

Chapter 20 Recycling of composite materials

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1. Introduction

A composite material can be defined as a combination of matrix and reinforcement resulting in a material with superior properties. The reinforcement is mainly used to increase strength and stiffness. The matrix sustains the reinforcement in the designed orientation. Three composite types - polymer-matrix composite (PMC), metal-matrix composite (MMC), and ceramic-matrix composite (CMC) - are in demand in various sectors. Composites could also be classified based on reinforcement such as particulate composites, fibre-reinforced composites and structural composites. Of these, thermoset composites like carbon fibre reinforced polymers (CFRPs) and glass fibre reinforced polymers (GFRPs) dominated the markets [1]. In the UK, the composite market is well segmented where 81% are dominated by three main players: aerospace (36%), wind energy (33%), and automotive (12%). Both marine and industrial have 7% respectively, sports (2%), and others (3%) [1, 2]

Composite materials are rapidly gaining popularity and usage due to their combined lightweight, stiffness and strength features. Although it is difficult to find absolute statistics on the total global composite production, the composite market is projected to grow from £55 billion in 2016 to £87.47 billion by 2022 [3]. The first large-scale commercial applications of composite materials began in the military sector during World War II and in the late 1940s and early 1950s. Since then, global use of composite materials has grown rapidly from 158,800 tonnes in 1960 to 6.1 million tonnes in 2004 representing a 3,800% growth in the last 45 years [4].

Between all types of composite material, PMCs dominate the market with thermoset based products account for more than two third [1]. On account of this, most recycling technologies are focused on recovery of PMCs compared to MMCs and CMCs. It is estimated that around five to 10 million tonnes of GFRP are produced globally every year with the demand for CFRP forecasted at approximately 157,000 tonnes annually. The high demand leads to the issue of scrap waste during production [5]. The MMCs and CMCs have lower demand in terms of volume compared to PMCs. This leads to the limited availability of the MMCs and CMCs composites scrap for recycling [1]. Apart from manufacturing waste, end of life waste becomes significant at the product end of service stage

The heterogenous nature of composite causes recycling of the material to be highly challenging. As opposed to thermoplastic polymers, the thermoset polymers have difficulties in recycling due to its cross-linked nature. This is the reason research and development in recycling technologies focus on recovery of thermoset based products.

The situation of rising composite waste is made difficult due to the limited availability of waste processing centres. Recycling technology is still relatively new and progressing with recyclates hardly finding application in the market. Individual businesses that assume responsibility for their waste are often confronted with issues of economics of scale and transportation costs. From literature, most studies within the area of composite recycling only focused on reuse applications and strength of recycled fibre. The aspect of environmental impact is rarely considered.

This chapter explains available composite recycling methods from the aspect of process details, energy demand and recyclate mechanical properties. The focus is on PMCs particularly on GFRP and CFRP as these composites dominate the current market in terms of volume. Limited studies on PMCs and MMCs are reviewed in this chapter. Strength of recyclate and new composite product are also reviewed and compared between the methods. Possible reuse applications are also suggested. This chapter seeks to simultaneously analyses composite recycling initiatives from the aspect of feasibility, environmental and economic aspects. The analysis will be a vital decision-making tool regarding composite material sustainability in the future.

2. Composite waste

The increasing demand for composites has balanced effects on the generated wastes both nationally and internationally. Halliwell [6] reported that only 0.08% of composite waste is collected in Europe, a situation indicative of issues with a collection scheme that may not be well-designed or is affected by challenges in the waste post-treatment. There are two types of waste mainly associated with product output: manufacturing waste [7, 8] and end-of-life waste [6, 9]. CFRP represents about 40% of the UK composite production by value but corresponds to only about 2% by volume since the vast majority is GFRP [7, 10]. Numerous research have recently been conducted on CFRP recycling due to the cost differentials compared to virgin carbon fibres as well as the value of CFRP being ten times higher than GFRP [10].

The price for virgin GFRP and CFRP is in the range of £15.30-21.60 and £23.50-26.10 per kg respectively [11]. The Boeing company estimates the cost of manufacturing virgin CFRP to be in the range of £22 to £44 per kg, while the price range of the recycled version is only £12-18 per kg [12]. Although there is a difference in price, these are still within the current market range in the UK. The ELG Carbon Fibre Ltd compared prices between virgin and recycled CFRP materials and found the former to be £15/kg and the latter at £9/kg. Based on this capability, 2,000 tonnes of

CFRP waste are recycled every year at the cost of £0.60/kg, a pricing deduced from the electricity and gas costs involved [8]. The limited capacity has prevented many manufacturers from sending their composite waste to this centre.

By 2030, the GFRP waste is projected to reach an estimated 170,000 tonnes per annum [7]. Aerospace and wind energy are the main composite sectors in the country. The UK aerospace industry is the second largest in the world and supplies 17% of the global market and 75% of aircraft components [13]. Composite waste within the aerospace sector therefore would be significant with the use of composites especially in the mainframe of aircrafts steadily increasing over the years [14-16]. For example, the composite portion utilised in the Boeing-777 is 10% of the total weight of the mainframe. This composite usage increases to 22% for the Airbus A380 and up to 50% for the Boeing 787 Dreamliner.

It is reported that 6,000 to 8,000 commercial planes are expected to reach their end-of-life by 2030 globally [17]. Based on recent data, a total of 1,229 commercial aeroplanes and 1,102 military aircraft are currently operational in the UK [18, 19]. The maximum zero-fuel-weight for A380 is 369 tons; by taking 22% of composite percentage (as a medium composite use in Airbus A380), the composite weight per aircraft would be about 81.18 tonnes. Thus, the total estimated composite end-of-life from aircrafts alone can reach to about 190,000 tonnes.

For wind turbines, waste from rotor blades is projected to reach 225,000 tons by 2034 [20]. In 2015 alone, there were more than 6,037 operational wind turbines in 729 individual projects in the UK [21]. Since 23,597 wind turbines were deployed from 2005 to 2012 [22], the situation alerted the stakeholders to be ready for dealing with a huge amount of composite waste in the near future. While landfills are the common disposal route for composite waste, legislative pressure has prompted for a more environmentally sound solution to be found and practised. Some of the legislation and policies associated with composite-based products are discussed in the following sub-section.

3. Drivers for composite recycling

3.1 Legislation and policies

An important and growing body of literature has agreed on environmental legislation being an influential factor to recycling enhancements [23]. About 95% of these legislations are derived from the European Union [9]. The end-of-life legislation and policies most relevant to composite waste are discussed in this section [9, 24].

3.1.1 The Landfill Directive (1999/31/EC)

The UK government is considering restrictions on the landfilling of biodegradable and recyclable wastes, including landfill bans [25]. Stringent technical requirements for waste and landfills have been introduced to prevent or reduce the negative impact of waste [9]. The Landfill Directive stipulates that materials with high organic content such as composite (i.e. wind turbine blades with an organic content of 30%) are to find alternative end-of-life routes.

3.1.2 UK Landfill Tax and Operator's Gate Fee

Associated with landfill directive are the UK Landfill Tax and Operator Gate Fee, a payable charge for the disposal of waste at landfills. These measures are aimed as disincentives for landfill disposal of waste and to encourage instead the utilisation and practice of environmentally sound routes such as recycling, composting and recovery.

Two rates of landfill tax - a standard rate and a lower rate for inert materials – are implemented with most composite waste falling under the standard rate of taxation. Records indicate a sharp steady increase in disposal costs in the UK with rates climbing from £7 per tonne in 1996 to £72 per tonne in 2013, £80 in 2014 and £82.60 in 2015 [26]. Coupled with the Landfill Operator Gate Fee (£20/tonne), the total landfill disposal cost is currently at approximately £100 per tonne [7]. The total composite waste landfilling

cost in 2015 was almost £304 million, a significant cost driver for developing alternative waste management routes [27].

3.1.3 End-of-life Vehicles Directives (ELV) Directive (2000/53/EC)

Through Regulations 2003, 2005 and 2010, the UK is required to divert at least 95% (by weight) of ELV from landfills, including recycling at least 85% and energy recovery at an additional 10%. This has represented a noteworthy driver towards improving recycling performance in the automotive composite sector. A recycling route must be made available if composites are to significantly substitute for metals in this area [7, 28].

3.1.4 The Framework Directive on Waste (2008/0241 (COD))

The EU Waste Framework Directive (WFD 2008/98/EC), a backbone for the resource efficiency management efforts [29] has been transposed into the UK law as the “Waste (England and Wales) Regulations 2011”, the “Waste (Scotland) Regulations 2012” and the “Waste Regulations (Northern Ireland) 2011” [30]. These regulations legally enshrine the waste hierarchy such as prevention, reuse, recycling, and finally the least preferred disposal method of energetic valorisation (energy recovery) through incineration and landfill. The waste management hierarchy is shown in Figure 1.1.

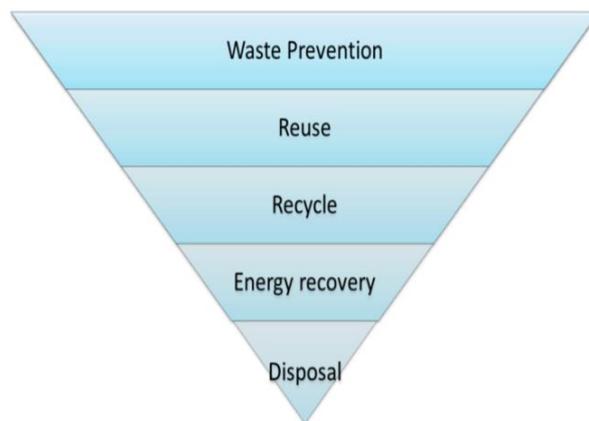


Fig. 1.1. Waste management hierarchy

3.2 Embodied energy and cost of virgin materials production

Virgin or original fibre has high embodied energy. Carbon fibre and glass fibre has a range of embodied energy from 183-704 MJ/kg and 13-45 MJ/kg [31, 32] respectively. The replacement of virgin fibre with the recycled precursor especially in short fibre or filler application can avoid usage of energy for virgin fibre production.

Replacing virgin fibre with recycled fibre can have a monetary advantage of around £403 per tonne [33]. Although there are changes of properties during the recycling process, the use of recycled fibre is still financially attractive.

High monetary value and embodied energy of carbon fibre is one of the reasons why most studies and industries are interested in recovering carbon fibre compared to glass fibre.

4. Challenges in composite recycling

Various obstacles have been discussed in the literature such as the difficulty to separate homogeneous particles from the composite without damaging their properties. The applicability of reclaimed materials in mainstream production is a crucial question yet to be fully answered [34, 35]. Numerous studies have attempted to explain other obstacles in composite recycling such as the low value of recyclates [7, 36] and variations in the feedstock [37]. All of these mentioned issues are related to the physical research of composite recycling with many efforts in place or progress to develop better recycling processes and the enhancement of recyclate quality. However, there has been limited research attempted on the non-physical challenges of composite recycling; for example, an issue acknowledged

in the literature is the insufficient amount of composite scrap collection for recycling operation [20] and logistics management issues [1]. These factors are also related to high capital, operating costs, and little supply chain pressure [7]. Research on these non-physical aspects should be as critical as the physical attributes due to each factor being interlinked and accountable during the implementation stage.

Since the technical and non-technical challenges are interlinked, it is crucial to examine both to arrive at a comprehensive solution. As could be seen, the logistic and scrap amount are often among the overlooked factors that may drive future recycling performance, especially once the recycling technology barrier has disappeared [38]. For efficient performance of recycling activities, hundreds of tonnes of composite waste are expected on a weekly basis [20]; to make this a reality, some composite wastes and proper methods of collection related study are started to take place and more study area are still need to be identified [39-41]. Thus, more research opportunities are available in the area of composites product take-back, composites reverse- supply chain, composites collection schemes and also composites recycling.

5. Composite recycling methods

Composite recycling methods can be classified into four main categories namely combustion or energy recovery, mechanical, thermal and chemical recycling technologies. Other methods are rarely investigated, such as high voltage fragmentation, biotechnological and electrochemical.

5.1 Composite recycling process chain

Process chain of composite recycling is shown in Figure 5.1. Pre-processes such as dismantling and size reduction are required for easy transportation and storage. Classification and treatment are essential post recycling stages to improve recycle quality for remanufacturing purposes.

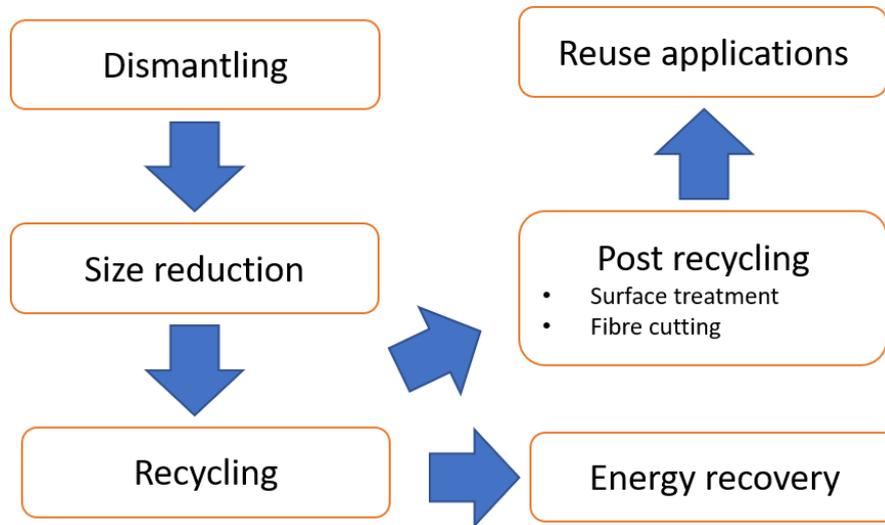


Fig. 5.1. Composite recycling process chain

Manufacturing waste and scrap do not need to undergo dismantling and downsizing steps. For the end of life composite waste such as aeroplane structure and wind turbine blade, foreign materials need to be separated first from the fibre reinforced composite part. Metal inserts and fasteners can be separated using magnetic fields. The separation process is time-consuming and requires human workforce as materials are sometimes mixed chemically or glued together, which makes physical separation without affecting the recyclate quality to be almost impossible. Size reduction is essential to reduce large composite waste into a smaller size to fit into a recycling machine, reactor or chamber.

During the recycling stage, reinforcement components of a composite (fibre or filler) can be separated from the matrix part. However, the challenge is to maintain the quality and mechanical properties of the fibre for reuse stage in the new composite product. The recyclate is typically in the form of fibre recovered with residual resin and filler. Thus, classification is usually used to separate the fibrous structure and powder rich fraction. For the fibrous fraction, chopping stage may be required to obtain desired fibre length.

In thermal and chemical recycling processes, the organic part of fibre reinforced composites is still recovered however the quality is degraded. The

use of recovered matrix is still under development as compared to the use of recovered fibre. The recovered can be formed into non-woven mats for prepreg and sheet moulding compound applications. Chemical treatment of recycled fibre surface is recommended to enhance interfacial bonding between the fibre surface and virgin thermoplastic matrix in new composite products. The treatment is to ensure good mechanical properties of the product, comparable to its original counterparts.

5.2 Combustion

Combustion or incineration aims to recover energy from the organic part of the composite waste. During the process, heat is used to break the bond of thermoset and thermoplastic matrices. The combusted polymer is the source of energy. The matrix has high calorific value and can be converted to other types of energy such as mechanical or electrical energy. The value depends on the type of composite. For example, oil and solid condensable product from pyrolysis process of glass fibre reinforced thermoset polyester was found to have around 27 – 33 MJ/kg of gross calorific value [42]. The high temperature from the incineration can be used as a source of heat energy for the steam boiler which is then used to spin a steam turbine for electricity generation. However, the conversion efficiency from heat to electrical energy is only about 35% [43].

Material recovery is rare since leftover after the process is in the form of ash residue, consists of incombustible fibre and fillers. The fibre and filler are not comparable to its virgin precursor. Because of this, some studies such as did not consider combustion as one of the recycling methods [1, 6]. However, the solid inorganic residue is possible to be as source material for cement production [44]. Another drawback is that certain types of filler absorb energy during incineration as the filler requires high temperature to decompose [45]. For instance, alumina trihydrate absorbs energy during incineration hence yield calorific value from the process can be reduced by 3.3% [46]. Apart from that, the leftover residues need to be disposed of in the landfill. The landfilling may lead to other environmental issues and additional cost

GFRP waste can be burnt in a cement kiln. The glass fibre and mineral fillers are raw materials for cement production while the matrix part can be combusted for energy source of the kiln. It was reported that the organic part of the waste can replace usage of fossil fuel and reduce carbon footprint by around 16% [47]. Besides, composite waste combusted with coal can minimise sulphur emission in a fluidised bed process [48].

The choice of composite waste to be incinerated depends on its percentage of organic proportion in the material composition. For sheet moulding compound (SMC), percentage of resin is only around 35% by weight which makes the SMC waste not suitable to be combusted. Disposal via incineration is not preferable for fibre with high monetary value as the recovered fibre and filler are not reusable. This explains why combustion process is preferred only for GFRP and not CFRP. Nonetheless, using heat to process composite waste for resin decomposition at a temperature can recover fibre with minimal quality degradation.

5.3 Mechanical recycling

Mechanical recycling is a process of reducing composite waste into forms of fibrous and filler fractions. The fractions can be reused as reinforcement in new composite products. Removal of foreign objects is vital in mechanical recycling to avoid damage of cutter in mechanical recycling machine.

Mechanical composite recycling usually loosely referred to as grinding although it can be confused with the abrasive grinding process using a wheel. Wittmann machines use the concept of milling using the hammer and IIT m-series machine crushes glass fibre composites between rollers and grinding ring to produce powder fractions. In terms of fibre length distribution, uniform distribution can be acquired when recycling using cutting mills. For long retained fibre, hammer mills can be used despite a faster rate of wear in cutting blades. Impact method in reducing the size of sheet moulding compound is desirable to retain long fibre length [49].

Composite waste typically comes in a bulk form, especially the end of life waste. Size reduction via cutting and shredding process is vital to ensure the waste can be fitted into a mechanical recycling machine. The smaller

size of waste can facilitate transportation of the material from the collection centre to the recycling facility. As a result of size reduction, coarse material is obtained from the waste with the size of 50 mm to 100 mm. The next stage is a further size reduction of the material using grinding or milling technique into a size of 50 μm to 10 mm. Typically, a milling or grinder machine consists of a screen with different aperture size. The aperture can be used as a guide to predict the maximum size of recyclates.

Based on literature and industrial practices, mechanical recycling is widely practised to transform GFRP waste into reusable recyclates. Shuaib and Mativenga [50] compared two granulator technologies of Wittmann MAS1 granulator (Figure 5.2) and Eco-Wolf grinder Model GM-2411-50 in terms of energy demand and recyclate quality. Based on the physical appearance of recyclate from both technologies, the recyclate from Wittmann and Eco-Wolf machines mainly consists of flake and fibrous structure respectively. Laboratory unit of Retsch SM2000 cutting mill was used to recycle shredding leftovers from a pultrusion company [51]. The milled waste is in form of fine and coarse material. AEG Co mini granulator was used for grinding and sifting process [52]. The granulator has two grinding disks where one disk is rotating and the other is fixed. The grinding process cut the composite into 10 cm x 10 cm pieces. After a classification step, only the fibrous fraction was used for analysis and mechanical tests.



Fig. 5.2. Wittmann MAS1 granulator

Mechanical recyclates consist of flake, fibrous and powder components with various sizes lower than aperture size of the machine. Size classification via sieving is necessary to separate the obtained recycle material for reuse applications. A 'zig-zag' classifier was used by controlling airflow to separate recyclates based on their density and particulate size [53]. Recycling processes at industrial scales used various separation technique, such as the combination of air classifier, screens and cyclone.

Recyclates in flake form is generally not reusable in thermoplastic-based composites. Such recycle may be reprocessed for further size reduction. Compared to other recycling methods, resin elimination on recycle surface for mechanical recycle is relatively low. During the recycling, resin from a part of the fibres is not completely removed. Residual resin is still a presence on fibre surface, hence preventing interfacial bonding to be formed within the new matrix. Post-treatment of recovered fibrous fractions is needed to obtain clean fibre hence ensuring good interfacial bonding with new matrix in reuse applications. Powder rich fraction can be used as a filler in cementitious products to replace the sand or fine aggregates. Mechanical recycling is not desirable to recycle high-value fibre such as car-

bon fibre as it is difficult to retain the long fibre. With a significant size reduction, fibres are far off from their original forms. Given the price of carbon fibre is around ten times higher than glass fibre, the mechanical approach is not preferable to process composite with high monetary value.

In recycling of metal matrix composite using the mechanical approach, ball milling process was used for downsizing of SiC particles reinforced aluminium turning chips [54]. The process transformed the waste into a powdered form. The powder was combined with molten aluminium to create a new, hybrid composite. Apart from that, compression process can be utilised in separation of the metal matrix and creation of a condensed composite [55].

Thermal recycling

5.3.1 Fluidised bed

In fluidised bed recycling, materials are fed through a hopper and placed on silica sand bed [56]. Heat which comes from a fluidised air passed over the bed with velocities ranges between 0.4 m/s to 1.0 m/s. Typical operating temperatures are around 450 °C to 550 °C. The heat causes degradation of resin and other organic constituents of composite materials and contaminants. The recovered fibre and filler are transported into a cyclone and rotating sieve for separation. The burnt gases are channelled through an exhaust for energy recovery. Past studies show potential in recovering glass fibres from thermoset composites [56, 57]. The temperature of 450 °C is an acceptable limit with minimal 50% of strength reduction. An advantage of this method is its high tolerance for mixed and contaminated materials [10].

5.3.2 Conventional and microwave pyrolysis

In pyrolysis processes, GFRP and CFRP waste is heated at 300-800°C in an inert atmosphere which results in the polymeric resin being volatilised while the fibres and fillers are recovered [1]. The temperature needs to be

controlled within an optimum range to maintain fibre strength. By-products of the process in forms of liquid and gases can supply energy or feed-stock for other chemical processes.

The ELG Carbon Fibre Ltd in England is one such example of a UK recycling company practising their patented thermal recycling method [58]. The recycled fibre is sold in milled, chopped and pelletised forms [8]. Other companies with this approach are Carbon Conversions in the USA, Karborek in Italy, CFK Valley Stade Recycling GmbH and Hadeq Recycling Ltd in Germany and Recycle Industry Co Ltd (Japan) [59].

Microwave pyrolysis uses microwave radiation as a source of heating. The rapid heating process reduces processing time hence reducing the possibility of fibre strength degradation. The radiation heats the material from the inner and not from the surface as in the conventional heating process. The diffuse nature of electromagnetic radiation is capable of heating waste uniformly [60].

5.4 Chemical recycling

Solvent such acid, alcohol or water is important in the chemical recycling method to break the chemical bond of the organic part of fibre reinforced composite materials [35]. During the process, the matrix is depolymerised into monomers. Polymeric resin is decomposed into solvent while the inorganic part (fibre and filler) are recovered for reuse applications in new composite products. The process can retain long fibre with excellent strength. The process has less thermal exposure at a lower temperature compared to most thermal recycling methods. Compared to mechanical recycling, the chemical approach has less fibre movement and agitation. However, this method is not yet commercially viable and mostly conducted in a laboratory-scale setup.

The processes are classified according to temperature and pressure states of the solvent used namely sub, near and supercritical. In terms of the critical state of the solvent, the supercritical fluid is preferable as the solvent has less impact on the environment and human health [61]. The state and

type of solvent are important parameters in controlling effect on fibre strength.

Past studies have carried out analyses on the hydrolysis process using water on glass fibre [62, 63] and carbon fibre composites [64-67]. Some studies used subcritical and supercritical alcohol in recycling carbon fibre reinforced epoxy waste [67-69]. While other studies focused on recovering the fibre, a study successfully formed unsaturated polyester resin from a yield of a supercritical alcohol process [70].

For metal matrix composite, the matrix can be separated from the reinforcement by adding a flux [55]. The addition can change interface energy between the two constituents.

5.5 Other recycling methods

Several recycling methods such as high voltage fragmentation (HVF), biotechnological and electrochemical are relatively new and only available at a pilot or laboratory scale. With low recycling capacity and high cost, these methods mainly focus on CFRP waste with the main aim to maintain fibre surface quality and fibre length.

Despite relatively new compared to other mature recycling technologies, HVF recycling shows potential for industrial-scale process with high retention of fibre strength and low energy consumption [71, 72]. HVF method uses the electrodynamic principle to disintegrate material into smaller parts, where generated high voltage electrical pulse passes through the material repetitively. The principle was initially used to break rocks in mining applications in the early 1960s [73]. The pulse creates a plasma channel which moves through the material particularly along the weakened material interfaces. As a result, tensile stresses are created in the material which causes internal fracture and material disintegration. Given that fibre reinforced composite is a multi-component material which consists of boundaries between fibre, matrix and filler, the shockwave generated by the electrical pulses in HVF process can be utilised for material fragmentation.

Past studies compared mechanical and HVF method by recycling GFRP waste [72, 74]. SELFRAG laboratory equipment was used to recycle chopped strand glass fibre reinforced with unsaturated polyester with fibre fraction by volume around 30% [74]. The study compared fibre characteristic and process energy demand between HVF and mechanical recycling processes. Potential of HVF over mechanical recycling is highlighted and the study recommended for upscaling of the current laboratory scale machine. Another study recycled sheet moulding compound waste by characterising residual resin content and fibre length distribution [72]. It was reported that the current HVF system has low recycling throughput because the machine is not yet optimised to recycle composite waste. In Cleansky Project, demonstrator setup is created with a recycling rate of 5.65 kg per hour with energy consumption of only 1.2 kWh (or 0.33 MJ) per kg for CFRP waste [71]. The setup has achieved the technology readiness level (TRL) 6 and was found to be economical. Besides the potential in HVF, it was reported that using high voltage electrical treatment in separating resin from fibre in unidirectional CFRP laminate can lead to a weight loss of the composite and resin elimination [75].

Researchers at Hohenstein Institute in Germany uses a microbiological system to degrade polymer part of a composites via biochemical approach [76]. Minimal fibre damage was reported and the fibre is suitable to be used in new products. The method has low material throughput and currently being used only to recycle CFRP waste.

The electrochemical method can also be utilised to degrade matrix in composite waste and recover fibre with a clean surface. A study experimented by immersing CFRP waste in a different solution of NaCl and different applied current [77]. Concentration of the solution or electrolyte needs to be controlled since higher concentration can cause severe fibre damage. Despite high retention of fibre strength at 80%, the recycling throughput was low and currently only available at a laboratory scale. The process requires 21 days to break down a matrix of 2 grams CFRP waste. Another study developed a method named electrically driven heterocatalytic decomposition (EHD) method which used the electrochemical principle at room temperature and atmospheric pressure [78]. Fibre strength retention was reported to be excellent (90% compared to virgin fibre) which improvement of 27% can be observed for fibre interfacial shear strength. Other advantages of

EHD method is non-toxicity nature and no size requirement of the waste. There is no presence of other environmental issues compared to other methods. such as dust in mechanical recycling, by-product gases (pyrolysis) and solvent (solvolysis). A study by Kamavaram et al [79] investigated the potential of using electrolysis in ionic liquids to recycle aluminium based metal matrix composites. It was reported that high purity of aluminium (more than 98%) can be recovered however this depends on the concentration of the liquid and applied voltage.

6. Environmental impact of composite recycling processes

Environmental impact of composite recycling processes can be assessed by considering the resource consumption. Impact of manufacturing processes mostly dominated by electrical energy consumption which usually expressed in terms of kilowatt-hour (kWh) or megajoule (MJ). The power source is the main contributor to the overall energy footprint of the process or product. Apart from minimising the cost of electricity, reducing energy in the composite recycling process is important to ensure less collateral damage [59]. Concerning material embodied energy, CFRP recycling is more favourable than GFRP as the production of carbon fibre is ten times more energy-intensive as compared to the production of glass fibre [32].

Figure 6.1 shows electricity energy demand for composite recycling processes found in scientific literature and industrial report. Differences in energy demand can be attributed to several factors such as recycling rate, reactor efficiency and type of waste.

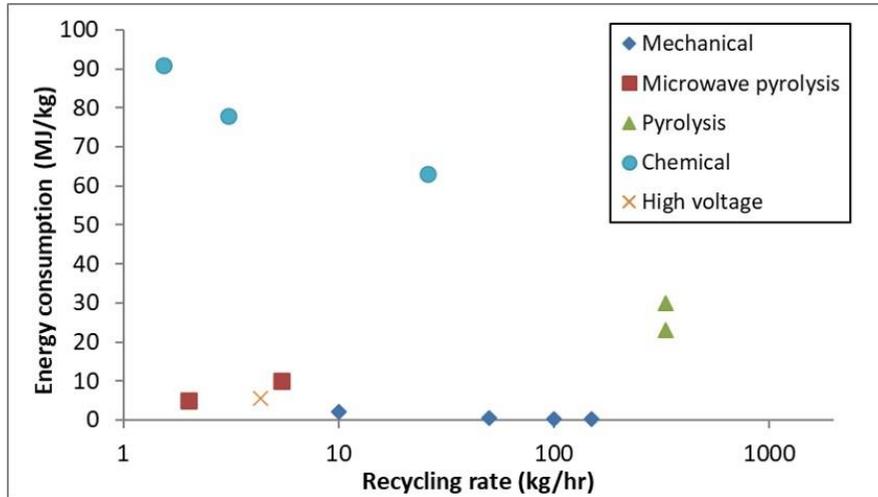


Fig. 6.1. Relationship between energy consumption and processing rate for different composite recycling methods

Comparing the pyrolysis methods, heating using microwave radiation has an advantage of low energy demand compared to the conventional heating. Selective and bulk heating by microwave radiation leads to a lower residence time of the sample in the reactor, therefore reducing the energy demand [80]. For mechanical process, the range of energy demand is not influenced by the type of material. This is because energy requirement in mechanical recycling machine is mainly dominated by its basic power to run the machine in comparison with the actual energy used for cutting (tip energy). Energy demand reported for pyrolysis process does not consider energy recovered from combustion liquid and gaseous by-product. The recovery is estimated to be around 33 MJ to 36 MJ [42, 81] however the value depends on the type of the decomposed polymer.

Generally, the energy demand of recycling processes is lower than the embodied energy of virgin material. Recovery of the material for reuse applications can give environmental benefits as the use of virgin material can be avoided. For instance, recycling of aluminium metal matrix composites using electrolysis process used energy between 11 MJ/kg and 24 MJ/kg [79] of aluminium. The range is notably lower than the embodied energy of pure aluminium which is in the range of 196 MJ/kg to 257 MJ/kg [32].

It should be noted that the resource consumption depends on the nature of each recycling method. Each method has its mix of energy sources. Mechanical recycling depends only on electrical energy usage. Thermal and chemical recycling processes typically involve resource in the form of material usage. For pyrolysis method, continuous nitrogen flow is a must to ensure inert atmosphere and extract gaseous by-products out from the reactor. Chemical recycling requires usage of solvent such as acid nitric and alcohols. Natural gas is an input for fluidised bed process alongside with the use of electricity for cyclone, air heater, draught fan and afterburner. From literature, the use of energy for auxiliary units is sometimes not reported. The energy requirement for auxiliary units may not be significant for a laboratory-scale process. For industrial-scale process, the resources used by the auxiliary units is significant and cannot be simply neglected.

Other than that, the process by-products can be a major contributor to environmental impact. For example, a mixed solution and waste-water from chemical and high voltage fragmentation process respectively. In the thermal recycling process, liquid and gaseous by-product would an environmental concern if they are not used for energy recovery. Heat loss is highlighted in an energy flow analysis of fluidised bed process [82]. The aspect of environmental credentials is rarely considered in the literature as most of studies focused on mechanical properties of recovered fibre and reuse applications. This is where life cycle assessment (LCA) plays an important role to assess the environmental impact of rec recycling processes holistically.

7. Quality of recycled fibre and new composite product

The main objective of composite recycling is to ensure composite recyclate can be reused in potential applications. From literature, most studies focused on recovering glass fibre or carbon fibre from the composite waste. Recovery of the matrix is challenging and not feasible as the polymer structure is broken during the recycling process. Typically, the matrix or organic part is combusted for energy recovery of used as a chemical feedstock.

Quality of recyclate is determined by the comparison of physical and mechanical properties of single fibre between recycled and virgin counterpart

[83]. The aim is to recover fibre with properties comparable to the virgin fibre. This is important to ensure the commercial viability of the recycled fibre. Single fibre properties commonly reported in the literature are tensile strength and tensile modulus.

Table 7.1 shows the single fibre tensile strength reported in the literature. The ratio of strength between recycled and original fibre is highly influenced by processing parameters and type of material. Glass fibre is more sensitive towards heating temperature and residence time in the reactor. This explains why recovered glass fibre has poorer fibre strength retention of in comparison with carbon fibre in thermal recycling. For pyrolysis process, the ratio is 17-67% for glass fibre [84-86] and 80-96% for carbon fibre [87-89].

Table 7.1. Recycled fibre tensile strength compared to virgin counterpart

Process	Ratio of tensile strength of recycled fibre to virgin fibre	Sources
Chemical	0.47 – 0.99	[62-65, 67, 90-92]
Electrochemical	0.80	[77]
Fluidised bed	0.54 – 0.74	[56, 93]
High voltage fragmentation (HVF)	0.88	[72]
Mechanical	0.80 – 0.82	[53, 72]
Microwave pyrolysis	0.79	[89]
Pyrolysis	0.17 - 0.96	[84-88]

High operating temperatures of 400 °C to 600 °C can significantly degrade the strength of glass fibre. A study treated glass fibre in a temperature range of 150 °C to 650 °C for two hours [94]. The study found that the strength reduction occurred around 150 °C to 250 °C, which is below the

processing temperature used in most thermal recycling methods. The damage on the fibre also influenced by the residence time in the thermal reactor. Because of this, it is desirable to use a rapid heating technique such as microwave heating to process glass fibre based composite waste to minimise degradation of recycled fibre strength. In terms of elasticity, exposure to heat below 650 °C did not significantly change the modulus of glass fibre [94-96].

Figure 7.1 compares the mechanical performance of new composite product made from recovered fibre for each recycling method. For thermal recycling processes, recycled products are reported to have inferior performance compared to the virgin counterpart. This is attributable to fibre sizing degradation during recycling process hence leads to poor interfacial bonding between the recovered fibre and new polymer as a binder. Besides, resin elimination may not be 100% and there is residual resin on the fibre surface. Coupling agents can be used to enhance bonding. For mechanical recyclates from glass fibre waste, silane was added in resin to improve adhesion with recycled fibre or filler [51, 97, 98].

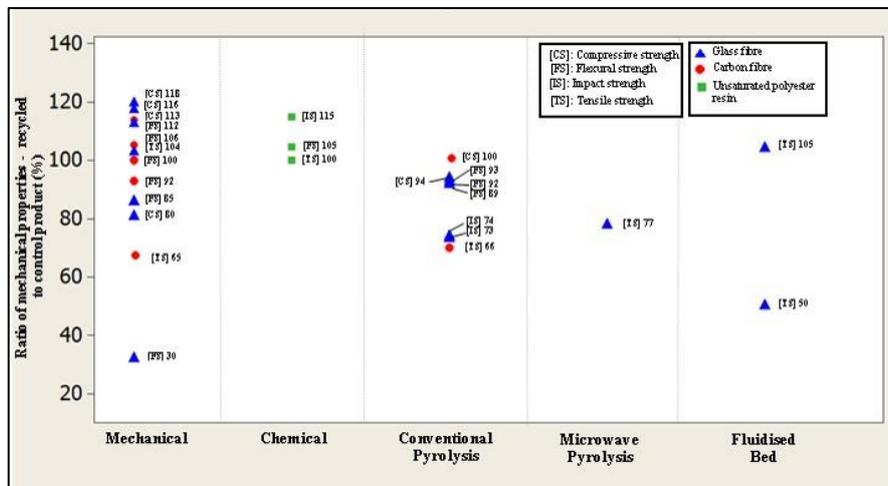


Fig. 7.1. Comparison of mechanical properties between recycled and control products adapted from Shuaib and Mativenga [80]

More reuse applications are found in a mechanical process than the other recycling methods. This is because mechanical recycling is available at industrial scale to recycle GFRP waste [99]. Besides, the method has a high

recycling rate with minimum energy consumption. It is expected that chemical, microwave pyrolysis and fluidised bed processes have fewer re-manufactured products as most of the studies only used small or laboratory scale processes.

8. Application of composite recyclate

Material costs can be reduced by nearly 45 and 50% with recycled production and end-of-life (EoL) glass and carbon fibre wastes, respectively [100]. Therefore, several end products have been manufactured from composite recyclates from retired composite products. These end products include, but are limited to ladders, table tennis bats, house doors, laser pointer case, among others [83]. In other words, there are many potential applications for ground FRP composite recyclates, as investigated and reported. For instance, a desirable natec pedal crank has been fabricated with recycled carbon FRP composites [101].

Besides, composite recyclates are sometimes applied as reinforcements: fillers, particulates and short/chopped fibres for manufacturing of some improved structural and non-structural composite components [100]. The properties and applications of the newly recycled composite materials depend on the nature of recyclates and recycling methods. For simplicity, numerous applications of composite recyclates in different manufacturing industries are subsequently classified and elucidated.

8.1 Automobile sector

With the leading and increasing application of composites in automobiles [102], automobile industries have taken a strong initiative and developed the strongest interest to add more recyclable components in automotive structures. For examples, a mixture of polymer materials/recyclates have been identified to produce a few automobiles separating functional units, including dashboards and bumpers. New bumpers have been developed from either recycled polycarbonate (PC) or polybutylene terephthalate (PBT) bumper, while recycled thermoplastic olefin bumpers have produced

other automobile parts, such as bumper fascias, air dams, splash shields as well as air claddings, after preferably reinforced with recyclate/fibre.

In addition, brackets that accommodate car radio antennae, small under-the-hood-parts and splash shields have been manufactured, using recycled acrylonitrile butadiene styrene (ABS) and polyester or PC alloys. Also, power-train applications, air conditioning evaporative housings, fender liners as well as vents have been produced from recycled polypropylene (PP), while polyethylene terephthalate (PET) has been recycled and used in headliners and car engine covers [102, 103]. An iStream hybrid structural FRP composite chassis has been developed. It was simple, has low cost steel tubular members and 14 composite ipanel, compared with conventional stamped steel chassis with typically 100s of stamped metal panels [104]. Each rCF ipanel cost nearly €30, while that of conventional woven fabric prepreg cost €300/panel. Now, is it possible to manufacture a car from 100% recycled materials in the future: 2030 or 2050 [1]?

8.2 Aerospace industry

The aircraft fleet recycling association (AFRA) (formed by Boeing and 10 other aerospace companies), Airbus process for advanced management of end-of-life aircraft (PAMELA), materials innovation technology (MIT), recycled carbon fibre limited (RCF) have been committed for more than a decade to improve management of retired aircrafts. By 2029, 10-15 million pounds of CF recyclate have been estimated to be generated by the aerospace industry from manufacturing and EoL aircrafts [1].

8.3 Wind energy

Wind energy is one of the global and increasing renewable energy supply. It is produced by wind turbines, with 40% of total blade mass. Nearly all turbine blades are produced from glass fibre/epoxy composites, with a glass-epoxy ratio of 60%. Both manufacturing and EoL turbine blade of average lifetime of two decades generate a huge amount of composite wastes, with an estimated quantity of more than 1 million tonnes over the

next two decades [105]. Due to the fact that the key turbine parts are made up of thermosetting composite materials, recycling and disposal of new and EoL scrap is a growing global challenge. Unfortunately, there is no commercial operations to embark on recycling of the new and EoL wind turbine composite materials.

8.4 Electrical and electronic industry

Milled recycled carbon fibres (rCFs) are used to manufacture conductive materials that provide electrical conductivity and antistatic behaviours in polymeric composites and coatings. For example, epoxy antistatic floor coatings and integrated circuit trays [104].

8.5 Other applications

- *Subsea buoyancy* – Milled rCF are used in deep sea oil and gas exploration [104]. They have the largest volume market for milled CFs.
- *Manufacturing (Additive/3D printing and net-shape)* – Milled rCFs are used to produce filaments for 3D printing or additive manufacturing (AM) processes [104]. For instance, these materials can be used to fabricate prototyping objects, drones and cars. The first 3D- printed car from Local Motors contained 20 and 80% of rCF and ABS plastic [104], respectively. Additionally, chopped rCF can be used in net-shape manufacturing of parts, through the application of preforming for resin transfer moulding or stamp forming process.
- *Building/construction company* – Pulverished waste FRP composite products were converted to artificial woods experimentally [106]. The woods can be nailed, drilled, notched and sawn, showing their similar properties when compared with natural woods. Also, ground glass reinforced polyester (GRP) has been used diversely: GRP/plastic lumber with bolted tenon joint to solid wood and GRP/wood flake blend particleboard [107].

- *Compounding* – The mechanical properties (essentially stiffness) of some injection moulded milled rCF polymeric compounds. This could be chopped or pellet rCF used in sheet and bulk moulding compounds (S/BMC), especially in applications where long fibres are not suitable for complex geometry, small products and exact requirements of surface quality.

Importantly, in addition to the cost reduction, the mechanical properties of the reCF reinforced products increase [104].

Finally, further improvement on development of more economical processing, optimisation techniques, the performance of recycled composites as well as the general acceptance of recycled composite products by educating the markets is required to increase applications of the composite recyclates, as there are no end-users for the recycled materials [1]. In addition, formulation of models for predicting the quantity of recyclates, recycled material value and effect of the variance of processing parameters on recyclate quality, cost and environmental impact are recommended for future works, probably towards vision 2030, 2050 and beyond.

9. Conclusions

Rising of demand for the composite material, particularly on the fibre reinforced type comes together with manufacturing and end of life waste problem. The heterogeneity and cross-linked nature of thermoset polymer lead to a challenging task for the recovery of material during recycling stage. Legislation and the high value of fibre are drivers for composite users to recycle their waste.

Studies on composite recycling methods are reviewed in this chapter. Most recycling technologies mostly developed to recover polymer based composites on account of its high demand and waste volume hence raises the urgency for recycling. Mature recycling process such as mechanical and pyrolysis are operated at an industrial scale and commercially available. Recycling methods such as microwave pyrolysis, high voltage fragmentation and others are relatively new and only conducted at a pilot or laboratory setup. In terms of environmental impact, the process energy demand for

recycling is far lower compared to the embodied energy of the original fibre. The energy demand is highly driven by the processing rate.

Recovered fibre and new composite product made from the recyclate have comparable performance with virgin counterparts. Potential applications in the automotive, aerospace and renewable energy sector are suggested in this chapter. The commercial viability of composite recycling only be realised if virgin fibre can be substituted by recycled fibre in both active and passive applications. Recycling of fibre reinforced composite waste is important to avoid loss of valuable materials. Using the concept of circular economy, recyclate can be returned into the market via closed loop or cross-sector applications.

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