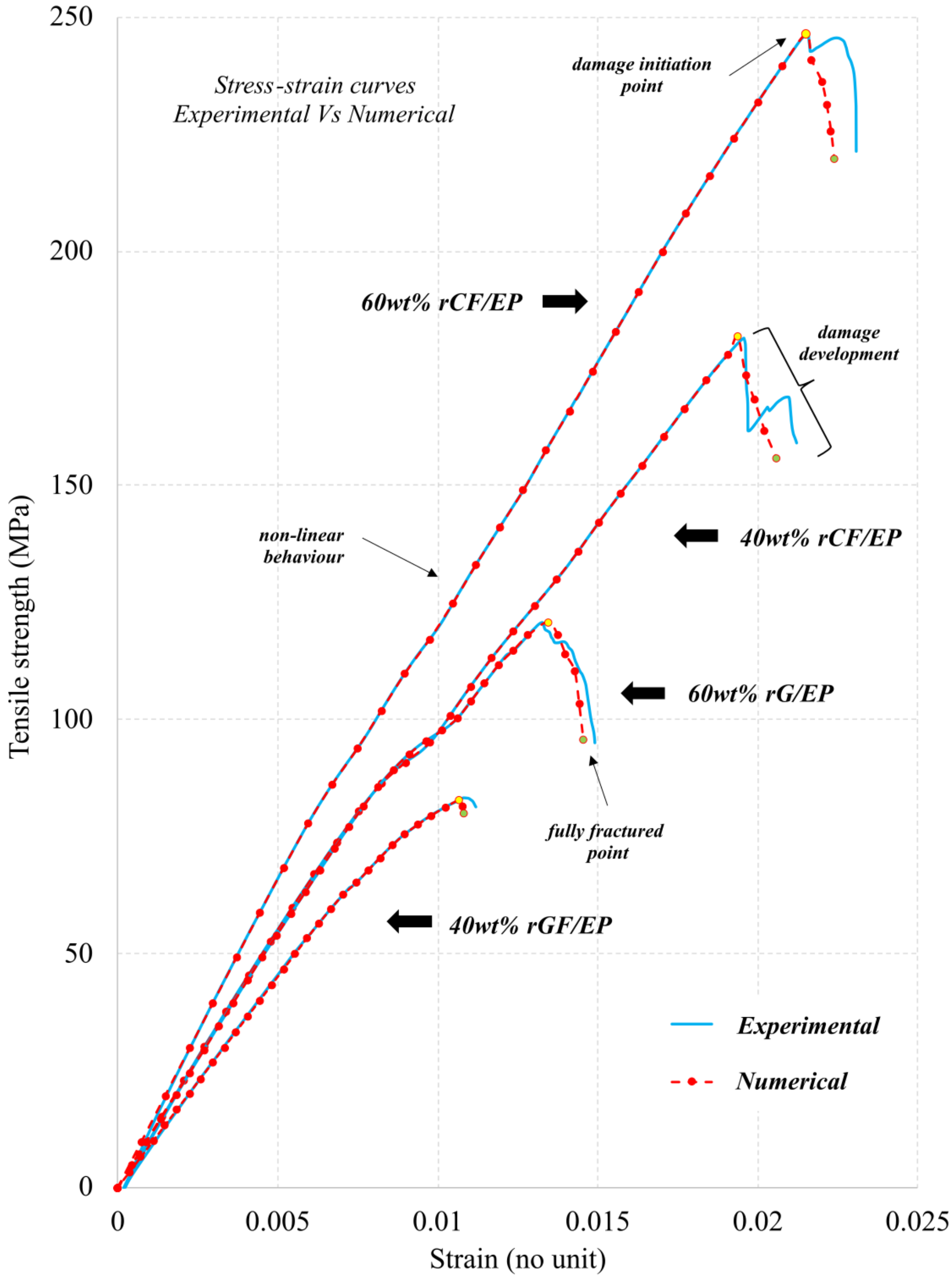


Figure 24. Tensile strength of the recycled composites

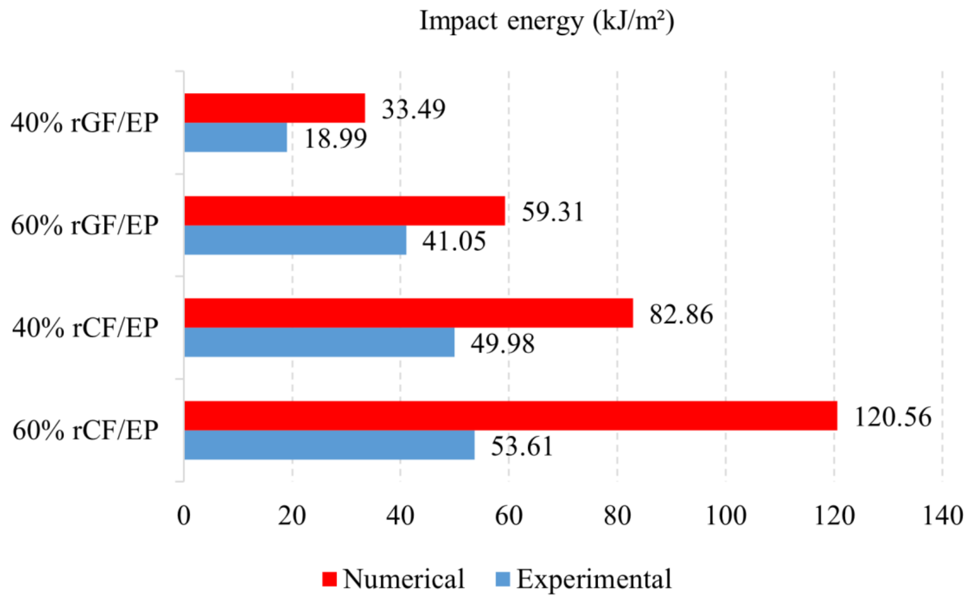


Figure 25. Impact strength of the recycled composites

Figure 26 presents the results of the energy observed by numerically tested rCFRP and rGFRP composite samples from Article III. The measured values represent energy observed from the point of impact to the point of fracture (highest value). These measured values are highly sensitive to the impact velocity of the hammer. The hammer velocities ranged from 4–8 m/s depending on the composite type with rCF/EP in higher-order and rGF/EP sample fracture occurred at lower velocities. During simulation, three parameters such as the mesh size of the composite samples, the applied hammer velocity and the time to execute simulation were considered significantly interconnected with the final results.

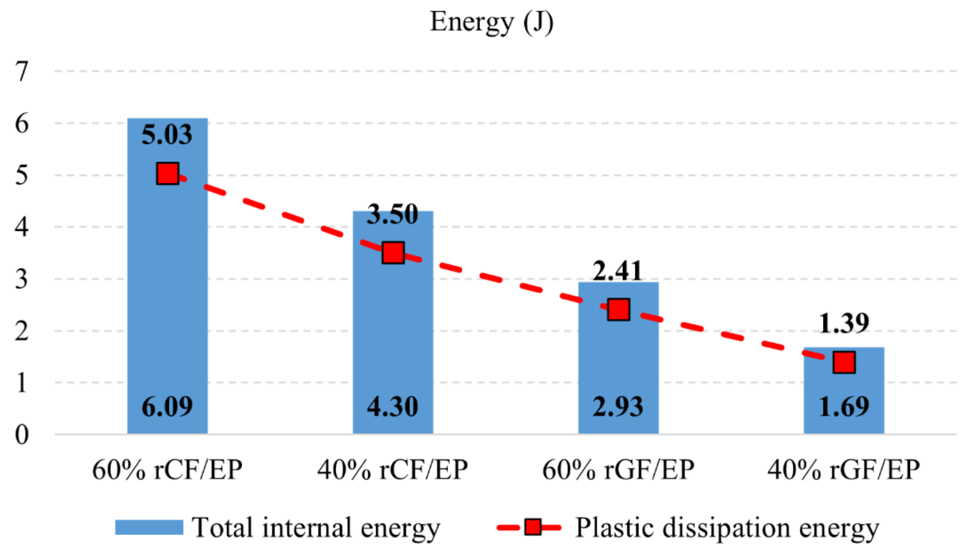


Figure 26. Energy observed by the sample during the numerical impact test

4.4 Environmental impacts of the waste management options

Figure 27 and Figure 28 illustrate the summarised outcomes from Article IV. The results represent the conducted LCA study using the data obtained from Articles (I–II). It estimates the environmental impacts using LCA indicators, namely global warming potential (GWP) in kg CO_2-eq and abiotic depletion potential (ADP) in MJ for 1 kg of CFRP and GFRP composite wastes. The developed thermal recycling process from Article II was investigated along with the traditionally available waste management options, namely landfill, incineration and co-incineration in cement kiln. The results include the environmental impacts, including the possible substitutions within each waste management option.

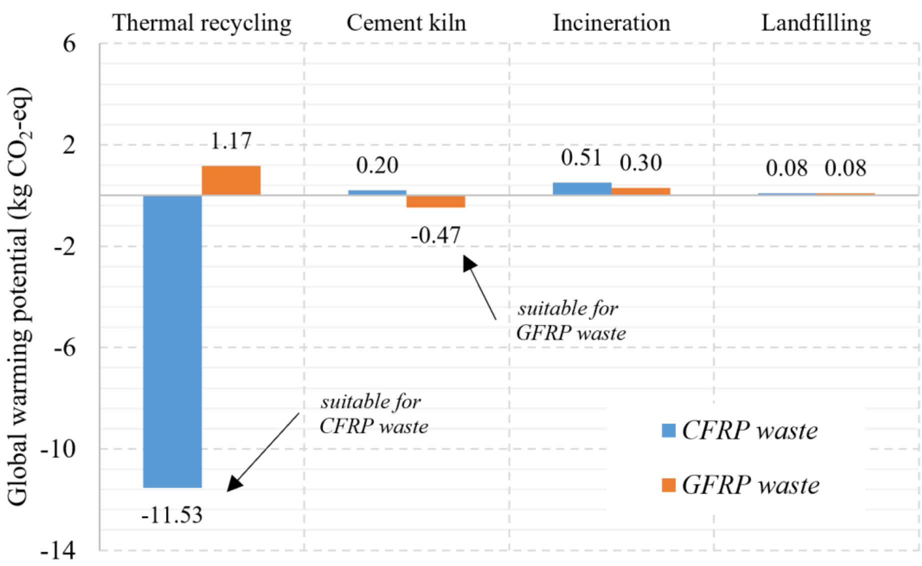


Figure 27. GWP results of various CFRP and GFRP waste disposal routes

As seen in Figure 27, the climate change results for CFRP wastes were favourable to the adopted thermal recycling process compared to the traditional waste management options. This was primarily due to the high energy-intensive carbonising processes (up to 3000 °C) involved in producing vCFs with emissions of 14.11 kg CO_2-eq, which will be substituted using the rCFs. As the recycling process only involves 3.06 kg CO_2-eq, closing the life-cycle loop of these wastes will directly result in a circular economy, helping to replace the production of vCFs with rCFs. On the other hand, the carbon emissions for GFRP wastes from the adopted thermal recycling process (1.17 kg CO_2-eq) were not as efficient as the co-incineration process (-0.47 kg CO_2-eq), in which the wastes were size reduced and used as feedstocks for cement production. As the vGF product is not energy-intensive, the presence of bauxite engaged to a favourable substitution during cement production. Still, the difference in the emissions can be reduced by further optimising the energy consumption during the thermal recycling process. However, the rGFs seems to possess lower mechanical properties, as seen in Articles (II–III), making it challenging to consider as a replacement of their counterpart.

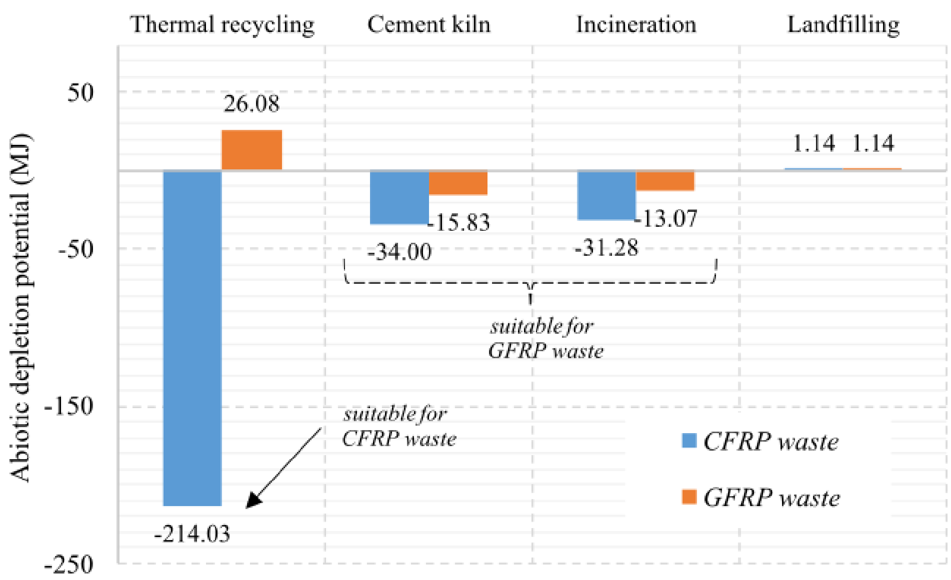


Figure 28. Suitable waste management option

As seen in Figure 28, the fossil fuel consumptions were lowest in the adopted thermal recycling process for the CFRP composite wastes at -214.03 MJ. The advantages of substituting the vCFs with rCFs have been strongly reflected in the results compared to the other waste management options without material recovery. Compared to the energy recovery processes like incineration and co-incineration to the material recovery process, the thermal recycling process possesses 6.5 times lower fossil fuel consumption in the CFRP waste management route. In contrast to CFRP, the GFRP wastes showed lower fossil fuel consumption for non-material recovery processes like incineration (-31.28 MJ) and co-incineration (-34 MJ) compared to the adopted thermal recycling process. The energy consumption to thermally recycle the GFRP wastes was 56.57 MJ before substitution. It can be concluded that thermal recycling processes are not beneficial for material recovery in GFRP waste management routes.

4.5 Waste management approaches

As closing the life-cycle loop of CFRP and GFRP composites by recycling, remanufacturing and reusing gains popularity, new waste management approaches within the sustainable routes, namely closed-loop recycling and open-loop recycling, were proposed by various studies [28,50,51,84,92]. The primary difference between the approaches is the end application. As its name implies, the material reused within a similar application represents a closed-loop. When it is reused in various other applications different from their waste of origin represents an open-loop. Figure 29.

illustrates a closed-loop recycling approach for CFRP and GFRP composite waste. The wastes associated with aeroplanes and wind turbines are recycled, remanufactured, and reused into a similar application. This approach is highly feasible only when the recycled composites possess mechanical properties similar to their virgin counterparts at a higher order of >95%. Additionally, the fibre's structural integrity, such as length and orientation, should be maintained throughout the recycling process to close the life-cycle loop multiple times successfully.

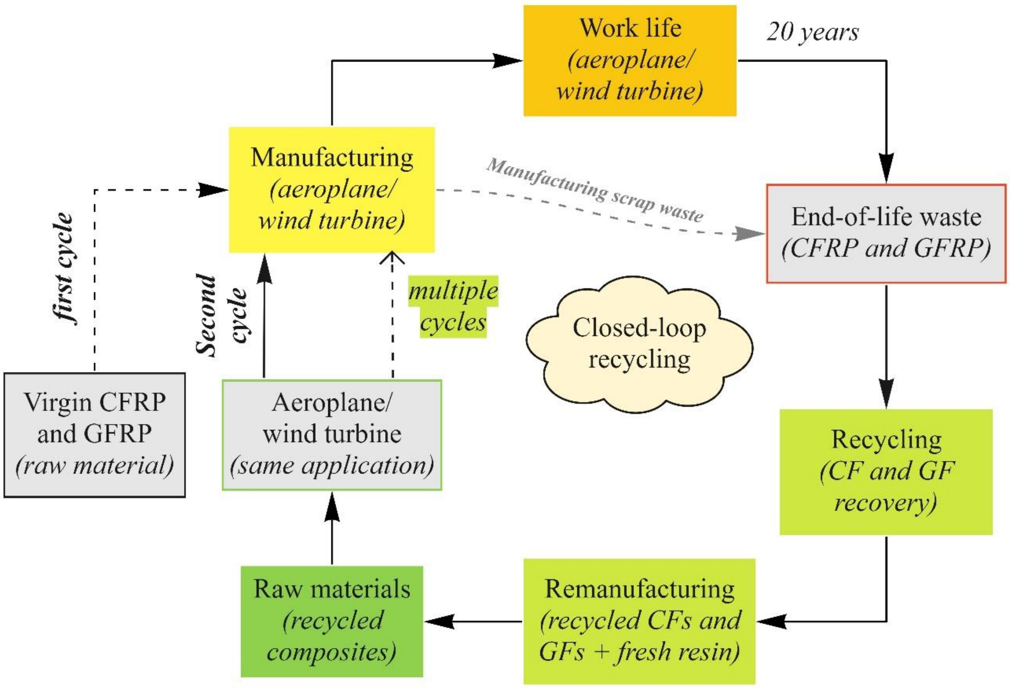


Figure 29. Closed-loop recycling approach

Figure 30. illustrates an open-loop recycling approach for CFRP and GFRP composites. The wastes associated with aeroplanes and wind turbines are recycled, remanufactured, and reused into various applications such as automobiles, construction, etc. This approach has been popular in recent decades as the mechanical properties of the recycled composites can be in an acceptable range of >65% compared to the virgin composite's mechanical properties. Also, the structural integrity of the recycled fibres can be compromised as there are applications that require short fibre and can act as a direct replacement for chopped virgin fibres. However, it can possibly reduce the life of the CFs and GFs as they are size reduced drastically, reducing their potential to reuse multiple times compared to the closed-loop approach.

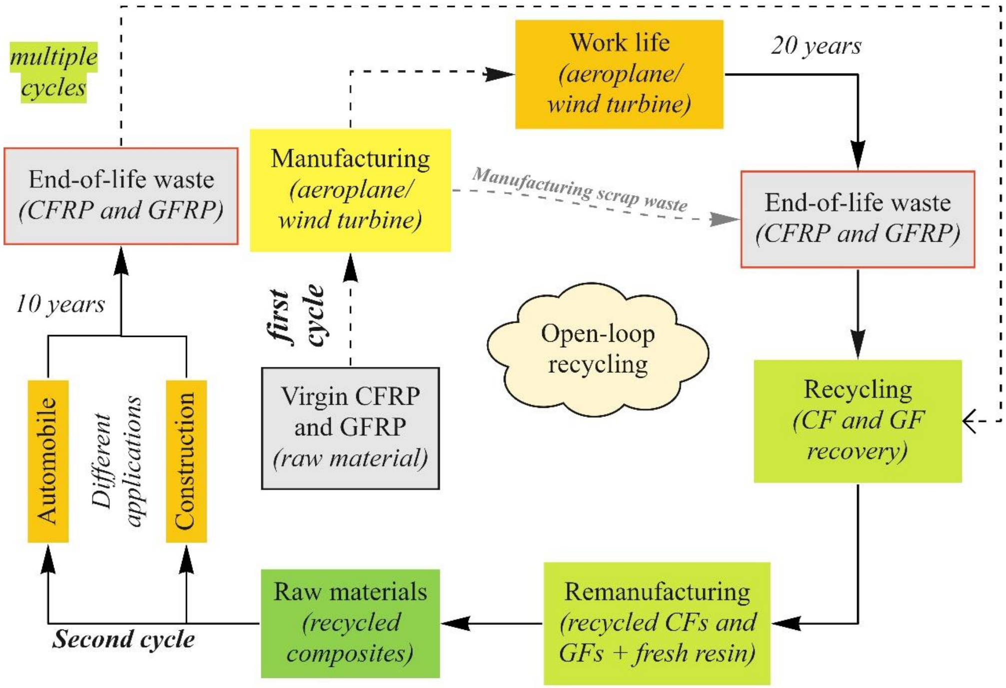


Figure 30. Open-loop recycling approach

4.6 Selection criteria to achieve a closed-loop recycling approach

The material recovery methods significantly impact the sustainable waste management options to close the life-loop of CFRP and GFRP composite wastes. Various studies have explored the different recycling methods—namely thermal recycling, chemical recycling and solvolysis using water and mild solvents at supercritical and subcritical conditions—to separate the valuable fibres from their resin system. Figure 31. illustrates the overall material recovery approach, primarily focusing on the recycling processes. Additionally, post recycling processes like secondary treatments and remanufacturing were highlighted as they play a significant role in the overall outlook involved in CFRP and GFRP composite waste. It is to be noted that the reclaimed fibres undergo secondary heat and chemical treatments after the primary recycling process to remove the residual resins deposited on the fibre surface and enhance the mechanical properties of the recycled fibres. At the post recycling phase, the recycled fibres possessing long fibres are arranged into woven formatting with additional processes. However, short and randomly oriented fibres are likely to be made into a non-woven arrangement. Therefore, selecting the proper recycling method is essential to utilise the maximum potential of these CFRP and GFRP composite wastes.

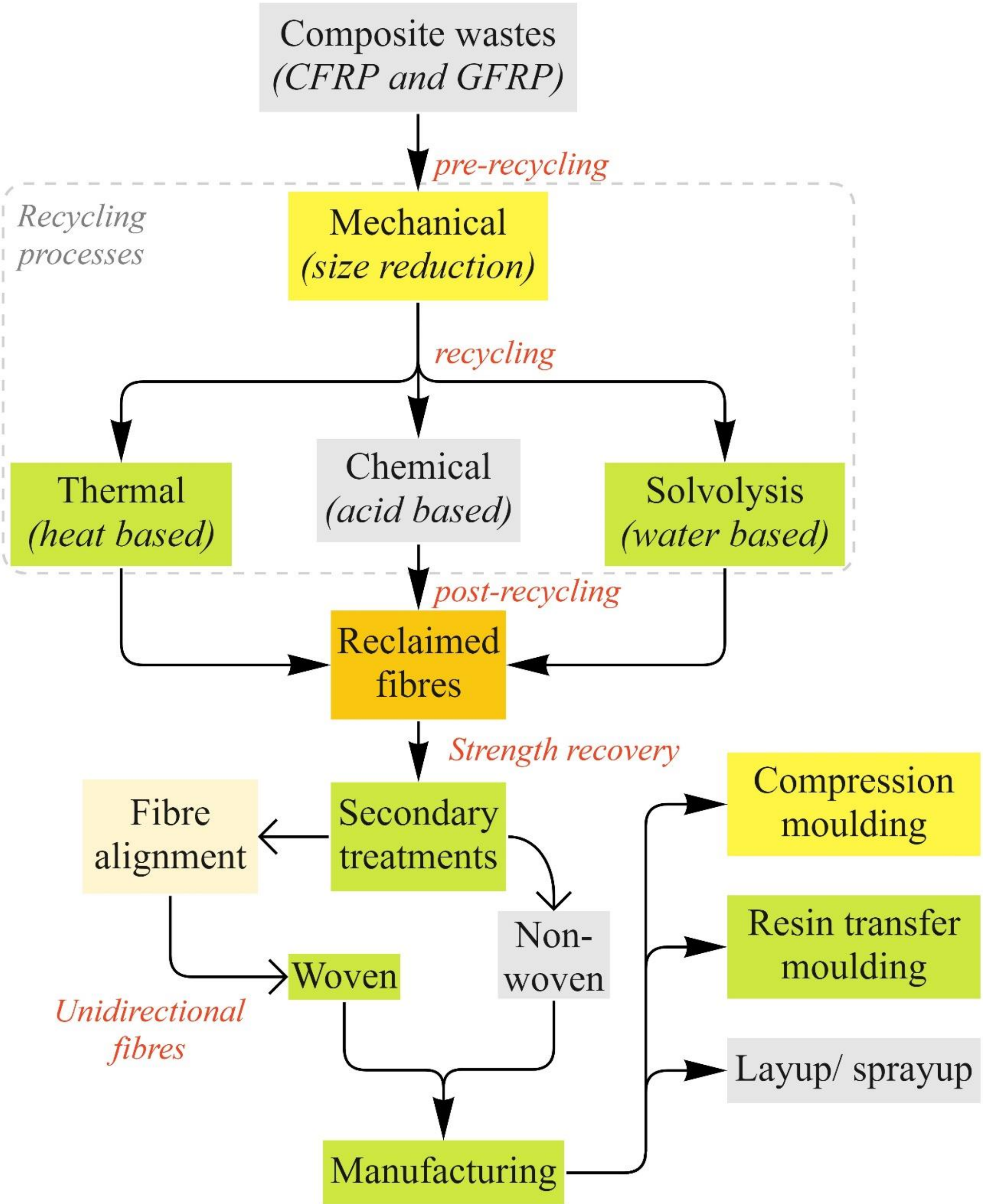


Figure 31. Outlook of material recovery from composite wastes to remanufacturing

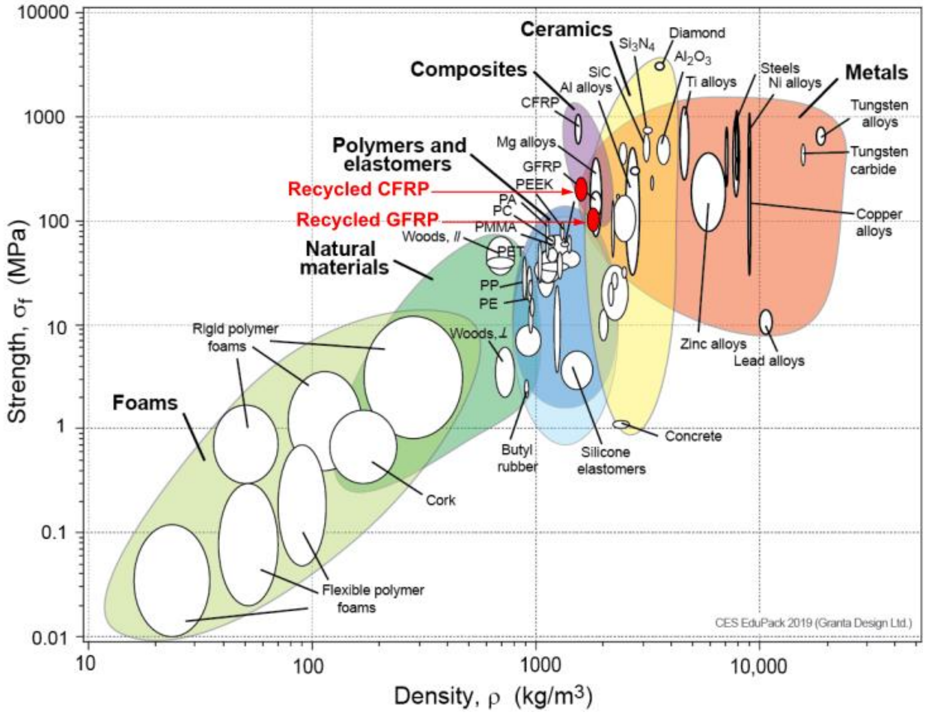


Figure 32. Recycled composites placement in Ashby chart for tensile strength (MPa) (chart created using [93] CES EduPack 2018, Granta Design Ltd.)

The recycled and remanufactured CFRP and GFRP composites possess tensile strength similar to the recycled composites from literature. However, compared to virgin materials available, it provides a better understanding, as seen in Figure 32. The measured tensile strength from the experimental and numerical test results are represented in Ashby's chart for tensile strength (MPa) vs density (kg/m^3). As noticed, the rGFRP composite lay within the range near the vGFRP composite. Still, the rCFRP did not completely fulfil to replace the vCFRP composite due to their minor defects and lack of secondary property enhancement treatments. However, the rCFRP composites are placed in a critical position capable of matching with other composite materials.

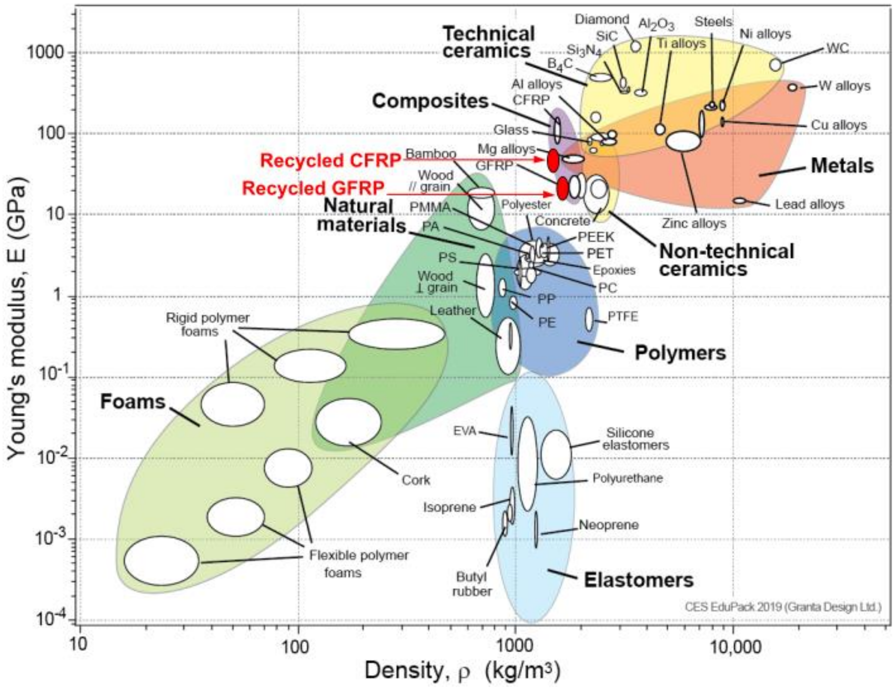


Figure 33. Recycled composites placement in Ashby chart for Young's modulus (GPa) (chart created using [93] CES EduPack 2018, Granta Design Ltd.)

Similar to the tensile strength properties, the rCFRP and rGFRP composites were compared with Ashby's chart for Young's modulus (GPa) vs density (kg/m^3), as seen in Figure 33. The recycled composites occupy the area within other composite materials, highlighting the possibility of replacing other composite materials in various applications if possible. Particularly, when compared to their virgin counterpart, rGFRP composites possess similar values to the GFRP composites. Once again, the rCFRP composites were not possessing properties like virgin composites but were placed in a closer range. The UD, long and continuous fibre arrangement played a significant role in such mechanical properties comparable to the other composites.

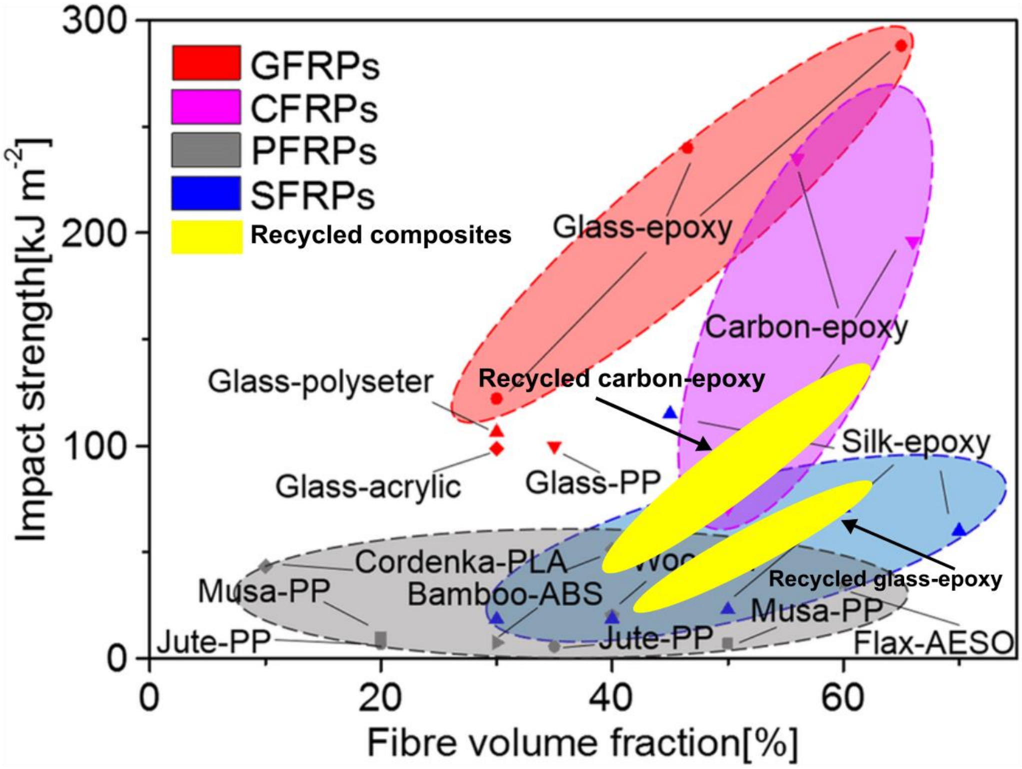


Figure 34. Recycled composites placement in impact strength chart adopted from [94]

Finally, the rCFRP and rGFRP composites were compared with similar FRP composites in terms of impact strength (kJ/m^2) vs fibre volume fraction (%). The mechanical properties of the experimentally and numerically measured values of both the recycled composites are placed in the chart, as seen in Figure 34. The recycled composites fall within their virgin composite ranges. However, the values are lower compared, which could be due to their observed defects like poor wettability and char formation. As the impact test occurs perpendicular to the fibre directions, the defects significantly affect the mechanical properties. Still, as observed, the recycled composites can be reused in an open-loop recycling approach.

5 Conclusions

This doctoral dissertation focuses on closing the life-cycle loop of the CFRP and GFRP composite wastes. A brief synthesis was performed combining four peer-reviewed research articles to successfully recycle the valuable carbon fibres and glass fibres from their CFRP and GFRP composite wastes and remanufacture utilising compression moulding process by employing a fresh resin system. The developed recycling process was investigated using LCA to estimate their environmental impacts and compared with the traditionally available CFRP and GFRP composite waste management routes. The newly produced composites were investigated under uniaxial tensile loading and impact resistance conditions to analyse their mechanical performance. The recycled fibres and composites were also studied under a SEM. The measured mechanical properties of the recycled composites, namely rCFRP and rGFRP, were subjected under FE methods to understand the fracture mechanics of the newly developed unidirectional, continuous and long-fibred composite materials.

Looking at the outlook of recycling composite wastes, it was evident that the government policies have an enormous contribution in forcing material recovery as a primary waste management option for incinerating and landfilling. Plus, considering the price of these virgin CFs and GFs along with the environmental impacts of producing them highlights the importance of recycling processes to recover these valuable fibres actively enabling a circular economy. Various studies have theoretically and practically explored the possibilities of recycling CFRP and GFRP composite wastes. However, only a few studies have considered closing the life-cycle loop of these composites and highlighted the significance of a closed-loop recycling approach. This dissertation has briefly explored the impacts of recycling these composite wastes in various aspects. Initially, developing a novel recycling process adopting optimised principles and parameters from existing processes, tailor-made to fit the CFRP and GFRP based composite wastes with properly recovering the fibres without disturbing their structural integrity. As longer and more aligned the fibres are, the better it is to recycle for multiple cycles. It also helps to explore the maximum potential of these valuable fibres.

Secondly, exploring the mechanical properties of the rCFRP and rGFRP composites have provided insights into the reliability of the developed recycling process and the adopted remanufacturing process. If replacing the virgin composites with recycled composites possesses various environmental and economical benefits, it is significant to reclaim the recycled composite's mechanical properties close to their virgin counterpart. This study has successfully showcased the results to highlight the high possibility of replacing virgin composites with recycled composites. Additionally, the results can be still improvised by employing secondary treatments after the fibre recovery. Such possibilities were clearly seen from the numerical analysis results. As the FE models predicted the failure behaviour of the rCFRP and rGFRP composites from the experimentally recorded material behaviour, it was evident that the defects from recycling and remanufacturing played a significant role in fracture behaviour.

Further upscaling the developed process will allow optimisation of the recycling and remanufacturing processes, eliminating the known defects such as poor wettability in rCFRP and char formation in rGFRP with overall improvised mechanical properties. Plus, such evidence can be observed from the currently available industrial-scale pyrolysis process. Still, the environmental assessment towards the developed process has provided additional insights into replacing virgin using recycled composites, with a detailed investigation comparing the traditional waste management options. Once again, as highlighted, the advantages of upscaling the process could reduce the energy consumption involved in the adopted recycling process, which will eventually reflect in the overall carbon emission and fossil fuel consumption. As noticed in the rGFRP environmental assessment, results showed the benefits of co-incineration to be a more favourable waste management route than the material recovery by thermal recycling process. Considering EU legislation’s recycling and reusing principles, further upscaling the rGFRP can also be positively involved in material recovery recycling processes. Thus, it can be concluded that landfilling and incineration waste management approaches with higher environmental impacts have to be stopped as soon as possible to promote the sustainable opportunity of using waste as a valuable raw material.

Based on the literature review, developed thermal recycling process, investigated mechanical properties of the recycled composites and environmental assessments on the recycling process, it is evident that recycling is the only sustainable alternative to traditional waste management options with a higher chance to replace the virgin composites by recycled once promoting circular economy. Furthermore, recycling approaches like open-loop and closed-loop will allow the recycled composites to be utilised multiple times to use their maximum potential. Plus, with sufficient proof, it can be possible to employ these recycled composites in high-performance applications such as aerospace, aviation, automobile, sports and windmill without compromising the quality and performance, simultaneously achieving a circular economy with a negative carbon footprint.

There are certain limitations present in this dissertation, as the raw materials obtained from the companies are actual composite wastes, their virgin properties are not fully known. As a result, the mechanical properties of the recycled composites were compared to the virgin properties from the literature. Secondly, the recycling and remanufactured defects were identified and discussed briefly compared with results from similar studies in the literature. Therefore, it was not required to entirely repeat the processes to measure the optimised properties as they are out of scope. Thirdly, non-traditional FE methodologies were implemented considering the limited measured mechanical properties of the recycled composites. Additional properties such as compression and shear modes were required to perform a higher-order FE modelling using composite-based damage or user-defined modelling using subroutine (UMAT). Finally, the developed thermal recycling process consumed higher electricity. With further optimising the electricity input, the possibility of rGFRP composite to adopt a material recovery process instead of their now favourable co-incineration is highly achievable.

Further studies are required in various stages of the proposed closed-loop recycling approach to provide insights into researchers and composite recycling industries to successfully establish an industrial-scale recycling plant to achieve the targeted carbon-neutral goals. As the primary recycling processes were thoroughly explored, it is required to focus on improving the secondary treatments in removing the resin residues and regaining the mechanical properties of the recycled fibres. Furthermore, a deep investigation into the fibre alignment systems is required as the targeted fibre outcomes are long and unidirectionally oriented recycled fibres to fully close the life-cycle loop of the newly produced composites multiple times, utilising their maximum potential. Considering the current demand for CFRP and GFRP composites, a mass remanufacturing process maintaining the recycled fibre's structural integrity is required so that the recycled composites can replace virgin composites and satisfy the global need for these high-performance composites.

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Publication I

Karuppannan Gopalraj, S., and Kärki, T.

A review on the recycling of waste carbon fibre/glass fibre-reinforced composites: fibre recovery, properties and life-cycle analysis

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Review Paper

A review on the recycling of waste carbon fibre/glass fibre-reinforced composites: fibre recovery, properties and life-cycle analysis

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Abstract
The growing use of carbon and glass fibres has increased awareness about their waste disposal methods. Tonnes of composite waste containing valuable carbon fibres and glass fibres have been cumulating every year from various applications. These composite wastes must be cost-effectively recycled without causing negative environmental impact. This review article presents an overview of the existing methods to recycle the cumulating composite wastes containing carbon fibre and glass fibre, with emphasis on fibre recovery and understanding their retained properties. Carbon and glass fibres are assessed via focused topics, each related to a specific treatment method: mechanical recycling; thermal recycling, including fluidised bed and pyrolysis; chemical recycling and solvolysis using critical conditions. Additionally, a brief analysis of their environmental and economic aspects are discussed, prioritising the methods based on sustainable values. Finally, research gaps are identified to highlight the factors of circular economy and its significant role in closing the life-cycle loop of these valuable fibres into re-manufactured composites.

Keywords End-of-life · Carbon fibre · Glass fibre · Recycling technologies · Life-cycle loop · Circular economy

1 Introduction

The materials commonly used in well-established sectors—such as aircraft, energy, sports, infrastructure, medical, defence, electronics, and automobile are fibre-reinforced polymer composites (FRPC) [1–3]. Especially carbon fibre reinforced polymers (CFRP) [4] and glass fibre reinforced polymers (GFRP) [5] are incredibly applicable due to their outstanding material properties [6, 7]. So far, industries have been rapidly utilising these materials without proper awareness about their disposal methods. For decades, landfill and incineration were the two popular disposal methods adopted by composite industries. These methods have led to increasing environmental awareness to identify a sustainable dispose method and provide a solution to prevent the cumulating wastes [8–14].

In recent decades, various studies have assessed market requirements for new composites and the amount of cumulating wastes to avoid the inevitable negative consequences. By 2020, The US market for fibre-reinforced composites (FRC) will reach an estimated value of $12 billion, with an annual growth rate of 6.6% [15]. Similarly, by the same year, the annual global demand for carbon fibres (CF) is expected to increase from 72,000 tonnes to 140,000 tonnes, and the CFRP global revenue expected to increase from $28.2 billion to $48.7 billion [16]. To keep up with such a drastic demand for virgin carbon fibre (vCF), the cumulating CFRP waste should be recycled efficiently to reduce environmental impacts and satisfy the need [17]. Indeed, recycling CFRP into a valuable resource is a challenging issue affecting the future of the fibre-based recycling industry [18].

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The cumulating composite wastes are more prominent than the needed new composites. A staggering 62,000 tonnes of unused end-of-life (EoL) and CFRP production waste will be cumulating every year in spite of the existing demand for new fibre composite [16]. The aircraft [19] and wind energy [20] sectors contribute to a major share of this. In the aircraft sector, an estimated value of 23,360 t/y of EoL CFRP will be accumulated if left unrecycled by 2035. Also, due to developments in modern aviation, the cumulative amounts from the following world regions are estimated to be available for recycling by 2050: North America (162,083 t), Europe (144,724 t) and Asia (102,500 t) [19]. Similarly, the EoL wind turbines will be cumulating 483,000 tonnes of CFRP waste, consisting of North America (95,000 t), Europe (190,000 t) and Asia (146,000 t) by 2050 [20]. Additionally, by the same year, wind energy will be replacing traditional electricity production and will account for 50% of the EU's total electricity consumption [21]. However, despite their green credentials, wind turbine blades made from GFRP have a lifespan of approximately 20 years. This means that, by 2030, an estimated 100,000 tonnes/year of wind turbine blades will be cumulated. Plus, currently, only a few recycling techniques are available to treat such an enormous quantity [21, 22]. After wind turbines, waste printed circuit boards (WPCBs) represents one of the fastest-growing global waste streams [23], contributing a significant share to overall electronic waste and consisting of 27.4–45.55 wt% glass fibres (GF) [24, 25]. Recycling WPCBs to recover GFs is a challenging process due to the presence of toxic heavy metals and organic compounds, along with the GFs themselves [26].

Current landfilling percentages recorded in the UK stand at 35% for CF and 67% for GF, with only 20% of CF and 13% of GF recycled and a small amount 2% CF and 6% GF—being reused [27]. Furthermore, 2000 t/y of CF waste (20% of the total CF made in the US), if rescued from landfills and properly recycled, can be worth up to €14.7 million of recycled carbon fibre (rCF) considering €10/Kg as the rCF market price. This value can increase dramatically to over €50 million by 2020, arguably due to a stable 15% annual increase in vCF production worldwide [28]. To develop new markets, an economically sustainable recycling model for CFRP and GFRP waste should be utilised [27, 29, 30], opening up both direct and indirect job opportunities and contributing to economic development [31].

In the current situation, a complete recovery of fibres (direct structural recycling approach) is considered to benefit the composite sector. The recycled fibres from this approach have an added market value because of the low usage of natural resources, energy, and labour-power, together with near-virgin fibre quality [1]. Numerous methods, especially mechanical, thermal and chemical-based recycling approaches, have been studied and established so far because method selection depends on the type of material to be recycled and the application in which it will be reused [2]. Also, identifying one standard recycling method among various methods is difficult.

In the research field, recycling CFRP and GFRP waste have progressively become an area of interest, as shown when assessing the overall literature studies of the past 20 years—highlighted in Fig. 1. There has been a steady growth in the recycling of both types of fibres. However, after 2011, studies related to CF recycling show an exponential growth without any deviation. Indeed, focusing on review articles, in particular, shows that previous studies have concentrated on either CF or GF or their Life-cycle analysis. But not all three factors combined into a single study. For the present scenario, such review articles are needed to compare various phenomena and to identify research gaps.

The method adopted in this study is a standard narrative review structure proposed by Green et al. [32]. The challenging literature search was conducted primarily in the Scopus database, and three additional databases were used as secondary supporting platforms such as Web of Science, Science Direct and Research Gate. The search was limited to the last 20 years. During the search, keywords such as "carbon fibre", "glass fibre", "Kevlar", "Twaron", "waste carbon fibre", "waste glass fibre", "waste Kevlar", "waste Twaron", "recycling", "recycle", and "recycled" were used. In the end, relevant articles were carefully selected based on an analysis of the title, aim, and novel findings from the research. Considering the limited information available on the topic of recycling Kevlar and Twaron waste, both the fibres were eliminated from this study.

However, all the relevant articles under the recycling of CF waste (360 articles) and GF waste (85 articles) were reviewed and organised into a private database using Microsoft Excel. A further article selection was made based on the following criteria: a focus on recycling methods; the mechanical properties of recycled

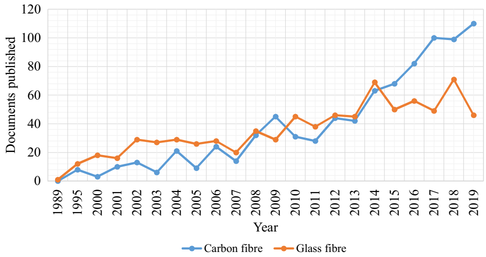


Fig. 1 Number of articles published based on CF and GF recycling for the past 20 years

fibre; and the economic and environmental analysis of the recycling methods. Articles that deviated from the framed selection criteria were eliminated. Overall, this literature review is narrowed solely focusing on summarising the significant techniques to recycle CF and GF waste. Based on the effective recycling circumstances to recover clean fibres, on the characteristics of the recovered fibres, on the life-cycle analysis of the recycling process. Finally, 153 articles were used in the reference list.

Overall, this review article will provide insights into the current status of recycling methods available to recover both the carbon fibre and glass fibre from their composite wastes. The study will also prioritise the available recycling methods based on the following factors—such as environmental impact, commercial value, quality of the recovered fibre and recyclability on an industrial scale. Additionally, the study highlights the process conditions associated and the characteristics of the resulting fibres to justify the necessity to replace virgin fibres with recycled fibres.

This paper is organised based on the available recycling methods in three sections, as shown in Fig. 2. The traditionally used mechanical recycling techniques and their latest alternative approaches are presented in Sect. 2. Thermal recycling techniques, including the fluidised bed process and pyrolysis process, are presented in Sect. 3. The chemical recycling techniques and their latest alternative approaches, along with solvolysis, are discussed in Sect. 4. Subsequently, Life-cycle analysis comparing all the significant factors are discussed in Sect. 5. After a brief discussion in Sect. 6, the paper ends with the conclusions in Sect. 7.

2 Mechanical recycling

In general, mechanical recycling is a technique used to reduce the size of scrap composites into smaller pieces known as recyclates. Normally, slow-speed cutting or crushing mills are used to reduce the material size to 50–100 mm but, when the scrap composites are homogeneous without any metal components, high-speed milling will be adapted to reduce the size between 50 µm and 10 mm [33]. The recyclates are classified based on coarse recyclates (higher fibre content) and fine recyclates (higher resin content) using cyclones and sieves. The effective reusing of recyclates is based on particle size [33, 34]. In the current situation, this has been used as a pre-recycling process for various methods [35, 36].

Even though the mechanical recycling process is capable of recycling both CFRP and GFRP, most of the research focuses on GFRP [2, 33, 37]. Discontinuous recyclates and their re-incorporation with low-value applications like fillers or reinforcements can provide the main reasons for such research variation [4]. Besides, CFs are expensive compared to GF. Disrupting their physical integrity by mechanical recycling can lead to economic and fibre property loss. Since the early development of the process, serious drawbacks have been involved, even though studies like Mou et al. [38] showed the improved flexural strength of concrete after the addition of GF recyclates as

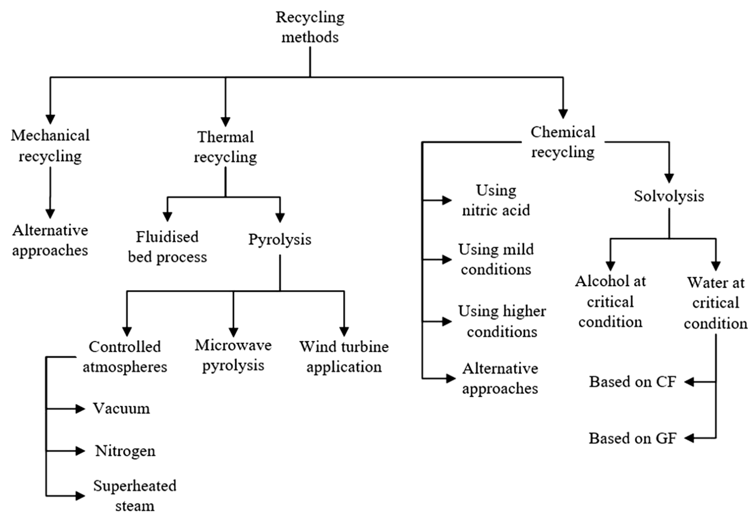


Fig. 2 The adopted recycling methods

filler materials. Studies like Pickering [33], however, noted that the GF recyclates used as fillers are not commercially feasible due to the availability of alternative cheap virgin fillers such as calcium carbonate or silica.

To overcome the limitations, recent studies show acceptable improvements in the process; for example, studies like Meira Castro et al. [39], which used computational intelligence for optimisation. This involved waste GFRP recyclates being used as aggregate and filler replacement in a concrete-polymer composite to showcase improvements in compressive and flexural strength. Moreover, this optimised process was cost-effective when compared to the thermal and chemical recycling process. Also, Shuaib and Mativenta [40] improved both the yield and quality of the GF recyclate by using low energy consumption. Their study found that reducing the screen size to obtain a fine recyclate will result in increased energy consumption and processing time. To overcome such energy loss, the clearance gap between blades and screen was decreased. In comparison, further increasing the yield without any residue, Kočevar and Kržan [41] separated 70% of the GF using a normal hammer mill. The remaining 30% of waste was used as filler material for thermoplastics.

As mentioned previously, the limitations of recycling CFRP waste were challenging and even visible in the latest study by Li and Englund [42], where aerospace industry scrap was size-reduced using a hammer mill followed by shredding. The recyclates are compression moulded into flat pallets and subjected to mechanical testing, which showed a minimum 50–60% decrease in mechanical properties compared to the original composite. However, the study pointed out that, as the CF recyclate particle size decreases, the mechanical property increases.

2.1 Alternative approaches

Recent studies have focused on alternative size-reduction approaches using high voltages. One such method was electrodynamical fragmentation (EDF) in which a high voltage pulse between 50 and 200 kV was passed into ionised water to break down the CFRP waste into smaller pieces [43, 44]. Similarly, high voltage fragmentation (HVF) was carried out using a high voltage pulse of 160 kV to breakdown the GFRP waste. This method produces clean and long fibres. Moreover, HVF can be a promising alternative to mechanical recycling [45]. However, recently, Oshima et al. [46] pointed out two drawbacks in this approach, using high voltage to remove resin from CFRP waste will result in a severe weight loss in the actual composite and also decreases the rate of resin removal.

3 Thermal recycling

In a thermal recycling process, heat is used to break down the scrap composite. Due to a higher operating temperature (450–700 °C), the insignificant volatile materials are likely burnt, leaving the valuable fibres behind. Usually, the process temperature depends on the type of resin utilised in the scrap composite. Improper temperature can either leave char on the fibre surface (undercooked) or result in reducing the diameter of the recovered fibres (overcooked) [2, 47]. Thermal recycling can be classified into three types [33], as shown in Fig. 3.

The basic principle for decomposing the scrap composite using heat remains the same, though, the results are different for each process. Since polymeric compounds have certain calorific values, electricity can be produced by converting the waste composite into heat [1, 2]. However, a major drawback in the combustion (incineration) process is the ash by-product, which can only be landfilled as inert waste, e.g. 92 €/tonne in France. This complication harms the progress of a circular economy. Besides, it is only possible to achieve a 35% efficiency rate when converting the heat to electricity. Overall, coal in the furnace is a much better option than incinerating CFRP. Recent studies have focused on complete fibre recovery using thermal recycling processes like fluidised-bed process (FBP) and pyrolysis [48].

However, instead of completely combusting CFRP, controlled resin decomposition at optimum temperature can result in CF recovery with negligible surface damage. In a study by Matielli Rodrigues et al. [47], the versatile compounds (resins) were thermally disintegrated at 450 °C for 2 h and CF recycled without damaging much of the surface integrity. The decomposed epoxy resins are derived from the diglycidyl ether of bisphenol-A (DGEBA), which is difficult to recycle due to its cross-linked structure from resin curing. Therefore, recycling with minor surface damage is a better option than landfilling completely.

Resin decomposing using optimal thermal conditions is not as efficient for GF as CF, but post-chemical treatments of recycled glass fibre (rGF) help to a certain extent in regaining their properties [49]. Unlike CF, the thermal recycling of GF in high-heat operating conditions (300–600 °C)

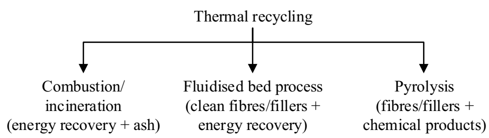


Fig. 3 Classification of thermal recycling

reduces the strength of the resulting GF up to 80% and is difficult to further reuse because of its low reinforcement potential [50, 51]. To overcome such phenomena, Yang et al. [50] investigated two chemical treatments: chemical etching and post-silanisation to treat decomposed GF (80% decreased tensile strength) at 500 °C for 30 min. The post-chemical treatment retained 30–70% of the lost mechanical properties in rGF. Thomason et al. [49] recovered 75% of the strength-loss by immersing the GF in 3 M NaOH solution for 10 min at 90 °C, followed by neutralising with HLC. The treatment is known as short-hot sodium hydroxide solution treatment. Pender and Yang [52] introduced catalysts: CuO, CeO_2, and Co_3O_4 to boost the resin decomposition. As a result, the processing time was reduced by 20 min, along with a 40% reduction in energy consumption. Among the three catalyses, CuO at 375 °C had maximum efficiency in removing the resins, while CuO and CeO_2 increase 20% of GF's strength retention capacity.

3.1 Fluidised bed process

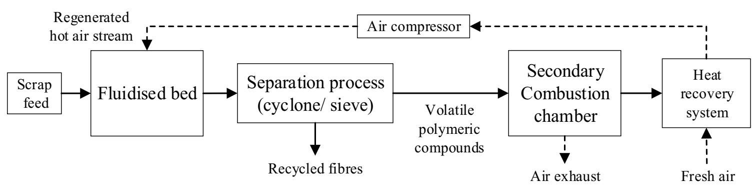
In a typical fluidised bed process (FBP), a rapid stream of hot air is passed through a bed of silica sand to decompose the chopped scrap composite at a low temperature. Usually, a fine silica sand of 0.85 mm particle size is used as a bed, which is then converted into a fluidised bed by passing air in the velocity range 0.4–1.0 m/s. The polymeric matrix scrap composites are chopped to 25 mm and fed separately into the fluidised bed. The operating temperature is between 450 and 550 °C. Inside a fluidised bed, the scrap composite separates into fibres and fillers (volatile compounds), which are carried out by the air stream as individual particles [33, 53].

Furthermore, suspending the individual particles in a high-temperature (1000 °C) secondary chamber will result in oxidising the volatile compounds, leaving the fibres alone [33, 53]. Figure 4 represents a diagram of the FBP. The process is capable of recovering both CF and GF [33] and is especially favourable for recycling EoL waste composites [2]. However, during the initial development of the process, limitations like fibre strength and length degradation became apparent. Also, the recovered fibres are fluffy [4].

The limitations are noticeable in Pickering et al. [53], as their research investigated the ability of FBP to recycle GF and was only able to achieve a 67% fibre yield. Furthermore, the process retained only 50% of the tensile strength of the rGF compared to virgin GF (vGF). However, the study included a cost analysis for commercialising the process and estimated that for 9000 tonnes/year, the net annual profit would be 0.002$ million/year. Besides the limitations, the author's contribution to the process has laid the foundations for modern-day FBP [4]. In contrast with Pickering et al. [53] results, Zheng et al. [55] claimed to have a novel FBP approach to recycle WPCBs, achieving a 94.8 wt% GF recovery rate and a 95.4 wt% purity rate. The recycling approach seems to be almost similar, except for the WPCB were finely chopped than regular chopping size (25 mm) before feeding. However, the results were validated based on weight loss and SEM micrographic image observations. Plus, no information regarding the strength of the rGF's was mentioned. To improve the yield of rGF, Pender and Yang [56] used CuO as a catalyst, with the results improving the yield from 59 to 70%.

A similar study to Pickering et al. [53] but replacing GF with CF was carried out by Yip et al. [57]. The findings showed that recycling CF by using identical operating conditions was much more efficient compared to GF recycling. The rCF had fully retained Young's modulus and 75% of tensile strength when compared to vCF. The study also mentioned that the rCF length depends on the initial length of the CF waste, and it was possible to recover a fibre length of 5.9–9.5 mm despite the non-uniform orientation. In alignment with previous studies, Pickering et al. [54] systematised the idea of commercial-scale FBP with its ability to recover CF and also designed a commercial scale FBP suitable for recycling mixed and contaminated CFRP waste. The rCF only had an 18.2% tensile strength decrease and no tensile modulus reduction. The study claimed that the design has the highest efficiency compared with any other rCF using FBP. Also, the energy consumption to recover CF via FBP is only 5–10% of the total energy needed to manufacture vCF.

Fig. 4 Fluidised bed process (modified from Pickering et al. [53, 54])



3.2 Pyrolysis process

The process is efficacious for recycling both CF and GF [15]. Unlike other thermal recycling processes, the scrap composites are heated in the absence of oxygen. Under this condition, the decomposing matrix produces oil and gas, along with fibres and fillers (solid products) [2, 33]. In a typical polymeric pyrolysis process, the operating temperature varies in the range of 400–700 °C depending on the nature of the scrap composite [1, 4]. The liquid produced from the process contains aromatic compounds and has a calorific value of 37 MJ/Kg, similar to fuel. The gas produced can be regenerated into the pyrolysis reactor. Overall, the oil and gas products produced can prove a significant chemical feedstock for any other process. Nevertheless, in spite of all that is produced, the process is still capable of retaining fibres with higher mechanical properties [1, 4]. This clarity of effect makes pyrolysis the most studied thermal recycling process [2], Fig. 5 presents a flowchart of the process.

Like any other recycling process, pyrolysis also suffers from certain limitations, with the possibility of char formation on the resulting fibre surface considered the most challenging of all [4]. A significant percentage decrease in the mechanical properties can be observed in the recovered fibres due to the char. Methods such as chemical treatment [26, 58] and post-heating the fibres result in reducing the char formation, but only to a certain extent [1]. Recent studies have used carbon dioxide (CO_2) and water vapours to remove the char formation from CFRP [59]. Also, oxidising the fibre surface will result in the formation of an oxygen-rich surface, improving the adhesive nature of the fibre with resins [60].

To completely avoid such char formation on the recovered CFs, a UK milled carbon group was successful in designing and implementing a commercial-scale semi-open continuous belt furnace with a controlled atmosphere [4, 61]. Similarly, Germany's CFK Valley Stade Recycling GmbH and Co. KG uses a continuous pyrolysis process. Both companies are capable of recycling various types of CFRP waste. Moreover, the large furnace and

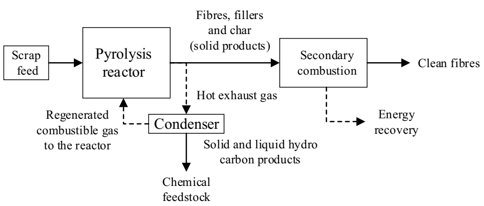


Fig. 5 Pyrolysis process (modified from Pickering et al. [33])

continuous flow allow them to recover longer and cleaner CFs [4]. Evidence for such a phenomenon can be seen in an earlier study from Meyer et al. [62] on recycling aircraft manufacturing CFRP waste, in which low-efficiency lab-scale pyrolysis was optimised to a semi-industrial scale with the help of the company ReFiber. The authors were able to synthesise semi-industrial plant operations using a larger oven, with rCF retaining 96% of its original tensile strength. However, a secondary heating system was implemented to eliminate the residue char, though the rCF was effective enough to replace vCF. Overall, a controlled atmosphere in the pyrolytic reactor can influence the process outcomes.

3.2.1 Pyrolysis: controlled atmospheres

The development of pyrolysis processes resulted in sophisticated and controlled atmospheric conditions being added to separate fibres from the solid pyrolysis products. Various literature works published since 2010 include evidence of such an approach. The primary principle of pyrolysis is unaffected. However, the atmospheric conditions inside the pyrolytic reactor have changed constantly, aiming for higher yields. Three commonly-used atmospheric conditions during pyrolysis are vacuum, nitrogen and superheated steam.

3.2.1.1 Vacuum atmosphere Inside a vacuum pyrolytic reactor, the organic vapours' residence time is shorter due to a low decomposition temperature, which is sufficient enough to recycle GF without disturbing surface integrity [63, 64]. In an early study focused on recycling WPCBs by Zhou and Qiu [63], vacuum pyrolysis (VP) was judged as an alternative approach to mechanical recycling. The liquid and gas products (can be used as chemical feedstock) along with solid products (GF and metal components) were found to increase the interests of researchers when compared to mechanical recycling [24, 63, 64]. Their study consists of a two-step pyrolysis method, such as VP and vacuum centrifugal separation (VCS), to separate GF from WPCBs. Similarly, Long et al. [24] used VP, which involved the GF being separated from metal parts using gravity separation (GS) method and non-metal parts using a calcination process. The studies focused on recycling WPCBs using a vacuum atmosphere did not include any investigation on the mechanical properties of rGF. However, the yields from the process are tabulated in Table 1. This process is limited to GF as no suitable study was found concerning CF.

3.2.1.2 Nitrogen atmosphere Unlike vacuum pyrolysis, pyrolysis under a nitrogen gas atmosphere is capable of recycling both CFRP and GFRP [61]. Pyrolysis under

Table 1 Pyrolysis yields (wt%) from recycled CFRP and GFRP

References	Material	Solid yield	Liquid yield	Gas yield	Process parameters
Zhou and Qiu [63]	GF	75.7	20	4.3	a. VP: 600 °C for 30 min
Zhou et al. [64]	GF	72.2	21.45	6.35	b. VCS: 400 °C, 1200 rpm for 6–10 min
Long et al. [24]	GF	74.7	15	10.3	a. VP: 550 °C for 120 min
					b. GS + calcination: 600 °C for 10 min
Onwudili et al. [65]	CF	77	20.6	2.4	NA: 400–500 °C < 30 min + SC: 450 °C for 2 h. (35 MJ m^{-3} calorific value of gas)
Onwudili et al. [61]	CF	73.1	23.8	2.3	NA: 500 °C for 45 min with 5 dm^3/h nitrogen + SC: 500 °C for 30 min at 20 °C min^{-1}
	GF	65.9	25.7	8.4	

nitrogen atmosphere (NA) was the popular pyrolysis process used to recycle WPCBs until Zhou et al. [64] 's study came in 2010. The study used vacuum pyrolysis to recycle WPCBs, achieving a maximum GF recovery. However, later analysing the advantages of pyrolysis under NA, in 2013, Onwudili et al. [65] further developed the process and successfully recycled CFRP waste—in which 98 wt% of the solid products (rCF) were recovered efficiently. The study stated that a reaction time increase could cause a decrease in the mechanical properties of the recycled CF. Also, insufficient reaction time could result in char formation on the fibres. The yield from the process is tabulated in Table 1.

In line with previous research, in 2016, Onwudili et al. [61] investigated the mechanical properties of both the recycled CFRP and GFRP waste using a nitrogen atmosphere in a semi-batch reactor. The study found that chemical modification on the surface of recycled CF and GF could improve fibre properties. Additionally, both studies [61, 65] used secondary combustion (SC) to oxidise minor resin impurities and char formation on the pyrolysed fibres. When comparing the results, oxidised fibres retained the most mechanical properties when compared to non-oxidised fibres.

3.2.1.3 Superheated steam atmosphere In general, superheated steam is produced as a result of heating saturated steam at constant pressure. During the transition phase, the steam is heated several times above its saturation point to achieve superheated levels. The use of superheated steam atmosphere in pyrolysis increases heat transfer, which results in heightening the thermal degradation and supporting an oxygen-free atmosphere inside the pyrolytic reactor [66]. Recycling CFRP using superheated steam results in a high retaining of the CF mechanical properties when compared to any other pyrolysis atmospheres [67, 68]. Also, a significant amount of the char formation on the resulting CF and GF can be reduced using chemical treatments [58].

Using pyrolysis under superheated steam, Shi et al. [67] investigated the mechanical properties of rCF. They found that the recycled fibres had a substantial amount of char on the rCF surface, which prevented the fibres from being completely reusable. The study also stated that the lower the pyrolysis temperature, the higher the value of recycled CF. Furthering their previous studies, Shi et al. [58] extended their research to CFRP and GFRP, eliminating the char formation on both recycled CF and GF using chemicals such as detergent, acetone and N-methyl-2-pyrrolidinone (NMP). As a result, the bending strength of rCF increased from 49 to 78% after treatment with NMP, and the bending strength of rGF increased from 26 to 94% after treatment with acetone. Recently, Jeong et al. [69] recycled CF and showed 66% of tensile strength and 100% of tensile modulus retained from an original value using rapid pyrolysis.

In an attempt to retain maximum tensile strength in the recycled fibres, Ye et al. [70] developed an optimised steam thermolysis process combining vacuum pyrolysis and mild gasification to recycle waste CFRP. The process retained 90% of tensile strength in both laboratory and semi-industry scales. The study stated that a degradation increase in the polymer matrix resulted in a decrease of the fibres' tensile strength. However, Kim et al. [68] retained 90.42% of tensile strength compared to the vCF by using a fixed bed reactor at 550 °C for 60 min. The study mentioned that increasing the steam pyrolysis conditions resulted in improving the removal of char deposited on the fibre surface. In a recent study by Kim et al. [71] superheated steam was used to remove minor resin residues after pyrolysis with carbon dioxide (CO_2) and recovered above 80% of the strength compared to vCF.

3.2.2 Microwave pyrolysis

In microwave-assisted pyrolysis, the conventional heating source was replaced with microwave radiation. This change has increased the rate of thermal transfer with minimal energy consumption, without disturbing the primary principle of pyrolysis. The process is capable of recycling both CFRP and GFRP with fibres retaining higher mechanical properties [2]. In a recent study, Obunai et al. [72] achieved a 100% resin elimination ratio using a 700 W, 2.45 GHz, argon atmosphere, and a 2.5 L/min flow rate after 300 s. The rCF had only a 0.7% decrease in tensile strength compared to vCF. Similarly, Jiang et al. [73] recycled CFRP at 500 °C for 30 min with a 0.70 m^3/min nitrogen flow. The rCF possess a clear fibre surface with mechanical properties like virgin fibres.

3.2.3 Pyrolysis on wind turbine application

Wind energy is a widely-used renewable energy source [74], for which GFs make an important material contribution to wind turbine manufacturing [3, 5, 75]. In general, the life cycle of an individual wind turbine only lasts from 20 to 25 years [76]. Studies focusing on GF recycling from wind turbine applications have frequently preferred pyrolysis [77]. Apart from the basic GF recycling, studies have focused on retaining their mechanical properties to complete the life-cycle loop by reusing them in various other applications [5].

Looking at early studies related to wind turbine recycling after 2010, studies like Åkesson et al. [76, 77] were successful in retaining 75% of rGF tensile strength by using microwave pyrolysis in a nitrogen atmosphere. Their study proposed that a loss in tensile strength can be overcome by reusing it as a hybrid composite, combining rGF with vGF. In line with the previous study, Åkesson et al. [78] further investigated improving the rGF properties by reinforcing them with polypropylene (PP). The study stated that the use of maleic anhydride grafted PP along with coupling agents improved the mechanical properties, including the flexural strength, tensile strength and Young's modulus.

Studies like Pico et al. [75] and Beauson et al. [74] have highlighted the limitations and mentioned that rGF from wind turbine blades are short, fluffy and randomly oriented. Also, the rGF tensile strength is lower when compared to vGF. However, minor organic contents of the rGF surface are removed by oxidising. As a counter-statement, Jensen and Skelton [3] highlighted the circular economy as a promising way to reuse the recovered fibres in various applications. The study mentioned that, as an outcome of the GENVIND project (a 4-year project carried out by a Danish innovation consortium), a wind turbine will no longer only be renewable in terms of producing energy but also in material reuse. A collaboration between the material supplier, the wind industry and the EoL sector can make this happen easier. A recent study has estimated that, in the future, chemical recycling will be capable of recycling GF, hybrid and CF wind turbine blades with higher efficiency [9].

4 Chemical recycling

In a chemical recycling process, the polymeric matrix present in the waste composite is disintegrated by dissolving it into any chemical solution, such as acids, bases and solvents. Normally, suitable chemicals and solvents are chosen based on the nature of the polymer substrate [4, 79], while the solid composites are mechanically grounded before chemical recycling to increase the surface area. Once the polymer matrix is dissolved, the recycled fibres are washed to remove minor surface residue [1, 79]. Fibres recovered using chemical recycling have retained long fibres with maximum mechanical properties. Plus, the process has a higher resin decomposition ratio [4].

In modern chemical recycling, resin degradation is either achieved using solvents (solvolysis) or water (hydrolysis). In solvolysis, solvents in different conditions (reaction time and concentration) are used to depolymerise or break the polymeric part of a composite. In hydrolysis, resin degradation takes place because of water [1]. Generally, the use of hazardous and concentrated chemicals results in environmental impact [4], so the harmful chemicals are replaced with water and alcohol at supercritical conditions (SC). Besides, the drawback of improper fibre alignment in discontinuous rCF with a length of more than 5 mm can be suppressed using a centrifugal alignment rig concept [80] or by using calendaring through rollers with 0.10–0.15 mm gap at 110 °C [81].

4.1 Classification of chemical recycling

Classifying the chemical recycling process on the basis of previous studies depends on structuring significant factors according to its priorities. In early literature, Morin et al. [82] classified chemical recycling (solvolysis) based on temperature; solvolysis at low temperature using solvents like acid solutions, alcohols, etc., at 90 °C and solvolysis in SC of solvents like acids and water. However, recent research by Oliveux et al. [2] classified solvolysis based on higher temperature and pressure (HTP) with temperature > 200 °C and lower temperature and pressure (LTP) with temperature < 200 °C. In this study, chemical recycling is organised based on studies of nitric acid, together with other chemicals in mild conditions (T < 100 °C), and others at higher conditions (T > 100 °C).

4.1.1 Recycling using nitric acid

Nitric acid is suitable for recycling both CFRP and GFRP. In the early-stage recycling of CFRP using acids, Liu et al. [83] found that nitric acid performed better in decomposing thermoset epoxy resin and recycled cleaner CFs compared to both sulfuric and hydrochloric acid. Also, orthogonal experimentation was used to prove the reusability of recycled CFs, while even stronger resins like amine cured epoxy were decomposed using nitric acid—for which Ma et al. [84] achieved a 99.18% resin decomposition ratio. The study resulted in an 85% CF cleanliness rate with minimal damage to the fibre surface. Similarly, Lee et al. [85, 86] recycled CFs with only a 2.91% tensile strength decrease compared to vCF. The detailed process conditions are mentioned in Table 2.

GFRP decomposition was also more efficient with nitric acid when compared to concentrated sulfuric acid [87]. In an early study, Yuyan et al. [87] achieved a 99% rate of resin decomposition. The rGF only has a 3.5–15.1% tensile strength decrease and a 2.5% decrease in interlaminar shear strength (ILSS) compared to the original fibres. The study stated that an increase in temperature and acid concentration would increase the decomposition rate. Another study by Dang et al. [88] investigated the corrosion resistance of T-GF and E-GF and stated that T-GF has higher corrosion resistance in nitric acid solution compared to E-GF.

4.1.2 Recycling using mild conditions

The chemical recycling of scrap composite using mild conditions increases resin degradation [2]. Studies related to a lower temperature (T < 100 °C) and mild acids have also achieved relatively higher results. In Li et al. [89] study, a 90 wt% epoxy decomposition ratio was achieved using a self-accelerating oxidative decomposition system (acetone + peroxide hydrogen [H_2O_2]). The authors mentioned that acetic acid is used to pre-treat the waste composite for expansion, producing a larger surface area. The rCF retained more than 90% of tensile strength compared to the original composite and showed that recycling under mild conditions is effective. The detailed process conditions are mentioned in Table 2.

Similarly, Xu et al. [90] achieved a 90% epoxy decomposition rate using N,N-dimethylformamide (DMF) and H_2O_2 mixed solution. The rCF only shows a 5% tensile strength decrease compared to the original composite. The single fibre tensile test only showed a 2% tensile strength loss compared to the vCF after a 10 °C process temperature increase. In a recent study, Das et al. [91] showed a 97% resin decomposition ratio from a single-stage oxidation process using an aqueous mixture (AM) of peracetic acids. The rCF retained 94% of its original strength. This process is limited to CF, and no proper study was found concerning GF. However, recent studies used solvents such as polyethylene glycol [92], dimethylformamide (DMF) [93] and water (1% NaOH) [94] to recycle WPCBs under mild conditions.

4.1.3 Recycling using higher conditions

Studies carried out since 2000 have adopted higher process parameters for recycling waste composites with higher resin decomposition, with chemical recycling using super and sub-critical conditions (SCC) playing an essential role in this [2]. However, recycling using super and sub-critical fluids, especially water and alcohol, have been significantly narrowed down (summarised in a separate section). Based on the literature, studies since 2010 have focused more on improving the mechanical properties of recycled fibre. The process parameters, conditions and outcomes of the research are listed in Table 3, with the outcome being

Table 2 Process conditions

References	Year	Process conditions
Liu et al. [83]	2004	70–90 °C
Yuyan et al. [87]	2006	90 °C, 8 M nitric acid, 2 g/100 mL feedstock ratio and 5 h
Ma et al. [84]	2009	95 °C, 8 mol/L nitric acid and 23 h
Le et al. [85]	2010	90 °C, 6 g:100 mL (weight CFRP: nitric acid) for 5 h
Lee et al. [86]	2011	90 °C, a hexahedral circulating system at a 1 cm/s linear flow rate, 100 g:1.8 L (waste composite: 12 M aqueous nitric acid ratio)
Li et al. [89]	2012	60 °C for 30 min
Xu et al. [90]	2013	90 °C for 30 min, hermetic reactor
Das et al. [91]	2018	65 °C for 4 h. AM: 95 vol% 14 M acetic acid and 5 vol% 9 M H_2O_2

Table 3 Recycling using high conditions after 2010

References	Year	Recycled	Solvents	Parameter	Outcome
Lee et al. [85]	2010	CF	Organic: tetralin (T) and diethylene glycol monomethyl ether (DGME)	350 °C, 2 Mpa for 2 h DGME, 193 °C, atm pressure (AP) for 10 h	3.67% decrease in tensile strength compared to vCF
Yang et al. [95]	2014	CF/GF	Polyethylene glycol/NaOH	200 °C for 4 h with 0.1 g NaOH/g	84.1–93% decomposition efficiency CF and GF 4–6% decrease in original tensile strength
Yamaguchi et al. [96]	2015	CF	Hydrochloric acid (HCL)	130–150 °C, 2 h	rCF like vCF
Nie et al. [97]	2015	CF	Molten KOH	285–330 °C, AP	Retained 95% tensile strength
Wang et al. [98]	2015	GF	Aluminium chloride/acetic $AlCl_3/CH_3COOH$	180 °C for 9 h	Retained 96% of tensile strength
Wang et al. [99]	2015	CF	Aluminium chloride/acetic $AlCl_3/CH_3COOH$	180 °C, 15 wt % solvent for 6 h	Retained 97.77% of tensile strength
Liu et al. [100]	2017	CF	$ZnCl_2$/ethanol catalyst system	190 °C	Retained high mechanical property
Oliveux et al. [101]	2017	CF	Water and acetone in 20:80 vol ratio	320 °C, 180 ± 10 bars in a 5 L batch reactor	rCF like vCF
Wu et al. [102]	2019	CF	Molten $ZnCl_2$	360 °C, 80 min	Retained 95% tensile strength

the retaining of maximum mechanical properties with maximum resin decomposition efficiency.

Besides the focus on property improvement, studies have also focused on comparing the recycling mechanisms involved in the chemical recycling process. For example, Ma et al. [103] compared the depolymerisation and also the acid digestion of amine-cured epoxy resin, finding that the resin dissolution rate was influenced not only by the chemical reaction rate but also by the diffusion rate. Working from their previous research, Ma and Nutt [104] were able to implement their findings on both CF and GF to disintegrate amine-cured epoxy resin. The study showed that acid digestion with acetic acid/H_2O_2 at 110 °C was effective when compared to depolymerisation with benzyl alcohol/K_3PO_4 at 200 °C for highly crosslinked amine cured epoxy resin. Also, in the case of acid digestion, minor degradation was found on the rGF surface. Plus, exposing GFRP waste above the elevated temperature will result in rGF strength loss [105].

From an economic point of view, WPCBs can be recycled cost-effectively using higher conditions (260 °C for 10 min). Moreover, the ionic liquid ethyl-3-methylimizadolium tetrafluoroborate [$EMIM^+$][BF_4^-] used to dissolve the WPCB resin is capable of multiple regenerations and reuse [106, 107]. Recently, cheaper solvents such as water and 1 M aqueous solution of sodium hydroxide (NaOH) have been used to separate WPCBs at higher conditions (280 °C for 15 min) [23].

4.2 Alternative approaches

As with every other process, novel chemical recycling approaches have been studied. By unaltering the overall principle involved in the process, studies have made minor changes for better results. As an early example, Kamimura et al. [108] used microwave-enhanced depolymerisation and recycled 51 wt% of pure GF from FRP waste. In their study, N-Methyl-N-propylpiperidinium bis(trifluoromethylsulfonyl)imide was used as an ionic liquid at 340 °C for 2 min to achieve a maximum chemical yield of 80%. The author mentioned that using microwaves resulted in a high fibre yield in a short process time.

Among the recent studies, Sun et al. [109] investigated electrochemical recycling (ECR) using waste CFRP as an anode and a stainless–steel plate (SSP) as a cathode, with a NaCl solution as the electrolyte. A voltage of 2.6 V was passed across the electrodes for 21 days to soften the CF with a 3% concentration of NaCl. After softening, the fibres are washed under ultrasonic and dried for three days in a 50 °C environment. The authors highlighted that the energy consumption for recycling CF using the electrochemical process is 2–10 kWh/Kg, and the energy consumption to manufacture vCF is 55–165 kWh/Kg. Similarly, using ECR, Zhu et al. [110] and Chen et al. [111] both retained mechanical properties similar to vCF; this method is inexpensive and suitable for large-scale application [111]. Recently, a mechanochemical process (MCP) similar to ECR has been tested [112], but limited information is available regarding the rCF quality.

Recent approaches also involve ultrasonics, Das and Varughese [113] investigated a sonochemical recycling process by reacting the CFRP waste with dilute nitric acid and H_2O_2 under ultrasonics at 60 °C, achieving a 95% decomposition ratio. The use of ultrasonic increased the decomposition ratio thrice when compared to the same process without ultrasonics. Similarly, Jiang et al. [114] used ultrasonics so that the waste CFRP could be pre-treatment with a nitric acid solution. The pre-treated CFRP was subjected to Macrogel 400 and a potassium hydroxide (KOH) system at 160 °C for 200 min. The resins are decomposed at a 95 wt% resin removal rate, and the rCF retained a 95% tensile strength compared to vCF. The authors highlighted that the process is highly efficient for recycling thermosetting composite materials.

4.3 Solvolysis using critical conditions

A fluid (solvent) adopts a high ability to diffuse a soluble substance when its critical temperature and pressure has been reached. It also performs new chemical reactions for decomposition and partial oxidation [115]. Solvolysis using supercritical fluids (SCF) is an emerging waste composite recycling technology, with the recycling process applying to both CFRP and GFRP. The two commonly used fluids in their subcritical and supercritical conditions are water and alcohol [82]. Solvolysis using alcohols is focused on recycling waste CFRP to dissolve the resin and to recover CF. However, there are fewer studies involving GFRP waste recycling.

It is easy to achieve a supercritical state with alcohols as opposed to water [116]. Supercritical alcohols possess good recycling capabilities when used with waste polymer composites. Among all the supercritical alcohols, propanol is better than ethanol and methanol. When comparing methanol, ethanol, acetone, and 1-propanol, research showed that methanol has a low mass-transfer rate under subcritical conditions. On the other hand, 1-propanols three atoms of carbon and high solvation capacity performs better than methanol and ethanol [115, 117].

4.3.1 Alcohol at critical condition

In an early study of CF recycling, Piñero-Hernanz, García-Serna et al. [117] investigated the resin elimination percentage of four alcohols: methanol, ethanol, 1-propanol, and acetone. The study resulted in 95% resin degradation under 15 min, with the rCF retaining 85–99% of its tensile strength. Among the four alcohols used, acetone performed better at a lower temperature. At higher temperature (450 °C), ethanol, 1-propanol and acetone had a maximum resin elimination of 78.8 wt%, and methanol had only 60.2 wt%. However, a later study conducted by Okajima et al. [118] showed that supercritical methanol performed much better when compared to previous studies. The recycled CF using methanol had a crack-free surface with only a 9% decrease in tensile strength and a 20% decrease in interfacial strength when compared to vCF. The solvent and process conditions involved in recycling CF are tabulated in Table 4.

In line with their previous research, Okajima et al. [119] studied the effect of eight supercritical solvents—methanol, 1-propanol, 2-propanol, 1-butanol, 2-butanol, tert-butanol, acetone, and methyl ethyl ketone—on dissolving the resin (amine-cured thermosetting epoxy) and recycle CF. Among the eight supercritical solvents, acetone had a higher resin elimination. Moreover, rGF using supercritical acetone obtained 95–99% resin degradation and retained 89% of its strength compared to vGF [120]. The resin decomposition efficiency improved with increasing

Table 4 Alcohols at critical conditions (CC)

References	Year	Solvent	Conditions
Piñero-Hernanz et al. [117]	2008	Methanol, ethanol, 1-propanol, and acetone	250–400 °C, solvent flow rate of 1.1–2.5 kg-alcohol/kg-fibre/min and 0.016–0.50 M alkali catalyst (NaOH, KOH, and CsOH), 15 min
Okajima et al. [118]	2014	Methanol	Thermosetting: 270 °C in a batch reactor 8 Mpa for 90 min
			Thermoplastic: 285 °C in a semi-flow type reactor 8 Mpa for 80 min
Okajima et al. [119]	2017	Acetone	320 °C with molar density 3.64 mol/L after 20 min
Sokoli et al. [120]	2017	Acetone	260–280 °C, 55–60 bar, 1.24–2.1 g/mL of composite/solvent ratio
Okajima and Sako [121]	2019	Acetone	350 °C, 14 Mpa, 60 min, 4.35 mol/L density of acetone
Jiang et al. [115]	2007	Propanol	300 °C, 50 bars and 10 min
Marsh [116]	2009	Propanol	Semi-continuous flow system at 350 °C, KOH catalyst flow of 1.1 kg of alcohol per 1 kg of fibre/min
Jiang et al. [123]	2009	n-Propanol	Semi-continuous flow reactor at 310 °C, 5.2 Mpa
Yan et al. [124]	2014	1-Propanol	320 °C, feedstock ratio 2 g epoxy resin for 0.2 L 1-propanol, 60 min
Yan et al. [125]	2016	1-Propanol	320 °C, 90 min, no catalysist

reaction pressure and acetone density [121]. Similar to Okajima et al. [119], Cheng et al. [122] analysed the degradation capability of different supercritical fluids. The study showed that supercritical n-butanol had the highest resin degradation and retained 98% of tensile strength compared to the original CF. Supercritical acetone holds the second position for resin degradation followed by ethanol and n-propanol. However, at a temperature range between 340 and 360 °C, n-propanol showed a greater capability for resin degradation compared to ethanol. Overall, both supercritical methanol and isopropanol performed poorly in resin degradation.

Among all the alcohols, propanol is a short chain and affordable. CF recycled using propanol achieves a 98% resin removal rate [116]. The rCF have only a 1% decrease in interfacial shear strength and a 5.43% decrease in tensile property compared to vCF [115]. Plus, the rCF offers properties equal to vCF and can be used as its replacement [116]. Like propanol, both n-propanol and 1-propanol also showed promising results. The rCF using n-propanol only shows a 0.3–2% decrease in tensile strength compared to the initial material used [123], whereas the rCF using 1-propanol had the same tensile strength compared to the CF prior to recycling. Also, adding 1 wt% of KOH, along with 1-propanol, enhances the decomposition ratio of the resin and improves the mechanical stability of rCF [124]. Adding catalysis is not necessary, as the rCF using 1-propanol as a solvent retained 90–95% tensile strength compared to CF before recycling without any catalyst addition [125].

4.3.2 Water at critical condition

Unlike solvolysis using alcohols, waste CFRP and GFRP are both capable of recycling using water at CC. Moreover, considering the number of studies, this section is divided into two to separate those based on CF and those based on GF. Recycling waste CFRP and GFRP using water at CC has resulted in a higher resin decomposition rate along with higher mechanical properties.

4.3.2.1 Based on carbon fibre Following their CF recycling studies using alcohols at CC [117], in the same year (2008), Piñero-Hernanz et al. [126] recycled CF using water at its supercritical condition. The study resulted in a resin removal efficiency of 95.4 wt% with only a 2–10% tensile strength decrease of rCF compared to vCF. The addition of alkali catalyst (NaOH) increases the resin removal efficiency [126, 127]. In 2012, when repeating the same approach without any change in the process parameters, Knight et al. [128] achieved a resin decomposition rate of 95.9–99.2 wt% and recycled woven based CF using supercritical water (deionised water). The rCF shows no tensile strength decrease compared to the original composite. However, in studies focused on maximum resin decomposition efficiency, Yuyan et al. [129] achieved the highest resin decomposition rate of 100 wt% by reducing water from a supercritical state to its subcritical state. The rCF only show a 1.8% tensile strength decrease compared to vCF. Similarly, Kim et al. [130] were able to achieve a 99.5% resin removal efficiency.

Contradicting the studies with higher resin decomposition ratio, Bai et al. [131] research concluded that the decomposition ratio increase above 96.5 wt% in recycling CF would decrease the recycled fibre strength, indicating the damage to the fibre surface after complete resin removal. In that study, the authors managed to obtain an 85 wt% decomposition ratio by using supercritical water and adding oxygen as a catalyst. In recycling CF using water at CC, catalysts play an important role in boosting the resin decomposition ratio [126]. To analyse, this phenomenon, Okajima et al. [132] investigated the resin degradation with a catalyst (2.5 wt% potassium carbonate) and without a catalyst using subcritical water as a solvent. The catalyst process had a better outcome with only a 15% tensile strength decrease of rCF compared to vCF. However, recent studies have avoided the use of a catalyst, increasing temperature and pressure to maintain higher process efficiency instead [130, 133]. The studies based on the catalyst are summarised in Table 5.

The process development after achieving a higher decomposition ratio resulted in studies comparing various solvents with water, in which Yildirir et al. [134] compared ethylene glycol (EG)/water mixture to EG at its near-critical condition. The resin removal was 97.6% with EG/water and 92.1% with EG. The rCF had tensile strength similar to vCF. Similarly, Ibarra et al. [135] studied the resin degradation system to compare water and benzyl alcohol at SC and SCC. The authors observed an initial delamination in the system with thin layers of resin-binding the fibres together and stated that the increased reaction resulted in the complete degradation of these thin resin layers to recycle CF. The decomposition ratio achieved was 90% from benzyl alcohol and 80% from water. In the recent scenario, a mixture of acetone/water at CC is used as a solvent, with the results showing a maximum resin elimination of 95% and successfully closing the rCF life-cycle loop [133, 136, 137].

4.3.2.2 Based on glass fibre In the recycling of GF using water at CC as a solvent, the solvents degree of fractionation increased whenever there was a substantial increase in any of the following three conditions: catalyst/solvent ratio, solvent/FRP ratio, and reaction temperature [138]. Similarly, the increase in reaction time resulted in a decrease in the mechanical property of the recovered GF. At-lower reaction temperature, the strength loss in rGF is lower and gradually raised with an increase in reaction temperature. A final

Table 5 The catalyst used to recycle CF

References	Year	Catalyst	Condition
Piñero-Hernanz et al. [126]	2008	KOH (alkali catalyst)	400 °C, 28 Mpa, 0.5 M of KOH
Knight et al. [128]	2012	KOH (alkali catalyst)	410 °C, 28 Mpa for 120 min and 0.5 M KOH catalyst
Yuyan et al. [129]	2009	Sulfuric acid	260 °C, 1 M sulfuric acid, solvent feedstock ratio of 1:5 g/mL for 105 min
Kim et al. [130]	2019	No catalyst	400 °C, 280 Mpa reactor pressure
Bai et al. [131]	2010	Oxygen	440 °C, 30 Mpa for 30 min
Okajima et al. [132]	2011	Potassium carbonate	400 °C, 20 Mpa, 2.5 wt% potassium carbonate for 45 min
Yildirir et al. [134]	2014	Water	400 °C ethylene glycol (EG) near-critical condition glycol/water ratio (mL/mL) of 5
Keith et al. [133]	2019	No catalyst	320–380 °C, 20–30 Mpa for 120–150 min
Keith et al. [136]	2019	0.05 M $ZnCl_2$ and $MgCl_2$/0.005 M $AlCl_3$	300 °C for 45 min

wash to the recycled GF with organic solvents is required to improve the feasibility of the process [139], as well as to increase the delaminating mechanism in recycling WPCBs to allow for GF recycling. A minor per cent (7 vol%) of water below its critical point can result in higher resin decomposition efficiency [140].

Like any other recycling process for GF, this process also suffered from char formation on the surface of the fibre, due to the gas condensation during hydrolysis. However, initial physicochemical mechanisms such as osmotic cracking, swelling and delaminating can be used to overcome the limitation [141]. In addition to the limitation, the resin structure of the waste GFRP is capable of influencing the rate of hydrolysis, and the hydrolysis mechanism tends to become unstable with varying results [142]. To control the hydrolysis mechanism, Liu et al. [25] used H_2O_2 (9.04 mL)/NaOH (0.21 g) along with supercritical water. The process resulted in 95.14% resin decomposition efficiency with stable hydrolysis throughout the process.

A recent study by Sokoli et al. [120] showed a contradictory use of water as a solvent for recycling GF below its critical point. The author explains that water reacts with alkali oxides on the GF surface and produces micro-cracks. The rGF had a 50–65% strength loss compared to vGF. Findings also showed that supercritical acetone is capable of completely degrading resins at 260 °C with 60 bar and c/s ratio up to 2.1 g/mL. The rGF retained 89% of its tensile strength in comparison to vGF.

5 Life-cycle analysis

Compared to CF, GF has contributed to the most global fibre production, which means the percentage of GFRP waste for recycling is higher compared to the percentage of CFRP waste. Surprisingly, studies have focused more on recycling CFRP waste due to its expensive price range and ability to retain maximum mechanical properties after recycling [7, 143]. A recent study by Hermansson et al. [144] suggested that, in the future, replacing polyacrylonitrile (PAN) with lignin as a raw material for CF will result in lowering the energy use and environmental impact at the time of recycling the waste CFRP.

However, looking at the existing CFRP waste recycling methods, the approaches can be divided into two major techniques. A cheap option; namely landfilling and incineration (approach 1), in which the fibres are not recovered and cause a high negative impact on the environment, including an economic loss in neglecting to reuse the valuable fibres. The second type (approach 2) is a profitable and fibre recovery method such as mechanical, chemical and thermal recycling [4, 48]. Even though the second approach needs capital investment and specific technologies to preserve sustainability, it has a lower environmental impact with a maximum recovery rate of fibres from waste composite. The studies related to the second approach can also be implemented to recycle GFRP waste, as both forms of waste (CFRP and GFRP) involve similar polymer structures [48].

LCA studies have emerged from comparing various recycling methods. Among all the LCA indicators, three examples, namely global warming potential (GWP), human toxicity (USETox) and acidification (AE), are identified as essential [145]. However, a GWP indicator is directly associated with environmental change. It was also noted that the cost of recycling and GWP impacts are inversely proportional to each other. When the GWP decreased, so did the cost [146]. In an attempt to analyse the relationship between GWP and cost, Dong et al. [48] studied the waste management of CFRP concerning economic and environmental aspects. The study highlighted that none of the recycling techniques were able to reduce both the recycling cost and GWP impacts simultaneously.

5.1 Alternative to landfilling and incineration

Industrial-scale recycling highlights that the energy (0.27–2.03 MJ/kg) required to recycle CF by mechanical recycling is lower compared to the energy required (183–286 MJ/kg) to manufacture vCF. Also, mechanical recycling via the milling process has a low environmental impact compared to vCF manufacturing [147]. In addition to this, when comparing landfilling and incineration with mechanical recycling methods, landfilling suffers from several taxations and incineration has a high environmental impact due to its massive carbon emissions. The carbon footprint in mechanical recycling is low compared to landfilling and incineration and, even though mechanical recycling involves high recycling cost with low revenue, targeting a higher rCF market can improve its revenue [11]. Moreover, to have an immediate rGF solution from wind turbine blades [9] and EoL WPCBs [8] mechanical recycling can be adapted, which will reduce 90% of the landfilling net impact [9].

Recent studies have attempted to compare pyrolysis [10, 14], chemical recycling [12], water at CC [10, 13] and EDF [10] with landfilling and incineration. These studies have chiefly focused on showcasing the environmental and economic values of using recycling methods involving fibre recovery. The LCA indicators strongly support pyrolysis over landfilling and incineration. In particular, the low environmental impact and low energy consumption to rCF, which even supports rCF over vCF [10, 14]. Results from the ReCiPe midpoint method supports chemical recycling over landfilling, with an average of 80% in all possible LCA indicators [12]. Results from CML-IA baseline and ILCD 2011 midpoint LCA methods show that recycling CFRP under landfilling and incineration possesses a 25–30% environmental impact and 25% additional energy consumption compared to recycling using water at CC [13].

5.2 Pyrolysis and chemical recycling process

Further recycling process development focusing on fibre recovery leads to comparisons of environmental and economic aspects within the approach to identify various recycling methods that support sustainability. In 2010, to replace train car bodies in South Korea with CFRP, Lee et al. [85] investigated the identification of environmentally-friendly recycling methods and compared chemical recycling with pyrolysis. The study concluded that the energy footprints of chemical recycling (7.62 MJ-eq) are six times less compared to pyrolysis (47.88 MJ-eq), and the greenhouse gas (GHG) emissions of chemical recycling (1196.22 g CO_2-eq) was five times less (5916.08 g CO_2-eq). Overall, these findings support chemical recycling over pyrolysis.

Similarly, results from an LCA study of La Rosa et al. [148] was favourable for recycling CF using chemical recycling, which involved less energy consumption and lowered environmental impacts compared to manufacturing vCF. The study stated that reducing material and energy consumption during product manufacturing can be a game changer for environmental benefits. The EoL wind turbine blades are also favourable for the chemical recycling process. However, if the recyclate value drops to 47% or processing energy increases above 35 MJ/kg, then using chemical recycling is worthless [9].

5.3 Pyrolysis and solvolysis using SCF

In contrast to Lee et al. [85], a Khalil [149] 2018 study used GaBi LCA software and showed opposite results, in which conventional thermolysis via pyrolysis and solvolysis using supercritical water (SCW) was compared. The study showed that the solvolysis process had a 78 times greater human health impact, 76 times greater ecotoxicity, 17 times greater carbon footprint (global warming) and 3 times greater ozone depletion when compared to pyrolysis. Also, the quantitative evidence proves that CFRP recycling via pyrolysis had the advantage of positive environmental and human health value compared to solvolysis using SCW.

Furthermore, Khalil [150] analysed different solvolysis methods using GaBi LCA software. The study covered supercritical fluids such as water, methanol, ethanol, 1-butanol, 1-propanol, acetone, ethylene glycol and binary mixtures of solvents and water. The results showed that binary mixtures of solvent and water were capable of recycling quality CF and had lower production costs, environmental and human health impacts compared to the use of pure solvents at CC. Plus, this could be a promising method for closing the life-cycle loop of CFRP waste.

5.4 Fluidised bed process

Several LCA studies on FBP by Meng et al. [16, 151–153], in 2017 and 2018, resulted in a better understanding of the process. The studies showed that replacing vCF with rCF can reduce global warming potential (GWP) by 33–51% and primary energy demand (PED) by 32–50% [153]. Similarly, replacing traditional materials (steel and aluminium) with rCF in the automotive (components) application showed potential improvement and environmental benefits [152]. As well as the environmental benefits, a hypothetical commercial-scale FBP design with conditions: 100–6000 t/y plant capacity and 3–12 kg/h m^2 feed rate can recover CFs at a rate of 5 USD/Kg, which means the recycling costs of CF are 15% of the overall vCF manufacturing cost. Therefore, it is clear that reusing has a positive environmental outcome along with economic profit [151]. Finally, when comparing the FBP to pyrolysis and chemical recycling using LCA indicators, FBP has a lower PED, GWP and power consumption. Overall, a significant amount of environmental and cost-benefits can be achieved by adapting FBP [16].

6 Discussion

In the current scenario, waste has become a valuable raw material. Based on the literature analysis of rCF and rGF, the scientific community and composite industries are highly focused on recycling techniques to recover fibres completely. Government policies play a significant role in such a drastic change. Strategies such as implementing a hefty tax on composite industries for environmental degradation, the EU policy towards commercial EoL components, economic-based calculations over composite waste and quantity-based estimations over composite waste from the US and the UK, are guiding the composite market to close the life-cycle loop of waste fibres through sustainable recycling. Additionally, EoL composite waste based on applications of the aeronautical/-aerospace sector, the renewable energy sector, and the automotive sector is enormous in volume. These cumulating wastes should be recycled in an industrial-scale.

There is a considerable need to determine such an industrial-scale process and further develop it into a sustainable method to achieve a higher yield in fibre recovery. This can be gradually achieved by briefly analysing the previous studies. In which, economic-based literature proposes that the energy consumption in fibre recycling using any recycling technique is lower when compared to the energy required to manufacture virgin fibres. The profitable selling price for both the recycled CF and GF with only a minor compromise in the quality of the fibres can compete with the expensive virgin fibres available in the market. Supporting this statement, Recycling-based literature proposes that replacing virgin fibres with recycled fibres is highly possible in various applications. On the other hand, environmental-based literature takes a controversial approach in stating that environmental values are inversely proportional to recycling industry profit margins. Overall, considering recycled fibres as an alternative to virgin fibres is a sustainable way to manage the cumulating CFRP and GFRP waste.

When considering the reusability of recycled fibres in various applications, the recycled fibres are either used directly or reinforced with a minimal percentage of virgin fibres (hybrid composite) to negotiate minor strength loss. These scenarios are entirely dependent on the end application and the commercial possibilities of a composite market. Recent studies focus on applications that function based on a complete replacement of virgin fibres by recycled fibres. The outcomings will benefit the growth of the recycling market and reduce the exponential growth in tonnes of EoL waste.

Overall, specific research gaps have been identified as a result of this study.

1. Alternative approaches mentioned in mechanical recycling and chemical recycling, such as recycling using high voltage and electricity. Further research in these studies can lead to cost-effective recycling processes in short time intervals, along with a low environmental impact. It can also be used as a pre-recycling process for initial separation during commercial-scale recycling.
2. In the thermal-based recycling process, using microwaves as an alternative source of heat reduces both the energy consumption and the recycling time in half compared to traditional heat sources available. Further research in the field can avoid unwanted heat-loss in industrial-scale recycling, considering huge structures to be recycled such as aeroplane and windmill parts.
3. Extensive researches are needed to transforming laboratory-scale solvolysis using water at CC (solvent) and binary solvent mixture (water + solvent) at CC into a fully functional commercial scale. This will benefit the recycling industries.
4. LCA studies contribute to major factors in understanding the pros and cons of the various recycling process, further studies to support the circular economy to show re-manufacturing as a significant player in closing the life-cycle loop of the recycled fibres are required. After all, reusing the recovered fibres in actual applications is the only possible way to suppress the need to manufacture virgin fibres.

7 Conclusion

The main goal of the current study is to determine various recycling techniques for CFRP and GFRP waste and prioritise the sustainably identified recycling methods based on economic and environmental values. A critical comparison was carried out based on factors such as process conditions, process outcomes, mechanical properties, ease of reuse, environmental impact and cost-effectiveness. However, the research has been narrowed down further to focus on completely recovering the valuable fibres by adapting recycling techniques such as mechanical recycling, thermal recycling, and chemical recycling. Plus their advantages and disadvantages were discussed briefly. Furthermore, to support the credibility of these techniques, LCA studies were included in comparing the environmental and economic aspects of the recycling techniques. Overall, this study promotes a circular economic approach to close the life-cycle loop of both CF and GF composite wastes into re-manufactured composites.

Currently, the fate of scarp and EoL CFRP and GFRP is mostly to become landfill or to be sent for incineration, being the methods adopted by conventional waste disposal industries. However, taking on board factors such as climate change, global warming, and a sustainable alternative and circular economy, waste disposal industries have recently shifted to complete fibre recovery methods. This change is occurring due to the immense contribution of studies focusing on recycling CFRP and GFRP waste. These studies are briefly discussed in this review article, and the outcomes are summarised below.

1. Landfilling and incineration methods of disposing of CFRP and GFRP waste are not sustainable approaches anymore and have to be completely shut-down without any further analysis.
2. Mechanical recycling techniques have reached their maximum exploration, losing status as a primary recycling method and becoming a pre-recycling process for other techniques such as thermal and chemical recycling. Also, alternative approaches to breaking down the waste composite using high voltage are becoming more attractive than traditional mechanical recycling.
3. Commercial-scale FBP is capable of recycling clean and high-quality fibres with a fraction of the energy consumption needed to manufacture virgin fibres. Also, the process has a low environmental impact and a decent commercial-scale profit margin. However, the fibres are fluffy and discontinuous, limiting attitudes toward ease of reuse.
4. Commercial-scale pyrolysis has been successfully adopted in Germany and the UK. The process implements green values by recycling fibres along with products such as gas and liquid, which can be further used as feedstocks. However, char formation in rGFs and retaining the complete mechanical properties of both CF and GF are challenging. Besides, the recycled fibres need a secondary heat/chemical treatment to eliminate minor resin impurities.
5. Traditional chemical recycling using strong acids or solvents at various conditions has severe environmental impacts. Even though the process is capable of recycling high quality and clean fibres with a crack-free surface and low energy usage, the disposal of such strong solvents is challenging. However, alternative approaches such as electrochemical recycling look promising but are still at the laboratory stage.
6. A chemical recycling process using solvents such as water at CC and a binary fraction of mild solvents with water at CC is considered to be the future of recycling both CFRP and GFRP waste. The maximum resin elimination ratio, the higher retention of mechanical properties in recycled fibres and the use of cheap and sustainable solvents make the process distinct from any other recycling process. However, the process is not yet commercialised, and critical operating conditions tend to consume additional energy.

The future aims of our research team are to study both recycled CFRP and GFRP from industrial waste. The research will be focused on analysing the recyclability of the recovered fibre, on the mechanical properties after reuse, on the possible applications to implement the remanufactured composite and, finally, on an analysis of how to close its life-cycle loop. The composite wastes will be recycled by thermolysis in a pyrolytic reactor. Also, compression moulding method will be adopted to re-manufacture the covered fibres with polymers.

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Author's contribution Both the authors contributed to the study conception and design. Material preparation, data collection and manuscript writing were performed by Sankar Karuppannan Gopalraj. Data selection and structuring were performed by Timo Kärki. Comments and corrections from Timo Kärki are incorporated with all previous versions of the manuscript written by Sankar Karuppannan Gopalraj. This final draft of the manuscript has been read and approved by both the authors without any conflict of interest.

Compliance with ethical standards

Conflict of interest The authors declare that they have no competing interests.

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Publication II

Karuppannan Gopalraj, S., and Kärki, T.

A Study to Investigate the Mechanical Properties of Recycled Carbon Fibre/Glass Fibre-Reinforced Epoxy Composites Using a Novel Thermal Recycling Process

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A Study to Investigate the Mechanical Properties of Recycled Carbon Fibre/Glass Fibre-Reinforced Epoxy Composites Using a Novel Thermal Recycling Process

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Abstract: Manufacturing-based carbon fibre-reinforced polymer (CFRP) and glass fibre-reinforced polymer (GFRP) wastes (pre-consumer waste) were recycled to recover valuable carbon fibres (CFs) and glass fibres (GFs), utilising a novel thermal recycling process with a cone calorimeter setup. The ideal conditions to recycle both the fibres occurred at 550 °C in atmospheric pressure. The processing time in the batch reactor to recycle CFs was 20–25 min, and to recycle GFs it was 25–30 min. The recovery rate of the recycled CFs was 95–98 wt%, and for GFs it was 80–82 wt%. Both the recycled fibres possessed a 100–110 mm average length. The resin phase elimination was verified by employing scanning electron microscopy (SEM). Furthermore, the fibres were manually realigned, compression moulded at room temperature, and cured for 24 h by a laminating epoxy resin system. The newly manufactured CFRP and GFRP composites were continuous (uniform length from end to end), unidirectionally oriented (0°), and non-woven. The composites were produced in two fibre volumes: 40 wt% and 60 wt%. The addition of ≈20 wt% recycled CFs increased the tensile strength (TS) by 12%, young modulus (YM) by 34.27% and impact strength (IS) by 7.26%. The addition of ≈20 wt% recycled GFs increased the TS by 75.14%, YM by 12.23% and the IS by 116.16%. The closed-loop recycling approach demonstrated in this study can effectively recycle both CFRP and GFRP manufacturing wastes. Preserving the structural integrity of the recycled fibres could be an advantage, enabling recycling for a specified number of times.

Keywords: manufacturing waste; carbon fibre; glass fibre; thermal recycling; compression moulding; mechanical properties

1. Introduction

Fibre-reinforced composites (FRCs), popularly known for being lightweight and having extraordinary mechanical properties, include carbon fibre-reinforced composites (CFRCs) and glass fibre-reinforced composites (GFRCs) [1–4]. They have an advantageous low weight to strength ratio; however, the fibres are expensive [1,3]. The presence of such valuable carbon fibres (CFs) and glass fibres (GFs) makes their accumulated FRC wastes into marketable raw materials. After decades of employing them in high-performance applications, industrial progress towards recycling such enormous wastes has opened up new, affordable disposal methods [1].

In the current scenario, achieving the closed-loop recycling (CLR) [5–8] approach is a primary target for recycling industries, leading to significant attention from researchers globally. Typically, for CLR, the valuable fibres from the composite wastes are reclaimed, remanufactured, and reused within a closed-loop (see Figure 1), promoting the circular economy [6]. In the past two decades, various studies [1–4], [9] have widely explored the recycling techniques that can be used to effectively recover fibres (CF and GF) from different waste sources, so that they can be reused into new composites. Modern studies are working on improving the mechanical properties of the recycled composites, on matching those of their virgin form [1].

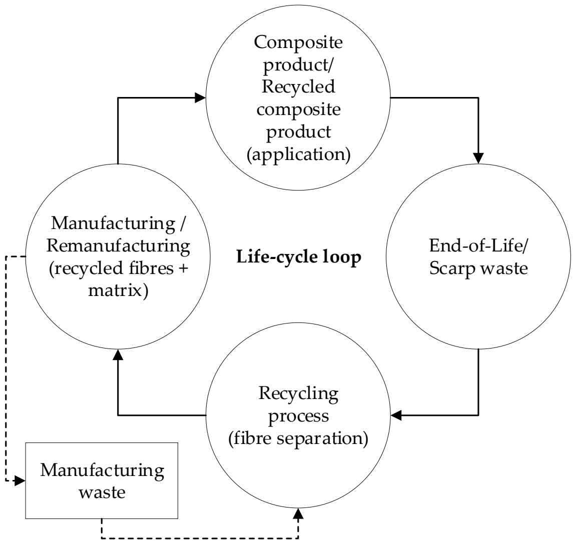

Figure 1. Life-cycle loop of carbon fibre-reinforced polymer (CFRP) and glass fibre-reinforced polymer (GFRP).

Within the enormous quantity of CFRC and GFRC wastes being produced, manufacturing-based waste contributes 40%, among which 60% of the wastes are trimmings and off-cuts [2]. Such wastes (pre-consumer waste) are challenging to recycle due to their irregular profile, dimension, fibre distribution, orientation, matrix phase, and volume. To successfully close their life-cycle loop, it is better to recycle long, continuous, and aligned fibres, so that later they can be recycled a further n-number of times. The thermal recycling process (pyrolysis) appears to be suitable for recycling manufacturing waste. In a typical thermal recycling process, the matrix phase evaporates from the reinforced fibres (core material) because of its higher calorific value [2,3,9,10]. However, this results in recovering discontinuous, short and randomly oriented fibres. Additionally, selecting a single favourable process to recycle both CFRC and GFRC wastes appears to be complicated.

This study attempts to recycle both types of fibres (CF and GF), utilising a single process. A novel thermal recycling approach is used which adopts incineration and combustion principles in a controlled environment. The process employs an open chamber heat radiation in a batch reactor. The recycled carbon fibres (rCFs) and recycled glass fibres (rGFs) are reused into recycled carbon fibre-reinforced epoxy (rCF/EP) composites and recycled glass fibre-reinforced epoxy (rGF/EP) composites. Furthermore, the new composites are investigated to study their mechanical properties. Overall, this study aims to achieve a CLR approach to recycle CFRC and GFRC manufacturing wastes, recover CF and GF fibres, remanufacture rCF and rGF fibres, and examine their mechanical properties.

2. Materials and Methods

This study adopts a two-stage CLR approach. The initial stage consists of thermal recycling, focusing on fibre recovery from the composite wastes. The second stage involves remanufacturing of the recovered fibres into new composites. In addition to this approach, the study primarily focuses on characterising the newly produced composites using standard test procedures to measure their tensile and impact properties. All three methods are briefly outlined in this section. The methodology used in this study is shown in Figure 2.

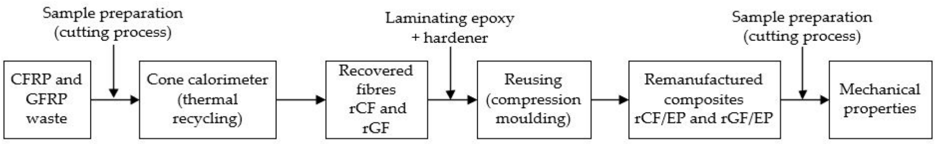

Figure 2. Schematic overview of this study. Abbreviations: CFRP = carbon fibre-reinforced polymer; GFRP = glass fibre-reinforced polymer; rCF = recycled carbon fibre; rGF = recycled glass fibre; rCF/EP = recycled carbon fibre-reinforced epoxy; rGF/EP = recycled glass fibre-reinforced epoxy.

2.1. Fibre Recovery

2.1.1. Materials

The materials used in this research work were pre-consumer wastes (discarded materials). These consisted of trimmings and off-cuts from commercially available carbon fibre-reinforced epoxy composite (CFEC) and glass fibre-reinforced polyester composite (GFPC) and were generously provided by Exel Composite Oyj, Finland (Figure 3). Both composites had a distinctive profile and fibre orientation. The CFECs possessed a C-shaped profile with a density of 1.81 g/cm^3. They featured a unidirectional (UD) fibre orientation (0°) with 55.5 wt% virgin carbon fibres (vCFs) (type unknown) and 44.5 wt% epoxy resin matrix. On the other hand, the GFPCs were rectangular plates with thicknesses ranging between 1.60 and 2.30 mm. They had a profile density of 1.52 g/cm^3 and featured a laminated thin-ply profile with UD (90°) fibre oriented virgin glass fibres (vGFs) in the middle enclosed by randomly oriented (chopped strand mat) vGFs on both sides. The composite contained 44 wt% vGFs (E-glass) reinforced with 56 wt% polyester as the matrix. Apart from the properties mentioned above, other properties were unknown.

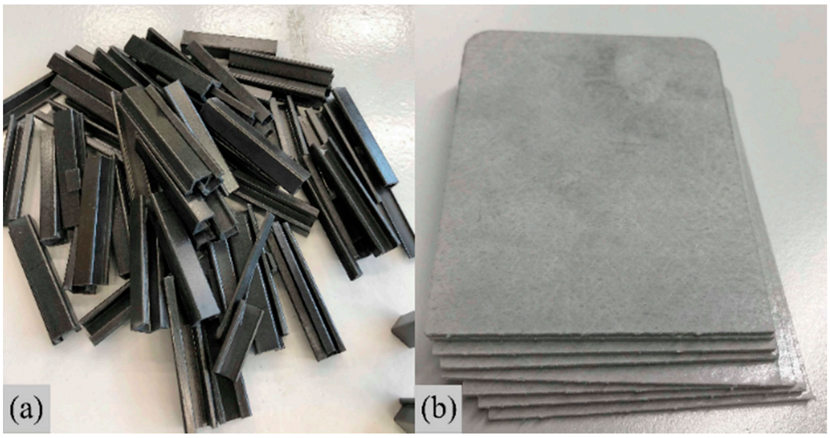

Figure 3. Discarded composite waste: (**a**) carbon fibre-reinforced epoxy composite (CFEC) and (**b**) glass fibre-reinforced polyester composite (GFPC).

2.1.2. Process Setup

A cone calorimeter (CC) setup was utilised for recycling the waste composites. Figure 4 presents a schematic arrangement of the CC. The setup consists of a load cell to measure the mass change, a material holder, a conical radiant heater with a heating coil to generate heat flux, and an exhaust duct to remove smoke throughout the process.

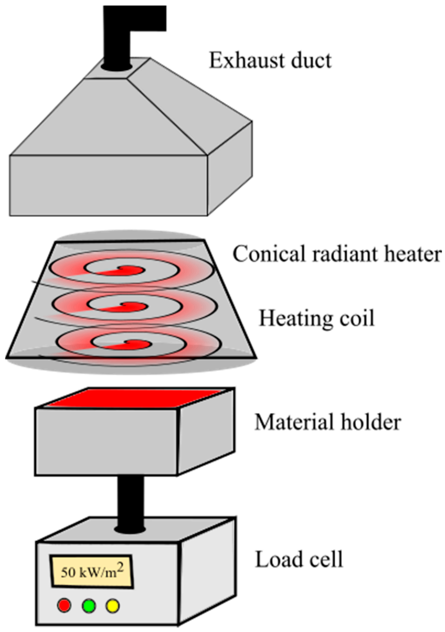

Figure 4. Fibre recovery experimental setup.

The recycling process was performed in batches, separately for CFEC and GFPC. Both the composite wastes were prepared to fit the material holder's (refractory) internal dimensions—110 (l) × 110 (b) × 10–20 (h) mm. A series of experiments were trialed before finalising the isothermal process condition. The temperature for cured epoxy to fully evaporate was in the range 450–600 °C [11,12], and for thermoset polyester, it was in the range 400–700 °C [11]. The optimal temperature for both the materials was fixed based on the processing time. Apparently, the temperature and process duration were inversely proportional to each other. Therefore, various possibilities were trialed with temperature and time ranges of 550–800 °C and 45–15 min, respectively.

The processing time was calculated between times of inserting and removing the material holder into the heat flux region. Once the materials were introduced to the heat flux chamber, the change in mass over time was recorded in the load cell placed beneath the material holder (as shown in Figure 4). Based on trial experiments, the heat radiation was kept constant at 50 kW/m^2 for both the composites, throughout the actual recycling process. However, the operating period varied for CFECs and GFPCs, depending on their mass.

During the recycling, smoke appeared from the material holder, indicating the start of the process, followed by high flaming. After a noticeable period, the flame extinguished gradually, indicating the end of the process. The generated heat flux at 50 kW/m^2 was equivalent to 750 °C, however, this was the coil's surface temperature. The monitored isothermal surface temperature on the composite wastes was 550 °C. At this temperature and atmospheric pressure, the matrix phase evaporated entirely, leaving the fibres in the material holder. The respective mass change over time was recorded using ConeCalc software. The recycled fibres (rFs) were carefully removed from the material holder and prepared for the remanufacturing stage.

2.2. Remanufacturing

2.2.1. Fibre Alignment

The existing composite structure in the waste CFECs was unidirectionally aligned non-woven CFs. The recycling process only removed the epoxy matrix phase, without interrupting the fibres' UD arrangement. Similarly, in the case of GFPCs, heat removed the polyester matrix phase, leaving the GFs. The rGFs consisted of 60 wt% unidirectionally aligned non-woven fibres, and 40 wt% randomly oriented fibres, which were manually aligned later. Highly aligned and continuous fibres mean the composite is capable of possessing a higher fibre volume fraction, of up to 60 wt% [11]. Continuous fibres mean the fibres are extended from one end of the sample to the other end without any discontinuity in the fibre system [13]. The rFs of both the rCF and the rGF possessed an average fibre length of 100–110 mm. They were continuously aligned and unidirectionally oriented (0°), meaning that they were ready to be reused.

2.2.2. Composite Production

Table 1 presents information about the different types of composites which were designed to be manufactured. The designed composites consisted of two primary composites (PCs) and one secondary composite (SC). The first PC was manufactured utilising rCF reinforced with an epoxy matrix (rCF/EP) and the second PC utilised rGF reinforced with an epoxy matrix (rGF/EP). Furthermore, both the PCs were manufactured using two recipes (as seen in Table 1), based on their fibre volume (V^f) and resin volume (V^r). The SC was manufactured with pure epoxy resin (P-EP), with no fibres, to evaluate the remanufacturing process.

Table 1. Designed composites for compression moulding.

Composite Recipes	Types of Composite	V^f (wt%)	V^r (wt%)	Density (ρ) (g/cm^3)	Fibre Length (mm)
rCF/EP	2	60 ± 2	40 ± 2	1.52	105 ± 2
		40 ± 2	60 ± 2	1.64	105 ± 2
rGF/EP	2	60 ± 2	40 ± 2	1.77	105 ± 2
		40 ± 2	60 ± 2	1.85	105 ± 2
P-EP	1	0	100	1.45	-

Abbreviations: V^f = fibre volume, V^f = resin volume, rCF/EP = recycled carbon fibre-reinforced epoxy, rGF/EP = recycled glass fibre-reinforced epoxy; P-EP = pure epoxy resin.

The compression moulding (CM) technique was adopted to remanufacture the designed composites. Both rCFs and rGFs were reused by reinforcing them with a new epoxy resin system. Commercially available laminating epoxy + hardener (EP) with a combined density of 1.5 g/cm^3 was used in a 2:1 resin to hardener ratio. The EP system was easy to handle, with an operating time of up to 20 min and 8 h of curing time at room temperature.

A dismantlable plywood mould was custom-made to manufacture the designed composites. To maintain uniformity, the inner dimensions of the plywood mould (110 × 110 × 10 mm) were similar to those of the material holder dimension from the CC. The inner-surfaces of the mould were waxed (3–5 wax layers) for the compression moulded composites (CMCs) to be able to slip easily. The hand layout method was adapted to align the rF bundles layer-by-layer (LBL), with EP manually poured between each layer (Figure 5).

Figure 5 presents the LBL CM arrangement of the recycled fibres and epoxy system. Higher fibre volume fractions required higher moulding pressure. However, aligned and continuous fibres demanded < 1 MPa moulding pressure [11]. Therefore, the CM pressure was calculated based on the mould position. The composites were designed to be manufactured into 110 × 110 × 10 mm dimensions. Therefore, respective positions, based on volume, were pre-marked on the plywood mould (seen in Figure 5). The density–volume relation was used to calculate the mass of the designed composites.

The compression was applied until the position markers aligned into a straight line, and the pressure was maintained until the composites cured entirely. The CMCs was cured at room temperature for 24 h. Overall, five composites for each recipe (25 composites in total) were produced, and their average densities were measured (see Table 1).

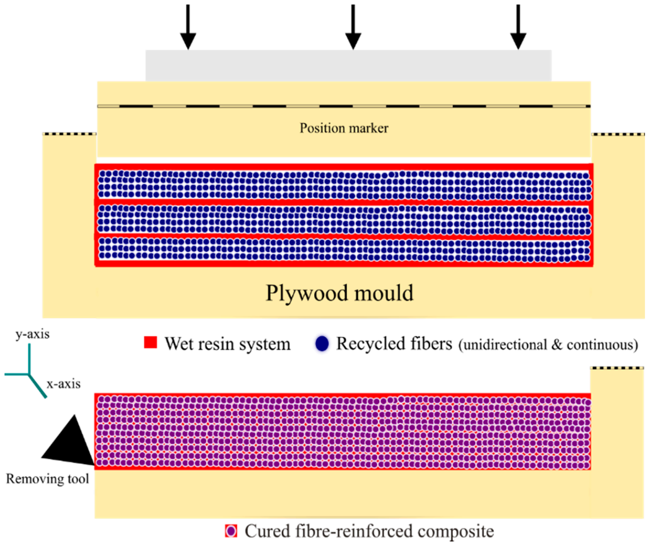

Figure 5. Compression moulding arrangement.

2.3. *Composite Characterisation*

The compression moulded square samples were thoroughly cleaned using a dry cloth to eliminate minor wax residue from their exterior surface. They were further dried at +23 °C and 50% relative humidity (RH) for 24 h, before being subjected to further tests. ISO standard procedures were adopted to prepare the test samples.

2.3.1. Tensile Strength

ISO 527-2 [14] was followed to measure the tensile strength (TS) and young modulus (YM) of the CMCs. The samples were prepared according to small specimens, type 1BA. Initially, utilising an electric circular saw system with water as a coolant, rectangular outer profiles were made. Later, dogbone shaped patterns were precisely cut using an automatic cutting machine. A minimum of 15 samples for each composite recipe (overall five types, as seen in Table 1) were prepared and subjected to a 24 h rest period at +23 °C and 50% RH before the actual testing. The experiments were performed using a Zwick Roell (Z020) tester at a test speed of 2 mm/min. With the guidance of an extensometer, the experimental data were collected and processed automatically using testXpert II software.

2.3.2. Impact Strength

ISO 179-1 [15] was followed to measure the impact strength (IS) of the CMCs. The samples were prepared according to type 1 test specimens—80 ± 2(l) × 10 ± 0.2(b) × 4 ± 0.2(h) mm—under charpy unnotched impact strength, utilising an electric circular saw system with water as a coolant. A minimum of 15 samples for each type of composite were prepared and subjected to a 24 h rest period at +23 °C and 50% RH before the actual testing. The impact energy observed in breaking or in some cases damaging the samples was measured in kJ/m^2. The experiments were performed using a pendulum impact tester with a hammer size of 40 Kpcm for rCF/EP composites and 10/20 Kpcm for rGF/EP and P-EP composites.

2.3.3. Microscopy

The rFs (both rCF and rGF) and recycled FRCs (both rCF/EP and rGF/EP) were examined employing scanning electron microscopy (SEM) to investigate their morphologies. The examination was conducted using a Jeol JSM-5800 LV scanning microscope at various magnification. The selected materials were prepared and dried in a vacuum chamber for 24 h before testing. The fibre diameters (Φ^f) were measured at 4k magnification. The images of fibre alignment and tensile pull-outs were captured at 500, 1k, 2k and 3k magnification.

3. Results and Discussion

3.1. Thermal Recycling Approach

3.1.1. Process Outcome

Figure 6 presents the data collected during the isothermal process, analysing the relationship between mass stagnant (g) and time (min). As observed, smoking started immediately after introducing the composites to the heat radiation. Without any externally induced sparking, flames appeared subsequently, indicating the start of the combustion process within the material holder. During the process, the matrix phase for both the composite wastes (CFEC and GFPC) rapidly oxidised and evaporated in 5–10 min, followed by the flame turning-off, indicating the end of combustion. This process was marked as the combustion zone (CZ, Figure 6), and left the fibres untouched in the material holder. The smoke started initially after 15–20 s for CFECs and 10–15 s for GFPCs. For both the materials, flames started and stopped inside the marked CZ in which the entire recycling process took place. The overall process was terminated once the mass change became stagnant. The processing time to recycle CFECs was 20–25 min, and for GFPCs it was 25–30 min.

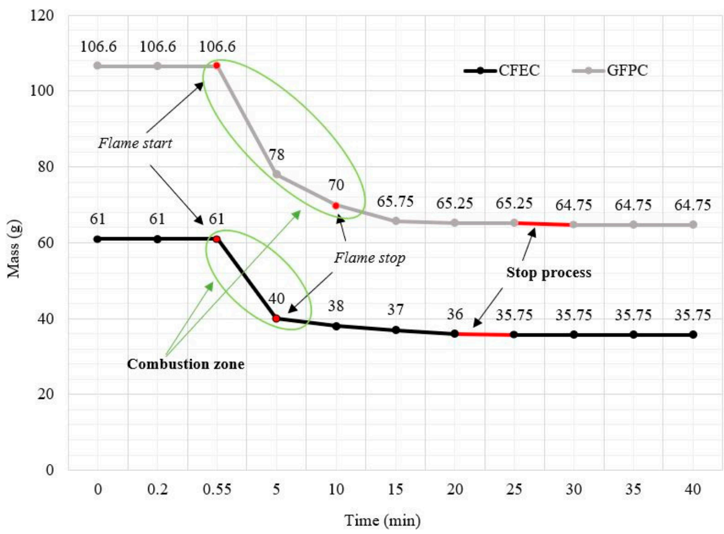

Figure 6. The relationship between mass (g) and time (min).

In a typical thermal recycling process, CF cannot be oxidised at temperatures <600 °C [16]. However, they become sensitive at higher temperatures >600 °C in the presence of air, making it easy to eliminate the resin phase from the reinforced-fibres rapidly, in a short period [17]. In GF recycling, temperatures >650 °C result in a massive drop in the mechanical properties of the rFs [18]. In this process, even though the generated heat flux at 50 kW/m^2 was equivalent to 750 °C, the monitored heat on the surface of the composite wastes during recycling was 550 °C. At this optimal temperature, the recycling process was rapid for both fibre types, and the maximum amount of fibres were able to be recovered, without any significant losses. Overall, this approach successfully combusted both the thermoset (epoxy and polyester) matrixes from the received manufacturing wastes.

3.1.2. Fibre Yield

Unlike typical thermal recycling processes, this process is capable of recycling bulk quantities. For a single batch, 35 ± 2 g (final mass) rCFs was obtained from 60 ± 2 g (initial mass) of CFEC wastes, and 65 ± 2 g (final mass) rGFs was obtained from 105 ± 2 g (initial mass) of GFPC wastes. The overall recovery rate of the process was 95–98% CFs recovered with 2–5% residue (negotiable) and 80–82% GFs recovered with 16–18% residue (char formation) + 2% ash (top surface).

The recycling process utilised in this study to thermally disintegrate the matrix possesses similar interpretations to the frequently used thermogravimetric analysis (TGA) approach. However, this process produces a higher yield. Additionally, the process can be up-scaled to any specific degree without limitations. By just increasing the area of the heating coil, recycling of longer and more intricate shapes can be achieved, without disturbing their structural integrity.

3.1.3. Fibre Evaluation

Figure 7 presents the SEM image of the rFs. The rCFs showed no evidence of any presence of residues (Figure 7a). The epoxy resin tends to leave no residue behind after the thermal process [11]. In addition, the higher surface purity results in better impregnation with new epoxy resin [19]. The average measured diameter (Φ^f) of the rCFs was 6.5 ± 0.2 µm. Apart from the SEM examination, the fibres visually looked fluffy and difficult to handle (realign).

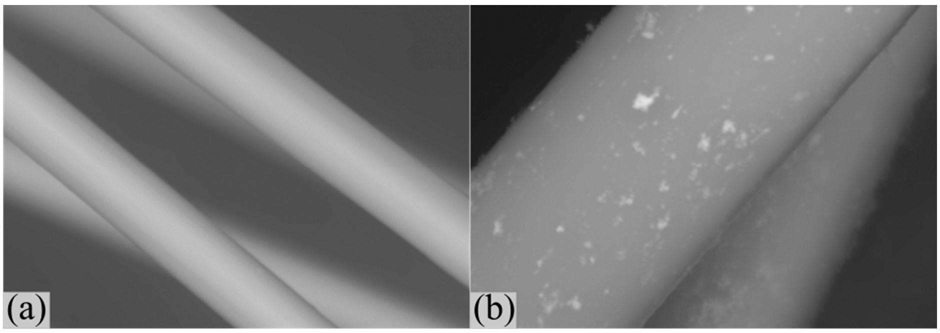

Figure 7. SEM images of (**a**) rCFs and (**b**) rGFs at 4k magnification.

Unlike rCFs, rGFs had evidence of impurities, including resin residues and minor char deposits on the fibre surface (Figure 7b). As mentioned previously, 18–16% of the rGFs contained residue from the recycling process. The average measured diameter (Φ^f) of the rGFs was 20.5 ± 0.25 µm. The handling of the fibres appeared to be easier compared to rCFs.

3.2. Mechanical Properties

Table 2 presents the results of the mechanical properties of the CMCs. An increase of ≈20 wt% fibre volume positively increased the mechanical properties of both the rCF/EP: TS (12%), YM (34.27%) and IS (7.26%) and the rGF/EP: TS (75.14%), YM (12.23%) and IS (116.16%) composites.

Table 2. Measured values of the remanufactured composites.

Type	Composition	Tensile Strength (MPa)	Young Modulus (GPa)	Impact Strength (KJ/m^2)
rCF/EP	40 wt%–rCF	210.34	45.28	49.98
	60 wt%–rCF	235.70	60.80	53.61
rGF/EP	40 wt%–rGF	65.42	27.37	18.99
	60 wt%–rGF	114.58	30.72	41.05
P-EP	100 wt%–EP	39.46	2.16	35.18

3.2.1. Tensile Properties

Figure 8 presents the measured average values of the TS and YM. The error bars display the calculated standard deviation for each composite type, and the obtained stress–strain curves appeared to be linear. The YM of all the tested FRCs exhibited a higher value range than their associated TS (seen in Table 2). The reason for this outcome is due to the absence of fibre discontinuity in the composites. The aligned continuous fibres increased their elastic region, resulting in stiffer composites.

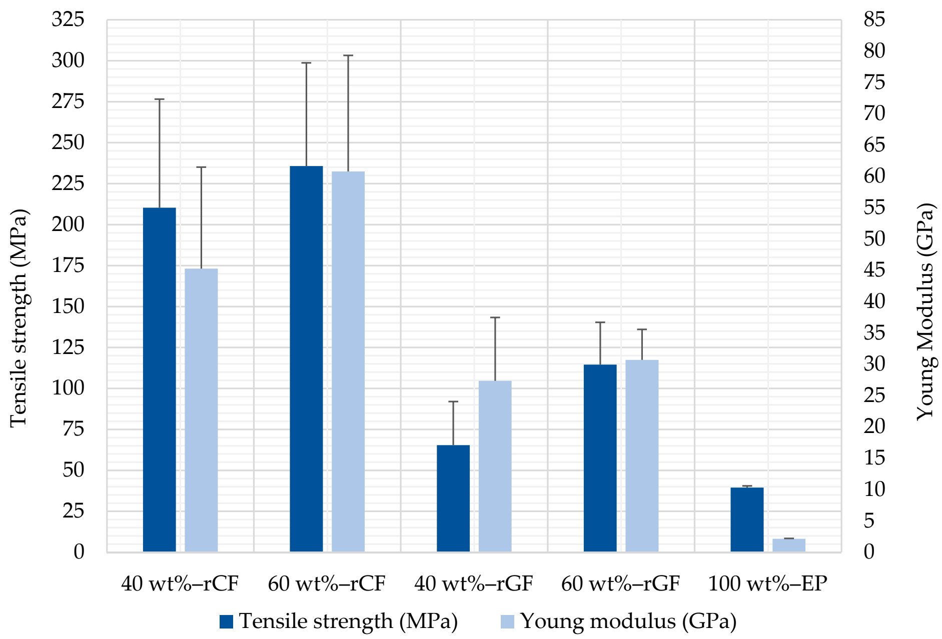

Figure 8. Tensile properties of the tested composites.

In rCF/EP composites, the increase in fibre volume (≈20 wt%) increased the YM, making the composites extra stiff, with a minor increase in the TS. During the tests, most of the rCF/EP composite samples failed in their midsection. However, a couple of cases exhibited cracking parallel to the tensile direction. With further investigation, an absence of a resin system was identified across the fibre bundles, causing the layers to slip with a minimal applied load. Such poorly wet fibres appeared fluffy, and can be related to a low electrostatic attraction between individual fibres. Two samples were excluded for exhibiting low wettability. Both the rCF/EP type composites (40 and 60 wt%) displayed fibre-dominated failure [20], confirming the failures occurred in the interface between the fibre and epoxy system. Overall, the rCF/EP composites were notably durable compared to the rest of the composites, without much variation in testing behaviour between the composite types.

In rGF/EP composites, the increase in fibre volume (≈20 wt%) essentially doubled the TS of the composites, with a negligible change to the YM. During the tests, all the samples failed in their midsection perpendicular to the direction of the tensile force, with no evidence for poorly wetted rGFs. The applied heat increased the rGF density, resulting in a much stiffer fibre and reduced ductility [21]. Both the rGF/EP type composites (40 and 60 wt%) displayed a highly brittle nature and resulted in brittle failure. Crack propagations were observed in both the fibre and matrix cross-sections. In addition, the tested samples showed multiple microbreaks and individual broken fibres distributed in various spots (Figure 9h).

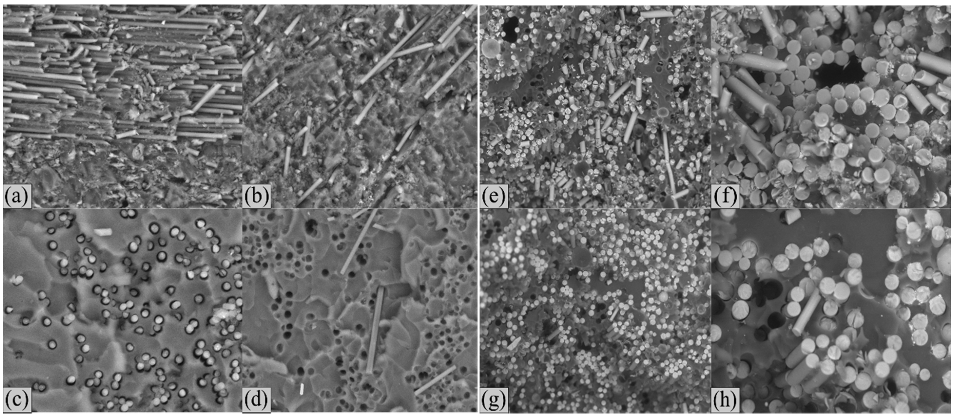

Figure 9. SEM images of (**a**–**d**) rCF/EP composites and (**e**–**h**) rGF/EP composites after tensile testing.

Figure 9 presents the SEM images of the tensile tested samples. Both rCF/EP and rGF/EP composites show evidence for defibrillation [22] and pull-out type failure. The rCF/EP composite sample pull-outs, associated with 60 wt% (Figure 9c,d) appear evenly distributed, with a dense fibre arrangement compared to the 40 wt% (Figure 9a,b) samples. Compared to rCF/EP composites, rGF/EP composites appeared to be extra brittle, with a three times higher fibre diameter and stable fibre-matrix interface (Figure 9e–h).

The P-EP composites appear to have a reasonable tensile strength to modulus proportion. Additionally, their impact strength was slightly lower compared to the 60 wt% rGF/EP composite. The composites appeared to be cured entirely, with no evidence of porosity. The tensile properties of the tested samples possessed a minimal coefficient of variation (CoV).

3.2.2. Impact Properties

Figure 10 presents the average values from the measured unnotched charpy impact test. The error bars display the calculated standard deviation for each sample type. The increase in ≈20 wt% fibre volume resulted in a 3.63 kJ/m^2 strength increase for the rCF/EP and 22.06 kJ/m^2 for the rGF/EP composites. The increase in the fibre fraction had a significant influence on the rGF/EP composites compared to the rCF/EP composites, in which the strength increase was negotiable. This variation in impact strength could be related to the size effect on the energy dissipation. Typically, a rougher microstructure results in composites with higher impact strength [23]. Another well-known reason is the interfacial strength between the fibre-matrix interface, in which a lower interfacial strength results in a higher impact strength [24]. The additional fibres made the rGF/EP composites tougher. However, this was absent in the rCF/EP composites. This could be due to a lack of fibres that were thoroughly wet by the resin system. Even though the thermal recycling of GF results in an overall decrease in mechanical strength, it does not degrade their stiffness properties after recycling [18].

3.3. Composite Investigation

3.3.1. Tested Composites

Figure 11 represents the defects observed in the CMCs. The remanufacturability of both the rCF/EP and rGF/EP composites was investigated using SEM after the mechanical tests. In the rCF/EP composites, boundary layers between fully cured fibres and unwet fibres (Figure 11a,b) and minor voids (Figure 11a) were observed. The boundary between fully cured and unwet fibres explains the manual LBL bundle-based fibre arrangement during the CM process, in which the resin system failed to spread evenly inside the aligned rCF bundles (Figure 11b). Such evidence was also present in the fibre pull-out samples, where the failure layers of the resin system are visible (Figure 11c).

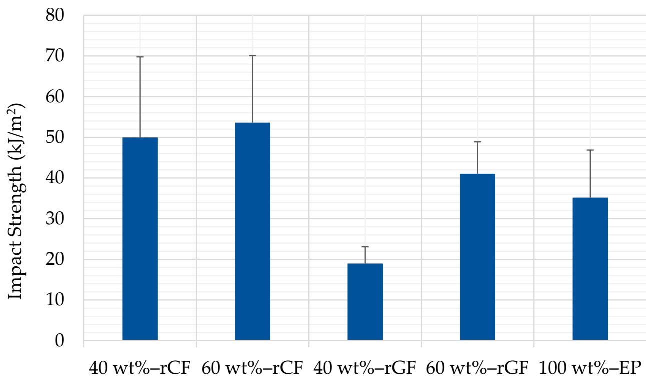

Figure 10. Impact strengths of the tested composites.

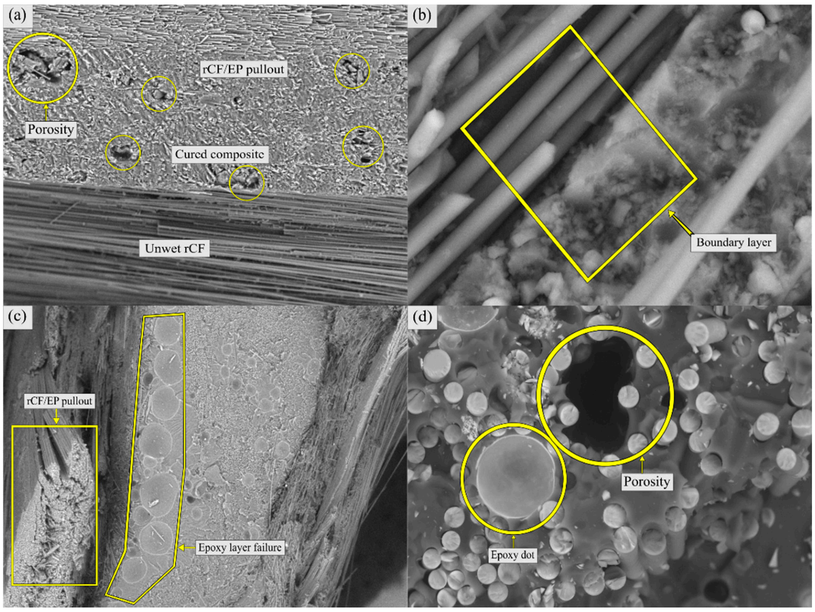

Figure 11. (**a**) Different layers in a single composite sample (60 wt% rCF/EP), (**b**) the boundary layer between un-wet fibres and the fully cured rCF/EP composite, (**c**) minor resin layers present in the rCF/EP composites, and (**d**) the presence of resin dots and porosity in the rGF/EP composite.

In the rGF/EP composites, the remanufactured composites appeared fully cured, without any signs of non-wet fibres. However, the samples showed evidence of microvoids and resin spots (Figure 11d). Utilising a higher heating rate will rapidly evaluate the matrix phase (volatile materials), eventually resulting in the formation of porous structures. Additionally, due to rapid volatilisation, void formation increases as a result of higher pressure [25]. Such evidence for the formation of minor porous structures was present in the SEM images for both the rCF/EP and rGF/EP composites (Figure 11a,d).

3.3.2. rCF/EP Composite

When comparing the measured properties of the rCF/EP composites from this study to similar studies in the literature, the values fall under the related limits. A study that adopted fluidised bed process (FBP), by Shah and Schubel [26], used an aligned non-woven mat of rCF recycled composites with a fibre volume of 27–34 wt%, a TS of 133–174 MPa, and a YM 19–32 GPa. The composites were remanufactured using liquid composite moulding. Similar aligned non-woven fibres with 30 wt% rCF composites were studied by Pimenta et al. [19,22], and had a TS of 194 MPa and a YM of 28 GPa. The study implemented pyrolysis to recycle the CFs, and resin film infusion to remanufacture them. Apart from the aligned composites, the random fibre alignment and discontinuous fibres also had values under those of the related limits. A study by Wong et al. [27] using compression moulding produced composites values of TS of 207 MPa and YM of 25 GPa, with 30 wt% rCFs. Another study by Feraboli et al. [28], with long and random fibre orientation, resulted in a TS of 196.5 MPa and YM of 29 GPa with 33 wt% rCFs.

The SEM images show various noticeable defects in the rCF/EP composites; apart from the visible defects, further investigation was performed to analyse the factors influencing the rCF/EP composite properties. The fibre alignment and length had a significant influence over the mechanical properties of the rCF composites [13]. These two factors can also be related to both the newly manufactured rCF/EP and rGF/EP composites.

The rCFs were manually realigned to produce both the rCF/EP composite types (40 and 60 wt%). Randomly oriented fibres lead to a non-homogenous outcome [26]. With a higher fibre content (60 wt% rCF) and continuous UD composites, even a minor fibre misalignment by 10° will result in a strength loss of up to 50%. This strength loss does not have an impact on the YM of the composites [29–32]. Even though the fibres appeared to be UD, the varying results between each sample from the population, as represented as error bars in Figure 8, suggest the possibility of fibre misalignment. To overcome such misalignments, recent studies by HiPerDiF team [33] of the University of Bristol proposed an automated realignment method, but they are suitable only for discontinuous fibre composites. Additionally, CM also induces a certain level of fibre misalignment due to the compressive forced applied to the composites before curing.

The employed long rCFs in the composites were intended to preserve the fibre integrity, so that it will be easy to recycle again after reusing. Even though longer fibres increase the lamination stiffness of the composite [13], employing them increases the overall brittleness of the composite [3,20]. The ≈20 wt% rCF addition increased the YM of the composites but at the same time made the composites more brittle, leading to an increased likelihood of brittle failure.

Apart from the fibre-based factors, the fibre-matrix interface can also influence the mechanical properties of the rCF/EP composites. The fluffy nature of rCFs makes it hard for the resin system to wet the fibres thoroughly. This can result in unevenly cured composites (Figure 11a,b). Such unwet fibre spots appear to be the origin for failure, making the composites behave non-linearly. This can be avoided by using shorter fibres [34], or by increasing the number of active atoms and functional groups on the surface of the inert rCFs [35] to improve the overall wettability for all fibre lengths.

3.3.3. rGF/EP Composite

When comparing the measured properties of the rGF/EP composites from this study to similar studies in the literature, the values fall under the related limits. A study which adopted thermal burning, by Fraisse et al. [36], used a UD-aligned non-woven mat rGF composites, recycled with a fibre volume of 46.8 ± 1.1 wt%, with a TS of 120 ± 20 MPa and YM of 45.1 ± 4.7 GPa. The composites were remanufactured using resin transfer moulding. Similar UD-aligned non-woven fibres with 63 wt% rGF composites by Yang et al. [24] had a TS of 85 MPa and YM of 14.3 GPa.

Recycling GF utilising thermal recycling processes has frequently resulted in a strength loss of the rFs [37–39]. Notably, both well-known thermal recycling processes, FBP and the pyrolysis process, result in strength loss in rGFs. The process temperature has a significant role in influencing rGF properties. Clean rGFs with minimal impurities are obtained between 400 and 600 °C. It has been observed that, at a higher temperature range, cleaner rGFs are obtained. However, GFs start to gradually lose their strength above 300 °C [38–40]. The strength decrease appears to be an effect of heat penetrating the GF, resulting in etching of the rGF surface. However, it can be reduced using secondary strength recovery treatments, resulting in a reduced rGF diameter and an increase in the overall mechanical properties [24]. After the commonly used thermal recycling processes, supercritical fluid-based solvolysis also results in a similar decrease in the mechanical properties of the rGFs, due to the influence of heat [37].

Another noticeable defect of thermal recycling of GF appears to be the formation of char (resin residue). Char can be associated with inappropriate materials burning during the recycling process. In thermal degrading polymer resins, char formation is inevitable. The formed chars possess low thermal conductivity and insulate the heat from completely oxidising the resin phase [41]. More char indicates a weaker performance of the recycling process. SEM images confirm the presence of minor char formation, with a shear-type failure in the composite (Figure 12). Such char formations can be removed by utilising supplementary treatments. However, this study does not focus on char removal and secondary strength recovery treatments for rGFs. The evidence of char formation was minimal in rCFs.

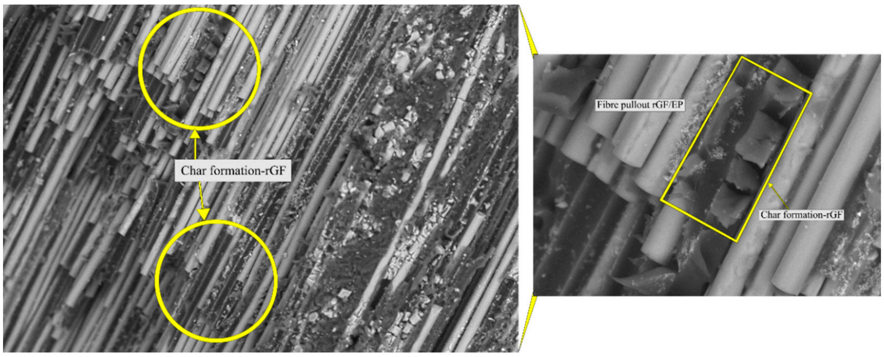

Figure 12. Presence of char formation in rGF/EP composite.

Alongside char located on the bottom layer of the rFs, a small quantity of ash appeared on the heat exposed surface. This could be the result of direct heat exposure. Despite some flaws, electrostatic attraction [36] between individual fibres was absent, making it easy to produce composites with a higher fibre volume fraction (up to 60 wt%). In the CMCs, fibres were uniformly distributed and thoroughly reinforced with the epoxy resin system, with less porosity (Figure 11d).

In addition to the thermal treatment, mechanical handling and fibre realignment during the remanufacturing of the rGFs also influence the mechanical properties [40]. As noted in rCF/EP composites, the rGF/EP composites were also influenced by fibre realignment. The error bar displays the variation from the measured sample population, which is not as strong as for rCF/EP composites. The 60 wt% rGF/EP composites had the minimum CoV between samples compared to other composites.

During the remanufacturing stage, rGF-reinforced with new thermosetting resins (e.g., epoxy) had decreased TS. However, the YM of the composites remained the same, compared to their virgin properties [4]. These behaviours can be seen in the measured mechanical properties (Table 2). For both cases (40 and 60 wt% rGFs), the YM of the CMCs was higher compared to its TS. The addition of ≈20 wt% rGFs improved the TS and IS, but the YM showed only a minor increase, indicating no change to the CMC's elastic region.

4. Conclusions

In this study, the composite wastes were recycled, remanufactured and investigated. The study utilised a novel thermal recycling process to recover the CFs and GFs and reuse the rCFs and rGFs into new fibre-reinforced epoxy composites via compression moulding. Finally, investigation on the rCF/EP and rGF/EP composites were conducted using standardised mechanical testing.

Both the CFs and GFs were successfully recycled from manufacturing waste (pre-consumer wastes) by a novel thermal recycling process. The recycled fibres, both rCF and rGF, showed evidence of a cleaner and resin-free fibre surface. The recovery rate of the rCFs was 95–98 wt%, while the rate was 80–82 wt% for rGFs, at an optimum temperature of 550 °C at atmospheric pressure. The processing time in the batch reactor was 20–25 min for rCFs and 25–30 min for rGFs. The recycling process can altogether remove the matrix phase of the composite without causing any damage to the fibres and their structure.

The newly produced composites rCF/EP and rGF/EP can be used in various applications. An increase in fibre content (≈20 wt%) in the rCF/EP composites did not cause a substantial increase in its TS. However, the TS of the rGF/EP composites almost doubled from its initial value. For the YM, the rGF/EP had no noticeable effect, but a significant rise was observed in the rCF/EP composites. The composites have certain factors which negatively influence their mechanical properties, among which poor wettability of resin to the rCF surface was a significant factor for the rCF/EP composites. Similarly, for rGF/EP composites, char formation (matrix residue) and fibre strength loss due to the thermal-based recycling were significant factors. To further utilise the recycled composites and establish continuous closed-loop recycling, these factors should be eradicated by implementing additional treatments.

Future research should investigate the mechanical properties of the remanufactured fibres using finite element methods (FEMs), and compare the values with experimental results. This kind of approach should also incorporate the investigation of the possible applications in which the newly produced composites can be utilised. Furthermore, performing life-cycle analysis (LCA) on the recycling process will provide insights into the sustainable aspects of the process.

Author Contributions: S.K.G. performed the laboratory work, prepared the samples, tested the materials, performed the investigation, analysed the results, and wrote this manuscript. T.K. contributed to the methodology section, provided feedback on the manuscript, and supervised the project. All authors have read and agreed to the published version of the manuscript.

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Publication III

Karuppannan Gopalraj, S., and Kärki, T.

A Finite Element Study to Investigate the Mechanical Behaviour of Unidirectional Recycled Carbon Fibre/Glass Fibre-Reinforced Epoxy Composites

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Article

A Finite Element Study to Investigate the Mechanical Behaviour of Unidirectional Recycled Carbon Fibre/Glass Fibre–Reinforced Epoxy Composites

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Abstract: Recycled carbon fibre–reinforced epoxy (rCF/EP) composites and recycled glass fibre–reinforced epoxy (rGF/EP) composites were numerically investigated to examine their mechanical properties, such as uniaxial tensile and impact resistance, using finite element (FE) methods. The recycled composites possess unidirectional, long and continuous fibre arrangements. A commercially available Abaqus/CAE software was used to perform an explicit non-linear analysis with a macroscale modelling approach, assuming the recycled composites as both homogenous and isotropic hardening. Five composite types were subjected to a numerical study based on the recycled fibre's volume fraction (40 and 60%) of rCF/EP and rGF/EP, along with (100%) fibreless cured epoxy samples. The materials were defined as elastoplastic with a continuum ductile damage (DUCTCRT) model. The experimental tensile test results were processed and calibrated as primary input data for the developed FE models. The numerical tensile results, maximum principal stress and logarithmic strain were validated with their respective experimental results. The stress–strain curves of both results possess a high accuracy, supporting the developed FE model. The numerical impact tests examined the von Mises stress distribution and found an exponential decrease in the stiffness of the composite types as their strength decreased, with the 60% rCF/EP sample being the stiffest. The model was sensitive to the mesh size, hammer velocity and simulation time step. Additionally, the total internal energy and plastic dissipation energy were measured, but were higher than the experimentally measured energies, as the FE models eliminated the defects from the recycled process, such as a poor fibre wettability to resin, fibre bundle formation in rCFs and char formation in rGFs. Overall, the developed FE models predicted the results for a defect-free rCF/EP and rGF/EP composite. Hence, the adopted modelling techniques can validate the experimental results of recycled composites with complex mechanical properties and damage behaviours in tensile and impact loading conditions.

Keywords: finite element methods; recycled composites; carbon fibre; glass fibre; elastoplastic material; ductile damage

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1. Introduction

Recent progress in recycling methods, such as thermal, chemical and solvolysis using water and mild solvents to recycle carbon fibre-reinforced polymer (CFRP) composite wastes and glass fibre-reinforced polymer (GFRP) composite wastes, have established a new era towards sustainable waste disposal. In particular, advanced recycling techniques, namely closed-loop and open-loop recycling, managed to reuse the recycled carbon fibres (rCFs) and recycled glass fibres (rGFs) repeatedly into either their identical or modified applications in order to close their life cycle loop, encouraging a circular economy [1]. The rCFs and rGFs possess mechanical properties similar to their virgin counterparts. In addition, only one-third of energy (in some cases, even less) is required for recycling composite wastes compared to manufacturing virgin fibres and composites. Hence, CFRP and

GFRP composite wastes have become valuable raw materials for recycling industries. As a result, laboratory-scale recycling processes with a higher fibre yield and recycling efficiency are transforming into industrial-scale recycling processes to recycle and reuse fibres in enormous quantities to satisfy the global demand, replacing virgin composites [1–7].

Further extensive investigations into the mechanical properties are required at the micro and macro levels to implement the recycled composites into an actual application, such as automobiles, aeronautics, windmills and other high-performance fields. Typical studies observed from the literature [1,2] primarily focus on testing recycled fibres without a matrix and with the matrix as fibre-reinforced polymer (FRP) composites in order to analyse the overall mechanical properties and failure modes in various testing conditions, such as tensile, compression, shear and impact.

Notable studies in recycled (r) rCFRP, such as Pimenta et al. 2010 [8], studied the mechanical properties and fracture behaviour of rCFRP composites to optimise the recycling process and develop the recycled composite design. Later, Pimenta and Pinho 2012 [9] briefly studied rCFs at both micro (as fibres) and meso-level (as composites) in tensile, compression and shear loading conditions, and compared the properties to their virgin form. It concluded that the stiffness of the recycled composites remains the same despite different loading conditions. Furthermore, in 2014, Pimenta and Pinho [10] studied rCFRP composites, highlighting the fibre bundle (defects from the recycling process), as it toughens the composites without reducing their stiffness or strength. The study also investigated the rCFRP at micro, meso and macro-level. Similar studies in rGFRP composites, such as Feih et al. 2011 [11], studied thermally recycled GFRP at micro and meso-scale. The study investigated the tensile strength reduction in the rGFRP due to the recycling temperature. Yang et al. 2015 [12] also studied rGFRP at micro and meso-scale to recover the strength of the rGFs using chemical treatments in tensile, flexural and impact modes. The study successfully regenerated the rGFs strength to achieve closed-loop recycling. However, such existing studies, observed from the literature, focused purely on experimental investigations. To understand the overall mechanical and fracture behaviours of the FRP composite, predominantly tensile and impact, it is required to investigate experimental testing along with numerical analysis [13–17].

Research involving finite element (FE) methods to analyse virgin (v) vCFRP and vGFRP composites have been well developed and established. Several studies have experimentally and numerically investigated the (tensile and impact) mechanical properties and failure behaviours. These can be found in literature reviews, such as Laffan et al. 2012 [18], Wang and Huang 2018 [19], Fatima et al. 2019 [20] and Müzel et al. 2020 [21]. However, these studies purely focused on virgin composites and recorded no significant studies involving recycled composites. Therefore, theories from virgin composite studies have to be analysed and incorporated to perform such studies over recycled composites. Based on the previous studies involving FE methods for FRP composites, both the elastoplastic material behaviour for composite modelling and a ductile damage model for fracture behaviours seem promising.

These numerical approaches were widely used for virgin FRP composites and can be observed in various stages of evolution in previous literature, such as Ju and Lee 2001 [22], who studied an unidirectional (UD) FRP composite's ductile behaviour in the matrix using an elastoplastic-based damage model and predicted the damage mechanism. The numerically predicted stress–strain (SS) behaviours have higher-order similarities compared with their experimental results. González et al. 2004 [23] studied composites using homogenised models, adopting elastoplastic material behaviour to investigate the tensile and shear properties of the composites. The study used experimental data as inputs for numerical analysis to define the non-linear behaviours at the onset of plastic deformation. Totry et al. 2010 [24] studied the in-plane shear response of the CFRP composites, considering the non-linear composite behaviour as elastoplastic behaviour, and validated the numerical model using experimental data. Melro et al. 2013 [25,26] studied UD continuous fibre-reinforced composites, similar to the materials used in this study, but in virgin form. Part I [25] studied the micro-scale material behaviour as elastoplastic with an isotropic damage constitutive model to investigate the composite failure behaviours. Part II [26] studied damage initiation and propagation in various loading conditions, such as transverse tension, compression, shear and longitudinal shear.

Further developments in adopting similar numerical modelling approaches can be seen in the latest studies, such as Ghayoor et al. 2019 [27] and Ahmadian et al. 2020 [28], who have investigated UD CFRP composite damage behaviours, highlighting resin-rich zones in the matrix (epoxy) using elastoplastic material behaviour. Ghayoor et al. 2019 [27] studied the effects of failure initiation at the resin-rich areas. The study concluded that composites fail at lower strain rates, as the failure initiation occurs at the fibre clusters. Ahmadian et al. 2020 [28] developed the study further by using a ductile damage model to study the composite failure development in tension, compression and shear modes. The study concluded that the resin-rich areas are susceptible to nucleation under tension loading, resulting in an overall strength reduction and failure. Liu et al. 2020 [14] used an elastoplastic damage model to predict the impact behaviour of the UD CFRP composites and validated the models using experimental impact test results. The study successfully managed to replicate the experimental impact damage behaviour using FE methods. Yadav and Thapa [29] studied GFRP and developed a strain-based continuum damage model to study the decreasing elasticity and stiffness degradation under fatigue damage, and validated the results. Khosravani and Zolfagharian [30] examined the tensile behaviours of fibreless polymers showcasing elastoplastic behaviour using experimental methods, and validated the non-linear behaviours using ductile failure models.

Based on the literature studies, a research gap appears, where previously no significant studies have numerically investigated the mechanical properties, especially the uniaxial tensile and impact resistance behaviours of UD and continuous and long rCFRP and rGFRP composites, and numerically predicted their damage behaviours by applying FE methods. As recycling technologies advance, and more rCFs and rGFs are available to replace virgin fibres, there is a need for such a study to explore these complex mechanical properties and failure behaviours by combining both experimental and numerical methods.

This study aims to numerically investigate the mechanical properties and damage behaviour of recycled carbon fibre–reinforced epoxy (rCF/EP) and recycled carbon fibre–reinforced epoxy (rGF/EP) composites, along with cured laminating epoxy (EP) materials, based on a macro-mechanical approach, embracing FE theories from virgin composite studies. Explicit non-linear analyses are performed to validate two standardised test methods: the tensile test (TT) and impact test (IT). The material modellings to map the non-linear behaviours are performed assuming the rCF/EP, rGF/EP and EP materials as elastoplastic. For damage, a continuum ductile damage model is used to predict their damage behaviours in uniaxial tensile and impact modes. The experimental TT results from the previous study [31] are processed as primary input data for all of the numerical test models. Finally, the numerical results from the TT and IT are validated and discussed for their respective experimental results. Hence, the study will provide insights into experimental—data processing and verification, numerical—material modelling, tensile damage prediction and the impact resistance behaviour of the rCF/EP and rGF/EP composites.

This paper is organised initially by describing the used composite types and their mechanical properties and damage behaviours during experimental testing in Section 2. Section 3 consists of the overall adopted FE methodology. Initially, a general composite behaviour is assumed. Then, equations defining the chosen material and damage modelling are presented. Subsequently, experimental data selection, calibration and processing as an input for numerical analysis are presented. Finally, FE—modelling, loads and boundary conditions are presented. Section 4 describes the calculated numerical input data and their results after numerical TT and IT. In addition, discussions involving experimental vs. numerical values, along with IT predictions, are presented. After the Results and Discussion, the paper ends with a Conclusion in Section 5.

2. Materials and Their Behaviour

2.1. Recycled Composites

The materials used in this research work were taken from the author's previous study [31]. It comprises of rCF/EP, rGF/EP and EP composite without fibres. These composites were obtained as a result of recycling CFRP and GFRP composite wastes containing valuable carbon and glass fibres using a novel thermal recycling process. The recycled fibres (rCFs and rGFs) were subsequently compression moulded using fresh EP. The newly produced rCF/EP and rGF/EP composites, along with the EP samples, were mechanically tested to measure both their uniaxial tensile properties using ISO 527-2 standard [32] and their impact resistance behaviour using unnotched charpy impact ISO 179-1 standard [33]. Table 1 presents the experimentally measured TT and IT results of the new composites. These values are used as primary data in this current study. As seen, the rCF/EP and rGF/EP composites consist of two types, based on the fibre weight fraction (V^f) 40 wt% and 60 wt%. The resin combined with its hardener in a 2:1 ratio occupies the composite's remaining volume (V^r). The recycled composites possess a uniform structure throughout the lamina. The composite types maintained a UD fibre orientation (0°) and continuous (uniform length from end to end) and long (105 ± 2 mm) fibres.

Table 1. Experimental results of the compression moulded recycled composites [31].

Composite RECIPES	V^f (wt%)	V^r (wt%)	Tensile Strength (MPa)	Young Modulus (GPa)	Impact Strength (kJ/m^2)	Fracture Strain (No Unit)	Density (g/cm^3)
rCF/EP	60 ± 2	40 ± 2	235.70	60.80	53.61	0.00683	1.52
	40 ± 2	60 ± 2	210.34	45.28	49.98	0.00827	1.64
rGF/EP	60 ± 2	40 ± 2	114.58	30.72	41.05	0.00272	1.77
	40 ± 2	60 ± 2	65.42	27.37	18.99	0.00156	1.85
EP	0	100	39.46	2.16	35.18	0.05810	1.45

2.2. Recycled Composite Damage Behaviour

The recycled composites damage behaviours are dynamic and heavily influenced by their recycling and remanufacturing processes. To understand such complex behaviours, it is required to compare them with well established and studied damage behaviours of virgin composites. Typically, in virgin UD FRP composites, the failure depends on the fibre, matrix and fibre–matrix interface [24], as all three contribute to the failure initiation and development. In particular, the matrix possesses a significant role in composite damaging, as the matrix strain to failure dominates the fibre strain. The fibre arrangement and direction and the loading conditions (tensile, compression, shear and impact) significantly influence the overall composite damage behaviour [34]. In this study, tensile and impact loading modes were taken for the investigation. Under uniaxial tension loading (see Figure 1), the UD fibres encounter multiple interlaminar breakages at random spots, decreasing the overall composite stiffness. Subsequently, debonding occurs at the fibre–matrix interface, leading to interlaminar composite delamination. As the tension develops, the stress propagates into the matrix, causing crack growth near the broken fibres and leading to a final fracture [35,36]. Similar damage behaviours were observed during the experimental testing [31] for the rCF/EP and rGF/EP composite samples.

Under unnotched charpy impact testing (see Figure 1), especially low-velocity impact, UD FRP composites experience plastic deformations, leading to the matrix cracking followed by a series of failures, such as fibre breakage and delamination. In an IT, composite failure initiation and development occur opposite (tension zone) at the impactor and samples contact point (compression zone). The fracture develops towards the impactor [14,17,20,37]. The UD FRP composites possess a low interlaminar fracture toughness due to their fibre arrangement, and, under impact loading, delamination occurs favourably, resulting in higher impact damage [38]. Similar failure behaviours were observed during experimental impact testing [31] rCF/EP and rGF/EP composite samples. However, the EP samples displayed an explosive impact fracture, as the samples are fibreless.

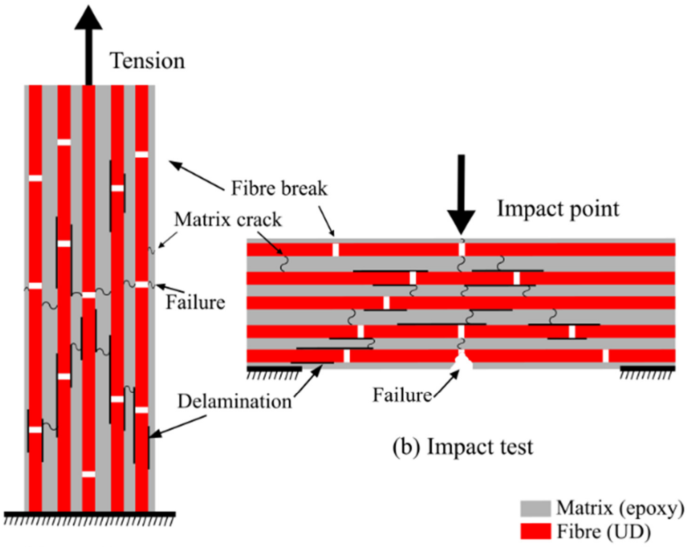


Figure 1. Damage sequence of UD FRP composites tensile mode (modified from [35]) and impact mode (modified from [39]).

Figure 2 presents the microscopic images of the rCF/EP and rGF/EP samples from their fractured region after experimental testing from the author's previous study [31]. These images were taken using a Jeol JSM-5800 LV scanning microscope at 4k magnification. As observed during the testing and microscopic images, most of the rCF/EP and rGF/EP samples failed normal to the fibre directions due to fibre damage, delamination, matrix cracking, fibre pullouts, internal matrix voids and, finally, matrix shear bands in resin-rich zones. Overall, the samples showed multiple matrix-dominating failures. In addition, evidence for ductile behaviour was noticed in the epoxy matrix failure zones. As seen in Figure 2a, ductile-damage-based void nucleation and growth were observed at the resin-rich matrix zones. In addition, in Figure 2b, ductile feathering was observed in multiple spots at the matrix.

Figure 3 presents the fractured sample's images after experimental tensile and impact testing from the author's previous study [31]. Figure 3a–e presents the images after TT and Figure 3f–j after IT. The images show that the tensile-tested samples broke without any neck formation, indicating a brittle failure. However, the SS values of the tested composites displayed a small elastic zone and a large non-elastic zone with failure initiation and evolution after the peak values, indicating ductile damage domination. Apart from the regular damage behaviours, the composites were influenced by external factors. In rCF/EP composite types, mostly in 60% rCF/EP, the poor wettability (defects from thermal recycling [31]) of the rCFs with the matrix created a weak interfacial strength and influenced the rCF/EP tensile and impact failure.

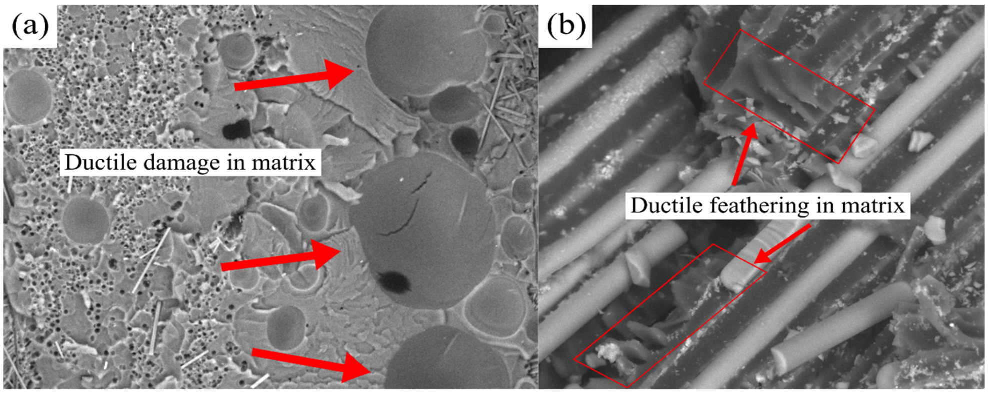


Figure 2. The evidence for ductile damages in matrix: (**a**) rCF/EP; (**b**) rGF/EP.

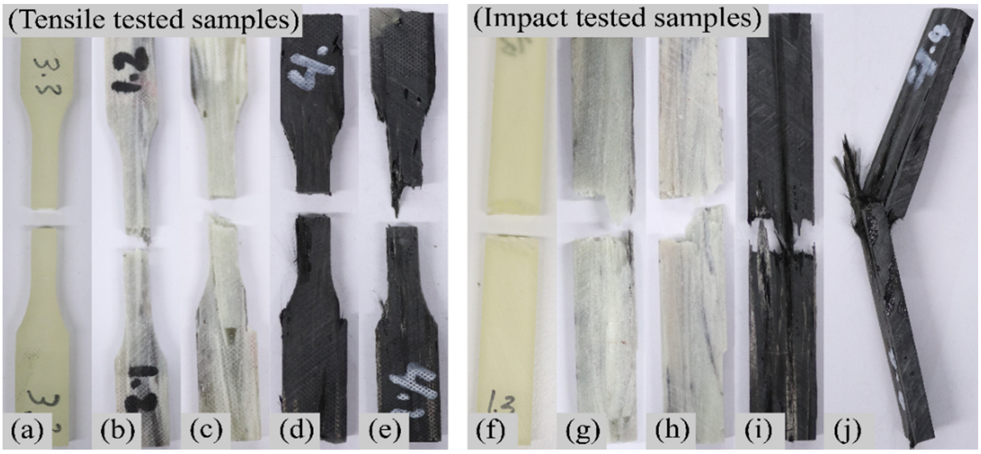


Figure 3. Sample fracture after experimental TT: (**a**) 100% EP; (**b**) 40% rGF/EP; (**c**) 60% rGF/EP; (**d**) 40% rCF/EP; (**e**) 60% rCF/EP; and after IT: (**f**) E-EP; (**g**) 40% rGF/EP; (**h**) 60% rGF/EP; (**i**) 40% rCF/EP; (**j**) 60% rCF/EP.

In UD FRPs, when the fibre properties are superior to the matrix, it creates a weak fracturable nature when aligned as normal to the fibre direction. It favours the crack distribution parallel to the fibre direction [40]. Furthermore, the stress concentrations are weak on the regions of unwet fibre clusters, promoting damage initiation and development towards the matrix zones [41]. Hence, it is known that the inter-fibre distance between each fibre has a heavy influence on the sample damaging [42]. Due to unwet rCFs merging into clusters, some samples developed resin-rich zones that influenced the crack propagation parallel to the fibre direction. Such poorly wet fibres were absent in rGF/EP types. However, the presence of char (resin residue from the recycling process [31]) caused a negative influence on the sample damage. The EP samples under TT and IT showed uniform results throughout the sample population.

3. Finite Element Methodology

Figure 4 presents the overall methodology adopted. Commercially available FE software Abaqus/CAE was used to perform both the numerical TTs and ITs. The tensile and impact models were developed based on explicit non-linear analysis using the experimentally measured TT data as inputs. The FE analysis was performed for all five composite types: EP (100%), rGF/EP (40 and 60%) and rCF/EP (40 and 60%).

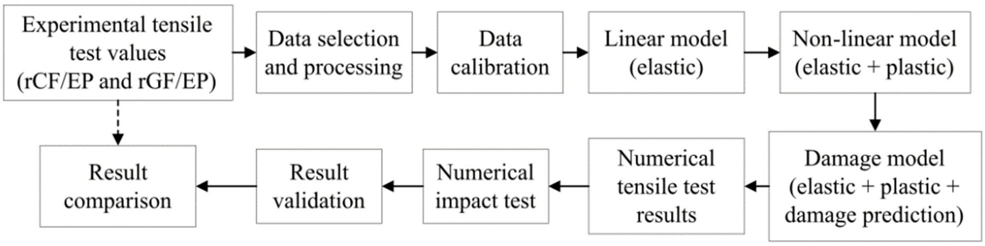


Figure 4. Implemented FE approach for the recycled composites.

3.1. Recycled Composite Assumption

In general, rCF/EP and rGF/EP composites possess complex mechanical behaviour compared to vCF/EP and vGF/EP composites. Both the recycling and remanufacturing processes significantly influence their mechanical properties, making it challenging to compare standard composite behaviours. In this study, a UD (0°) and continuous (end-to-end) and long (105 ± 2 mm) fibre arrangement was constantly maintained for the rCF/EP and rGF/EP composite types. Similar structured vCFRP composites lamina are transversely isotropic [15,43,44], or, in some cases, even anisotropic [21]. The vGFRP and EP behave as elastic isotropic solids [36], even though vGFRP microscopically behaves as transversely isotropic [13]. In general, composite assumptions were made depending on the adopted methodologies.

The UD FRP composites are homogenous along their fibre direction. In this study, the experimental uniaxial TT provided results for the composites concerning a single axis (tensile). However, the recycled composite's compression, shear and individual fibre (rCFs and rGFs) elasticity properties are unknown due to the limitations in handling the recycled fibres. Overall, a macroscale numerical modelling was performed considering all the composite types—EP, (40% and 60%) rGF/EP and (40% and 60%) rCF/EP—as homogenous and isotropic hardening to avoid complexity by empirically assuming the data and fully utilising experimentally measured values.

3.2. Elastoplastic Material Model for Recycled Composites

The experimental TT results (SS values) of EP, rGF/EP and rCF/EP composite types initially recorded a smaller linear elastic region. Furthermore, with the applied force (tension), the region developed into a larger non-linear inelastic region. Finally, the SS curves decreased and stopped after reaching their respective ultimate points, indicating the sample fracture. The elastic region (fibre behaviour) can be defined by the Young modulus and Poisson ratio in UD FRP composites. The non-linear region is described using Hahn and Tsai's [45] equation. However, due to the observed recycled composite behaviour, the non-linear response of the samples can also be explained by defining the composite matrix as elastoplastic behaviour [13]. Such elastoplastic behaviour is commonly observed in UD FRP composites and plays a significant role in predicting composite strength, failure and damage evaluation [19,25,46,47].

Based on overall observations, elastoplastic material modelling was used in this study. The model [48] assumes that the plastic deformation (F^{pl}) follows after elastic deformation (F^{el}) to form an overall deformation (F^{T}).

$$F^{T} = F^{el} + F^{pl} \tag{1}$$

The deformation identification and separation are based on an additive relationship between the strain rates, where the total strain rate (ε^{T}) is defined as the sum of elastic (ε^{el}) and plastic strain rates (ε^{pl}) [48].

$$\varepsilon^{T} = \varepsilon^{el} + \varepsilon^{pl} \tag{2}$$

The damage model involves elastoplastic material behaviour with ductile damage as dominant. Therefore, Equation (3) [28,49] represents the yield surface of the model, in which, the experimental SS curves are used as an input to determine the yield function $\sigma_{Y}\left(\bar{\varepsilon}_{eq}^{pl}\right)$, where, $\bar{\varepsilon}_{eq}^{pl}$ = the equivalent plastic strain.

$$f(\sigma) = q - \sigma_{Y}\left(\bar{\varepsilon}_{eq}^{pl}\right) \tag{3}$$

3.3. Continuum Ductile Damage Model for Recycled Composites

As the composite types are defined as elastoplastic behaviour, studies using similar materials [25,26] have proposed isotropic damage models to predict the sample damage evolution. Failure initiation usually occurs in the matrix in UD FRP composites, making the composites ductile-dominating failures [50,51]. Similarly, the experimental TT values highlight a ductile-based behaviour dominating the overall composite outcomes, which is numerically implemented using a continuum ductile damage model. Hooputra et al. 2004 [49] laid the foundation for the ductile-based failure of the cured epoxy matrix (brittle material) under tension loading. The matrix encountered a noticeable plastic deformation before failure initiation and development.

As seen in Figure 2a,b, the void nucleation, growth and coalescence were the foundation for developing and incorporating the damage model. The damage model [48,49] assumes that the onset of damage ($\bar{\varepsilon}_{D}^{pl}$) is a function of stress triaxiality ($\eta = -\frac{\sigma_m}{\sigma_{eq}}$) and the equivalent plastic strain rate ($\bar{\varepsilon}_{0}^{pl}$), as shown in Equation (4), where σ_m = stress state hydrostatic component and σ_{eq} = Huber–von Mises equivalent stress.

$$\bar{\varepsilon}_{D}^{pl}\left(\eta, \bar{\varepsilon}_{0}^{pl}\right) \tag{4}$$

The stress triaxiality is further defined in Equation (5) [48], where trace (T) = trace of the stress tensor, which equals the sum of principal stresses. For the performed uniaxial TT, the following stress conditions are considered, $\sigma_2 = \sigma_3 = \sigma_{12} = \sigma_{31} = \sigma_{23} = 0$. Finally, the stress triaxiality for uniaxial $\eta = -0.333$ and the equivalent plastic strain rate $\bar{\varepsilon}_{0}^{pl} = 0$, as the materials are strain-rate-independent.

$$\eta = -\frac{\frac{1}{3} \times \text{trace}\ (T)}{\sigma_{mises}} = -\frac{\frac{1}{3} \times \left(\sigma_{xx} + \sigma_{yy} + \sigma_{zz}\right)}{\sigma_{xx}} \tag{5}$$

For the damage to initiate, the equivalent plastic strain ($\bar{\varepsilon}_{eq}^{pl}$) reaches a threshold value known as fracture strain ($\bar{\varepsilon}_{0}^{pl}$), where the variable D = 0, in which, the plastic deformation increases monotonically as the state variable (ω_D) increases. The damage initiation occurs when Equation (6) is satisfied ($\omega_D = 1$) [48,49].

$$\omega_D = \int \frac{d\bar{\varepsilon}_{eq}^{pl}}{\bar{\varepsilon}_{0}^{pl}\left(\eta, \bar{\varepsilon}_{0}^{pl}\right)} = 1 \tag{6}$$

The state of stress at the point is defined in Equation (7) [28,48], where $\overline{\sigma}$ is the undamaged stress tensor:

$$\sigma = (1 - D)\overline{\sigma} \tag{7}$$

For the damage required to develop ($\overline{\varepsilon}_{eq}^{pl} > \overline{\varepsilon}_{0}^{pl}$), the variable D increases from 0 to 1. At this maximum value, the material loses its load carrying capacity for the equivalent plastic strain ($\overline{\varepsilon}_{eq}^{pl} = \overline{\varepsilon}_{f}^{pl}$). The damage development occurs when Equation (8) is satisfied [48,49].

$$\omega_D = \int \frac{d\overline{\varepsilon}_{eq}^{pl}}{\overline{\varepsilon}_{0}^{pl}(\eta, \overline{\varepsilon}_{0}^{pl})} \geq 0 \tag{8}$$

The damage evolution is specified using fracture dissipation energy (G_f) before the damage initiation and equivalent plastic displacement at failure (u^{pl}) after damage initiation. The fracture energy is measured based on the stress–displacement outcomes, as seen in Equation (9) [48].

$$G_f = \int_{\overline{\varepsilon}_{0}^{pl}}^{\overline{\varepsilon}_{f}^{pl}} L\sigma_y d\overline{\varepsilon}^{pl} = \int_{0}^{\overline{u}_{f}^{pl}} \sigma_y d\overline{u}^{pl} \tag{9}$$

The damage evaluation law [48,49] can also be defined using displacement at failure in two cases: linear form and exponential form. The governing equation for equivalent plastic displacement is

$$D = D\left(u^{pl}\right) \tag{10}$$

In this study, linear form is used, as the damage variable evolution is expressed in Equation (11) [48], where u_f^{pl} = the effective plastic displacement.

$$\dot{D} = \frac{u^{pl}}{u_f^{pl}} \tag{11}$$

The relationship between effective plastic displacement (u_f^{pl}) and energy dissipation (G_f) is defined in Equation (12) [48], where σ_{y0} = yield stress value after reaching failure criteria. All of the composite types used in the study stopped immediately after the fracture point. The displacement at failure (u^{pl}) is maintained at 0.05.

$$u_f^{pl} = \frac{2G_f}{\sigma_{y0}} \tag{12}$$

3.4. Numerical Material Modeling

The experimental TT results from the author's previous study [31] were used as input parameters for numerical analysis in this study. These experimental TTs were performed using Zwick Roell (Z020) tester. The extensometer was connected digitally using testXpert II software to record the composite's SS curves. For each composite type, 15 samples were tested. The tests recorded data for applied force (N) to its corresponding nominal strain (%). The force–strain readings ranged from 1250 per 100% EP sample (lowest) to 4800 per 60% rCF/EP composite sample (highest) per test. Overall, an average of 236,000 readings were recorded from the experimental TT.

In the measured experimental TT values, the Young modules represent the slope of the elastic zone. The yield points represent the transition phase from elastic to plastic and, finally, the plastic zone ended by reaching the ultimate points. The samples broke immediately after achieving their ultimate points, and the process ended with a minor value drop in the end. Furthermore, the data were processed. The force was converted into engineering stress (MPa) using the cross-section values of each sample, and the strain was kept unitless. Out of the obtained SS curves, 15 curves (minimum) per composite type—EP, (40 and 60%) rGF/EP and rCF/EP, a single SS curve per composite type has to be chosen instead of performing an average to combine the 15 curves per sample. One best sample per composite type, with the SS curve occupying the average position from the overall population, was selected. By doing so, real-time measured composite behaviours were preserved compared to averaging the SS values to obtain one unified curve (totally modified SS values).

After selecting five SS curves (one per composite type), standard formulas were used to convert them into true SS values. During the experimental TT, samples were held using pneumatic holders, causing the samples to experience micro compression. The sensitive extensometer also recorded such behaviours, clearly visible in the measured data as negative values. Hence, these values below zero were eliminated from the true SS data. Then, these data were further processed using Abaqus inbuilt material calibration tool. The true SS values were fed as data sets (input). A SS curve appears based on the input with strain as x-axis and stress as y-axis. Before initiating the actual calibration, curve smoothing was performed with 0.75 weight. Next, an elastoplastic isotropic behaviour was created to calibrate the inputs. A manually operated pendulum impact tester was used to perform the experimental charpy unnotched IT. Similar to the TT, 15 samples (minimum) per composite type were tested. However, only the TT data were further processed and used as an input for the simulation.

3.5. Modelling, Loads and Boundary Conditions

3.5.1. Tensile Test Model

The numerical TT simulation was performed using a dogbone-shaped sample. The sample was designed and extruded based on ISO 527-2 specimen type 1BA standard [32], similar to the samples used during the experimental TT. After the primary modelling, based on the overall sample length of 100 mm, it was divided into seven points, which were A_1, B_1, C_1, X_1, C_2, B_2 and A_2, and five cells, which were A_1–B_1, B_1–C_1, C_1–C_2, C_2–B_2 and B_2–A_2, in that order (see Figure 5).

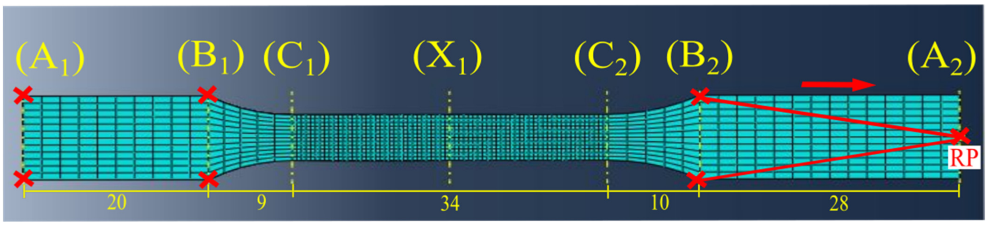


Figure 5. The modelled TT sample with boundary conditions and loads.

The sample meshing was performed at each cell based on the created seven datum planes on each point, perpendicular to the sample length. A fine mesh was created between points A_1 to C_1 and C_2 to A_2 with 4080 elements. These are the cells experiencing the least stress. The maximum stress will be experienced within the cell C_1–C_2, leading to damage at X_1 (expected point). A very fine mesh was created with 8160 elements, covering the sensitive area. An explicit mesh with linear geometric order was used, with full integration eliminating the hourglass effect throughout the sample.

The dogbone sample was fixed at the bottom cell A_1–B_1, restricting free movements in terms of displacement (U) and rotation (UR) in all three (x,y,z) axes (U1 = U2 = U3 = UR1 = UR2 = UR3 = 0). Similar restrictions were applied to cell B_2–A_2 at the opposite end (U1 = U3 = UR1 = UR2 = UR3 = 0), but allowing displacement (U2) in the direction parallel to the applied force (y-axis). This particular condition was assigned to the sample via a separately created reference point (RP) at point A_2. The cells occupying the space between points B_2 and A_1 were couples to the created RP. During the numerical TT simulation, the applied displacement (U2) will pull the cells uniaxially at RP and evenly distribute the force within the respective cells below.

3.5.2. Impact Test Model

The numerical IT was performed using a rectangular sample and an impactor (hammer). The sample was designed and extruded based on the dimensions from ISO 179-1 type 1 test specimens 80 mm × 10 mm × 4 mm under charpy unnotched impact strength [33]. The actual dimensions of the hammer were measured and modelled to achieve a closer simulation to experimental IT. After the primary modelling, based on the overall sample length of 80 mm, it was divided into seven points, which were A_3, B_3, C_3, X_2, C_4, B_4 and A_4, and five cells, which were A_3–B_3, B_3–C_3, C_3–C_4, C_4–B_4 and B_4–A_4, in that order (see Figure 6). The parts (sample and hammer) were then assembled by placing the hammer tip above X_2, maintaining a small gap between the parts.

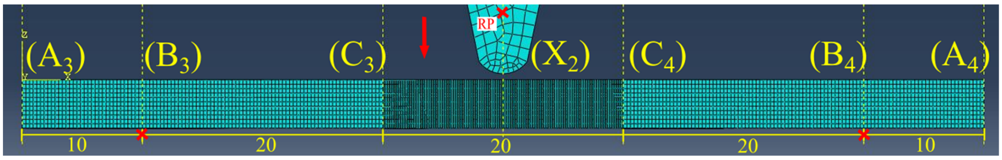


Figure 6. The modelled IT sample with boundary conditions and loads.

The sample meshing was performed at each cell based on the created seven datum planes on each point, perpendicular to the sample length. A fine mesh with 63,800 elements was created between points A_3 to C_3 and C_4 to A_4. The points cover four cells that were not exposed directly to impact, making it a less sensitive area. The hammer hits perpendicular to the middle cell at point X_2. This sensitive region of the sample experiences heavy damage, leading to a break. A very fine mesh with 63,800 elements was created at the middle cell C_3–C_4. An explicit mesh with linear geometric order was used throughout the sample. Full integration was used at the impact cell C_3–C_4, but reduced integrations were adopted at cells with no direct contact with the hammer in order to reduce the complexity. The hourglass was blocked from the entire model.

The meshed sample was pinned parallel to the impact surface at points B_3 and B_4. The displacement in all three (x,y,z) axes (U1 = U2 = U3 = 0) was restricted at the pinned edges. The hammer was defined as a rigid body by creating constraints. A normal mesh with 1786 elements was created for the hammer. A reference point (RP) was created at the centre of the hammer to apply uniform velocity perpendicular to the sample. Furthermore, contact modelling was implemented. The hammer (moving part) was adjusted until its surface fully covered the sample's surface (inert part) during impact. The datum planes were used as a guideline to align the parts. After fixing the positions, surfaces were created using designated cells. An explicit contact-based interaction was created to assign the surfaces. First, a frictionless contact was created on the surfaces. Then, for the numerical IT, a tangential behaviour was created on the surfaces, with a 0.3 friction coefficient, followed by creating a normal behaviour with hard contact for the hammer to hit the sample, due to the applied velocity.

4. Results and Discussion

4.1. Numerical Input Parameters

Table 2 presents the input parameters for the numerical simulation. The experimental TT results were calibrated as elastoplastic isotropic behaviour to model the non-linear behaviour of the recycled composites numerically. These calibrated values are primarily used as input parameters to perform numerical TT and IT. The elastic region (linear) was defined using the Young modulus and Poisson ratio. The plastic region (non-linear) was defined using values between yield stress and ultimate points. Furthermore, these obtained a plastic stress (yield stress), and the plastic strain values were processed by excluding the SS values below the yield point, as the values were already defined as the elastic region. The damage initiation and development occurs after the ultimate points.

Table 2. Primary input parameters for numerical testing.

Composite Types	Young Modulus (MPa)	Yield Point		Ultimate Point		Fracture Strain	Poisson Ratio
		Stress (MPa)	Strain (No Unit)	Stress (MPa)	Strain (No Unit)		
60% rCF	13,262	55.8332	0.00421	246.529	0.02151	0.00683	0.3
40% rCF	11,103	45.8554	0.00413	181.254	0.01952	0.00827	0.3
60% rGF	10,921.3	26.4294	0.00242	120.598	0.01321	0.00272	0.25
40% rGF	9011.50	22.0782	0.00245	83.16	0.01077	0.00156	0.25
100% EP	2004.46	10.1626	0.00507	41.7633	0.02825	0.05810	0.3

Therefore, damage prediction was included using non-calibrated parameters obtained from experimental TT results, such as fracture strain, stress triaxiality, strain rate and displacement at failure. These parameters were recorded during the experimental TT and further incorporated as input values for modelling. The stress triaxiality was −0.333 for all of the models. The strain rate was maintained as zero for the TT and as one for the IT. During TT, the displacement at failure for the rCF/EP and rGF/EP samples was maintained as low, in a range between 0.03–0.06, as the applied tension was parallel to the fibre direction. The displacement was raised to 0.15–0.17 for all of the composite types during IT, including EP during its TT (contains no fibre), as the damage occurred immediately at the composite's ultimate point. The calibrated values recorded after were removed, as the values displayed a minor drop (neglectable), indicating that the recycled composite samples experienced a break, and no further significant SS values were recorded.

4.2. Numerical Tensile Test Results

Figure 7 presents the fractured samples of all composite types after numerical TT. The sample damage occurred based on ductile damage modelling (DUCTCRT). As can be seen, the ductile damage criteria distribution diversified across the composite types. As the variable D increases from zero to one, the materials lose their load carrying capacity for the equivalent plastic strain. At this phase, the progress in break initiation and development appears across the samples, depicting the overall fracture. The samples were fractured based on the applied tensile displacement at the sample's reference point (RP) A_2 (see Figure 5). As the tensile strength of the rGF/EP and rCF/EP samples increased, the displacement to failure also increased, with an increase in strain rate to failure. Initially, multiple estimations were made to fracture the samples within the defined time step. A lower displacement failed to damage the samples, and higher values exceeded the experimental time frame, resulting in errors. Finally, the optimal values of the applied displacement are as follows: 0.55 mm (40% rGF/EP), 0.67 mm (60% rGF/EP), 1.05 mm (40% rCF/EP), 1.15 mm (60% rCF/EP) and 2 mm (EP). It can be observed that the fibreless EP samples required a higher displacement to failure, indicating the longer strain rate to failure.

As expected, the numerically fractured samples appear to have similarities compared to their experimentally fractured samples, shown in Figure 3. In the EP samples, the fibreless material showed minor necking at the fractured region. Such behaviour was not visible in the experimental TT sample, despite the EP samples being brittle. In the rGF/EP samples, the damage initiation and development were evenly spread throughout the sample's midsection (see Figure 7b,c), unlike other composite types (EP and rCF/EP), with damage behaviour in a particular spot. Such behaviour in the rGF/EP samples could be due to the thoroughly wet, long and UD rGFs evenly distributing the applied tension. However, as the rGFs volume increased (40% to 60%), the samples gradually transformed towards similar damage behaviour, such as EP and rCF/EP. In the rCF/EP samples, the resin-rich zones, which originated due to the poorly wet rCFs clusters, weakened the samples in uneven spots and created damage initiation in more than one spot at the sample's midsection (see Figure 7d,e). The numerical simulations assume the composites are defect-free, as no external errors are modelled, yet the input parameters are derived from the experimental results. The minor defects reflected within the experimental results are also visible to a certain extent in the numerical TT results.

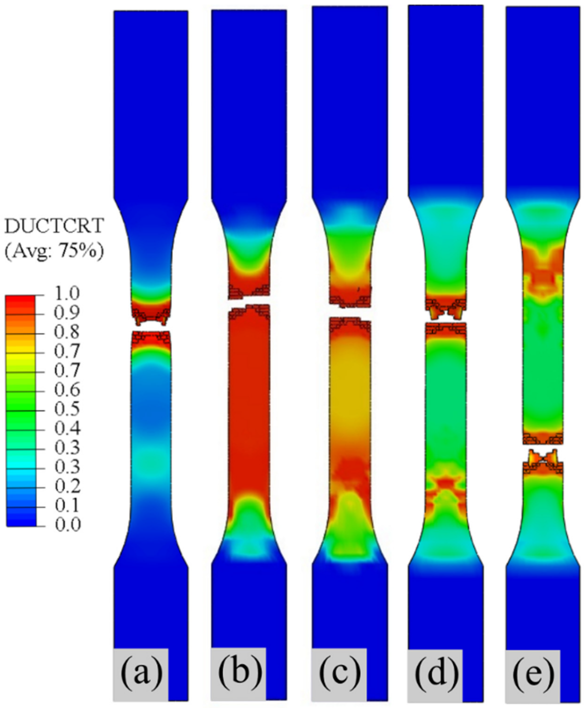


Figure 7. Fractured samples after numerical TT: (**a**) 100% EP; (**b**) 40% rGF/EP; (**c**) 60% rGF/EP; (**d**) 40% rCF/EP; (**e**) 60% rCF/EP.

Furthermore, the numerical tensile results of all of the composite types were analysed. The results from the FE models were recorded as frequencies, with 200 intervals recorded from the start to the end of the experiment based on evenly spaced time intervals. The recorded results for each composite type were extracted using the ODB field output. As the samples experienced maximum stress at the fractured area, one element was picked to plot their respective values. Finally, the maximum principal stress (y-axis) and respective logarithmic strain (x-axis) for all of the composite types at the damaged region were plotted and compared to their respective experimental tensile SS values.

Figure 8 presents the plotted SS values of experimental vs. numerical for the 100% EP composite sample. The experimental TT results (15 samples minimum) displayed a 2.69 coefficient of variation (CoV) for the tensile strength and 2.09 CoV for the tensile modulus; the SS curves in particular displayed uniform consistency. These results make EP a highly reliable composite type to validate the adopted numerical method. Typically, failure initiation for fibreless polymers under tension occurs as the strain rate increases, decreasing its overall stiffness [52]. Similarly, EP samples fractured at a higher strain among all of the tested FRP composite types. The numerically extracted SS values from the FE models were further processed by eliminating additional values after the fracture region. As can be seen, the numerical SS values fit both the linear and non-linear paths of the experimental SS values. The predicted damage values from the fracture initiation to development slightly deviate from the actual damage values. However, they lie within an acceptable range. From the results, it can be concluded that the adopted FE methodology is capable of recording the TT results numerically for highly consistent material.

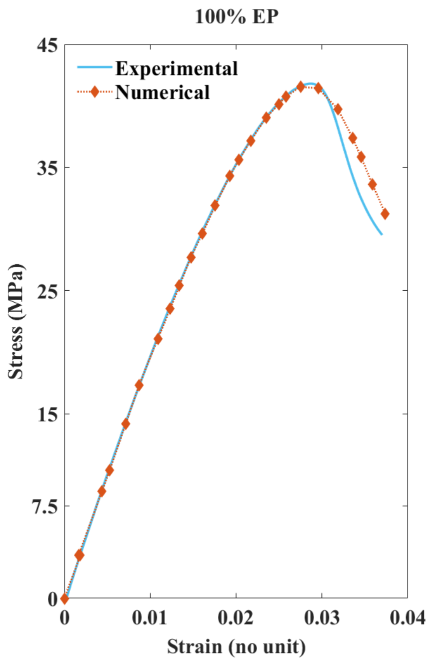


Figure 8. Comparing experimental and numerical SS curves of 100% EP composite.

Figure 9a,b presents the plotted experimental vs. numerical SS values for the 60 and 40% rGF/EP composite types. The composites possess a lower strain rate compared to the EP sample results. As the fibre volume increases, the strain rate decreases, resulting in a higher stress value. Figure 10a,b presents the plotted experimental vs. numerical SS values for the 60 and 40% rCF/EP composite types. In vCF/EP, composites containing a higher fibre volume (60%) exhibit a reduced inter-fibre distance. This increases the residual stress, causing the damage initiation at lower SS rates [42]. In addition, UD vCF/EP laminate with resin-rich spots fail at a lower strain rate and possess a lower stiffness, despite holding an average fibre volume [27]. Similarly, the poorly wet (40 and 60% rCF/EP) and higher fibre volume (60% rCF/EP) composite samples displayed damage initiation and development at a high stress rate, with a relative lower strain rate ratio when compared to EP and rGF/EP.

As seen in Figures 9 and 10, the numerical SS values fit the non-linear paths of the experimental SS values. However, the predicted damage values did not accurately follow the path of the experimental values. Regardless, they lie within the range. Such deviations are expected, as the rCF/EP and rGF/EP are complex materials. The numerical modelling did not incorporate the external defects, such as the resin-rich zone and unwet fibres in rCF/EP samples and char formation in rGF/EP samples, that significantly influenced the sample fracture behaviour during experimental testing. Alternatively, the FE models predicted a defect-free damage behaviour. In 40 and 60% rGF/EP, the predicted damage coincides with a higher accuracy than the rCF/EP samples. The damage predictions seem to be dropping directly from their ultimate points in (40 and 60%) rCF/EP and 60% rGF/EP, instead of following the non-linear experimental damage range.

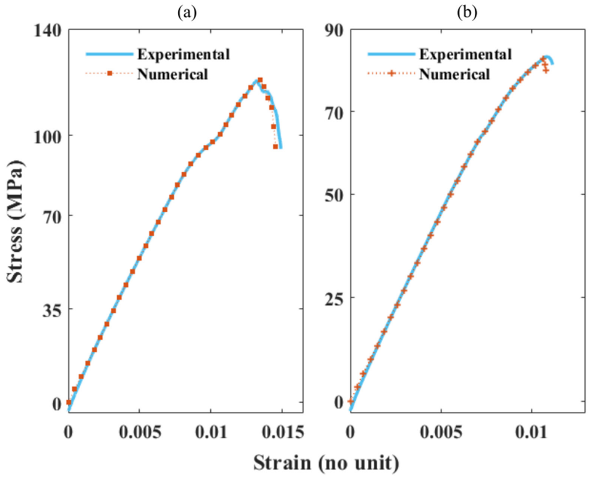


Figure 9. Comparing the experimental and numerical SS curves of rGF/EP composites: (**a**) SS curves of 60% rGF/EP; (**b**) SS curves of 40% rGF/EP.

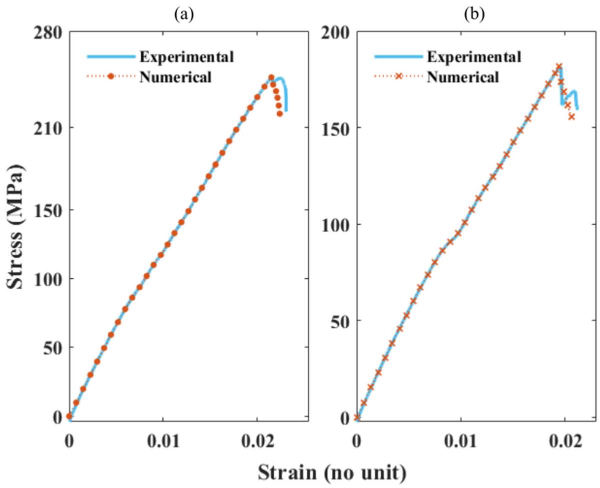
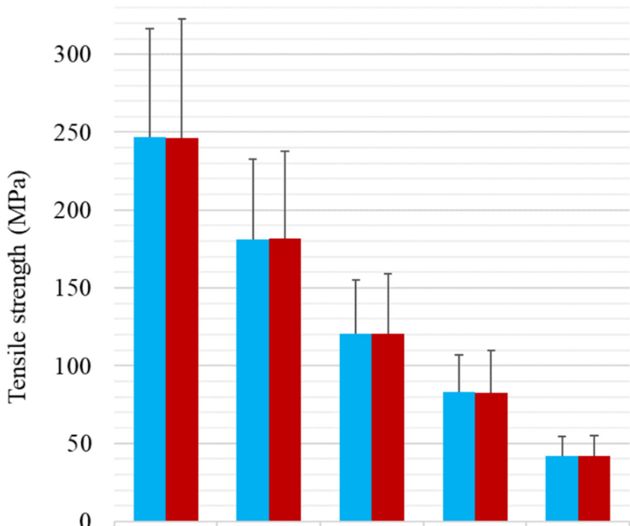


Figure 10. Comparing experimental and numerical SS curves of rCF/EP composites: (**a**) SS curves of 60% rCF/EP; (**b**) SS curves of 40% rCF/EP.

Figure 11 presents the overall TT results of experimental and numerical analyses for all of the composite types. The tabulated values were obtained from the true stresses. As can be seen, the numerically measured tensile strength appears to be similar when compared to the experimental values. Furthermore, standard deviation (SD) values were calculated from stresses of each composite type and incorporated as error bars. The SD of experimentally measured stresses and numerically obtained maximum principal stresses were compared. Despite the numerical SS, values were measured at particular intervals in order to reduce the computing complexity. The values seem within the range of higher-order SS values from the experimental test. Based on the results, it can be concluded that the adopted FE methodology is capable of recording the TT results numerically for inconsistent materials.



	60% rCF/EP	40% rCF/EP	60% rGF/EP	40% rGF/EP	100% EP
Experimental	246.79	181.45	120.73	83.26	41.8
Numerical	246.33	181.5	120.52	82.68	41.64

Figure 11. Measured tensile strength of the composites experimental vs. numerical.

4.3. Numerical Impact Test Results

Figure 12 presents the von Mises stress distribution across the numerical IT samples. As the hammer impacted the sample, variable D increased from zero to one, causing crack initiation and development at the hammer's contact point with the sample. Typically, von Mises stresses are used to estimate yield failures in ductile materials. As the ductile damage model (DUCTCRT) was used to study the recycled composites, the von Mises stress distribution projects an understanding of the plasticity for all of the tested composite materials. As seen from all of the composite types, the majority of the elements experienced mid-range stress. However, the scattered elements at critical points, such as pinned regions and inner fractured regions, reached the maximum stress value. The test was performed until the samples experienced a complete fracture. The samples displayed an exponential decrease in their overall stiffness in the following order: 60% rCF/EP (stiffest), 40% rCF/EP, 60% rGF/EP, 40% rGF/EP and EP (high-plasticity). The EP samples appear more highly flexible than fibre-reinforced composites (rGF/EP and rCF/EP), representing the absence of fibre (low-elasticity). Overall, as the fibre volume and the composite strength increased, the Young modulus increased, resulting in a stiffer composite.

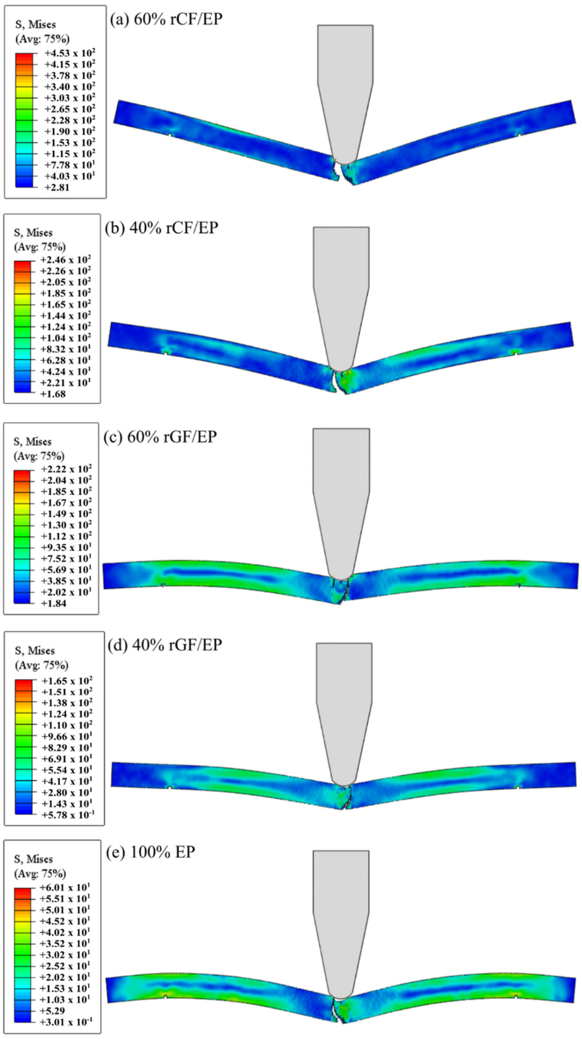


Figure 12. Numerical IT results using von Mises stress distribution: (**a**) 60% rCF/EP; (**b**) 40% rCF/EP; (**c**) 60% rGF/EP; (**d**) 40% rGF/EP; (**e**) 100% EP.

In the impact mode arrangement, the direction of applied force was perpendicular to the recycled fibre direction, similar to a three-point bending test. As seen in Figure 12, the composite types 40% rCF/EP and 60% rGF/EP experienced maximum stresses that resembled each other. However, the stress concentration for 60% rCF/EP was higher when compared to the rest of the composite types, especially 40% rCF/EP. The 20% increase in the fibre volume fraction increased the stress concentration by 84.14%, making it a robust composite in an impact mode. However, the 20% increase in the fibre volume for rGF/EP resulted in a 34.54% increase in the von Mises stress. As expected, the homogeneous EP samples failed, encountering a 63–84% lower stress range compared to the rCF/EP and rGF/EP composite types. However, it possessed a high stress distribution across the sample.

Figure 13 presents the total internal energy and plastic dissipation energy observed by the composite samples after the numerical IT. The energy measurement starts as the hammer contacts the sample (damage initiation). Further, as the hammer progressed based on the applied velocity and time step, the crack propagation parallel to the impact direction finally ends with a fracture. Thus, the recorded maximum energy at fracture was taken as the plastic dissipation energy observed by the samples. During numerical IT, the energy dissipation in the samples increases with respect to the hammer velocity, and is required to establish a boundary [53]. Such entities were also observed during the simulation process. The modelling failed with errors at higher velocities, as the sample energies were not fully recorded based on the established time interval, and at lower velocities, the samples remained undamaged. Finally, after repeated estimations using lower velocities, the hammer velocity for all of the composite types were fixed as follows: 8.1 m/s (60% rCF/EP), 7.1 m/s (40% rCF/EP), 6 m/s (60% rGF/EP), 4 m/s (40% rGF/EP) and 4.25 m/s (EP).

The model outcomes were susceptible to three significant parameters—the mesh size, applied velocity (hammer velocity) and time interval (step)—for the experiment to occur. Even minor modifications in these parameters influenced the overall energy values. After various combinations of remodelling the parameters to obtain results closer to the actual events, two parameters—time interval and mesh size—were fixed for all of the composite samples. The impact velocity of the hammer varied for each composite type, exponentially increasing from 40% rGF/EP, 100% EP, 60% rGF/EP and (40% and 60%) rCF/EP, in that order. In addition, during experimental IT, the hammer size increased exponentially as the fibre volume and the composite strength increased.

Figure 14 presents the impact energy results from experimental and numerical analyses for all of the composite types. The SD values from the experimental IT of each composite type—15 samples minimum per composite type—were incorporated as error bars. The numerical IT simulations are unified to eliminate the model's errors, and one test per material was performed, leaving no room for SD. As seen, the numerical results seem to be higher than the experimental results. As the fibre volume and sample strength decreased, the impact resistance of the samples also decreased gradually. The rCF/EP sample displays a high variation compared to other composite types. Such variation could be due to the local defects (poorly wet rCFs and high fibre volume) involved in the experimental IT samples. They possess a higher CoV between the sample population: 29.62 CoV for 60% rCF/EP and 38.40 CoV for 40% rCF/EP sample types. Meanwhile, the numerical IT was predicted based on the elastoplastic behaviour, in which, external defects were absent.

Similarly, the experimental IT rGF/EP samples, despite possessing a comparatively lower CoV of 18.48 for the 60% rGF/EP and 24.20 for the 40% rGF/EP sample types, contained random char distribution (local defects) across the sample. This significantly influenced fracture behaviour. Hence, the predicted IT also neglected such defects, exhibiting the impact results for defectless rGF/EP samples. In contrast, EP samples displayed explosive breaking during experimental IT due to their high hardness property, whereas the FE model tested the samples step-by-step, avoiding such explosive behaviour and recorded a lower impact behaviour than experimental IT.

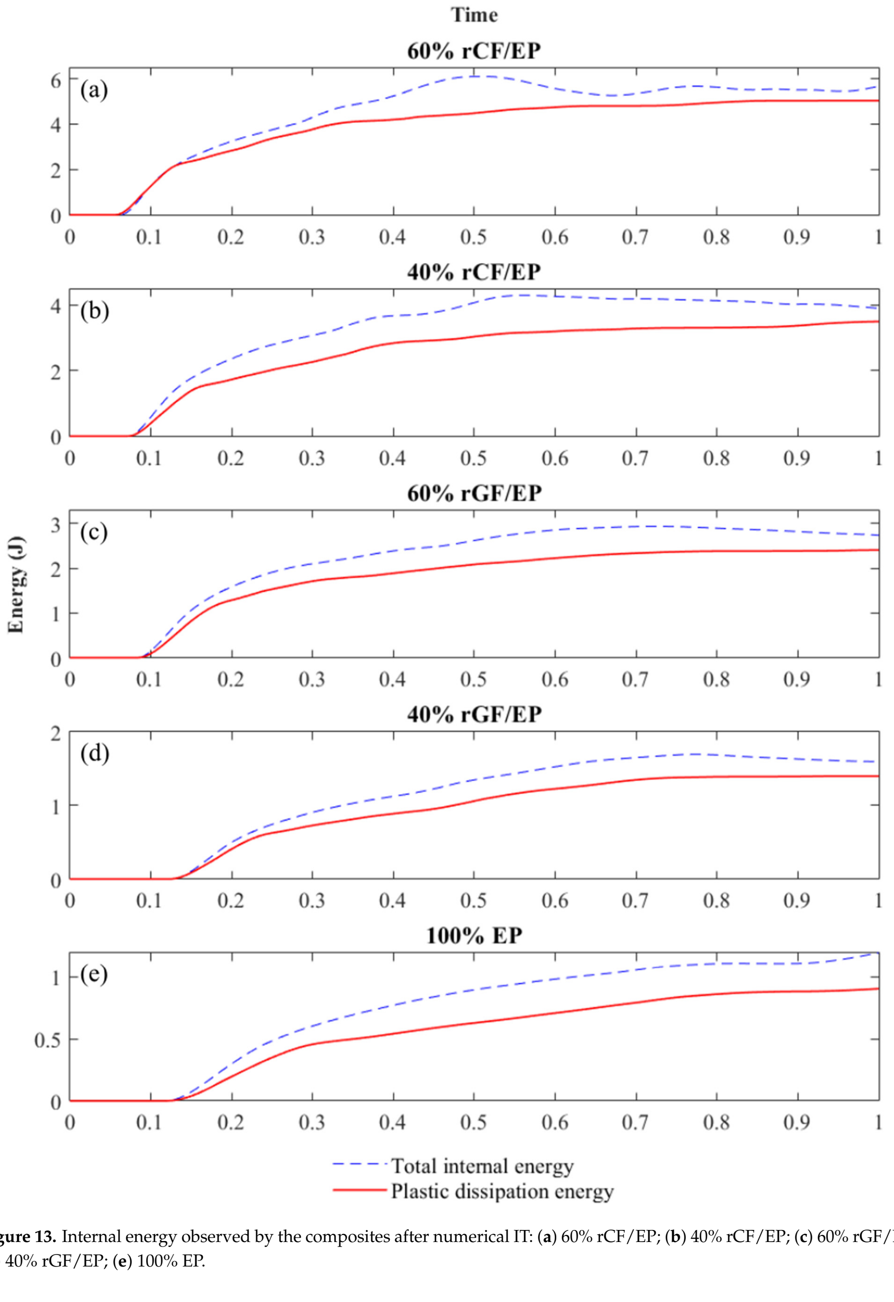
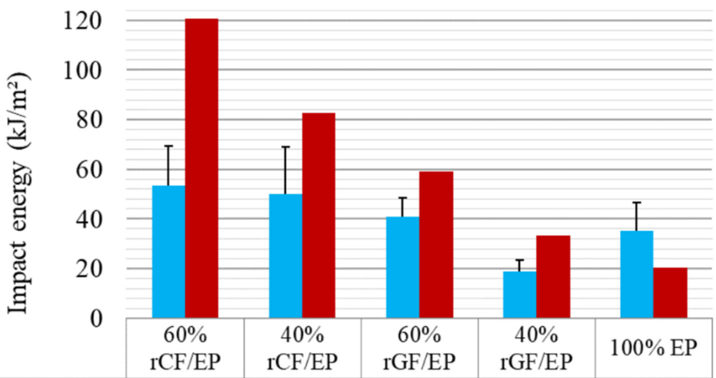


Figure 13. Internal energy observed by the composites after numerical IT: (**a**) 60% rCF/EP; (**b**) 40% rCF/EP; (**c**) 60% rGF/EP; (**d**) 40% rGF/EP; (**e**) 100% EP.



	60% rCF/EP	40% rCF/EP	60% rGF/EP	40% rGF/EP	100% EP
Experimental	53.61	49.98	41.05	18.99	35.18
Numerical	120.56	82.86	59.31	33.49	20.54

Figure 14. Measured impact strength of the composites experimental vs. numerical.

The energy difference within the 40% rGF/EP and 60% rGF/EP composite types in the experimental case is 22 kJ/m^2, and the numerical case is 25 kJ/m^2. However, the difference between 40 and 60% rCF/EP in the experimental case is 3.63 kJ/m^2, and in the numerical case is 37.7 kJ/m^2. Such a poor energy difference between 40 and 60% rCF/EP experimental IT can be related to previously mentioned defects, such as the poor wettability of rCF to reinforce with EP, as the energy pattern in other composite types seems to fall within a comparable order. However, the char-based defects on rGF/EP were reflected less. Overall, based on the energy variations in rGF/EP and rCF/EP, it can be concluded that the numerically observed energies are reliable in all composite types, and are defect-free.

Furthermore, when comparing the numerical IT results to literature-based experimental IT studies, the energy absorbed by 40% rCF/EP is 82.86 kJ/m^2, and by 60% rCF/EP is 120.46 kJ/m^2. Meanwhile, Caminero et al. 2016 [54] tested UD 66% vCF/EP using unnotched charpy IT and noticed a 189.01 kJ/m^2 internal energy. The study highlighted that, among various multidirectional vCF/EP laminates, the UD laminate possesses a high performance under impact and flexural testing. In addition, when comparing the energy absorbed by 40% rGF/EP, which is 1.4 kJ or 33.49 kJ/m^2, and 60% rGF/EP, which is 2.4 kJ or 59.31 kJ/m^2, to a similar study, Bazli et al. 2019 [55] tested UD 70.5% vGF/EP composites under unnotched charpy IT, which resulted in 5.6–7.1 kJ depending on the exposed temperature. The study also highlighted that UD vGF/EP displayed a higher performance in flexural and impact modes compared to a woven and randomly oriented fibre arrangement.

It can be noted that the numerical predicted IT results of the recycled composites are not identical when compared to literature studies with their virgin counterpart. Regardless, they are higher than their experimental IT results. This could be due to the lack of input parameters concerning the composite's shear properties. The impact direction is perpendicular to the fibre orientation, demanding interlaminar shear characters for higher-order modelling. In addition, the numerical IT model's sensitive status towards the mesh size, hammer velocity and time interval also significantly influence the results.

5. Conclusions

In this study, the recycled composites (40 and 60%) rCF/EP and rGF/EP, along with cured EP samples, were numerically investigated using FE methods. The study primarily investigated the uniaxial tensile and impact resistance properties of the recycled composites. The FE modellings were developed based on the elastoplastic behaviour and ductile damage failure. The experimentally measured uniaxial tensile results from the previous study [31] were utilised as input parameters for modelling. All of the composite types—EP, rCF/EP and rGF/EP—were successfully fractured using the developed models in tensile and impact loading conditions.

The FE models managed to record the non-linear behaviours of the recycled composites from the numerical TT. Simultaneously, it predicted the recycled composite's damage initiation and development under tensile loading. The numerical measured maximum principal stress and logarithmic strain from the dogbone-shaped samples showed that the applied displacement to failure increases as the overall strain rate increases. The fibreless EP samples possessed a higher displacement to failure (2 mm), which explains their higher strain rate and associated plasticity—followed by 60 and 40% rCF/EP and rGF/EP. The numerical results mapped the non-linear behaviour of the composites until damage initiation with a higher accuracy. However, the predicted damage behaviour did not precisely replicate the damage path. Regardless, they lie within an acceptable range.

The results from the contact-based FE models under impact loading conditions with rectangular samples and a hammer (impacter) were investigated using the von Mises stress distribution. The results showed that, as the fibre volume and the composite strength increased, the composites became stiffer, with the 60% rCF/EP sample being the stiffest and fibreless EP samples exhibiting a high plasticity. The 40% rCF/EP and 60% rGF/EP exhibited similar stress concentrations. Similarly, the impact velocity increased as the composite strength increased, making 60% rCF/EP samples the toughest and 40% rGF/EP the weakest to impact resistance. Furthermore, the damage behaviours for impact loading were investigated by the energy observations. The total internal energy and plastic dissipation energy were measured from the FE models. These observed energies were higher than their experimental impact energies, but lower than similar virgin composites from the literature. Such results were expected, as the FE models did not include any external defects from the recycling process that influenced the experimental results significantly, such as a poor resin wettability in the 40 and 60% rCF/EP samples, a bundle formation in high fibre volume fraction in the 60% rCF/EP samples and a char formation in the rGF/EP samples. Alternatively, the FE models predicted defect-free damage behaviours and energy absorption for the recycled composite.

The numerical IT was highly sensitive to three primary numerical parameters: sample mesh size, hammer velocity and experimental time (step time). A trivial change in these parameters will drastically influence the total internal energy absorbed by the samples. However, by fixing the mesh size and time step as constant and modifying the impact velocity based on the sample type, such defects can be overcome.

It is concluded that the FE methodology developed to numerically investigate the mechanical properties (tensile and impact) and predict the damage behaviour of rCF/EP and rGF/EP composites have shown substantial results. Hence, the adopted modelling technique can validate experimental results of recycled composites possessing complex and inconsistent mechanical behaviours. The study will provide insight towards investigating recycled composites containing defects from recycling methods numerically. Further improvements are required to overcome certain limitations. For meso-scale modelling, the recycled composite's material properties in compression, flexural and shear are required to obtain input parameters to define the elastic region of the fibres. By doing so, the composites can be defined as transversely isotropic and even anisotropic. For micro-scale modelling, the individual recycled fibre's tensile properties are required to define the FRP composites as heterogeneous solids in order to analyse the fracture mechanics under various loading conditions. The future research directions are aimed to optimise such limitations in order to map the mechanical properties and fracture behaviours of the recycled FRP composites closer to real-time.

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Publication IV

Karuppannan Gopalraj, S., Deviatkin, I., Horttanainen, M., and Kärki, T.
Life Cycle Assessment of a Thermal Recycling Process as an Alternative to Existing CFRP and GFRP Composite Wastes Management Options

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Article

Life Cycle Assessment of a Thermal Recycling Process as an Alternative to Existing CFRP and GFRP Composite Wastes Management Options

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Abstract: There are forecasts for the exponential increase in the generation of carbon fibre-reinforced polymer (CFRP) and glass fibre-reinforced polymer (GFRP) composite wastes containing valuable carbon and glass fibres. The recent adoption of these composites in wind turbines and aeroplanes has increased the amount of end-of-life waste from these applications. By adequately closing the life cycle loop, these enormous volumes of waste can partly satisfy the global demand for their virgin counterparts. Therefore, there is a need to properly dispose these composite wastes, with material recovery being the final target, thanks to the strict EU regulations for promoting recycling and reusing as the highest priorities in waste disposal options. In addition, the hefty taxation has almost brought about an end to landfills. These government regulations towards properly recycling these composite wastes have changed the industries' attitudes toward sustainable disposal approaches, and life cycle assessment (LCA) plays a vital role in this transition phase. This LCA study uses climate change results and fossil fuel consumptions to study the environmental impacts of a thermal recycling route to recycle and remanufacture CFRP and GFRP wastes into recycled rCFRP and rGFRP composites. Additionally, a comprehensive analysis was performed comparing with the traditional waste management options such as landfill, incineration with energy recovery and feedstock for cement kiln. Overall, the LCA results were favourable for CFRP wastes to be recycled using the thermal recycling route with lower environmental impacts. However, this contradicts GFRP wastes in which using them as feedstock in cement kiln production displayed more reduced environmental impacts than those thermally recycled to substitute virgin composite production.

Keywords: life cycle assessment; composite recycling; carbon fibre; glass fibre; waste disposal; thermal recycling

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1. Introduction

1.1. Background

Carbon fibre-reinforced polymer (CFRP) and glass fibre-reinforced polymer (GFRP) composites have been used in high-performance and lightweight applications such as renewable energy, automobiles, construction, aeronautics, aerospace, sports, and defence. The composite's incredible mechanical properties, especially, strength to weight ratio, attracted industries to utilise these composites in enormous quantities despite their hefty price range [1]. In 2020, the global composite market size reached USD 95.89 billion. By 2027, it is estimated to be at USD 160.54 billion. The majority of the shares were contributed by CFRP and GFRP composites in lightweight applications [2]. In particular, two applications, namely wind turbines (WTs) and aeroplanes, are notable for using CFRP and GFRP composites in higher volumes. Based on the 2021 report by the global wind energy council (GWEC) [3], it is necessary to achieve net-zero carbon dioxide emissions by

2050 in order to avoid climate change, and wind energy plays a significant role. Despite the impressive targets in the year 2020, the report suggests three times more WT installation requirements in the following years to reach such global targets.

Meanwhile, the cumulating end-of-life (EoL) WTs from 1980 need to be disposed of. As the new WTs and aeroplanes utilised a higher volume of CFRP and GFRP composites, they will be available for disposal after 25 years of usage. Overall, an estimation of 1 million tonnes of composite wastes will be generated by 2050 [4]. In these composites, the virgin carbon fibre (vCF) production is recognised as one of the most energy-consuming processes. It consumes 14 times more energy than conventional steel production [5]. However, it is indicated that replacing polyacrylonitrile (PAN) (commonly used precursor) with lignin can significantly reduce the environmental impacts and energy consumption to a certain extent [6]. However, the wastes have to be carefully recycled to reduce virgin fibre production. As the recycled carbon fibres (rCFs) have a higher potential to be reused in automobile industries. They have lower environmental impacts compared with materials typically used in automobile components, namely virgin (v) CFRP, aluminium, and conventional steel [7]. The virgin glass fibre (vGF) production is not as energy-intensive as CF production but is used in applications as GFRP composites in a heavier volume than CFRP composites [8]. However, the GFRP wastes are mainly used as feedstocks in cement production [9].

Earlier, there were no sustainable waste disposal methods studied and established. These composite wastes were typically landfilled and incinerated (energy recovery). After 2008, when the European Waste Frame Directive (2008/98/EC) [10] came into force, it initiated a benchmark on the waste disposal hierarchy, encouraging recycling as an exclusive waste disposal route. As a result, the landfilling taxation was increased. The cost of landfill in the UK stands at GBP 130–140 per tonne. Meanwhile, countries such as Germany and other EU states predominantly ban landfills and have promoted a circular economy in recent times [9].

1.2. Literature Review

Various studies have previously used life cycle assessment (LCA) to investigate the environmental effects of various disposal methods concerning CFRP and GFRP composite wastes. Witik et al. (2013) [11] studied three possible disposal methods for CFRP wastes, namely landfilling, incineration (energy recovery), and pyrolysis to recycle CFs (material recovery). They highlighted the advantages of replacing vCFs and vGFs with rCFs as it possesses similar mechanical properties and consumes less energy to recycle using pyrolysis than producing new fibres. However, the authors have assumed values for pyrolysis from various patents and reports making it highly uncertain. La rosa et al. (2016) [12] investigated a chemical recycling method using acetic acid to recycle CFRP wastes. Subsequently, the study used LCA methods, and highlighted that rCFs could substitute vCFs. However, the study failed to address the environmental effects of the hazardous chemicals used in the recycling process on an industrial scale. Plus, the fact that rCFs should be further treated to be reused (system boundary limitation). Li et al. (2016) [13] briefly studied mechanical recycling additional to landfilling and incineration. The study indicates that mechanical recycling is never a suitable CFRP waste disposal approach as the process is costly, 3000 EUR/tonne and does not have proper energy and material recovery paths.

Meng et al. (2017) [14] studied the fluidised bed process (FBP) to recycle CFRP wastes, and highlighted that recycling consumes 32–50% less energy and reduces 33–51% GWP compared with vCF production. Moreover, the recycling process has lower environmental and economical effects compared with traditional landfilling and incineration routes. However, the processes possess short and randomly oriented rCFs, limiting their reusing ability in various applications (substitution limitation). Subsequently, Meng et al. (2018) [15] investigated mechanical recycling, thermal recycling—pyrolysis and FBP and chemical recycling disposal methods. Overall, the study highlighted recycling CFRP wastes to recover the valuable carbon fibres from being the only sustainable alternative to landfilling and incineration.

The LCA results—GWP −19 to −27 kg CO_2-eq and PED −395 to −520 MJ/kg—indicate a substantial reduction in the environmental impacts. Later Meng et al. (2018) [16] continued the studies addressing the industrial-scale FBP, including the rCF's fibre alignment (drawbacks from the previous study). The study highlighted that only 15% of the energy from vCF production is required to operate an industrial-scale FBP with an overall recycling cost of 4.31 EUR/kg. Despite these well-established studies in FBP, a functional industrial-scale process has not been established.

The increase in the popularity of solvolysis at supercritical and subcritical conditions has also reflected an LCA-based study. Vo Dong et al. (2018) [17] briefly investigated the CFRP waste disposal method's economic and environmental aspects. They studied traditional methods such as landfilling, incineration and material recovery method—mechanical, thermal and solvolysis using supercritical water (SCW)—recycling processes. The study highlights the advantages of using thermal recycling and solvolysis waste management methods using the GWP indicator. Despite their higher price range than landfilling and incineration, the rCFs can substitute vCFs and vGFs with minimum environmental effects. However, there are many practical difficulties in establishing an industrial-scale solvolysis plant. Khalil (2018) [18] performed an in-depth LCA study and developed gate-to-gate recycling models keeping pyrolysis as baseline and solvolysis as an alternative approach. Overall, based on nine impact categories, the study concluded that pyrolysis possesses lower environmental and human health-based impacts compared with solvolysis to recycle CFRP wastes. Pillain et al. (2019) [19] performed a comparative LCA study. They analysed the sustainability aspects and highlighted the advantages of recycling the CFRP wastes by solvolysis using SCW parallel to pyrolysis, electrodynamic fragmentation (mechanical recycling) and pointed out the disadvantages of traditional disposal methods such as incineration and landfill. Similar to the LCA studies, Liu et al. (2019) [20] followed an ecoaudit approach utilising energy consumption to measure the environmental impacts to compare EoL options for WT blades. The study highlighted the solvolysis approach as the future of CFRP and GFRP waste management options for EoL WT applications.

A recent study by Meng et al. (2020) [21] utilised LCA to investigate the possibility to replace vGFs with aligned rCFs in aviation applications. As 500,000 tonnes of EoL aeroplane are expected by 2050, recycling the CFRP wastes using FBP and replacing the vGFs in the aviation application promotes closed-loop recycling with a higher reduction in environmental and economical aspects. However, Tapper et al. (2020) [22] reviewed the LCA studies to analyse the closed-loop CFRP waste disposal methods and pointed out pyrolysis as preferred to FBP in recycling CFRP wastes thermally. Furthermore, La Rosa et al. (2021) [23] studied both closed-loop and open-loop using LCA for recycling CFRP wastes using chemical recycling, and highlighted the advantages of the open-loop approach being more realistic and cost-efficient.

Overall, specific research gaps were noticed from the literature review. It can be seen that the majority of the LCA studies focused only on CFRP waste disposal options. Even though certain studies include discussions about substituting vGFs with rCFs. There are no significant LCA studies dedicated to analysing the GFRP waste disposal methods, as the modern recycling (material recovery) methods for CFRP wastes are similar to GFRP waste. Additionally, most LCA studies involving CFRP waste disposal possess inventory analysis taken from other studies and reports. Only a few works of literature have performed LCA studies for their personally developed recycling methods.

This study primarily aims to perform an LCA assessment on a recycling route that thermally recycles the CFRP and GFRP wastes and remanufactures the recycled fibres into recycled (r) CFRP and rGFRP composites employing a fresh epoxy resin system using a compression moulding process. Furthermore, LCA was performed over three traditionally practised waste management options: landfilling, incineration, and feedstock in a cement kiln (co-incineration). Finally, a comprehensive LCA assessment was conducted by comparing the LCA results using Global Warming Potential (GWP) and Abiotic Depletion Potential for fossil fuels (ADP_f) indicators to investigate the feasibility of the thermal recycling process as an alternative to the traditional waste management options. Overall, this study will provide insights into the waste management options of CFRP and GFRP wastes, with material recovery being the highest priority to close the life cycle loop of the composites and encourage a circular economy.

2. Methodology

2.1. Studied Waste

In this study, manufacturing wastes (pre-consumer) of CFRP and GFRP composites from domestic applications were subjected to study. The wastes were provided by Excel composites Oyj (Heinävaara, Finland). The CFRP wastes with a density of 1.81 g/cm^3 possessed 55.5 wt% of unidirectional vCF and 44.5 wt% cured epoxy resin. The GFRP wastes with a density of 1.52 g/cm^3 possessed a 44 wt% laminated thin-ply vGF structure and 56 wt% unsaturated polyester resin (UPR). Apart from the mentioned features, the mechanical properties of the composites were unknown. Overall, it was assumed that the share of carbon fibres and glass fibres in the composites was 55 wt% for this study. The rest was matrix polymers–epoxy resin in the case of CFRP and UPR in the case of GFRP. Furthermore, these composite wastes were recycled and remanufactured to produce rCFRP and rGFRP composites employing fresh resins with the rCFs and recycled glass fibres (rGFs) to close their life cycle loop.

2.2. Thermal Recycling Route

Figure 1 presents the overall thermal recycling route. The thermal recycling process used for this LCA study is from the author's previous research work [24]. The process involves incineration and combustion principles using heat radiation in a controlled environment. The developed process is capable of recycling both CFRP and GFRP wastes. The process uses heat flux at 50 kW/m^2 to recycle these composite wastes separately in an open chamber batch reactor in the presence of air. At 550 °C in atmospheric pressure, the epoxy resin from CFRP wastes was completely evaporated in 20–25 min, leaving clean rCFs in the reactor. Similarly, the UPR from GFRP wastes evaporated in 25–30 min, leaving clean rGFs. The evidence for maximum resin removal from the recycled fibre's surface was investigated by employing a scanning electron microscope.

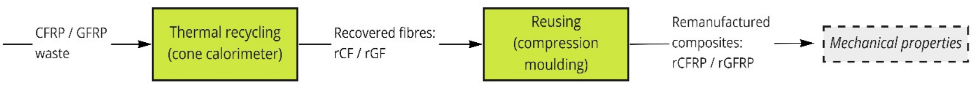

Figure 1. Performed thermal recycling route (modified from [24]).

The recycled fibres, both rCFs and rGFs, possessed a unidirectional (0), long (105 ± 2 mm) and continuous (end-to-end) fibre arrangement. These fibres were reused by reinforcing with fresh laminating epoxy resin and hardener in a 2:1 ratio using compression moulding. The newly produced composites were further experimentally [24] and numerically [25] examined. Table 1 presents the experimental mechanical properties measured from the produced composites. As seen, two types for each composite were produced based on the fibre (V^f) and resin volume fraction (V^r). Overall, the process is capable of recycling CFRP and GFRP composite wastes with a fibre recovery rate of 95–98 wt% for rCFs and 80–82 wt% for rGFs.

Table 1. Mechanical properties of the produced rCFRP and rGFRP composites [24,25].

Composite Recipes	V^f (wt%)	V^r (wt%)	Tensile Strength (MPa)	Young Modulus (GPa)	Impact Strength (kJ/m^2)	Fracture Strain (No Unit)	Density (g/cm^3)	Poisson Ratio
rCFRP	60 ± 2	40 ± 2	235.70	60.80	53.61	0.00683	1.52	1.52
	40 ± 2	60 ± 2	210.34	45.28	49.98	0.00827	1.64	1.64
rGFRP	60 ± 2	40 ± 2	114.58	30.72	41.05	0.00272	1.77	1.77
	40 ± 2	60 ± 2	65.42	27.37	18.99	0.00156	1.85	1.85

2.3. Life Cycle Assessment

LCA methodology was primarily performed based on the ISO 14040 [26] and ISO 14044 [27] standards to investigate the respective impacts in various disposal routes. The LCA was modelled using GaBi software (version 9.0.0.42, DP service pack 38) provided by Sphera Solutions GmbH, Leinfelden-Echterdingen, Germany [28].

2.3.1. Goal and Scope Definition

The goal of this LCA study was to model the climate change impacts and depletion potential of abiotic resources of the developed thermal recycling process for CFRP and GFRP waste. Furthermore, the study compares the impacts of this process with other traditional waste disposal methods practised for CFRP and GFRP wastes, namely landfilling, incineration with energy recovery, and the use as feedstock in the cement production. The function of the studied product system is to treat or dispose of CFRP and GFRP wastes. Therefore, the functional unit is 1 kg of CFRP waste and 1 kg of GFRP waste collected in Finland. The study utilises a gate-to-grave (or a bin-to-grave) approach: the assessment of the environmental impacts started from the point of waste generation, thus accounting for its transportation to the treatment facility and ended with the waste being treated. The system expansion approach was utilised to account for the multifunctionality of the studied product system with several scenarios.

Figures 2 and 3 presents the system boundaries and expanded systems for CFRP and GFRP waste's disposal scenarios. The scenarios are placed in the descending order of the EU regulations [10] for waste hierarchy, where recycling is the highest priority. The CFRP and GFRP wastes were considered carrying no burdens from their previous life cycle stages, i.e., so-called "zero burden" approach. For the CF recycling sector, GWP was considered an essential LCA indicator [29]. The life cycle impact assessment was performed for the GWP and ADP_f using the characterisation method of the product environmental footprint implemented in GaBi software as "EF 2.0 (Environmental Footprint 2.0)" [30].

The four scenarios concerning this product system were the thermal recycling process (scenario 4) from the composite wastes to rCFRP and rGFRP production. It consists of two system expansions, first to substitute with virgin composite production and the thermal energy produced during recycling to substitute with natural gas. Cement kiln production (scenario 3) involves hard coal mix for both the composites and an additional bauxite mix for GFRP wastes. The incineration (scenario 2) process utilises energy recovery and an expanded system for electricity and thermal energy substitutions. Finally, landfilling (scenario 1) is used without any possible substitutions.

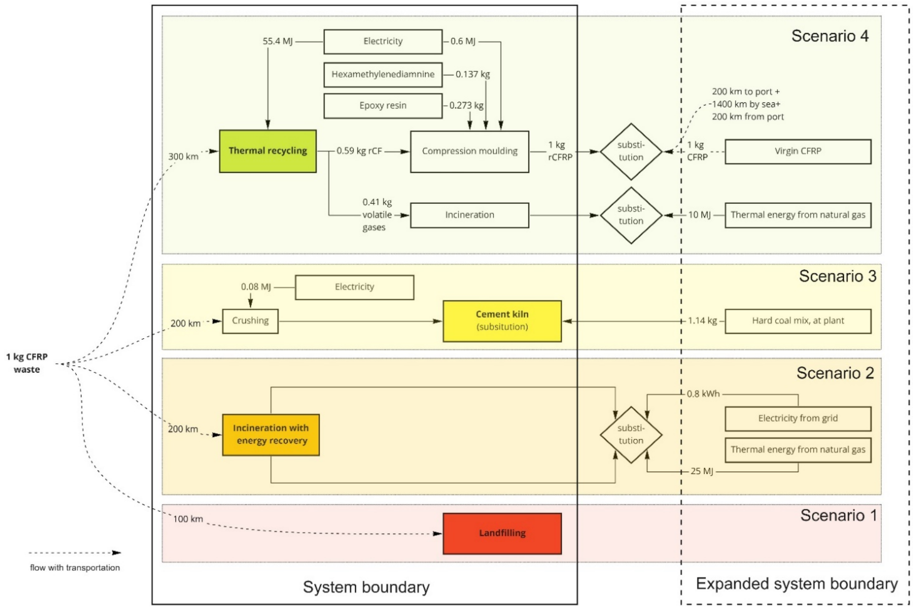

Figure 2. System boundaries of the studied and expanded systems for CFRP waste management.

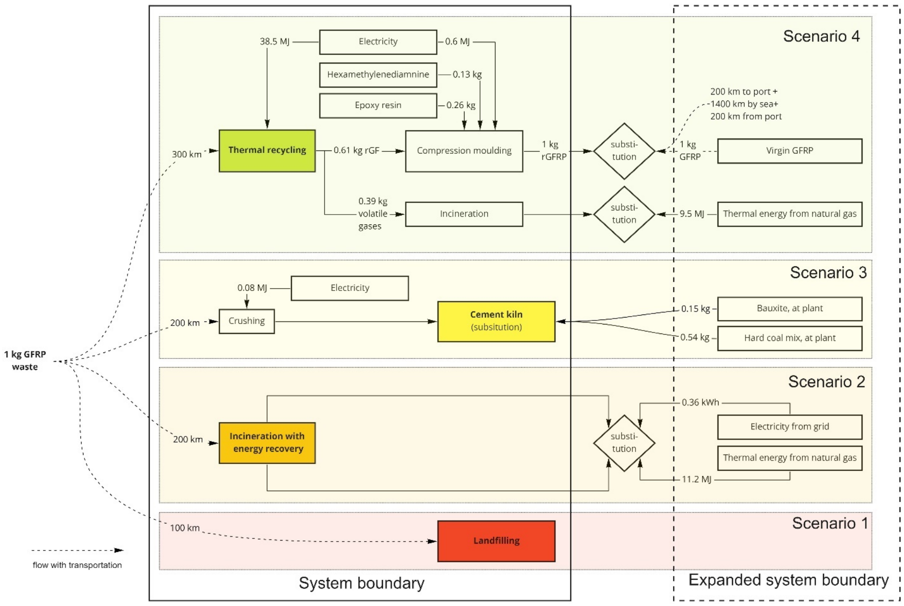

Figure 3. System boundaries of the studied and expanded systems for GFRP waste management.

2.3.2. Life Cycle Inventory

Thermal recycling of composite waste: The data on thermal recycling of CFRP and GFRP wastes were obtained from the laboratory-scale equipment. Recycling 1 kg of CFRP wastes consumed 15.4 kWh of electricity under stabilised process conditions and yielded 0.59 kg of rCFs. Recycling 1 kg of GFRP wastes consumed less electricity than CFRP wastes, 10.7 kWh, and yielded 0.61 kg of rGFs. The rest of the resin system's mass in the composite wastes was gasified and removed from the system. The gases were not captured in the laboratory conditions but were expected to happen on an industrial scale to regenerate the heat within the system. Therefore, the incineration of the gases was modelled in this study. The process was modelled using the unit process "FI: Plastics (unspecified) in waste incineration plant". This process is expected to represent the impact from incinerating the resin system vaporised during the process. The thermal energy generated during the incineration process was substituted with thermal energy from natural gas using the process "FI: Thermal energy from natural gas".

Once the fibres (rCFs and rGFs) were recycled, they were used to produce rCFRP and rGFRP composites. These recycled fibres were mixed with laminating epoxy resin and hardener in a 2:1 ratio. The produced recycled composites were assumed to have 60 wt% recycled fibre volume fraction and 40 wt% epoxy resin. A low energy-consuming compression moulding technique was used to produce the new composites. The process required 0.167 kWh to produce 1 kg of rCFRP or rGFRP. The process used for epoxy production was "DE: Epoxy resin (EP) mix". The hardener was modelled using the process "EU-28: Hexamethylenediamine (HMDA); from butadiene via adiponitrile". The produced rCFRP and rGFRP were compared with the virgin polymers.

Production of virgin CFRP: To produce vCFRP, CFs needed to be produced first. Their production was modelled using the data reported elsewhere [5,31]. The inventory of the production process and the unit processes used are presented in Table 2.

Table 2. Life cycle inventory of the carbon fibre production process [5,31].

	Amount	Unit	Unit Process
Inputs			
Amonium bicarbonate	0.02	kg	RER: market for ammonium bicarbonate ecoinvent 3.6
Epoxy resin	0.01	kg	DE: Epoxy resin (EP) mix
Polyacrylonitrile fibres	1.89	kg	EU-28: Polyacrylonitrile Fibres (PAN)
Polydimethylsolixane	0.01	kg	GLO: market for polydimethylsiloxane ecoinvent 3.6
Potassium permanganate	0.1	kg	GLO: market for potassium permanganate ecoinvent 3.6
Sulphuric acid	0.02	kg	EU-28: Sulphuric acid (96%)
Water	2.77	l	EU-28: Process water from surface water
Electricity	20.2	kWh	EU-28: Electricity from grid mix
Heat	98.4	MJ	EU-28: Thermal energy from natural gas
Outputs			
Carbon fibres	1	kg	-
Carbon dioxide	0.63	kg	-
Nitrogen monoxide	0.33	kg	-
Nitrogen dioxide	0.66	kg	-

The CFRP composite's composition was assumed in this study, corresponding to the results of the thermal recycling experiments. Thermal recycling of CFRP yielded 59% of rCF. However, a part of this mass is epoxy resin which did not gasify. Therefore, in this study, the share of carbon fibres in virgin CFRP was assumed to be 55 wt%. The rest of the mass was epoxy resin and hardener mixed in the ratio of 2:1, as is the case with the rCFRP production. The CFRP production was modelled using the low-pressure resing transfer moulding (LPRTM) process. Vita et al. [32] reported the energy consumption of the LPRTM process for producing one car CFRP hood. The energy consumption of the pre-forming, moulding, mixing, and metering stages of the process equals 2.85 kWh per 1 kg of CFRP produced.

Production of virgin GFRP: The production of vGFRP was modelled as mixing 0.55 kg of vGFs ("DE: Glass fibres") with 0.45 kg UPR ("DE: Polyester resin (unsaturated) (UP)"). Electricity consumption was modelled in the same way as in the production of the CFRP, which is equal to 2.85 kWh [32].

Use of composite waste in a cement kiln: Before the co-incineration of CFRP and GFRP wastes in a cement kiln, the waste is crushed to ensure suitable particle size. The electricity consumption of waste crushing was taken from the study by Witik et al. [11] and equalled 0.09 MJ per 1 kg of crushed composite waste. In the cement kiln, both composite wastes were assumed to substitute coal. The substitution was calculated based on the energy content of composite waste. The higher heating value (HHV) of CFRP was 32 MJ/kg, and the HHV of coal was 28 MJ/kg, so the mass of coal substituted with 1 kg of CFRP waste was 1.14 kg.

In the case of GFRP, the calculation took into account only the HHV of UPR since GFs are mineral in nature. The HHV of UPR was 33.5 MJ/kg, and accounting for its share of 45% in the GFRP waste, the mass of substituted coal was 0.54 kg. At the same time, GFs were assumed to substitute bauxite due to their high aluminium content. The aluminium oxide content in E-glass fibre is 8%, and that in bauxite is 50%. Therefore, 1 kg of GFRP waste can substitute 0.15 kg of bauxite in the process. The emissions from the cement kiln were modelled based on the carbon content of the fuels: 92% for CFRP, 61.4% for UPR, and 65% for coal. The substituted products were modelled using the following processes: coal—"FI: Hard coal mix", and bauxite—"EU-28: Bauxite". Coal transportation was included in the unit process, whereas bauxite was reported in a previous study by Khan et al. [33].

Incineration of composite waste: The incineration of CFRP and GFRP waste was modelled using available unit processes. The incineration of CFRP waste was modelled using the process "EU-28: Plastic (unspecified) in waste incineration plant" since its heating value and composition are similar to plastics. The incineration of GFRP was modelled according to the content of GFs and UPR. The GFs incineration was modelled using the process "FI: Inert material in waste incineration plant", whereas incineration of UPR was modelled in the same way as for the CFRP waste. The efficiencies of heat and electricity generation were adjusted to represent the specific condition of Nordic countries: 9.6% for electricity generation and 82.9% for heat generation.

Landfilling of composite waste: Landfilling of CFRP and GFRP wastes were modelled in the same way, unlike in the case of incineration because both waste types are not biodegradable are expected to behave similarly in the landfill. Landfilling was modelled using the process "EU-28: Plastic waste on landfill".

Transportation: Table 3 shows the distances and transportation modes used in the study. The distance to the disposal facilities was assumed based on their availability: landfills are the most common disposal sites, so they have the shortest distance, whereas a thermal recycling facility would be the most scarcely located.

Table 3. Transportation distances and modes.

Flow	from	to	Distance	Transportation Mode
CFRP waste/GFRP waste	Generation place	Recycling facility	300 km	Truck [1]
		Cement kiln	200 km	Truck [1]
		Incineration plant	200 km	Truck [1]
		Landfill	100 km	Truck [1]
rCFRP/rGFRP	Recycling facility	Customer	100 km	Truck [1]
vCFRP/vGFRP	Production	Port in Germany	200 km	Truck [1]
	Port in Germany	Port in Finland	1400 km	Sea-going container ship [2]
	Port in Finland	Consumer	200 km	Truck [1]

[1]—GLO: Truck, Euro, 5, 28–32 tonne gross weight/22 tonne payload capacity; [2]—EU-28: Container ship ocean incl. fuel, 27,500 dwt payload capacity, ocean-going.

2.3.3. Sensitivity Analysis

The sensitivity analysis was performed for the composites by varying the parameters concerning electricity consumption involved in the production process and the product substitution. Amongst the impacts to produce rCFRP and rGFRP composite, the electricity consumption to thermally recycle the composite wastes is expected to be high and will significantly influence the impacts. In CFRP composites, the possibility to replace 100% of vCFRP with rCFRP (1:1) is practically not possible. Therefore, various possible ratios were taken under consideration. Similarly, in GFRP composites, the vGF production is less energy-intensive than producing rGF and vCF. Hence, the possibilities in reducing the energy consumption to produce rGF were taken under consideration. Overall, the sensitivity analysis was conducted using break-even point analysis.

3. Results

3.1. Impacts from Recycled Composite Production

3.1.1. rCFRP Production

Figure 4 presents the GWP and ADP_f impacts from thermally recycling the CFRP wastes and rCFRP production. As seen, the overall carbon footprint of producing 1 kg of rCFRP was 5.68 kg CO_2-eq. The largest share of impact 3.07 kg CO_2-eq (53.87%) came from electricity consumption to generate the designated heat flux of 50 kW/m^2 in order to evaporate the epoxy resin from the composite wastes. The second-largest contribution of 2.30 kg CO_2-eq (40.5%) was from the production of epoxy and hardener (resin system) to be employed to produce rCFRP composites. Electricity consumed by compression moulding possessed a minor impact of 0.03 kg CO_2-eq (0.53%) on the results. Finally, the incineration of exhaust fumes from thermal recycling generated was 0.94 CO_2-eq. (16.55%). However, it was substituted by energy produced from natural gas, whose production emits 0.65 CO_2-eq. The ADP_f results to produce 1 kg of rCFRP composites were 122.31 MJ. Similar to GWP results, most of its impact, 81.42 MJ (66.56%), comes from the electricity involved in the recycling process. Subsequently, the resin system holds 50.88 MJ (41.60%). Finally, the lower impact contributions were from electricity consumption in compression moulding at 0.88 MJ (0.72%) and the incineration of exhaust fumes at 0.23 MJ (0.19%). The substituted energy produced from natural gas was 11.10 MJ (9.07%).

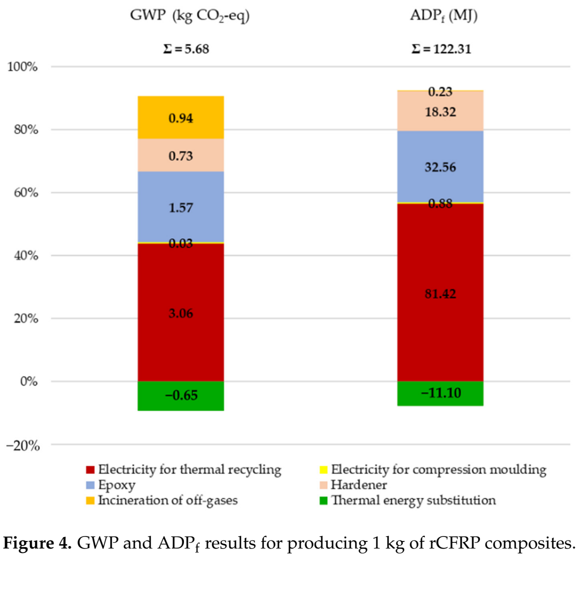

Figure 4. GWP and ADP_f results for producing 1 kg of rCFRP composites.

3.1.2. rGFRP Production

Figure 5 presents the GWP and ADP_f impacts from thermally recycling the GFRP wastes and rGFRP production. As seen, the overall carbon footprint to produce 1 kg of rGFRP composites was 4.62 kg CO_2-eq. The maximum impact was from the epoxy and hardener (resin system) production 2.19 kg CO_2-eq (47.40%). At the same time, the electricity consumption in the thermal recycling process possesses similar emissions of 2.13 kg CO_2-eq (46.10%). The emissions from the incineration of exhaust fumes from the recycling process were 0.94 kg CO_2-eq (20.34%) and were substituted by 0.65 kg CO_2-eq of energy produced by the natural gas. Finally, the minimum emissions were 0.03 kg CO_2-eq (0.65) electricity consumed by compression moulding. The ADP_f results to produce 1 kg of rGFRP composites were 95.50 MJ. The electricity consumption during the recycling process possess the maximum impact of 56.57 MJ, and impacts from resin system production were 48.40 MJ. The percentage of contribution for both these impacts was 59.23% and 50.68%. The incineration of off-gases and electricity to compression moulding holds the lowest impacts at 0.22 MJ and 0.88 MJ. At the same time, the gas emissions were substituted with 10.56 MJ energy produced from natural gas.

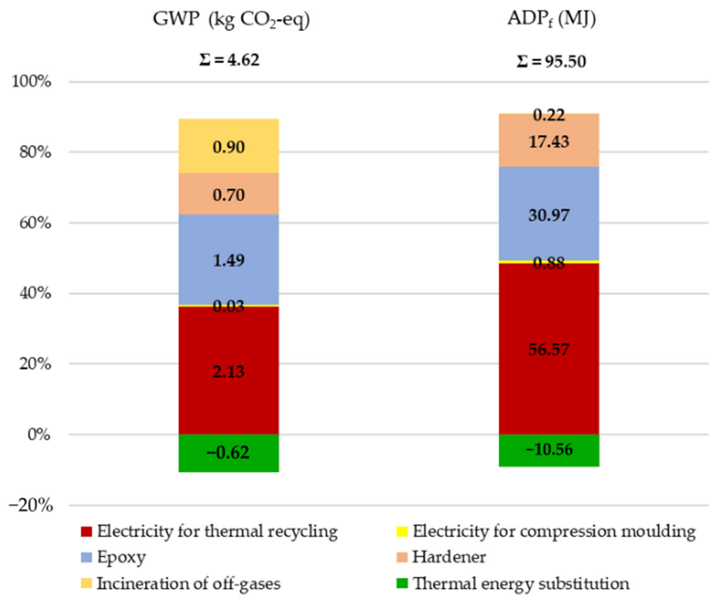

Figure 5. GWP and ADP_f results for producing 1 kg of rGFRP composites.

3.2. Environmental Impacts: Gate-to-Grave

3.2.1. CFRP Waste Disposal Methods

Figure 6 presents the GWP results for 1 kg of CFRP waste disposal using thermal recycling, cement kiln, incineration, and landfill. As seen, the thermal recycling route appeared to have an immense potential to reduce the impacts of climate change and preserve fossil resources compared with other disposals scenarios. Such high reduction potentials were enabled through the substitutions from vCFRP production. The production of 1 kg of vCFRP has a GWP of 17.20 kg CO_2-eq. The impacts from producing vCF alone hold 14.11 kg CO_2-eq (82.03%), and the remaining impacts were contributed from the epoxy resin system and electricity for the resin transfer moulding process. Therefore, producing

1 kg of rCFRP reduces the overall GWP by 11.53 kg CO_2-eq, including the direct emissions from recycling and the avoided impact.

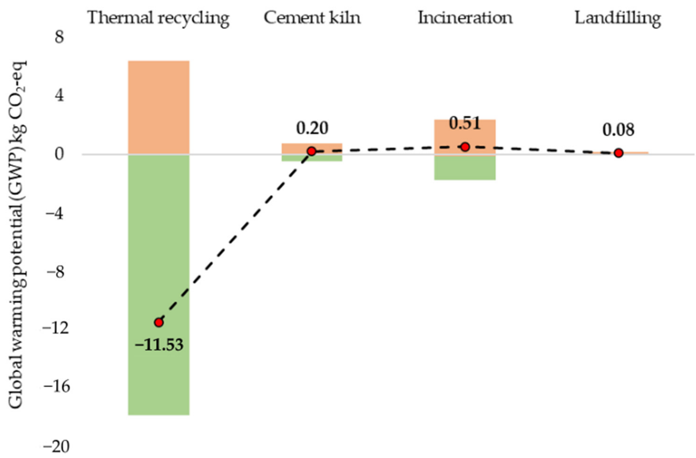

Figure 6. GWP of various CFRP waste disposal scenarios.

The GWP of the other disposal scenarios were relatively close, ranging at 0.08–0.51 kg CO_2-eq. Amongst which landfilling has the lowest impact of 0.08 kg CO_2-eq per 1 kg of CFRP waste disposal. No landfill gas was generated. The emissions were from the working machinery in landfills. The emissions from incineration were 2.30 kg CO_2-eq. It was almost twice higher than the 0.94 kg CO_2-eq emissions from incinerating exhaust fumes generated during the thermal recycling. The energy recovered from incineration could substitute 1.80 kg CO_2-eq in electricity and thermal energy but completely burn the CFRP waste leaving only ashes behind. Finally, the cement kiln route possessed 0.20 kg CO_2-eq emissions with 0.47 kg CO_2-eq hard coal substitution. Overall CFRP waste disposal route via thermal recycling seems to be more efficient and sustainable than other disposal scenarios. Based on the sensitivity analysis, the rCFRP composites should be capable of replacing $\geq$ 30% vCFRP composites, as the overall GWP will be 0.51 kg CO_2-eq, equal to the impacts from the incineration disposal route (non-sustainable). Any substitution ratio < 30% will result in higher impacts compared with other traditional disposal routes.

Figure 7 presents the ADP_f results for 1 kg of CFRP wastes disposal using various scenarios. Similar to GWP results of the thermal recycling route, the ADP_f is reduced by 214 MJ per 1 kg of rCFRP, primarily due to avoided production of vCF from substitutions. After thermal recycling, cement kiln's hard coal substitution have reduced the impacts by 34 MJ. Similarly, the energy substitutions from incineration have reduced impacts by 31.28 MJ. Landfilling impacts were 1.14 MJ without any possibility for substitution.

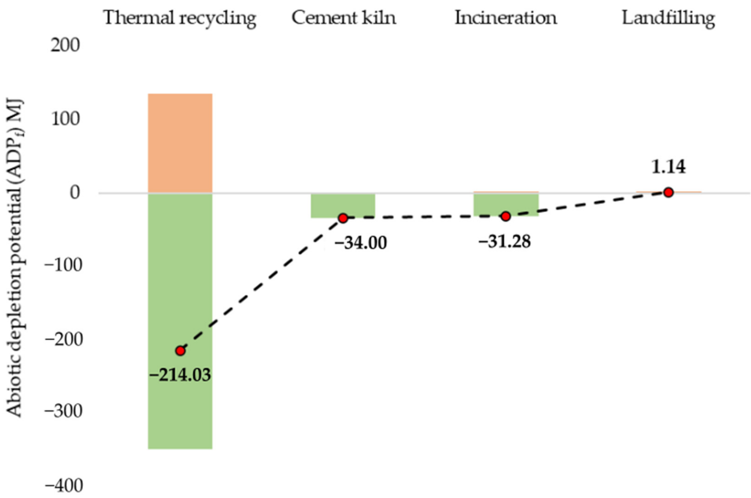

Figure 7. ADP_f of various CFRP waste disposal scenarios.

3.2.2. GFRP Waste Disposal Methods

Figure 8 presents the GWP results for 1 kg of GFRP waste disposal using thermal recycling, cement kiln, incineration and landfill. As seen, the cement kiln possesses a reduced GWP impact of 0.47 kg CO_2-eq due to GFRP being favourable for cement kiln production along with hard coal and bauxite substitution. The impacts from the thermal recycling route were 1.17 kg CO_2-eq, including the substitutions from vGFRP production. Despite the 30.71% lower energy consumption in thermal recycling GFRP wastes compared with CFRP wastes, the higher impact values were reflected due to the lower substitution values from vGF production. As the vCF production was highly energy-intensive with 17.20 kg CO_2-eq, substitutions with rCFRP composites 5.38 kg CO_2-eq significantly reduced overall emissions, whereas in the case of vGF production, the GWP emissions were 3.43 kg CO_2-eq compared with the GWP emissions of 4.62 kg CO_2-eq from producing rGFRP. Overall, 34.70% additional emissions were created by producing rGFRP composites. The emissions from incineration were 1.09 kg CO_2-eq, whereas the reductions from the energy substitution were 0.80 kg CO_2-eq. Finally, landfilling with 0.08 kg CO_2-eq emissions was similar to the landfill emissions from CFRP wastes.

Based on the sensitivity analysis, reducing 40% of emissions from energy consumption for the thermal recycling process will result in impacts similar to incineration. Reducing $\approx$ 77% impacts results in emissions similar to the cement kiln route. However, for scenarios such as incineration and cement kiln, when end-of-life WTs are used as GFRP waste sources, high energy-intensive processes such as shredding (size reduction) should be used to reduce the enormous GFRP wastes size from 60–70 m to 5–6 mm. Overall, the cement kiln route seems to be more sustainable than the thermal recycling route.

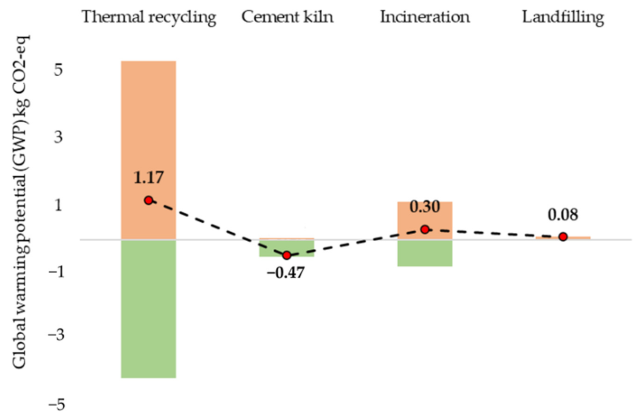

Figure 8. GWP of various GFRP waste disposal scenarios.

Figure 9 presents the ADP_f results for 1 kg of GFRP wastes using various disposal scenarios. As seen, the thermal recycling route displays a higher impact of 26.08 MJ, including the substitutions from vGFRP production with a UPR system. Subsequently, cement kiln 15.83 MJ and incineration 13.07 MJ had comparable reduced impacts after their respective substitutions. The landfill possessed 1.14 MJ.

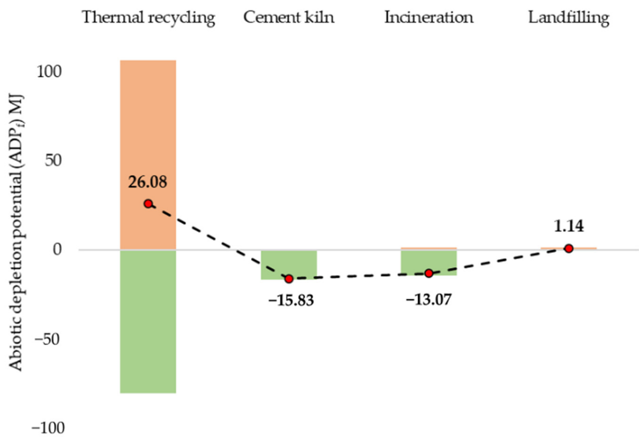

Figure 9. ADP_f of various GFRP waste disposal scenarios.

4. Discussion

The traditional waste disposal routes, namely landfill and incineration (energy recovery), have unfavourable environmental impacts. As mentioned in the literature [1,22,34], the hefty landfill taxation has established waste management industries to avoid landfills altogether. Furthermore, considering the price of vCFRP and vGFRP composites, the popularity of disposal via incineration has also reduced drastically. In particular, the low energy recovery rate with higher emissions and piles of ashes after incineration made it an undesirable disposal route for CFRP and GFRP wastes. The LCA results for CFRP waste disposal via incineration were higher than GFRP wastes. Most CFRP grades are thermal insulators, and heat-based approaches require added effort to react with CFRP composites. Such behaviours are visible even in the adopted thermal recycling route with higher energy consumption to recycle CFRP wastes than GFRP wastes.

The LCA results for CFRP and GFRP waste disposal via cement kiln were expected. The CFRP wastes are not suitable to use as feedstocks in cement production, but perform better than incineration with comparatively lower environmental impacts. On the other hand, the results for GFRP wastes were surprisingly positive compared with the proposed thermal recycling route with material recovery. GFRP composites majorly consist of E-glass made from aluminium borosilicate, calcium-based fillers and resin. When used in the cement kiln, the resin evaporation provides heat, and the minerals are used as feedstocks in cement production. Overall, GFRP can be disposed of without any residues. However, the presence of boron makes it challenging to replace 100% of cement kiln fuel with GFRP wastes but can be used to a certain extent. Moreover, the GFRP wastes must not contain impurities or metals, and the waste should be size reduced to fit in [35,36], which is highly energy consuming when considering EoL WTs with 60–70 m-long components.

The GWP and ADP_f results to produce rCFRP and rGFRP composites highlighted the maximum impacts from energy consumption during the recycling phase. The measured electricity consumption to thermally recycle 1 kg of CFRP wastes was 55.44 MJ, and GFRP wastes were 38.52 MJ. However, considering an upscaling to pilot process and further towards industrial-scale process, the thermal heat produced during the resin evaporated will be regenerated into the system. Hence, reduce the electricity consumption to a certain extend. Additionally, replacing the electricity source with renewable energies will result in maximum sustainable outcomes with lower environmental emissions. Still, at this laboratory scale, it is required to compare the LCA results with currently dominating recycling processes as highlighted from the previous study [1], namely pyrolysis and solvolysis using supercritical/ subcritical water or mild solvents. LCA studies involving pyrolysis had mostly adopted inventory data from industrial-scale pyrolysis operated by ELG carbon fibres (UK), at present named Gen2carbon. The pyrolysis energy consumption was 30 MJ per kg of CFRP waste recycling with a 2000 tonne/year capacity, resulting in GWP 2.88 kg CO_2-eq [15]. For solvolysis, a recent study by Liu et al. 2021 [37] recycled 1 kg of CFRP wastes via solvolysis using supercritical n-butanol at 49.21 MJ/kg. The LCA study by Nunes et al. 2018 [38] highlighted the energy required to recycle 1 kg of CFRP waste via steam thermolysis to be 54 MJ. Additionally, 17.64 MJ energy was spent to pre-process (cutting) 1 kg of CFRP waste.

Furthermore, various studies have compared the LCA results of pyrolysis and solvolysis. Khalil (2018) [18] highlighted the energy required to recycle 1 kg of CFRP waste via pyrolysis to be 12.53 MJ with 0.96 kg CO_2-eq GWP and 25 MJ via solvolysis with 16.2 kg CO_2-eq GWP. However, an additional 145 MJ of energy is required for solvolysis to produce the supercritical state for water (4.67 kg) to recycle 1 kg of CFRP waste. These results have significantly lower environmental impacts compared with the thermal recycling route from this study. However, a significant LCA study by Vo Dong et al. (2018) [17] showed −19 to −22 kg CO_2-eq GWP impacts to dispose of 1 kg of CFRP waste via pyrolysis and solvolysis SCW, including the substitutions with vCFRP production. In comparison, the results from this study have impacts of −11.53 kg CO_2-eq GWP, including the substitution from vCFRP production. He et al. (2020) [39] studied pyrolysis and solvolysis to recycle CFRP waste. They focused on the energy demand during pre-recycling and post-recycling phases and highlighted that energy conservation is possible if the recycled carbon fibre's structural integrity is maintained. Similar to the fibre arrangements from the thermal recycling process [24], resulting rCFs and rGFs were unidirectional, long and continuous. Overall, as noticed from the LCA studies, no significant studies were available to compare the disposal routes of GFRP composites. However, some studies [11,17,21] have proposed rCF to replace vGFs. Such research gaps can be understood from the LCA result of this study as they could be related to the advantages of the cement kiln process (energy recovery), the defects (char formation) from recycling GFRP wastes, and rGFRP's reduced mechanical properties failing to replace vGFRP composites, even to a minimum extent.

There are certain limitations faced in this study, such as the vCFRP and vGFRP production being considered perfect without any wastage, which is practically not possible. In incineration modelling, CFRP and GFRP wastes were assumed to be plastic, as no proper process was available for those wastes. The End-of-life WTs are enormous and require an energy-intensive mechanical shredding process to reduce the size of the waste. Finally, a 100% fibre recovery rate (yield) was considered from thermally recycling the composite wastes. However, the original fibre recovery rates were 95–98 wt% for rCFs from CFRP wastes and 80–82 wt% for rGFs from GFRP wastes. Despite these limitations, the results obtained from this study seems to be realistic and coherent when compared with the similar LCA studies from the literature.

5. Conclusions

This study analysed various waste disposal methods for CFRP and GFRP wastes using the LCA methodology, especially, focusing on a thermal recycling route developed from the previous study [24] to recycle and remanufacture CFRP and GFRP wastes into rCFRP and rGFRP composites. Three more traditional waste disposal scenarios were analysed: landfill, incineration with energy recovery, and feedstock in cement kiln production. The climate change results and fossil fuel consumption for CFRP waste disposal through the thermal recycling route seem sustainable, with a significant potential to substitute vCFRP. When rCFRP replaces vCFRP with a ratio of 1:1, i.e., 100% of recycled composites to virgin composites, the combined GWP emissions will be −11.43 kg CO_2-eq. Additionally, taking into account the high mechanical properties (>90%) of rCFRP reported in various studies [1,40,41] and lower emissions (3.06 kg CO_2-eq) from recycling the pricy CFs. The rCFRP substitutions with vCFRP played a significant role in emphasising circular economy by reducing virgin composite production and encouraging the reuse of recycled composites.

The climate change results and fossil fuel consumption for GFRP waste disposal as feedstocks in cement kiln seems sustainable with −0.47 kg CO_2-eq. These results are better than the proposed thermal recycling process results of 1.17 kg CO_2-eq, including the substitution with vGFRP production. The high environmental impacts occurred significantly due to the energy consumption (38.52 MJ/kg) to thermally recycle the GFRP wastes. Additionally, vGF production possesses more limited energy consumption, and the benefits from substituting vGFRP with rGFRP composites were not possible. However, material recovery was totally absent despite the positive results of using GFRP wastes as feedstocks in cement kiln production. Incineration and landfill, straightforward and popular disposal options, were strictly unsuitable for treating CFRP and GFRP wastes. The incineration results imply higher environmental impacts with a lower energy recovery rate and zero material recovery possibility.

It is necessary to further study an optimised thermal recycling route with industrial-scale operating conditions, especially for GFRP waste disposal. The possibility to regenerate the heat within the recycling system will result in reducing the energy consumption at the same time, increasing the recycling capacity to satisfy the demands in material recovery and substituting their virgin counterparts.

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