Proceedings of the 4th International Conference on Materials Chemistry and Environmental Engineering
DOI: 10.54254/2755-2721/60/20240859

# Recovery and reuse of thermosetting carbon fibre reinforced composites

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**Abstract.** With the widespread application of carbon fibre composites in sectors such as automotive, aerospace, and construction, recycling and reusing these materials have become crucial for reducing environmental impact and resource waste. This article summarises the potential of recycled carbon fibres in additive manufacturing by describing the main techniques, tools, and benefits and drawbacks of mechanical, thermal, and chemical recycling of carbon fibre composites. Notably, oxidation in air at 400–450°C for 15-20 minutes can greatly improve the tensile characteristics of thermally treated carbon fibres, offering promise for thermal recycling. In terms of chemical recycling, using recyclable subcritical and supercritical fluids can improve carbon fibre recovery rates and result in smoother-surfaced, higher-performing fibres. Additionally, recycled carbon fibres show potential in additive manufacturing due to their lightweight, high strength, and high stiffness. The purpose of this study is to accelerate the widespread use of carbon fibre composites across industries by providing an overview of carbon fibre recycling methods. Research and innovation in this field are crucial for sustainability and the circular economy.

**Keywords:** Recycling, Carbon Fibre, Additive Manufacturing.

## 1. Introduction

In recent years, carbon fibre reinforced polymers (CFRP) have been used more and more in marine, aerospace, automotive, construction, wind turbine blades and other fields. A 2019 analysis on the carbon fibre industry projects that the worldwide carbon fibre market will develop at an annual growth rate of 12.5%, from $4.7 billion in 2019 to $13.3 billion by 2029, owing to the exceptional qualities of carbon fibre reinforced polymer composites (CFRPCs), which include their high modulus and strength, low density, light weight, resilience to heat and corrosion, etc [1].

CFRPC is usually composed of carbon fibre and substrate, of which the resin-based composite material with an epoxy functional group has the most outstanding performance, with impact resistance, humidity and heat resistance, etc., and can reduce the weight of the structure by about 25% [2]. However, carbon fibre is very expensive and consumes a lot of energy (183-286 MJ/kg) [3], which hinders its use in larger quantities in industries such as automobiles. Most carbon fibres are formed by high-temperature carbonization of PAN (polyacrylonitrile) precursors under a specific gas atmosphere, which is not only cumbersome to manufacture but also carries high equipment maintenance and safety measures due to the toxicity of PAN [4]. More importantly, the substrate of CFRPC is a thermosetting material, although the mechanical properties of this material are higher than that of a thermoplastic substrate, it has strong chemical inertness and corrosion resistance, so it is difficult to separate from carbon fibre materials or cause greater wear and fracture of carbon fibre during the separation process, which will lead to reduced performance of recycled carbon fibre. In addition, taking the aircraft field as an example, due to the rapid iterative update of technology, a large number of abandoned fuselage wings will continue to be produced in the future, according to statistics, 30%-40% of carbon fibre will be landfilled because it cannot be reasonably recycled.

In recent years, the rapid development of new energy fields such as wind power has increased the demand for carbon fibre manufacturing, and the loss of energy has made few manufacturers master mass production, the biggest producers in the world include Teijin, Hexel, SGL Carbon, Mitsubishi Chemical Carbon Fibre and Composites, Toray of Japan (31% of worldwide output), and Teijin, they account for 87% of global carbon fibre production [5]. Therefore, the recycling and reuse of CFRPC become a hot topic in various countries, with the continuous progress of 3D printing technology, recycled carbon fibre (rCF) with low energy consumption, high performance, sustainability and other advantages quickly occupies the market, has become one of the key technical means widely used in carbon fibre, has the great potential [6]. Composites filled with rCF are already used in electrodes in lithium-ion batteries because they require lower energy consumption than virgin carbon fibre (vCF), which provides an opportunity to improve battery performance. The roof and headlamp components in the BMW i3 models are also used to form a wear-resistant material with a PEEK substrate to improve load-bearing performance and wear resistance, as well as reduce weight and improve fuel efficiency. It solves the garbage problem, brings huge environmental benefits, and reduces manufacturing costs.

This article will introduce and evaluate three mainstream recycling methods (the overall framework is shown in Figure 1) [7]: the mechanical method is mainly cutting and shredding, which may cause wear and tear on carbon fibre; The effect of pyrolysis method is better, but excessive energy consumption and high temperature will produce carbon black substances on its surface; The performance of chemical methods is the best, but the environmental pollution problems brought by them cannot be ignored. The practical application of rCF's additive manufacturing technology (AM) will also be explained, and finally, suggestions and prospects in the field of carbon fibre recycling will be put forward, helping people better understand and value the recycling of carbon fibre and related environmental issues.

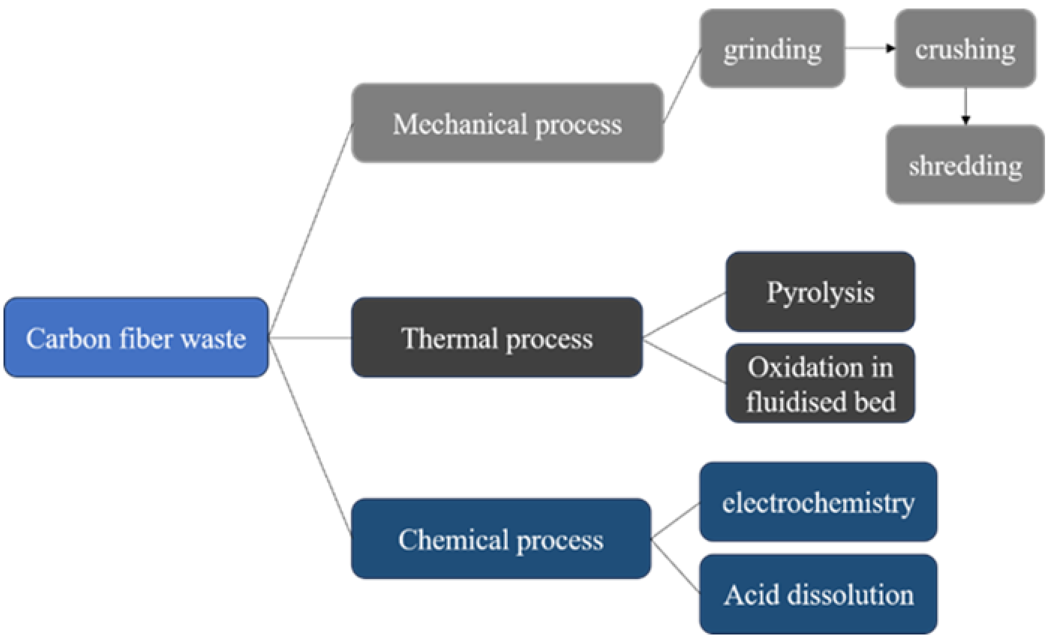


**Figure 1.** Three main recycling methods for carbon fibre composites.

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## 2. Recycling methods of carbon fibre composites

### *2.1. Mechanical process*

Mechanical recycling of carbon fibre composite materials involves several steps aimed at converting these materials into reusable forms, including carbon fibres and thermosetting plastic matrices. Here is a detailed description of the mechanical recycling process:

The first step is to cut the carbon fibre-reinforced thermosetting composite material into smaller chunks. This step is designed to reduce the volume of the original composite material for easier handling and processing. Next, the cut chunks undergo a grinding process to transform them into powder. Typical equipment used in this process includes a granulator, which helps to polymerize the cut material into pellets to make it easier to handle; actually, a crusher, which is mainly used to further refine the particles and turn the composite material into a finer powder; and then a classifier, which is used to separate and classify the powder according to the size of the particles. The powder separated from the classifier is divided into two parts according to the degree of grinding: most of it consists of fibres, and this part mainly contains carbon fibres or other fibre materials. These fibre powders have relatively large particle diameters and are therefore of high value in the recycling process and are suitable for use in the manufacture of new reinforced composites. A portion consists of a thermoset plastic matrix, which consists primarily of thermoset plastic matrices, usually resins or adhesives. Although these matrices play a vital role in composites, their value in the recycling process is relatively low. Carbon fibre materials obtained through mechanical recycling are often used for blending with other fillers to create new reinforced composites, which helps to reduce resource waste and improve material sustainability. However, it is worth noting that mechanical recycling methods can cause severe damage to the fibres. During the grinding process, fibre length may decrease, and the fibre surface may undergo abrasion, potentially leading to reduced performance and strength in the recycled materials.

Furthermore, mechanical recycling methods are not universally suitable for all types of carbon fibre composite materials. Some composite materials may have complex structures or use specific adhesives, making it challenging to effectively separate fibres from the matrix through mechanical means. Therefore, when selecting a recycling method, it’s essential to consider the characteristics and structure of the composite material to determine the most suitable recycling approach for achieving sustainable resource reuse and reducing environmental impact. Despite its limitations, mechanical recycling remains an important recycling pathway in the carbon fibre composite materials industry, offering a significant way to contribute to sustainable development.

### *2.2. Heat recovery*

*2.2.1. High-temperature pyrolysis.* The steps involved in the pyrolysis method primarily begin by heating the high-temperature pyrolysis reactor to a temperature range between 800°C to 1500°C, ensuring that the reactor has proper atmosphere control, typically achieved using inert gases such as nitrogen to prevent oxidation of carbon fibres. In this atmosphere, the polymer matrix begins to volatilize into lighter-weight molecules, while carbon fibres and various fillers remain inert and solid.

The gases produced during pyrolysis are then cooled and separated into liquid hydrocarbon compounds through a condenser. The gases are purified to remove impurities and harmful substances, ensuring their safe reusability. After pyrolysis, the solid residue typically contains residual carbon char on the surface, which requires an appropriate oxidation reaction to ensure the purity of the recovered carbon fibres. However, this oxidation process can lead to a reduction in fibre thickness, which may affect their strength. Finally, the carbon fibres undergo mechanical cleaning, acid treatment, or other chemical processes to remove residues and impurities. The specific process flow is illustrated in Figure 2.

Although high-temperature pyrolysis is an effective method for carbon fibre recycling, it also presents some drawbacks and challenges, including high energy consumption and a decrease in carbon fibre performance [7]. Therefore, strict control of reaction time and heating temperature is crucial.

Studies have shown that the tensile strength of carbon fibres typically decreases by 50% after pyrolysis, but an appropriate oxidation process can enhance the performance of recycled carbon fibres. Oxidation of pyrolysis residues is more effective at temperatures between 400°C and 450°C. However, at lower temperatures, the resulting carbon fibres may have a rougher surface and more residual carbon char. If the oxidation time is too short, it may also lead to incomplete removal of surface carbon black. An oxidation time of 15-20 minutes can yield carbon fibres with the cleanest surface and the highest tensile strength [8].

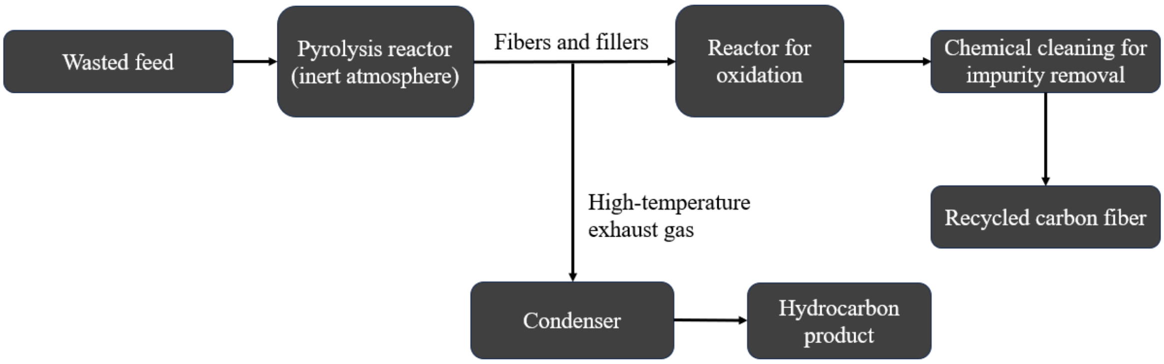


**Figure 2.** Pyrolysis process.

*2.2.2. Fluidized Bed Pyrolysis Method.* One process that shows promise for recovering carbon fibre composite materials is fluidized bed pyrolysis. The materials for the carbon fibre composite are initially chopped into appropriate sizes in this procedure so they may be processed further. Subsequently, the fluidized bed reactor is heated to a temperature range of approximately 400-500°C. The bed material typically consists of fine granular materials, such as sand, ceramic particles, or other thermally stable materials. Next, the prepared carbon fibre composite materials are introduced into the fluidized bed [9].

Under high-temperature conditions, the polymer matrix begins to decompose, transforming into gaseous products such as carbon gas and other volatile substances, while the carbon fibre's carbon structure is preserved. To ensure a successful recovery, it is essential to carefully control the pyrolysis temperature and reaction time to ensure complete decomposition of the polymer matrix without burning the carbon fibre. Subsequently, the carbon fibres need to be separated from the gases and particulate materials.

During this separation process, airflow and vibration are often used to assist in separating the decomposed carbon fibres from the gases and particulate materials. This can be achieved through appropriate exhaust and gas-solid separation devices, allowing the gaseous products to be discharged while effectively collecting pure carbon fibres. Following this, subsequent cleaning steps are performed, with the aim of removing surface residues, similar to the pyrolysis method.

Despite the potential of fluidized bed pyrolysis for carbon fibre recovery, it also comes with some significant drawbacks. Firstly, this method requires a substantial amount of energy because heating the bed material to high temperatures consumes a considerable amount of energy, and maintaining a stable bed temperature also requires significant electrical or other energy sources, leading to high energy consumption and operational costs. Secondly, the complexity of the equipment is quite high, with fluidized bed systems being relatively complex in themselves, requiring precise temperature control, airflow control, and bed material handling. This increases the complexity of equipment maintenance and operation, often necessitating specialized technical expertise and equipment.

For these reasons, establishing and maintaining a fluidized bed recovery system requires a significant initial investment, making it less feasible for small-scale or emerging recycling projects. However, despite these challenges, fluidized bed pyrolysis remains a promising carbon fibre recovery method, especially in conditions of large-scale production and efficient energy utilization.

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### 2.3. Chemical Recycling

Chemical recycling is currently one of the most popular methods undergoing research and development in various fields. Some mainstream methods involve using subcritical and supercritical water in batch reactors to dissolve the epoxy resin base of carbon fibre composite materials, aiming to recover pure carbon fibre. Supercritical water refers to water existing under temperature and pressure conditions above its critical point (374°C and 22.1 MPa). Under these conditions, the properties of water change significantly, making it a useful solvent and reaction medium with many unique advantages [10]. It has high diffusivity, aiding in the dissolution of reactants and increasing reaction rates. Supercritical water reactions typically do not require organic solvents or catalysts, thus reducing the use of organic solvents and waste generation, and contributing to environmental pollution reduction.

Supercritical water also has efficient thermal conductivity and is highly sensitive to heat transfer, rapidly transferring heat during the reaction process, maintaining a uniform temperature distribution, avoiding the formation of hotspots or cold spots, and helping to achieve higher-quality recycled carbon fibres. Research has shown that under conditions of 28 MPa and 673 K, a recovery rate of 79.3 wt% can be achieved in supercritical water, and the addition of alkaline catalysts can enhance the mechanical properties of the recycled carbon fibres to some extent. More specific solvents such as supercritical acetone [11], which has similar dissolution parameters to epoxy resin, are favourable for decomposition. Compared to other supercritical fluids such as methanol, acetone has better solubility for high molecular weight decomposition products. It was discovered by examining the kinetics model and the impact of reaction pressure on the breakdown of epoxy resin that, during the first 60 minutes in a batch reactor operating at 350°C and 2–14 MPa, the decomposition efficiency progressively rose before declining.

## 3. Potential and Applications of Recycled Carbon Fibre in Additive Manufacturing (AM)

Additive Manufacturing (AM), sometimes called 3D printing, is a manufacturing technique that allows products to be constructed layer by layer [6]. In contrast to subtractive manufacturing methods, which typically involve removing excess material from bulk materials to obtain the desired shape, AM can convert materials almost entirely into the final product, reducing material waste, and making it a powerful tool for sustainable manufacturing. This is of significant importance in reducing resource waste and manufacturing costs.

Recycled carbon fibre holds exciting prospects for a wide range of applications in the field of AM. Firstly, its sustainability attributes make it an outstanding representative of sustainable manufacturing. Recycled carbon fibre is derived from recycled carbon fibre composite materials or waste carbon fibres, providing an essential solution for the manufacturing industry's environmental goals [12]. Secondly, the lightweight properties of recycled carbon fibre make it highly potential for manufacturing lightweight components. This is crucial for industries such as aerospace, automotive, and energy, as it can reduce component weight, improve fuel efficiency, and lower transportation costs while maintaining strength and rigidity.

The third key advantage of recycled carbon fibre is its ability to adapt to complex structures. AM technology itself can create components with complex geometric shapes, and the introduction of recycled carbon fibre further enhances this potential. This has enormous value in fields such as healthcare, aerospace, mechanical engineering, as it allows for the manufacturing of higher-performance and more complex products.

Cost-effectiveness is also a critical advantage of recycled carbon fibre. Compared to fresh carbon fibre, recycled carbon fibre is typically more economical, reducing manufacturing costs and enhancing product competitiveness. This is particularly important for businesses as it can improve profitability and increase market share.

The controllability of recycled carbon fibre's performance makes it a powerful tool for multi-field manufacturing. Process parameters can be adjusted to meet the requirements of specific applications. This means that material strength, rigidity, and other performance characteristics can be adjusted as required to ensure that the product meets the specific needs of its intended use.

## 4. Conclusion

This paper has delved into the recycling methods of carbon fibre composites, including mechanical recycling, thermal recycling, and chemical recycling, as well as the potential of recycled carbon fibre in the field of additive manufacturing (AM). These recycling technologies have made significant progress in achieving sustainability and resource reuse, but they also come with limitations and challenges.

In terms of mechanical recycling methods, although this approach is relatively straightforward, it can easily lead to damage and performance degradation of carbon fibres. During mechanical processing, carbon fibres may become shorter and experience surface wear, thereby reducing their performance and strength. Furthermore, mechanical recycling is not suitable for all types of carbon fibre composites, especially those with complex structures or adhesives, which are difficult to separate effectively by mechanical means.

Secondly, in the description of thermal recycling methods, including high-temperature pyrolysis and fluidized bed pyrolysis, these methods are highly effective in decomposing composite materials and recovering carbon fibres. However, they also face challenges of high energy consumption and equipment complexity. High-temperature pyrolysis requires a substantial amount of energy, and maintaining the stable temperature of the fluidized bed material also requires significant electrical power or other energy sources, resulting in high operational costs. Additionally, the complexity of the equipment is relatively high, requiring precise temperature control, airflow control, and bed material handling, which increases the complexity of management and operation. Therefore, establishing and maintaining these fluidized bed recycling systems require a high initial investment, which may be less feasible for small-scale or emerging recycling projects.

The third chemical recycling methods, particularly those involving subcritical and supercritical water, have been discussed. These methods can achieve the recovery of carbon fibres under milder conditions while maintaining high performance. Supercritical water possesses unique advantages, such as high diffusivity, no need for organic solvents or catalysts, and efficient heat transfer capabilities. However, they still require specialized reactors under high-pressure and high-temperature conditions, which need cost reduction and lower energy consumption for widespread application.

In the latter part of the article, the focus has been on emphasizing the potential of recycled carbon fibres in the field of additive manufacturing (AM). AM is a manufacturing technology with tremendous potential that can reduce material waste and achieve sustainable manufacturing. Key advantages of recycled carbon fibres, including sustainability, lightweight properties, adaptability to complex structures, cost-effectiveness, and performance controllability, make them highly promising in AM. They can reduce the weight of manufactured components, improve fuel efficiency, and lower transportation costs, all while maintaining strength and stiffness. Recycled carbon fibres also have the ability to adapt to complex structures, enabling the production of higher-performance and more complex products in fields such as healthcare, aerospace, and mechanical engineering. Furthermore, the cost-effectiveness of recycled carbon fibres makes them a competitive advantage in the manufacturing industry, improving profitability and market share for businesses. Their performance controllability makes them a powerful tool for multi-domain manufacturing, allowing for the adjustment of material properties according to specific application requirements.

Despite the existing challenges and limitations, research and development in recycling technologies will continue to receive attention as the widespread application of carbon fibre composites in various fields increases. In the future, innovative methods and equipment may emerge to improve the efficiency of carbon fibre recycling and reduce energy consumption. This will help drive the sustainable utilization of carbon fibres, promote resource recycling, and provide new ideas and opportunities for sustainable manufacturing and high-performance product development. Therefore, while there are currently some challenges, the future outlook is promising, and further breakthroughs in carbon fibre recycling and reuse are expected to occur.

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