Recycling of composite materials

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In this report, an overview of the most common recycling methods for composite materials is created. It describes the process and recyclate characteristics, as well as the (dis)advantages of each method and the environmental and technological development. To draw conclusions on the current feasibility of composite recycling, the gathered data is analysed on the subject of application, quality, economy and sustainability. The report ends with a future outlook, and what implications this might have.

1. Introduction

1.1 Composite materials

Composite materials are materials which are constructed of two or more materials to be able to benefit from a combination of material properties. One material acts as binder, also known as the matrix, which surrounds and binds fragments of the other material, the reinforcement (Royal Society of Chemistry, 2015), which can be a fibre or particle. Sometimes, filler materials are added as well for additional benefits, such as cost reduction or fire retardance. Composite materials are widely used in automotive, aerospace and renewable energy industries, as these materials have a higher strength, lower weight and less maintenance compared to conventional materials, as well as a long lifespan (Yang et al., 2012).

For most applications the type and proportion of resin, reinforcement and filler are tailored to the particular end use, which means that there are many types of composites (Pickering, 2005). Classifying composite materials can be done in two ways, namely structure (Figure 1) and material use (Figure 2). The structure describes the way in which a composite is build up, whereas the material use describes which materials are used for both the matrix and the fibre.

This report will focus on fibre-reinforced and structural polymer composites, using a thermoset or thermoplastic matrix with glass or carbon fibres.

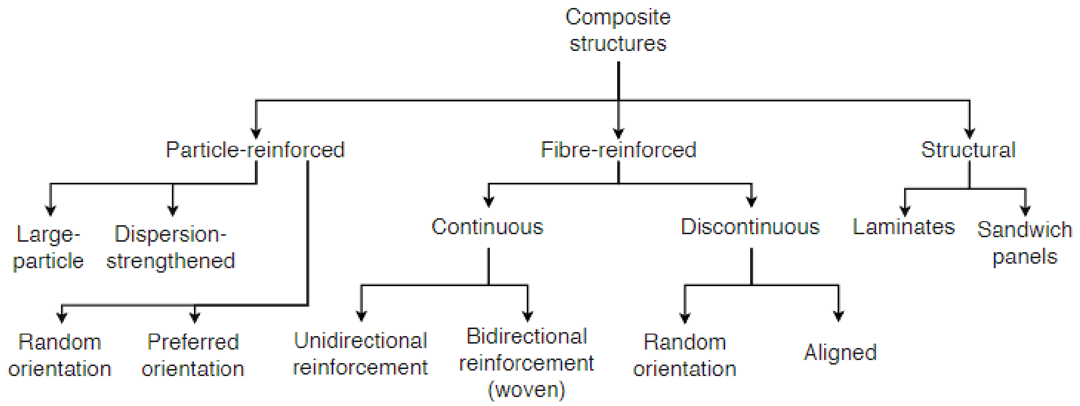

Figure 1: Classification of composite materials based on structure (Jayaram & Lang, 2013; Park & Seo, 2011)

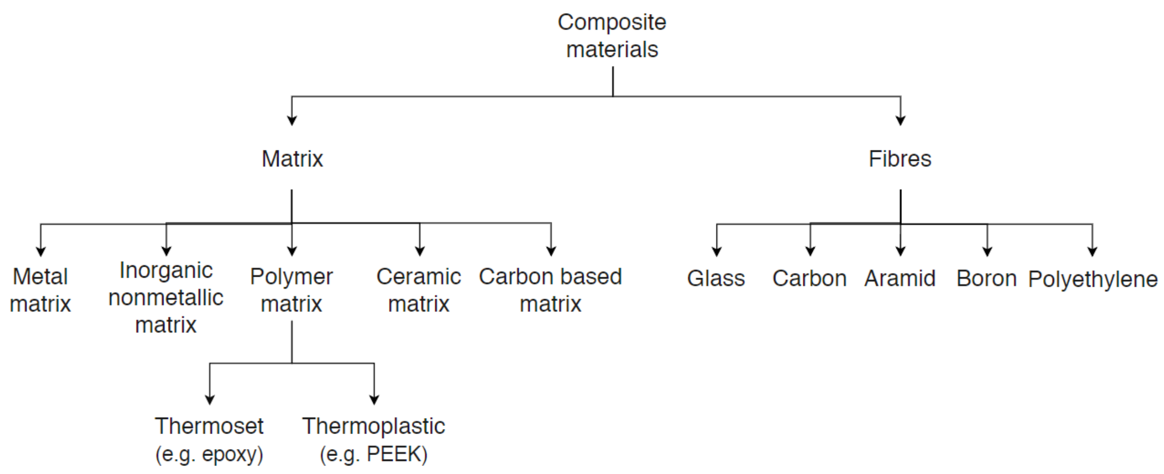

Figure 2: Classification of composite materials based on material use (Aravind et al., 2013; Wanhill, 2017)

1.2 Recycling

Although composite material products usually have a long product lifetime (Ribeiro et al., 2016; Yang et al., 2012), the first generation of composite products such as windmills, are reaching their end of life (Cherrington et al., 2012; Liu & Barlow, 2017). This poses a new problem, namely the recycling of composite waste. The material is constructed in such a way that it is strong, durable and non-homogeneous, which makes it inherently difficult to recycle (Job et al., 2016). Also, composite materials are often used in combination with other materials such as foam cores (Pickering, 2005), which leads to mixed waste streams. As the use of composite materials is expected to grow (Job et al., 2016), it is important to come up with a solution to be able to deal with the increasing waste. Currently, most composite waste ends up as landfill (Rybicka, Tiwari, & Leeke, 2016). However, when considering the movements towards a more sustainable future with developments, such as landfill bans in European countries and end of life directives (Job et al., 2016), not having a satisfactory way to recycle composite materials may inhibit future applications. There is a lot of research and testing on how these materials can be best recycled, which will be explored in the research section of this paper.

2. Method

2.1 Aim

The aim of this report is to provide an overview of the current recycling options for composite materials, the technological readiness level of these methods, and how feasible it currently is to recycle composite materials. Also, it sketches a future outlook, and briefly mentions what implications this could have for the current approach.

2.2 Approach

This research report consists out of three research topics, namely composite materials, recycling theory, and the practical application of composite recycling, followed by a future outlook and future implications.

For the composite material and recycling theory study, first an analysis was made of the characteristics of composite materials, as well as the current composite market. This was made by perusing a number of papers and market reports that were found searching using composite market related keywords.
Then, a literature review was made to create an overview of all the possible recycling methods and to get an idea of future developments. In this second review, a base set of overview papers was used, from which there was then expanded into interesting directions using additional papers. The main papers used were:

1. Recycling of composite materials (Yang et al., 2012)
2. Recycling technologies for thermoset composite materials - current status (Pickering, 2005)
3. Current status of recycling fibre reinforced polymers: Review of technologies, reuse and resulting properties (Oliveux, Dandy, & Leeke, 2015)
4. Composites recycling: Where are we now? (Job et al., 2016)
5. Technology readiness level assessment of composites recycling technologies (Rybicka et al., 2016)

To analyse how composite recycling held up in practice, the previously gathered data for each recycling method and virgin material data were put together and compared. Additionally, it was researched if there are other options for end-of-life composite materials other than recycling it, and how feasible these options are, as well as a short note on the role of composite materials in a circular economy.

For the future outlook, the found data was extrapolated. This was done by both finding evident data and conductive research in expected directions, as well as using educated guesswork. Then, using the future outlook, the implications for the future were shortly discussed, and a composite lifecycle diagram was constructed.

2.3 Relevant frameworks

This section introduces three frameworks that will be used and referred to in this report, namely the waste management hierarchy (WMH), technological readiness level (TRL) framework, and the circular economy. The waste management hierarchy and technological readiness levels will be used to indicate the sustainable desirability and technological development of the recycling methods discussed in this report. The circular economy is relevant context to illustrate why it is important that end-of-life composite scrap gets recycled back to a valuable material with viable applications. At the end of this paragraph, a list of definitions and abbreviations used in this report can be found as well.

2.3.1 Waste management hierarchy

The waste management hierarchy (European Commission, 2016) shown in Figure 3 was developed to illustrate the priority order for dealing with waste. It shows that only prevention works to prevent waste, whereas the following steps only serve to reclaim as much value from the waste as possible. The lower the level, the less value is recovered. The framework is part of the Directive 2008/98/EC on waste, which also requires that Member States adopt waste management plans and waste prevention programmes.

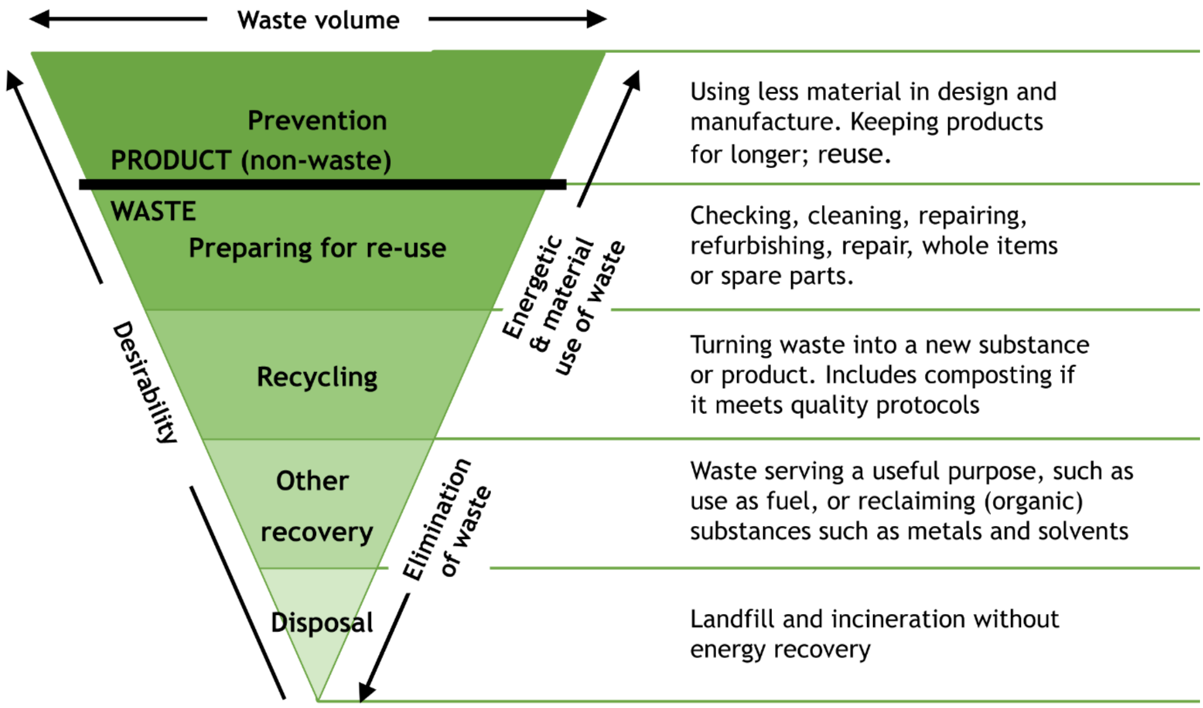

Figure 3: The waste management hierarchy (DEFRA, 2011; European Commission, 2016; Ziegler et al., 2006)

Other relevant waste directives are the Landfilling Directive (1999/31/EC), the Packaging Waste Directive (94/62/EC), and the Directives on end-of-life on vehicles (2000/53/EC), on (waste) batteries and accumulators (2006/66/EC), and on waste electrical and electronic equipment (2012/19/EU) (Davies, Lee, & Braasch, 2018). More information on legislations and directives can be found in section 4.2.2 Legislation.

2.3.2 Technological Readiness Level

The Technology Readiness Level (TRL) framework is used in many variations across industries to measure the technology maturity of a project (Nakamura, Kajikawa, & Suzuki, 2013). It can also be adapted to support the understanding of capabilities and resources that are needed to develop technologies at different stages of the development (Rybicka et al., 2016). The most extensive TRL framework is the NASA framework developed by Conrow (2011) as shown in Table 1. A more comprehensive version adopted can also be used, defining TRL 1-3 as lab scale, TRL 4-6 as pilot scale and 7-9 as commercial scale (Rybicka et al., 2016).

Table 1: Technology readiness level (TRL) NASA definition (Conrow, 2011)

TRL	Definition
9	Actual system ‘‘mission proven’’ through successful mission operations (ground or space)
8	Actual system completed and ‘‘mission qualified’’ through test and demonstration in an operational environment (ground or space)
7	System prototyping demonstration in an operational environment (ground or space)
6	System/subsystem model or prototyping demonstration in a relevant end-to-end environment (ground or space)
5	System/subsystem/component validation in a relevant environment
4	Component/subsystem validation in a laboratory environment
3	Analytical and experimental critical function and/or characteristic proof-of- concept
2	Technology concept and/or application formulated
1	Basic principles observed and reported

2.3.3 Circular economy

The circular economy concept has been around for a long time, but it has become more popular since the late 1970's. The goal of the circular economy is to (re)build economic, natural and social capital, using renewable energy. Also, it recognizes the importance of the economy to work effectively at all scales, from individuals to large organizations and businesses, both locally and globally.
There are three important principles in the circular economy:

1. Design out waste and pollution
2. Keep products and materials in use
3. Regenerate natural systems

The value circle in Figure 4, also known as the butterfly diagram, illustrates how enhanced and continuous flows of goods and services can be created by cycling materials in a technical and biological cycle (Ellen MacArthur Foundation, 2017).

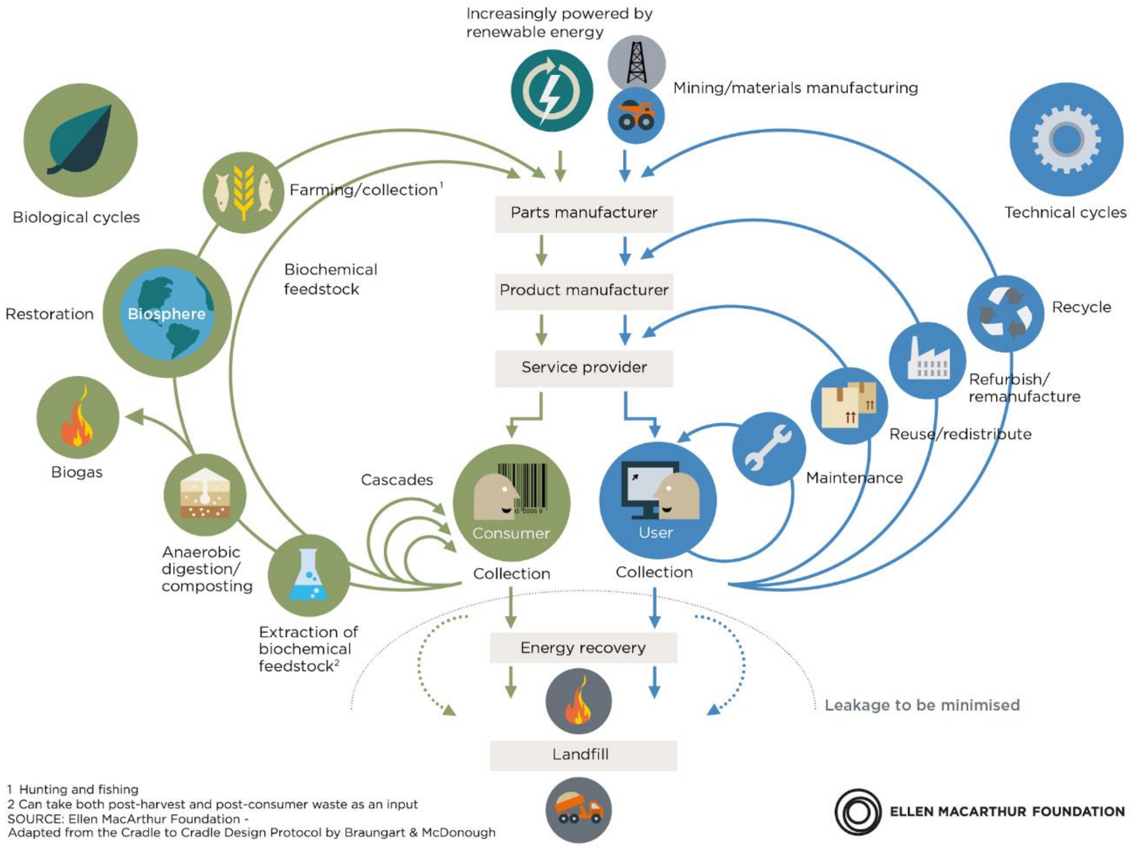

Figure 4: The value circle/butterfly diagram of the circular economy (Ellen MacArthur Foundation, 2017)

When placing composite materials in the context of the circular economy, it can be seen that it is important to realize that the material can move through the flow like any other material. Until this has been realized, the composite material will have no place in the circular economy, regardless of its sustainable benefits (e.g. saving weight and fuel). The difficulty in moving a composite material through these flows lies in the fact that the fibres and fillers are embedded in the resin material, and that these are hard to separate. In this report, there will be discussed what attempts have been made so far, and what future development is needed to correctly recycle composite materials so that they can have a place in the circular economy.

2.4 Terminology

Below, a list is given with standard definitions and abbreviations used in this report and in the field of composite recycling. This list has been extended using the list of definitions for circular product design (composed by J. Joustra and R. Balkenende of the IDE Circular Product Design department of the TU Delft).

Term	Definition
CF/GF	Carbon fibre/Glass fibre
CFRC/GFRC	Carbon fibre reinforced composite/glass fibre reinforced composite
EfW	Energy from waste, common with waste incineration plants
EPR	Extended producer responsibility; expected future legislation along lines of life-cycle thinking. This would include recycling and recovery targets for manufacturers
Glass rovings	Large diameter filaments of ≥10 µm consisting of multiple strands, used for reinforcement in structural composites (Chawla, 2012, p. 16; Liddell et al., 2017)
Glass yarns	Flexible <10 µm filaments, that are generally woven into fabrics (Liddell et al., 2017)
Lifecycle	The functional life of a product; starts when released for use and ends when definitively disposed or dismantled. Lifecycles of individual components can continue in new products.
Use cycle	Starts when released for use and ends when the user of the product changes or when the lifecycle ends. A single product lifecycle can consist of multiple use cycles, for example through sharing, resale or remanufacturing.
Repair	Correcting specific faults to restore the product to satisfactory working condition. Also referred to as corrective maintenance.
Repurposing	Utilizing a product or its components in another function than in its original use. Might be taken into account in the initial design
Reuse	All actions where a product are used again, with the same purpose or little changes to the original.
Recyclate	The material product that is generated as output of a recycling process
Recovery	On the level of products: returning a product to usable (functional) condition through for example reuse, repair, refurbishment or remanufacturing actions. On the level of parts: Parts harvesting On the level of materials: recycling (which might be upcycling or downcycling) In the Waste Framework often used to describe energy recovery (through incineration) of materials. Energy recovery is explicitly considered not to be a Circular way of handling resources as it incurs complete loss of materials.
Parts harvesting	Disassembly, cleaning, testing and stocking of product components to prepare them for a next use cycle, often as spare component in another product
Recycling	Preparing materials for a next lifecycle through reprocessing.
Up/downcycling	Recycling materials into new materials with higher/lower performance or functionality. Also referred to as primary/secondary recycling
Refurbishing	Returning a product to good working condition by (preventive) replacement of components by OEM or third parties, usually the start of a new product use cycle. (limited) warranties on product functionality may be given
Upgrading	Extending a product's original properties to offer improved performance
Adapting	Adjustments to a product to meet changing use conditions and demands, without adding new functions
Remanufacture	Disassembly, cleaning, testing and reassembly of a product resulting in a product that meets at least the original specifications
SCF	Supercritical fluid; the properties of supercritical fluids (SCFs) are in between liquid and gas phases, and benefit from low viscosities, high mass transport coefficients, and high diffusivities (Hyde et al., 2006)

3. Composites

This chapter will give a more in-depth view on what types of composite materials there are and what they are currently being used for. There is a large variety in composite material types that all have their own characteristics. Understanding these differences helps to see why recycling methods need to be flexible and adaptive to handle these mixed waste streams.

3.1 Types of composites

Figure 5 shows that in 2010, the raw materials shipped most for manufacturing composite materials were polyester resin and glass fibres (Gutiérrez & Bono, 2013). The global composites material market (fibres and resins) in 2016 was approximately 12 million tons. The glass fibre market determines the market size with 99% in volume, versus 1% in volume for the carbon fibre market (Effing, 2018).

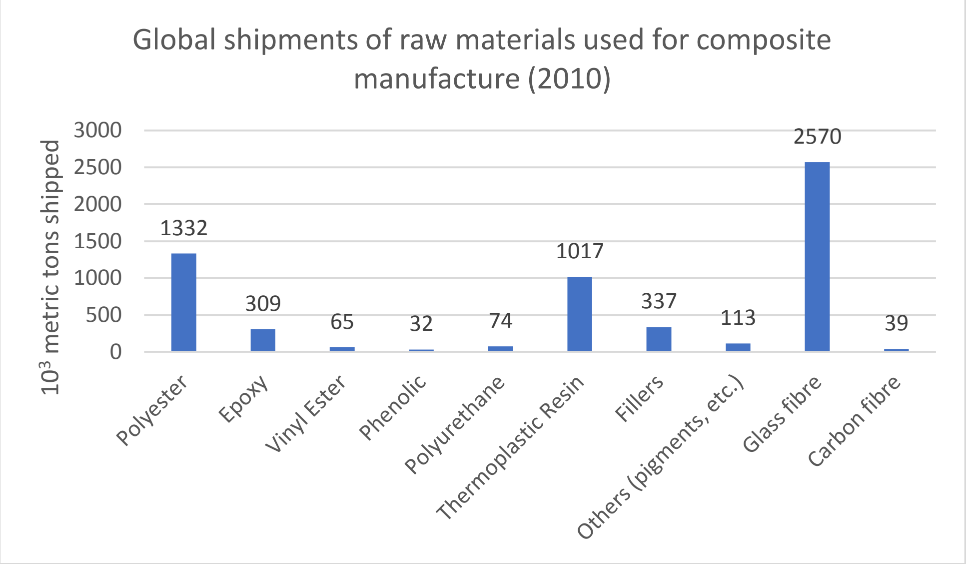

Figure 5: Global shipments of raw materials used for composite manufacture in 2010 (in 10^3 metric tons shipped Figure data retrieved from Statista (2018)

3.1.1 Fibre type

Glass fibre

Glass fibres are used the most in composite materials: 95% of all composites types use glass fibre as reinforcement (Effing, 2018; Gutiérrez & Bono, 2013; Witten, Thomas, & Michael, 2015). Glass fibres are cheap, and available in a variety of forms. Commonly used types are E glass, C glass and S glass. E (electrical) glass is a good electrical insulator, C (corrosion) glass has better resistance to chemical corrosion and S (high silica) glass can withstand higher temperatures than other glasses.
In general, glass fibres and have high strength and low density, resulting in a quite high strength-to-weight ratio. However, the modulus of glass fibres is not very high, which means that the modulus-to-weight ratio is only moderate. This led to the usage of more advanced fibres such as carbon. Glass fibres are also susceptible to strain fatigue, which means that they wear down when subjected to a constant load for an extended time period. This leads to failure over time at loads that might be safe with instantaneous loading. Furthermore, moisture also decreases glass fibre strength (Chawla, 2012, pp. 11–16).

In 2010, there was 3.82 million metric tons of glass fibre rovings being produced globally (Liddell et al., 2017). In Europe, the glass fibre reinforced plastics production volume was growing at a rate of 2.5% in 2015, reaching 1,069 megatons (Witten et al., 2015). In the US, the glass fibre market grew by 4% in 2017, reaching 2.5 billion pounds (equals 1.1 billion metric tons), and is expected to grow further with a compound annual growth rate of 3.4 percent (Mazumdar, 2018).

Carbon fibre

Advanced composites such as carbon fibres represent only 1.5 % of the total composite market by weight. However, they account for over 20% if the net market value (Gutiérrez & Bono, 2013) as they are expensive to produce due to high cost raw materials and a complex manufacturing process (Baker & Rials, 2013; Park & Seo, 2011). Carbon fibres are mostly engineered for strength and stiffness, but they are available in multiple grades. They can vary in electrical conductivity, and thermal and chemical properties, depending on the carbon content and fibre structure. Commercial carbon fibres are made by extruding organic precursor materials into filaments, which are then carbonized to turn them into carbon. Depending on the desired properties, different precursors and carbonization processes are used (Park & Seo, 2011). A commonly used precursor fibre is polyacrylonitrile (PAN), but purified rayon and pitches such as petroleum asphalt or coal tar are used as well (Chawla, 2012, pp. 24–36). PAN-based carbon fibres typically have a higher specific strength and lower specific modulus and are generally less expensive, whereas pitch-based fibres have higher electrical and thermal conductivity. (Chawla, 2012, pp. 24–36; Park & Seo, 2011).

In 2010, there was 33 thousand metric tons of carbon fibre being produced globally. The demand for carbon fibres has grown with a compound annual growth rate of 11.5% from 33 metric tons in 2010 up to 63.5 tons in 2016. A further positive grow of 10-13% is expected for the following years, reaching 117 metric tons in 2022, as shown in Figure 6.

Most of the carbon fibres produced was used for use in carbon reinforced composites. The demand for carbon fibre composites has grown with a compound annual growth rate of 11.98% from 51 metric tons in 2010 up to 101 tons in 2016. A further positive grow of 10-13% is expected for the following years, reaching 194 metric tons in 2022 (Witten et al., 2015).

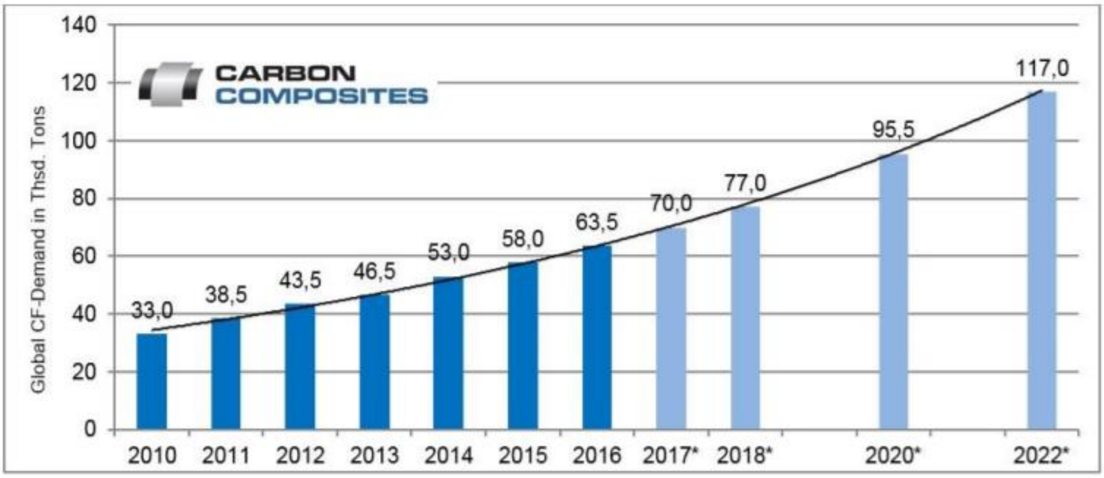

*Figure 6: Development of global carbo fibre demand in thousand tons from 2010 until 2022 (*Estimations, 09/2017;* Witten et al., 2015)

3.1.2 Matrix type

Thermoset matrices are most popular, with an estimated global market share of 83.3% in 2015. The most common polymer matrices are thermoset polyester and epoxy resins (Chawla, 2012, pp. 73–103).

Unsaturated styrene-hardened polyesters have the majority of low-to-medium performance applications, whereas epoxy and other more sophisticated thermosets have the higher end of the market (Singh, Goga, & Rathi, 2012).

Compared to thermoplastics, thermosets are stronger due to cross linking. Also, they have much lower creep, higher dimensional stability, and are suited for high temperatures (Grand View Research Inc., 2018; Ratna, 2012). However, they are brittle (Chawla, 2012, pp. 73–103) and cannot be remoulded after initial heating (Grand View Research Inc., 2018).

Thermoplastic composites are becoming increasingly attractive. They have high strain-to-failure values, compared to thermosets (Chawla, 2012; Singh et al., 2012), and they are less expensive, weldable, and non-toxic (Grand View Research Inc., 2018). Also, they are able to be recycled to some extent as they can be reheated and reshaped (Chawla, 2012; Grand View Research Inc., 2018). However, they are more difficult to process due to high viscosity and large expansion (Chawla, 2012; Singh et al., 2012). Also, to achieve reasonable levels of physical heat resistance, the softening (or glass) transitions must be relatively high which also influences the processing costs (Kandola & Horrocks, 2005). Thermoplastic resins are also more sensitive to organic solvents (Ratna, 2012). Currently, new development is being made to create new thermoplastic matrix materials with improved material properties (Singh et al., 2012).

3.2 Market outlook

3.2.1 Key players

The map in Figure 7 from the Suschem White paper by Devic et al. (2018) illustrates the main industrial and academic players in Europe, selected based on their recycling capacity and their activity level in developing new technologies. There are also a number of parties outside Europe, such as in the United States and Japan.

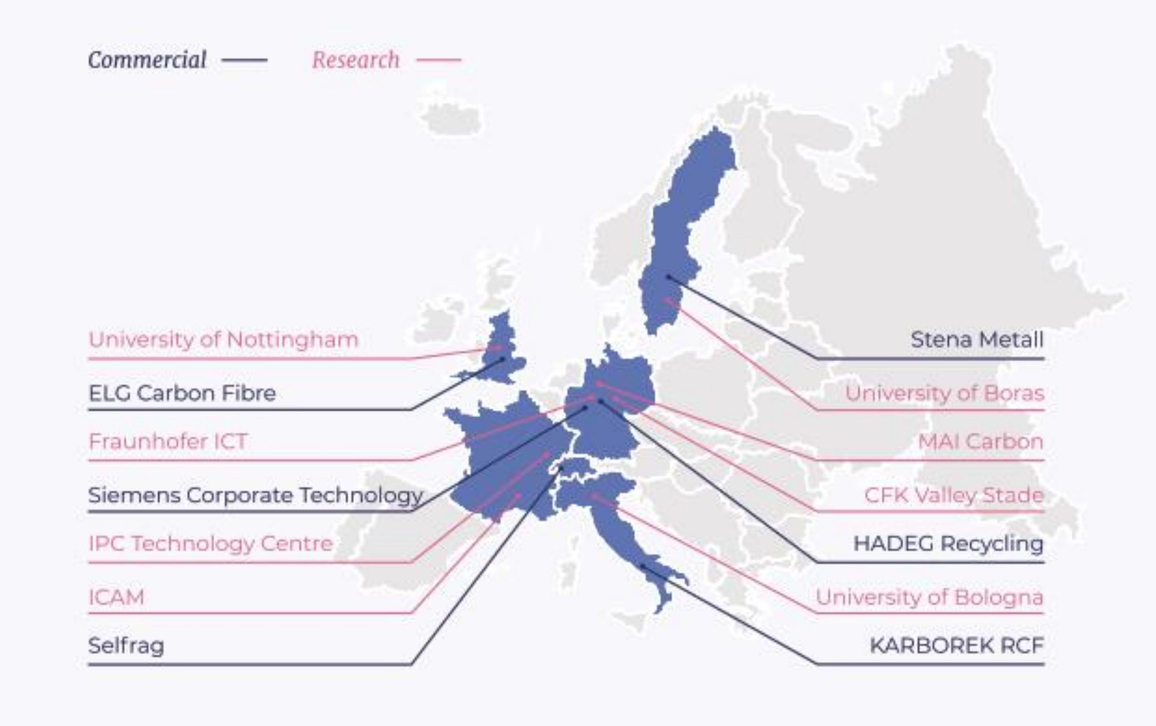

Figure 7: Key players in commercial and research applications.
Image retrieved from Devic et al. (2018)

3.2.2 Market shares

According to Hacıalioğlu (2018), the biggest industry sectors in 2016 for composite materials seen globally were logistics (28% of volume) and construction and building (18.7%; see Figure 8). The wind energy sector accounted for 6.8% of the volume. Interestingly, the defence and aviation industry held only 0.4% of the volume, but they accounted for 13.0% of the total market value. This can be assigned to the fact that there are differences between sectors in terms of the standard manufacturing processes and quality requirements (Holmes, 2014), which affects the material value levels.

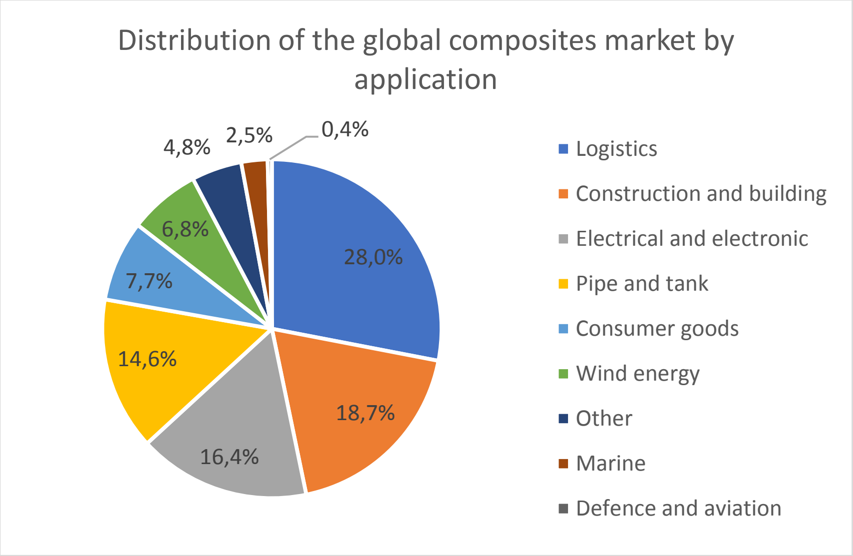

Figure 8: Distribution of the global composites market by application (Hacıalioğlu, 2018)

When looking more specifically at each fibre type, there can be seen that there are differences between the main applications of glass and carbon fibres. It is difficult however to find cohesive data on all the numbers and values of the composite market, and to find comparable data for glass and carbon fibre (composite). The data often varies in regard to e.g. fibre/composite type, geographic location, year, and data type (e.g. numbers vs. percentages). Additionally, market reports are often only available against a very high price, or offer only very limited insights into very specific aspects. As this report does not aim to research the composite market, the data found below is not conclusive and cannot be used to compare the fibre type, but serves only to illustrate the order of magnitude, applications, and areas of interest for the fibre and composite markets over the past years.

Glass fibre

In 2010, there was 3.84 million metric tons of glass rovings being produced globally, of which 790 thousand tons (20.6%) in North-America (JEC Group, 2011). Figure 9 shows the distribution of the glass fibre market in North America. Here, there can be seen that the majority of glass fibres is used for automotive purposes (220.4 million kg; Liddell et al., 2017).

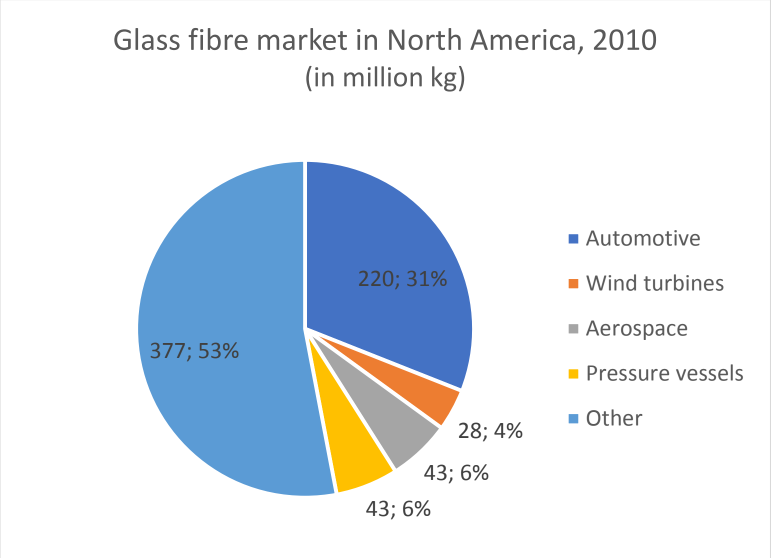

Figure 9: Estimated makeup in million kg of the 2010 glass fibre market in North America (glass rovings only)
Source: EERE, retrieved from Liddell et al. (2017)

The volume of GRP manufactured in Europe was 850 thousand tons in 2010 (JEC Group, 2011) and reached 1.118 million tons in 2017. Figure 10 shows the distribution of the different application industries, in which transport and construction are the biggest contributors. Production of thermoplastics, used primarily in the automobile industry, is generally still growing more strongly than production of most thermosetting materials (Witten, Sauer, & Kühnel, 2017).

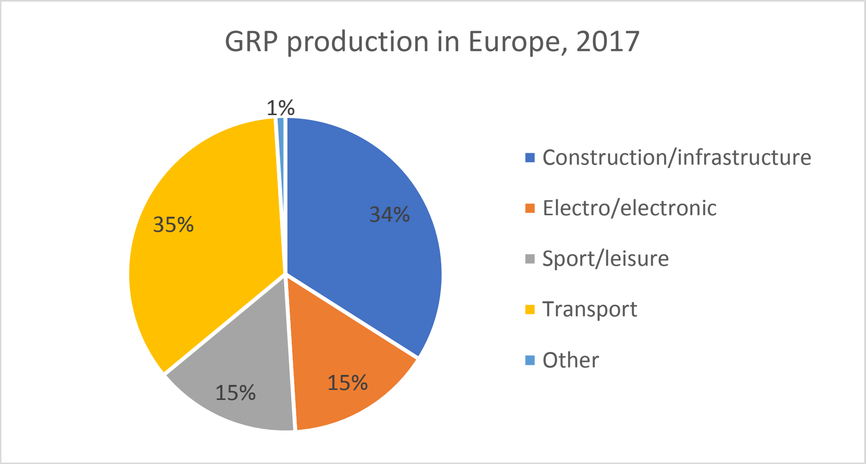

Figure 10: GRP production in Europe in 2017, by application industry (Witten et al., 2017)

Carbon fibre

Figure 11 shows the global carbon fibre demand in 2013, which reached a total of 46,500 tons. Compared to the previous year, aerospace and defence applications (13.9t, 30%) have grown significantly. This sector is now the largest consumer of carbon fibre as fuel-efficient and cheap travel require lightweight materials. The wind turbine industry is a considerable sector as well (14%). Global wind energy capacity has been expanding continuously since 2005, and the energy capacity of each rotor (and thus blade size) has been increasing as well. These multi-megawatt wind turbines are increasing the demand for carbon fibre as it is the only material suitable for constructing rotor blades with a length of 40 to 50 m. The automotive segment is also becoming increasingly important, with consumption of carbon fibre more than doubling over that past year. This was probably due to the ramp-up phase for the production of the BMW i-model cars (Grand View Research Inc., 2017; Holmes, 2014).

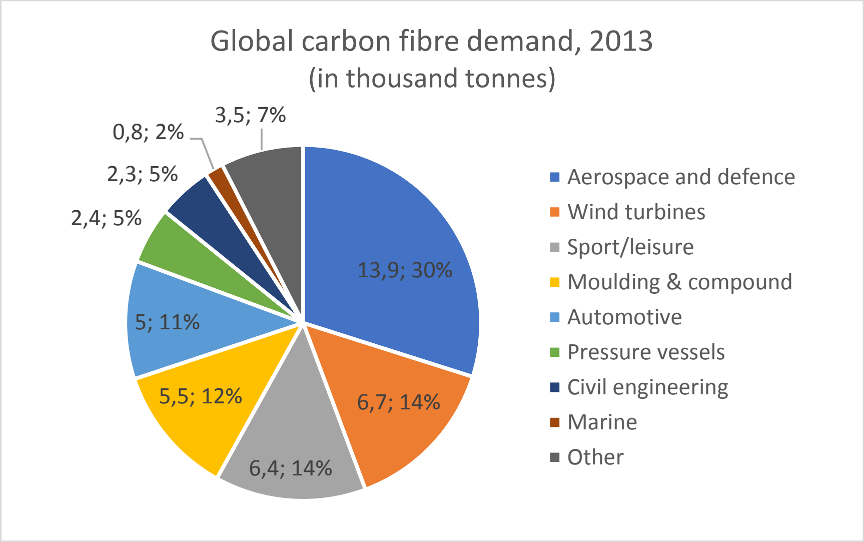

Figure 11: Global carbon fibre demand by application in thousand tonnes in 2013 (Holmes, 2014)

4. Recycling in theory

As stated before, moving towards a circular economy requires the ability to recycle composite materials. This chapter looks at what difficulties there are when recycling composite materials, as well as what incentives there are to motivate development in this field.

For recycling, the focus of this report will be shifted towards Europe. This because it has a better overview on the legislation and the available opportunities. Also, Europe is predicted to be the first to encounter the composite waste problem. For example, Figure 12 by Liu & Barlow (2017) shows the predicted wind turbine composite waste from the blades, where Europe precedes the other countries by almost ten years. Additionally, according to the Environmental Services Association (ESA) (2004), 95% of environmental legislation is driven from the EU (Cherrington et al., 2012). This will make Europe a leading example in composite recycling for the rest of the world to follow.

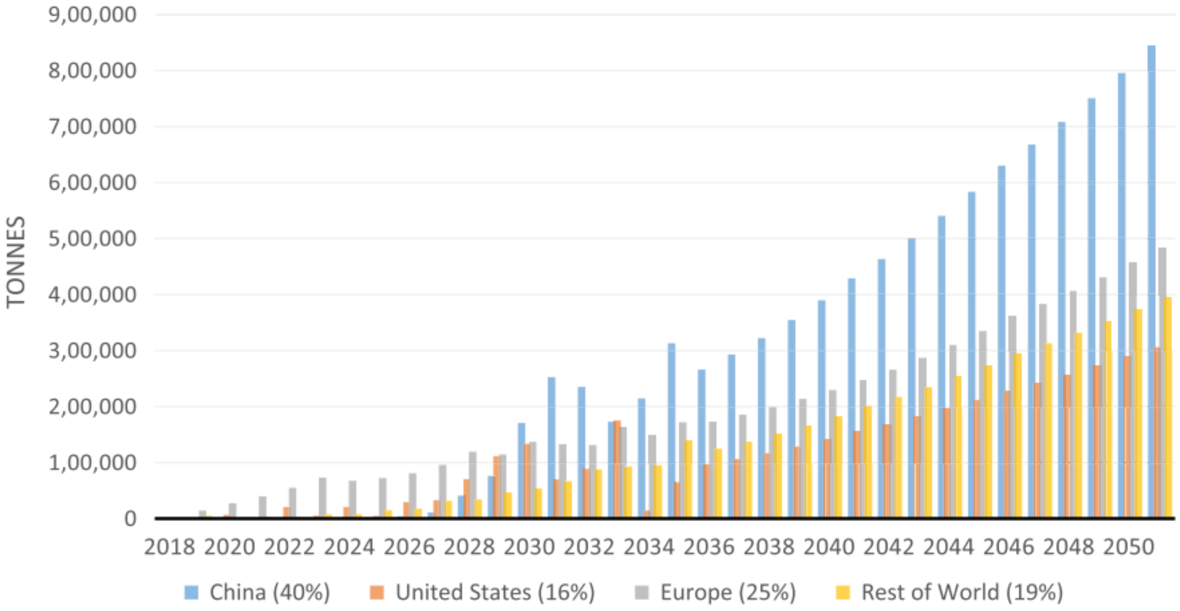

Figure 12: Regional wind turbine waste projection up to 2050 (Liu & Barlow, 2017)

4.1 Difficulties

The structure of a composite material makes it inherently hard to separate the different materials again. This creates a number of problems. Firstly, there is the monetary and energy costs of recycling the materials, compared to creating virgin materials. For glass fibre for example, the price of virgin glass fibres is so low that no process currently available can provide recycled glass fibres with the same characteristics as virgin fibres at a competitive price (Oliveux et al., 2015).

Secondly, the materials are always at a lower value level as they are decreased in quality due to the recycling process and lifetime wear, or reduced to their feedstock elements (Yang et al., 2012). This is also illustrated in the value graph made by Chawla (2012, p. 193) as seen in Figure 13. Here, the value change of the different constituents is plotted over the lifecycle of the materials. When cycling back, it can be seen that the value loss, illustrated with the arrows with the dashed lines, becomes bigger with each step. When looking to retain higher mechanical performance over consecutive recycling loops, it is important to optimise the fibre reclaiming process and avoiding fibre damage (Longana et al., 2016), both during product use and during the recycling process.

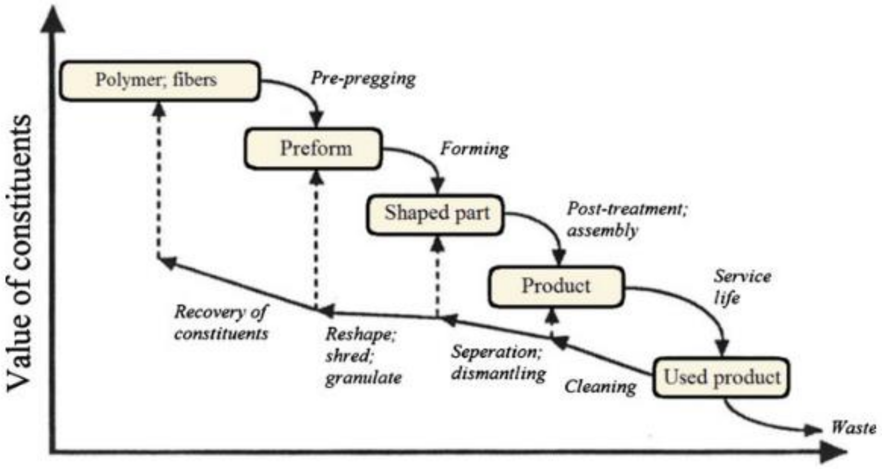

Figure 13: Change in value of the constituents with increasing life cycle (after Manson, 1994) (Chawla, 2012, p. 193)

Thirdly, as the material is of lesser quality, it has fewer application purposes and is more difficult to use than virgin materials. For example, recycled fibres are short and of a fluffy nature, which makes it difficult to incorporate it into a new material due to reduced bonding between the fluffy fibres and the new matrix (Oliveux et al., 2015).

Considering these drawbacks, it might not make sense from a sustainable perspective to use and recycle composite materials in the current manner. This would suggest that there needs to be a change in how the composite materials are currently constructed, used, and/or recycled. A desirable recycling method would deliver materials of near-virgin quality against a cost cheaper than creating virgin fibres.

4.2 Incentives

Successful composite recycling and use requires incentives, infrastructure, good recycling techniques, and viable markets for the recyclate materials (Tarverdi, 2009). This section will consider the incentives currently in place. According to Cherrington et al. (2012), there are three main motivators that define composite recycling incentive: environmental impact, legislation, and economic opportunity.

4.2.1 Environmental impact

The motivator environmental impact stems from a desire towards sustainable development. A much cited definition of this is the Brundtland definition (1987) which states that sustainable development “meets the needs of the present without compromising the ability of future generations to meet their own needs”. It is measured through impact assessments such as global warming potential (GWP; Cherrington et al., 2012). It is mainly an intrinsic motivation, but it is also used for promotion or brand positioning purposes.

4.2.2 Legislation

In the EU, the Landfilling Directive (1999/31/EC) is used to provide countries with the basic operational and technical requirements on waste and landfills to prevent or reduce as far as possible any negative effects on the environment (Council Directive, 1999).

Additionally, there is the landfill tax and the landfill ban. The landfill tax is a fee per ton of waste that needs to be paid when placing waste in landfill. Tax rates vary from 3€/t (LT) to more than 100€/t (BE), and for some countries the fee is dependent on the type of waste. The landfill ban poses additional measures compared to the requirements of Directive 1999/31/EC on the landfill of waste. In 2017, 24 EU Member States[1] have a landfill tax, and 19 EU Member States[1] adopted a landfill ban for certain waste streams (CEWEP, 2017).

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[1] As well as Norway and Switzerland

There are also other waste directives in place targeting specific waste streams, such as the Packaging Waste Directive (94/62/EC), Directives on end-of-life on vehicles (2000/53/EC), on (waste) batteries and accumulators (2006/66/EC), and on waste electrical and electronic equipment (2012/19/EU; Davies, Lee, & Braasch, 2018).

In 2008, the European Union (EU) climate change and energy package was adopted to reduce greenhouse gas emissions and reduce the dependence on energy sources located outside the EU. This package includes the EU Emissions Trading Scheme (EU ETS), the Greenhouse gas (GHG) emissions target and the Renewable energy sources (RES) 2020 target (Cherrington et al., 2012). Requiring more renewable energy sources would promote the use of composite materials, as this will increase the building of windmills which use composite material for construction. However, combined with the desire for a reduction of waste, it is necessary to also develop the legislation for sufficient recycling. Cherrington et al. (2012) expect that policy legislation for producer responsibility is highly likely to appear in the EU first, based on the fact that the EU has driven 95% of environmental legislation over the past 10 years. Seeing the developments around waste treatment and landfill bans, they state it can be assumed that future legislation will evolve along similar lines of life-cycle thinking. This extended producer responsibility would include recycling and recovery targets for manufacturers.

4.2.3 Economic opportunities

Economic opportunities are big drivers for companies as the economic bottom line determines the survival of the company. There are multiple possibilities, as listed below:

Funding and investments

There is a wide arrange of funding and investments for research and business on the topic of composite recycling on different scales. From the European Union, there is the Horizon 2020 project. This is a shared strategic framework aimed at achieving smart, sustainable and inclusive growth, funded by the EU as to secure Europe's global competitiveness (European Commission, 2013). Examples of funded projects are FiberEUse, a “large scale demonstration of new circular economy value-chains based on the reusing of end-of-life fibre reinforced composites” (FiberEUse, 2017), and ECOBULK, a large-scale collaboration of numerous partners which looks into rethinking and demonstrating circular applications, which involves the circular use of composite materials as well (ECOBULK, n.d.).

More locally, there are governments and institutes that provide funding. The UK Government through TSB and WRAP is investing in many projects that are directly involved in mixed waste recycling procedures. (PIM & RECCOMP Projects, 2009 & 2008) respectively (Tarverdi, 2009).

There are also research hubs that give out grants for conducting composite recycling research, such as the EPSRC Future Composites Manufacturing Research Hub that in 2018 funded two projects targeting the recycling of composite materials (Materials Today, 2018).

Paying/planning in advance

It is also possible to already plan the decommissioning costs in advance, and pay them beforehand to fund the end-of-life treatment. The purchase of a car for example includes a recycling tax. In Europe, these range from no (Germany) or very low fees (3-4 EUR/vehicle, Finland, Austria) up to 45 EUR/vehicle (Netherlands) and even 66 EUR/vehicle (Slovakia; Monier et al., 2014). This fee is redirected to recycling companies and organisations to fund their research and operation. An example is ARN Car recycling in Tiel, which is a non-profit organisation that is able to operate and invest in improving their processes due to these recycling taxes. Together with the car dismantling plant, ARN achieved a car recycling percentage of more than 96% (ARN, n.d.).

A similar structure is used for wind turbines. Currently, before the installation of any wind turbine project, a decommissioning programme is designed. This programme sets out potential costs and timing for disassembling and disposing of the turbine and any associated infrastructure after end use. Evaluating these future decommissioning costs is difficult due to several factors, such as the unpredictability of the salvage value of the material, the recycling costs, and the disposal costs (Cherrington et al., 2012). Therefore, a single security amount can also be used. In a research on paying security bonds for decommissioning by Aldén et al., a sum of around 30,000 Euro was used in a large number of the permits.

It varies per project when the decommissioning security costs are paid. In Sweden alone, fourteen different payment structures were recorded between 2010 and 2012. The most popular decision was to pay all the costs up front (33 out of 119 cases), but there was also a wide variation of paying in instalments such as paying part of the sum every five years (Aldén et al., 2014)[2]. Paying these decommissioning costs encourages to reuse and recycle parts and materials, as this lowers the decommissioning costs. For example, recycling the blades would be 6% of the total cost for disposal of a wind turbine and 0.14% the cost of a new wind turbine (Cherrington et al., 2012).

Deposits/lease model

Part of the difficulties of composite recycling lies in the reverse logistics (Ortegon, Nies, & Sutherland, 2013), whether this is to the original fibre producer or product manufacturer, or a recycling company. To encourage users to return the products and materials, economic benefits could be deployed. The most familiar example of this is the deposit fee, for example when buying plastic beverage bottles, glass (beer) bottles or aluminium cans. When purchasing these items a fee is paid, which is returned after handing the bottles back in again at the designated return points (European Parliament et al., 2011).

Another structure for product retrieval is the rent or lease model, where the consumer does not get full ownership of the product during use. There is often no purchase fee when acquiring the product, but instead a regular fee is paid for using the product (Tukker, 2004). At the end of life or an agreed period, the user returns the product back to the company, as the company is still the rightful owner. David Symons (consultancy WSP) states that as resources become more scarce, keeping ownership of the resources and leasing them out keeps costs down, while the rising price of raw materials and production makes renting even more attractive (Anderson, 2013). Deploying this strategy not only for products but also for raw materials could be an interesting development for funding the reuse and recycling of composite materials, and retaining the maximum residual value for as long as possible.

Retrieved material value

When looking at the retrievable monetary value in composites, it depends on the materials used. Carbon fibres are targeted for reuse due to their economic value which can amount up to €97 per kg (Yang et al., 2012). However, glass fibres and matrix materials do not have much economic incentive, as there is almost no economic value in them compared to the low price of virgin materials. Instead, they are mainly used for energy recovery (Oliveux et al., 2015).

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[2] Translated version of table available (McCarthy, 2015, p. 27)

5. Recycling methods

This section will explore the current state of composite recycling and what recycling methods are currently available. The methods are divided in three main categories: mechanical recycling, thermal recycling, and chemical recycling. Each method will be elaborated on in terms of process and output characteristics, advantages and disadvantages, and the waste management hierarchy and technology readiness level. This will allow for better understanding of the methods, and it makes it easier to compare methods in chapter 6.

5.1 Mechanical recycling

Mechanical processing of composite scrap is a common step for all recycling techniques due to the size of some industrial parts in relation to the size of reactors. With mechanical grinding and fragmentation however, the chunks are crushed or grinded further down into smaller pieces to produce the recyclate (Oliveux et al., 2015). This section will discuss two mechanical recycling methods: mechanical grinding and high voltage fragmentation.

5.1.1 Mechanical grinding

General

Mechanical grinding means to break down the composite scrap into small pieces to roughly separate fibre and resin fractions. Mechanical grinding has been investigated for both glass and carbon fibre reinforced composites, but the most extensive research has been done on glass fibre (Pickering, 2005).
The energy demand of mechanical grinding lies between 0.1-4.8 MJ/kg, depending on the used machinery and process scale. At maximum machine capacity, the drive motors can perform most efficiently, reaching the lowest energy demand. Powering the motor of the granulator or hammer mill machine makes up most of the energy demand. The pre and post recycling stages, such as shredding and sieving, are not as energy intensive as the actual recycling process (Job et al., 2016).

Process

In the mechanical grinding process, there are three main steps to be distinguished, as shown in Table 2.

Table 2: The mechanical recycling process (Oliveux et al., 2015; Pickering, 2005)

	1. Shredding	2. Grinding/milling	3. Classifying
Process	Reduce waste materials into pieces	Grind pieces into fragments	Separating the fragments into resin-rich powders and fibrous fragments
Particle size [a]	50 – 100 mm	10 mm – 50 µm	-
Equipment	Slow speed cutting or crushing mill	High speed cutting or hammer mill	Cyclones and sieves

[a] at the end of the step

First, the material is shredded down to chunks of 50-100mm. This makes it easier to remove metal inserts and, if done at the waste location, the volume reduction makes transport easier.
Secondly, the chunks are grinded further down into fragments of 10 mm – 50 µm using a cutting or hammer mill (see Figure 14; Pickering, 2005). Both have their own benefits: cutting mills give more homogeneous fibre length distribution and longer fibres, whereas hammer mills don't have blades that require sharpening, thus reducing wear and increasing the output. There is no significant difference between the two concerning the resin content of the recyclate (Schinner, Brandt, & Richter, 1996).
Thirdly, the fragments are separated on their contents and size using cyclones and sieves (Pickering, 2005). It is important to note that there is no complete separation of the two materials; the fragments will almost always consist of mixed materials.

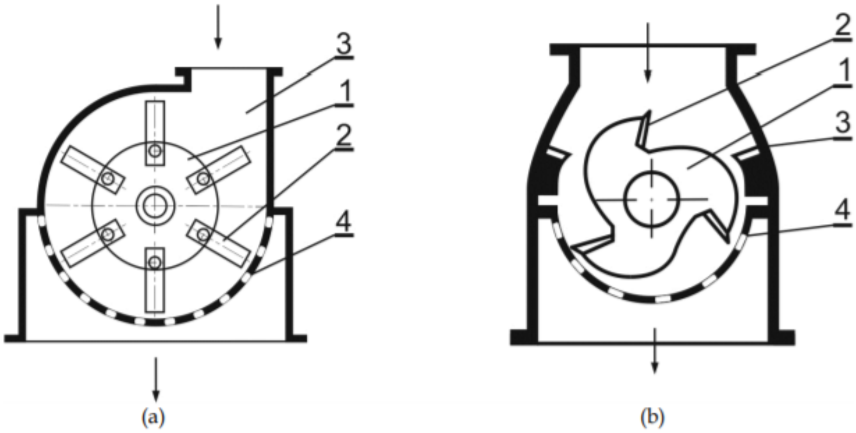

Figure 14: (a) beater mill: 1 – rotor, 2 – beater, 3 – hopper, 4 – screen
(b) cutting mill: 1 – rotor, 2 – cutter, 3 – counter-cutter, 4 – screen (Macko, 2012)

Output

The recyclate of the mechanical grinding are mixtures of resin, fibre and filler. In this, two main fractions can be distinguished: fine and coarse. The finer fractions are powders with a higher resin and filler proportion compared to the original composite, whereas the coarser fractions are more fibrous with a higher fibre content (Pickering, 2005).

Resin

To give an example of what the composition of a finer resin-rich powder might be, the PHX-200 filler fraction from Phoenix Fibreglass had a fragment size of 14 µm with 13% glass content and 87% filler and organic content (Pickering, 2005).
Resin-rich powder can be used as a filler, but this is not that commercially viable because of the very low cost of virgin fillers (e.g., calcium carbonate or silica). Also, the incorporation level as a filler is quite limited (<10 wt.%) because of the deterioration in mechanical properties and increasing processing problems at higher content due to the higher viscosity of the compound (Oliveux et al., 2015). However, the recyclate has a lower density than conventional fillers as it contains a significant proportion of low density polymer. This could lead to weight savings of 5% compared to using only calcium carbonate for example. Also, although filler substitute recyclates are more expensive by weight than traditional fillers, the lower density means they are cheaper per unit volume (Pickering, 2005). This could be favourable for industries where weight and cost savings are very important, such as the aerospace industry.

Fibres

The fibrous fragment can come in a wide variety of forms (e.g. powders, fibre-particulate bundles, fibre tows and woven platelets), which all partly consist of resin. This makes it hard to say what the exact recycled fibre properties are, so in most experiments the performance is mainly judged by integrating the material into new resin. This however makes it difficult to compare the recyclate to that of other recycling methods (Bream & Hornsby, 2001a). The length of the fragments varies as well, depending on initial fibre length, composite type (Bream & Hornsby, 2001a), and scrap feeding size. Bream & Hornsby (2001a) report that the structural integrity of the fibres is preserved, with fibres up to 10 mm being retained and 69% of the recyclate particulates greater than 1 mm in diameter. The fibres produced by ERCOM Composite Recycling in Germany range from <0.25 mm to 3-20mm particle size or fibre length (Pickering, 2005).

In general it can be said that the recyclate performance in composites is inferior to virgin fibres (Pickering, 2005). Even with low reincorporation rates, the resulting mechanical properties of the new composite material are significantly impaired due to a poor bonding between the recyclates and the new resin (Oliveux et al., 2015). To improve bonding, additional treatments such as grafting or coupling agents (Bream &

Hornsby, 2001b) or increased mixing times are needed (Oliveux et al., 2015). Finally, it is important that the remainder of the virgin fibres are replaced with longer fibres, as these will compensate for the deleterious effect of the recyclate (Pickering, 2005).

The fibrous recyclate can be used as filler, or as reinforcement for new composites in short-fibre applications such as bulk moulding compound (BMC), sheet moulding compound (SMC) parts, and as inclusions in injection moulded products (Mativenga et al., 2016). Using the recyclate as reinforcement represents a higher value route as the reinforcing value of the fibre is (partially) retained, whereas the filler replaces very low value materials and more energy is needed to grind to fine filler (Job et al., 2016).

(Dis)advantages

There are both environmental and economic advantages for mechanical recycling. As it does not use any chemicals, it does not produce any atmospheric or water pollution. Also, no high-end technical and expensive equipment is needed compared to the other methods, and it allows for processing of larger amounts of waste at higher throughputs (Ribeiro et al., 2016).
A big drawback however is that the recovered recyclate is of inferior quality compared to the virgin product, as previously mentioned. This makes it difficult to find market applications for the recyclate, as it is hardly competitive with virgin raw materials (Ribeiro et al., 2016; Yang et al., 2012). Additionally, a continuous flow of waste is required for setting up and running a mechanical recycling plant and for producing products from the recyclates. This makes mechanical recycling not a viable solution if there is only a minimum amount of waste (Vijay, Rajkumara, & Bhattacharjee, 2016). With the expected composite waste volume from for example end of life windmills (see Figure 12), the required waste volume will not be hard to achieve. However, without a viable application for the recyclate, going through the whole process might not be justified or even sustainable.

WMH Level & TRL

Waste Management Hierarchy level: Recycling (Bax et al., 2015; Devic et al., 2018; Rybicka et al., 2016)
Mechanical grinding recovers materials but at a much lower quality compared to virgin materials. It is not a true method for retrieving the base materials as they are still partly mixed, which makes it not a preferred recycling option.

Technological Readiness Level: 8-9 for GF – Commercial, 6-7 for CF – Pilot/Commercial (Bax et al., 2015; Rybicka et al., 2016)
For the Technological readiness level, it depends on the type of composite being processed. Due to the relatively low values of the recycled products, mechanical recycling is mostly used for glass fibre reinforced polymers. For glass, it has reached industrial scale, with ERCOM in Germany and Phoenix Fibreglass in Canada often cited as example. However, Phoenix Fiberglass ceased operating in 1996, and ERCOM and other similar operations not been able to find sufficient markets for recyclate to operate at commercially viable levels of production (Yang et al., 2012). This stresses the importance of not only technological development, but viable industrial application as well.
For carbon fibre composites, there is no known mechanical grinding process exploited commercially; it is only used for resizing composite parts into chunks before feeding it into another process, such as pyrolysis (Job et al., 2016).

5.1.2 High voltage pulse fragmentation

General

High voltage (pulse) fragmentation (HVF) is an electromechanical process that uses electricity to separate matrices from fibres (Devic et al., 2018). It originates from rock mining applications where it was used for fracturing rocks to easier separate high value minerals and crystals, such as in gold excavation (Mativenga et al., 2016). When applied to composites, the material is fragmented into smaller pieces which liberates the fibres.

The energy efficiency of the process depends on the process characteristics such as the amount of pulses and processing rate. During their experiments, Mativenga et al. (2016) found an average process efficiency of 18.1% for processing rates between 0.15-0.04 kg/hour. The specific energy demand found for 500, 1000, 1500 and 2000 electrical pulses was 17.1 MJ/kg, 35.6 MJ/kg, 60.0 MJ/kg and 89.1 MJ/kg, respectively. The lowest amount of pulses gives the best energy demand and efficiency, but this gives lower quality fibres. When compared to mechanical grinding (6.7 MJ/kg at 1.2 kg/h), the process is more energy intensive. For both processes it is important that they operate at maximum capacity to make use of the basic energy demand of the machine (Mativenga et al., 2016).

Process

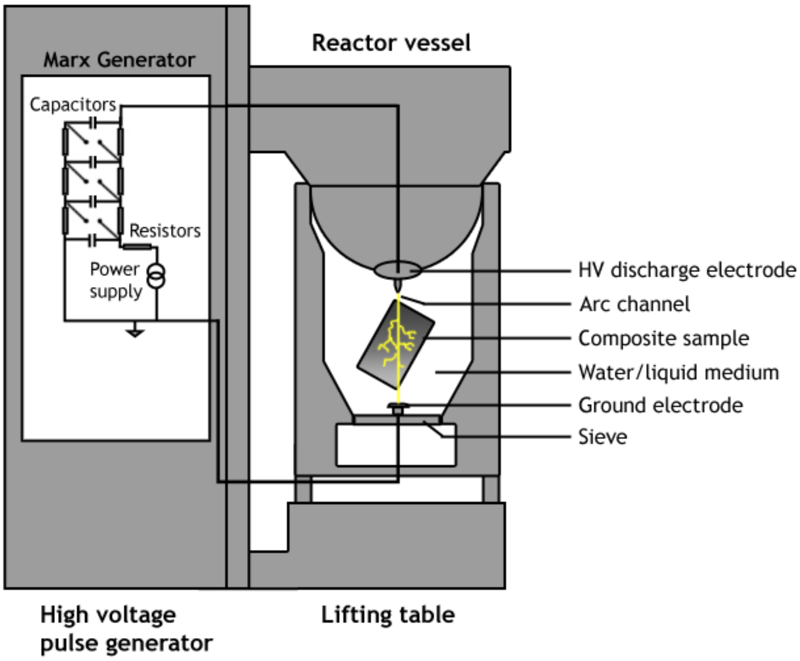

Figure 15: High voltage pulse fragmentation of composite material
Image constructed using Duan et al., (2015), Roux et al. (2014) and Van Der Wielen et al. (2013)

Figure 15 shows the set-up for HVF, which uses repetitive pulse electrical discharges within a dielectric liquid environment, normally water. During HVF, the sample material is placed in a gap between the electrodes. Pulses are then discharged in a very short period between two electrodes, using high voltages (100-200 kV) at a pulse rise time of less than 500 ns. This creates a spark channel that travels through the material along internal boundaries and weak regions such as pre-existing cracks. The spark channel in turn generates an intense shockwave with high pressure (around 10^9-10^{10} Pa) and temperature (> 10^4 K), and induces internal mechanical stress higher than the tensile strength of solid materials (Bluhm et al., 2000). This strips the matrix from the fibre, and propagates cracks (Bax et al., 2015), leading to the disintegration of the material.

Output

The HVF process produces fibres extracted in their original shape as well as matrix pieces in the solution (Bax et al., 2015).

Resin

In the experiment by Mativenga et al. (2016), the state of the removed matrix was not discussed, nor the possibilities for further processing or recycling. Only the residual resin on the fibres was investigated. The amount of residual resin for HFV varied between 32% and 37%, compared to 49-59% for mechanical grinding. A greater number of pulses leads to less residual resin. To remove the residual resin from the fibres, Mativenga et al. used temperature treatments of 2h at 400 °C.

Fibres

The number of pulses has an effect on the length, length distribution, and cleanliness of the fibres. In their experiment, Mativenga et al. (2016) found that a high number of pulses (2000) led to high amount of short fibres with narrower distribution compared to distribution using lower number of pulses. Additionally, the glass fibre recyclates that were recovered using a low number of pulses (500 and 1000) still contained poorly separated portions of resin, whereas the fibres processed with 2000 pulses were cleaner and separated. When comparing the HVF recyclate to that of mechanical recycling, there can be seen that fibres from HVF have a wider fibre length distribution, as shown in Figure 16. For both methods, the mean length was 2 mm and most recovered fibres were less than 5 mm long, but for HVF the maximum retrieved fibre length spread to up to 9 mm, compared to 5 mm for mechanical recycling. This, together with the higher cleanliness of the fibres, suggests that that HVF recyclates would have a higher degree of reinforcement in new composite materials compared to mechanical grinding recyclate.

HVF fibre recyclate is suitable for the same applications as mentioned for mechanical grinding: use in short fibre applications such as bulk moulding compound (BMC), sheet moulding compound (SMC) parts and as inclusions in injection moulded products (Mativenga et al., 2016).

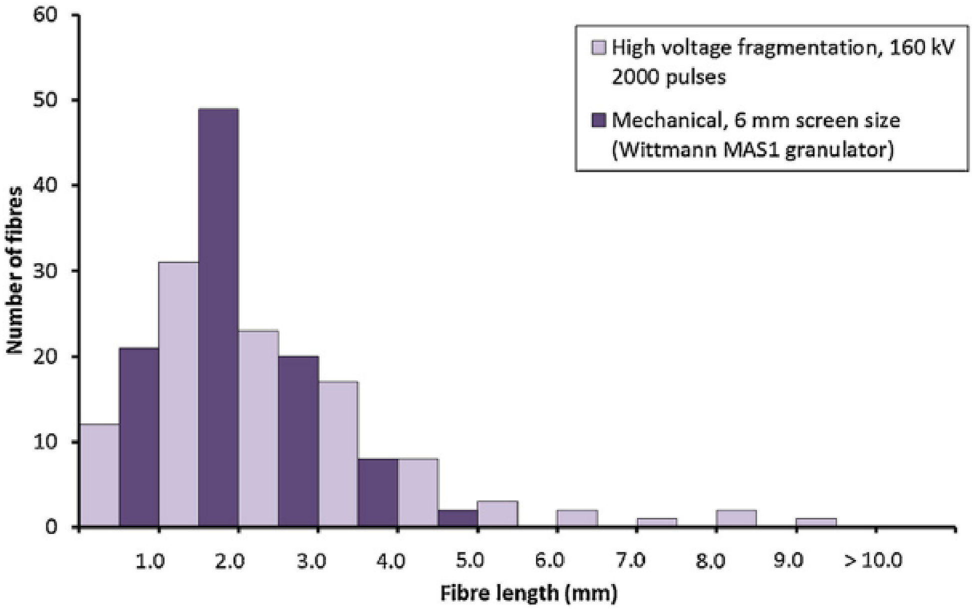

Figure 16: Fibre length comparison between fibres recovered from mechanical and HVF methods

(Dis)advantages

The advantage of HVF is that it provides quite good separation of matrix and fibres at relatively low cost, both for initial investment and operation (Bax et al., 2015). Compared to mechanical grinding, it produces fibres of higher quality. The fibres are cleaner fibres and longer, with a larger fibre length distribution and a higher percentage of fibres at mean fibre length. Also, the fibres have lower residual resin content. This means the fibres are less downcycled than with mechanical grinding (Devic et al., 2018; Mativenga et al., 2016).

The disadvantage of HVF is that the process is quite energy intensive. For Mativenga et al. (2016), the specific energy for HVF was at least 2.6 times higher than mechanical grinding. For both processes, the energy demand should reduce when operating at higher recycling rates, due to more effective operation of the machines. Future research is needed to find the optimum mass loading for most efficient HVF (Mativenga et al., 2016). Another disadvantage is that the fibres are only of moderate fibre quality (around 20% reduction of mechanical properties) and that residues need to be rinsed multiple times during the process (Bax et al., 2015). Still, some improvements have been made regarding the length and quality of the recovered fibres, which seems to indicate that more can be achieved (Devic et al., 2018).

WMH Level & TRL

Waste Management Hierarchy level: Recycling (Bax et al., 2015; Devic et al., 2018)
High voltage pulse fragmentation separates the fibres and matrix, of which the fibre can be reused in short disaligned fibre applications. However, utilization of the matrix is currently not investigated, which could be an improvement to make the method more sustainable and circular.

Technological Readiness Level: 5-6 – Labscale (Bax et al., 2015)
In 2014, Selfrag AG (Kerzers, Switzerland) developed a lab scale operating fragmentation vessel for recycling of composites in collaboration with the Institute of Polymer Engineering (IKT; Windisch, Switzerland). This was done within the SELFRAG CFRP EC funded Clean Sky project (Bax et al., 2015).

5.2 Thermal recycling

In Table 3, an overview is created of the thermal methods that are currently deployed for processing composite material waste (European Parliament and Council, 2000; Job et al., 2016; Pickering, 2005; Yang et al., 2012). In the next section, each of these methods will be elaborated on. In practice, methods are often combined to reach better end results. For example, most pyrolysis processes are combined with gasification or combustion to achieve cleaner fibres (Chen et al., 2015).

Figure 17 further distinguishes the main differences between the thermal processes based on the process characteristics. The most determinate factor is the amount of oxygen available: without oxygen pyrolysis takes place, with a limited amount of oxygen[3] it becomes gasification, and with an excess of oxygen it becomes combustion or incineration (EnerSol Technologies Inc., 2008; Williams, 2005). Important to note is that although they are two different processes, the terms incineration and combustion are sometimes used interchangeably. In this paper, incineration refers to completely burning the waste at a high temperature yielding only ashes, which makes that it is only considered recovery and not recycling. Combustion on the other hand is more controlled burning process at a lower temperature that aims to recover fibres and fillers from the composite waste.

The process heat from exothermal recycling methods is often recovered through a waste-heat recovery system as a source of energy. The calorific value of a composite generally depends only on the calorific value and proportion of polymer, as most fibres and fillers are incombustible. Most composite resins considered have a calorific value of approximately 30,000 kJ/kg (Pickering, 2005), which is high compared to the general

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[3] Less than the amount of oxygen needed to oxidize all combustible elements

calorific value of municipal waste (ranges from 7,500 to 11,000 kJ/kg; Johnke, 2000). Some mineral fillers decompose and absorb energy during combustion, especially fire retardants, but the amount of energy absorbed is small (ca. 3-6%) compared to the calorific value of the resin (Pickering, 2005).

Table 3: An overview of the thermal processing methods used for composite materials (European Parliament and Council, 2000; Job et al., 2016; Pickering, 2005; Yang et al., 2012)

	Definition	Energy	Output	Types [f]
Incineration	Burning it completely Temperature: > 850 °C [a] or > 1100 °C [b]	Exotherm Retrieved energy is roughly equal to the calorific value of the resins (approx. 30 MJ/kg for most resins [e])	Energy recovery and ash	- Grate firing - Rotary kiln incinerator
Combustion	Controlled burning Temperature: 450-600 °C [c]		Fibre + filler recycling and energy recovery	- Fluidised bed combustion
Co-processing	Burning and incorporating the residue into the cement Temperature: 1050-2000 °C		Energy recovery and ashes + minerals that can be used as cement clinker feedstock	- Co-processing in cement kiln
Pyrolysis	Thermal decomposition of polymers (in inert atmosphere) Temperature: 300-800 °C [d]	Endotherm Energy costs are 23-30 MJ/kg (5-10 MJ/kg for microwave pyrolysis)	Solids (50-75%): fibres, fillers and carbon char Liquids (10-50%): mix of organic compounds Gas (5-15%): mix of CO_2, CO and hydro-carbons	- Chain conveyor - Rotary kiln - Microwave - Superheated steam - Catalytic - Gasification

[a] general waste (European Parliament and Council, 2000)

[b] in case of > 1% of halogenated organic substances (European Parliament and Council, 2000)

[c] depending on resin type and atmosphere; typically, 450 °C for polyester and 550 °C for epoxy (Yang et al., 2012)

[d] A higher temperature of 1000 °C can be applied but resulting fibre products will be seriously more degraded (Yang et al., 2012)

[e] polyester, vinyl ester, phenolic, and epoxy resin have a calorific value of approximately 30,000 kJ/kg, urea formaldehyde has a calorific value of 15,700 kJ/kg (Pickering, 2005).

[f] The most common types that will be discussed in this report

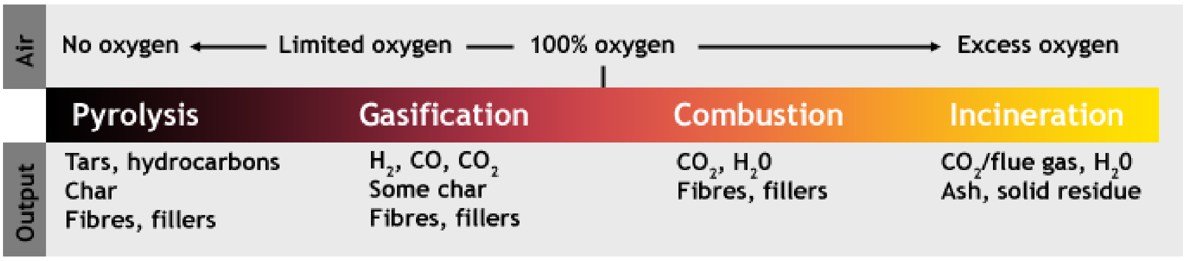

Figure 17: The difference between pyrolysis, gasification, and combustion.
Image constructed using EnerSol Technologies Inc. (2008) and Williams (2005)

5.2.1 Incineration

General

Mixing scrap composites at 10% loading with municipal solid waste has been shown to be a practical way to dispose of scrap if landfill is prohibited (Pickering, 2005). At almost all municipal waste incineration plants, the heat produced during incineration is used for steam generation (Johnke, 2000). In combined heat and power (CHP) plants, the heat from incineration is used to create electricity, and to feed a district heating system (Larsen, 2009). When processing composites, information on contents and the amount of materials in the composite materials are very important for the incineration plant as there are strict discharge regulations. It is also important to consider the fibre content of the waste. If fibres are not completely broken down, there is a risk of release of fibres, which can lead to all sorts of health and safety issues (Hedlund-Åström, 2005).

Process

There are various types of incinerators, including grate incinerator and rotary kiln (which are discussed below). The choice of incinerator type depends on the type, volume, and hazard of the waste to be destroyed (Holder et al., 2013).

Moving grate incineration

Grate incinerators are widely used for incineration of municipal waste, as they can be designed to handle large volumes of waste (Environment Agency, 2001). In Europe, approximately 90% of installations treating mixed municipal solid waste use grates. Other wastes commonly treated in grate incinerators, often as addition to solid waste, include commercial and industrial non-hazardous wastes, sewage sludges and certain clinical wastes (European Commission, 2006).

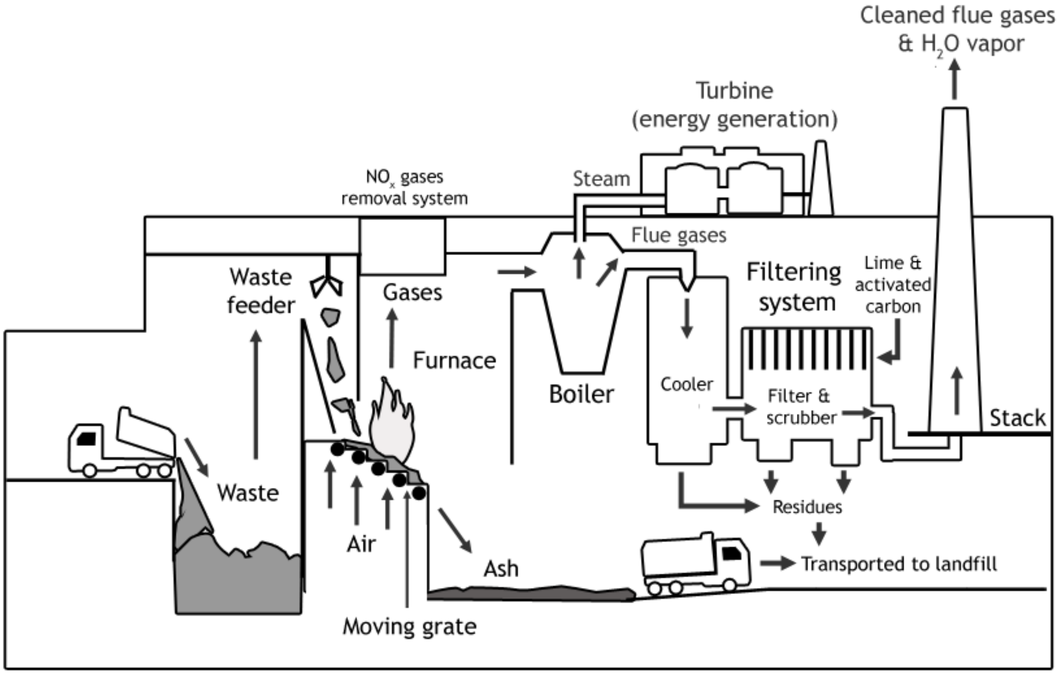

Figure 18: Grate firing incineration process for municipal waste
Image constructed using Holder et al., 2013, p. 1656; Indaver, n.d.-a; Olsen, 2006; Tokyo Environmental Public Service Corporation, 2012

In the grate firing process (seen in Figure 18), the waste is moved onto a grate which transports it into the combustion zone whilst providing a good distribution of air (European Commission, 2006). Newer grate systems are designed to agitate the waste to break it into smaller pieces during combustion. This creates a larger surface area of waste to air and high temperatures, assisting complete combustion (National Research Council (US) Committee on Health Effects of Waste Incineration, 2000). The residence time of the wastes on the grates is normally not more than 60 minutes (European Commission, 2006). The temperature of the gases the combustion chamber is required to be 850 °C for general and 1100 °C for hazardous waste, which is maintained using an auxiliary burner (European Parliament and Council, 2000).

After the combustion chamber, the solid discharge is collected at the bottom. The flue gases from the process go to a secondary combustion chamber to ensure complete oxidation of the volatile organic compounds (Crowder & Richards, 2003; European Commission, 2006). Then, the hot gases are used to power a steam boiler which produces electricity that is supplied to the grid. At the end of the boiler, the flue gases are cleaned using cooling, filtering and scrubbing methods, before being released into the atmosphere via the stack (Indaver, n.d.-a; Tokyo Environmental Public Service Corporation, 2012).

Rotary kiln incineration

In addition to grate firing, there are also rotary kilns systems (see Figure 19). Rotary kilns are very robust and almost any waste can be incinerated, regardless of type and composition. They are in particular very widely applied for the incineration of hazardous wastes (European Commission, 2006), but they can be used for industrial waste as well (Indaver, n.d.-b). Using a rotary kiln could be beneficial for use with composite material, as these achieve good burnout, provided waste residence time in the furnace is adequate (Environment Agency, 2001). Also, using a partial rotation (reciprocating) kiln can be used to reduce the generation of fine particles and dust (Environment Agency, 2001). To further prevent leakage of gases, unburnt waste and dust emissions, a negative pressure should be maintained in the kiln (Environment Agency, 2001; National Research Council (US) Committee on Health Effects of Waste Incineration, 2000).

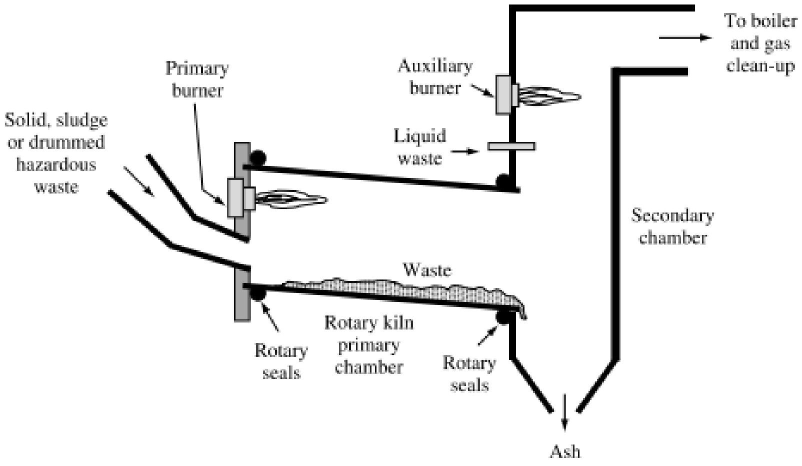

Figure 19: Rotary Kiln incineration process. Image retrieved from "Waste Treatment and Disposal" (Williams, 2005)

The general process of the rotary kiln is similar to that of the moving grate system. Additionally, rotary kilns use a rotational or oscillating motion of between two revolutions per minute to six revolutions per hour, depending on the type of waste and type of rotary kiln (European Commission, 2006; Williams, 2005). This causes good waste agitation which, together with the radiant heat from the kiln walls, creates good conditions for complete incineration (Crowder & Richards, 2003). The temperature in the rotary kiln is generally above 850 °C for conventional and 900 - 1200 °C for hazardous waste incineration. Normally, a residence time of between 30 to 90 minutes is sufficient to achieve good waste burnout (European Commission, 2006). After the kiln, the gases move to a secondary chamber, where temperatures are typically up to 1400 °C with residence times of between 1 and 3s, before moving to the boiler and gas clean-up (Williams, 2005).

Output

After burning, there is a solid discharge consisting of ash and residue (Crowder & Richards, 2003). Depending on the process and discharge consistency and contents, it is either collected dry and moved to landfill (Environment Agency, 2001; Larsen, 2009), or melted into slag (Tokyo Environmental Public Service Corporation, 2012). Melting into slag has as benefit that it is less susceptible to leaching of the dissolved materials (Williams, 2005), which is particularly suitable for hazardous waste incineration (Environment Agency, 2001). Sometimes, the solid discharge is sifted to recover various fractions for useful applications (Indaver, n.d.-a). It is common for some fine material (riddlings or siftings) to fall through the grate, and they get recovered in the bottom ash. These can then be recycled to the grate for repeated incineration, or it is disposed directly (Williams, 2005).

The incinerator bottom ash can be processed into aggregates or construction applications, but in some cases it is still landfilled (Job et al., 2016). Table 4 shows an overview of the recycling percentages and applications of incineration bottom ash, as performed by European countries in 2004 (Williams, 2005).

Table 4: Recycling percentages and applications of incineration bottom ash for European countries in 2004
Constructed using the data from "Waste Treatment and Disposal" (Williams, 2005)

Country	Percentage	Examples of bottom ash use
Germany	80%	Sub-base paving applications
Netherlands	> 90%	Granular base, in-fill for road bases, embankments, and noise and wind barriers, and also aggregate in asphalt and concrete
Denmark	90%	Development of a granular sub-base for car parking, bicycle paths and paved and un-paved roads, etc.
France	70%	
UK	> 50%	
Other countries (e.g. Austria, Switzerland, Portugal, Italy and Norway)	<10%	

(Dis)advantages

The advantage of incineration of composite materials as opposed to landfill, is that some of the residual material value gets recovered as energy which can be used to generate electricity. However, when compared to other recycling methods, the drawbacks far outweigh this advantage. The main drawback of incineration is that over 60% of the scrap is left behind as ash after incineration. For example, a typical GRP product is 40% glass, 30% inorganic filler and 30% resin. The glass and the filler do not burn, leaving 70% of the composite as a residue after incineration (Halliwell, 2006; Larsen, 2009). Due to the presence of inorganic elements in composites, this ash may be a pollutant, and is, depending on the type and post-treatment options, either dumped at a landfill or recycled as a substitute construction material. The inorganic loads also lead to the emission of hazardous flue gasses (Larsen, 2009). Also, as mentioned before, incinerating composite material with energy recovery can result in energy losses of around -400 kJ/kg (Halliwell, 2006).

Another problem of the process is that the fibres don't simply burn out, but shatter into fragments. If these fragments are not fully burnt, they are carried out of hot zones via thermal convection which can cause health, safety and performance issues. The fragments and particle dust can pass into the airways and lungs of workers, and cause health problems (Hedlund-Åström, 2005; Limburg & Quicker, 2016). The particles can also pass into the gas treatment installation, where they can cause problems at the filter, leading to hazardous emission (Larsen, 2009). Additionally, carbon fibre fragments retain their electrical conduction capabilities, which can lead to electrical failures in the gas treatment installation, thus disabling the whole incinerator (Hedlund-Åström, 2005; Limburg & Quicker, 2016). To prevent these issues, it is important to obtain complete fibre disintegration. Therefore, the material must be thoroughly shredded in combination with a high temperature and considerate oxygen supply (Hedlund-Åström, 2005). To address conductivity-induced problems, electrostatic precipitators are common, as well as measurement and control technology (Limburg & Quicker, 2016).

WMH Level & TRL

Waste Management Hierarchy level: Recovery (Bax et al., 2015; Devic et al., 2018; Rybicka et al., 2016)
For incineration, combined heat and power incineration is seen as recovery. The inorganic residues after combustion could be potentially used in the cement industry, raising the recovery value (Vijay et al., 2016; also see section 5.2.3 Co-processing in a cement kiln). Plain waste incineration however is considered as disposal, as no value is recovered.

Technological Readiness Level: 9 – Industrial scale (Bax et al., 2015; Rybicka et al., 2016)
For the Technological readiness level, incineration of waste is performed on an industrial scale. In 2016, on average 22% of all municipal waste was incinerated with energy recovery in Europe with Finland, Norway, Estonia and Denmark as largest waste incinerators (between 55-51%). Interesting to note is that incineration without energy recovery is generally rare both inside and outside Europe (around 2-3% of total municipal waste), with the exception of Germany and Italy that incinerated 7-8% of their waste without energy recovery (OECD, 2016).
Composite materials can be added to the waste stream of energy from waste incineration plants (Job et al., 2016), but it is difficult to find data on the specific amounts of composite material that enter the waste stream. However, due to the health drawbacks and the fact that the fibres do not always fully deteriorate, incineration plants are stopping to accept composite materials as mono fraction. To ensure that composite material doesn't enter waste incinerators via municipal waste, a separate collection system should be established, as done with electronic scrap for example (Limburg & Quicker, 2016). Still, the drawbacks of the process and the fact that incineration is no recycling process mean that incineration of composites is not a feasible long-term solution (Halliwell, 2006).

5.2.2 Combustion

With combustion, there is a more controlled burning than with incineration. As a result, the fibres are less damaged and can therefore be retrieved. A widely researched combustion method with promising perspectives is the fluidised bed combustion (Yang et al., 2012), as discussed below.

General

General fluidised bed technology has a wide range of applications in combustion and process engineering. Research at the University of Nottingham (UK), has been developing the process for recycling composite materials. It removes the thermoset polymer from the fibres by a thermal oxidative process, recovering glass and carbon fibre reinforcements that are suitable for recycling back into composite material (Pickering, 2005, 2009). The organic resins are used as energy source, and the combustion heat is recovered through waste-heat recovery system (Yang et al., 2012). This method has been investigated for both glass fibre and carbon fibre materials (Hedlund-Åström, 2005), although the University if Nottingham focused mainly on carbon fibre reinforced composites as these are potentially a more valuable source of fibre for re-use (Halliwell, 2006).

Process

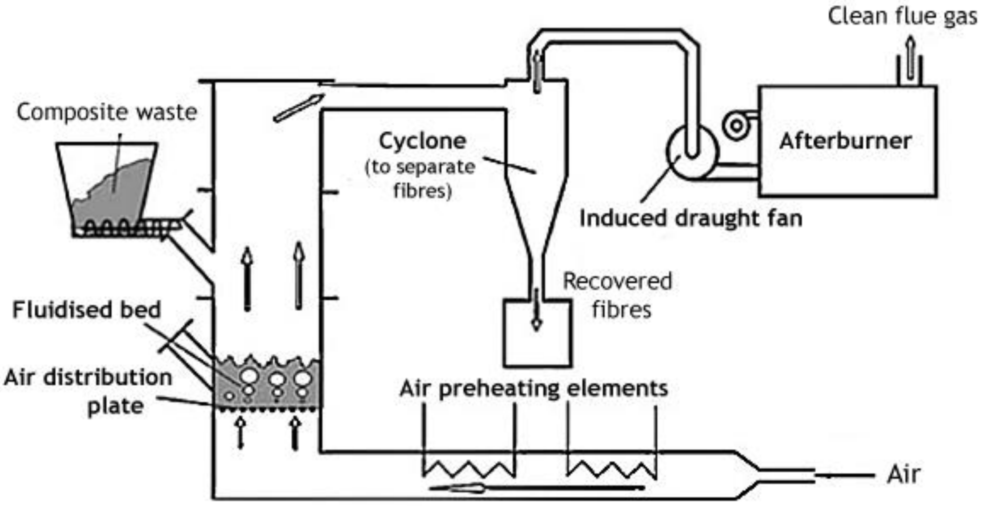

Figure 20: Fluidised bed combustion process. Image taken from (Pickering, 2005, p. 1210)

Figure 20 shows the fluidised bed combustion process. Before the composite material enters the reactor, it is first broken into 25 mm size (Yang et al., 2012). This is very important, especially for carbon fibre composites, to achieve a complete incineration that results in low emissions. Additionally, a high combustion temperature and good oxygen supply is important for carbon fibres as well (Hedlund-Åström, 2005).
The scrap is then fed into the fluidised bed reactor: a chamber of sand which acts like fluid when suspended in an airstream (Halliwell, 2006). The temperature is around 450 °C for polyester resin composites and up to 550 °C for epoxy resin (Yang et al., 2012). The temperature is very important for the resulting fibre quality: too low and the fibres will not be fully cleaned, any higher and the fibres suffer from a reduction in strength (Halliwell, 2006).
In the fluidised bed, the heat dissolves the matrix which releases the fibres and fillers. These are then carried out (elutriated) of the fluidised bed in the gas stream, and can be separated from the gas in a cyclone or other gas–solid separation device. The operation temperature of the fluidised bed does not fully oxidise the polymer, so after separation of the fibres and fillers, the gas stream passes through to a secondary combustion chamber where high temperature combustion achieves full polymer oxidation (Pickering, 2005, 2009, p. 68). Energy can be recovered from the high temperature exhaust gases and, if necessary, any pollutants can be scrubbed from the gas before being emitted to atmosphere (Pickering, 2009, p. 68).

Output

The recovered fibres are clean and have very little surface contamination, which indicates that the fibres have good potential for bonding to a polymer matrix if used in a composite (Pickering, 2005). The fibres have a mean length of 6–10 mm, and are of a fluffy nature. Recovered glass fibres suffer from strength reduction at the higher temperatures, with a 50% tensile strength reduction at 450 °C and a 90% reduction in strength at 650 °C. Carbon fibre has less degradation after the thermal treatment at with only 20% loss in strength at 550 °C. Both fibre types retain the same stiffness as virgin fibre (Pickering, 2005; Yang et al., 2012).

By optimising the fluidised bed with respect to temperature and process, mechanical properties can be improved (Hedlund-Åström, 2005). The discontinuous shorter length and fluffiness of the fibres however, together with the degradation in mechanical properties, will limit the use of the recycled fibres to short-fibre applications such as moulding compounds (Yang et al., 2012) or non-woven veil or tissue products (Pickering, 2005). For achieving more high-value uses, research needs to focus on improving fibre length, developing fibre alignment, high strength and modulus, as well as producing recyclate that fits easily into existing manufacturing processes (Halliwell, 2006; Job et al., 2016).

(Dis)advantages

The main benefit of combustion is that it is very tolerant of mixed and contaminated materials, which means that any resin type or mixture can be processed and that contaminations such as painted surfaces and foam cores are not a problem. Also, metal inserts moulded into a composite do not have to be removed beforehand as they are retained in the bed and could be separated by regrading the sand (Pickering, 2005). This was demonstrated by the RRECOM project, which showed that fibres recovered from processing a painted car boot lid (double-skin glass reinforced polyester, polyurethane foam core and metal inserts) were no different in quality from fibres recovered from processing pure composites (Halliwell, 2006).
When co-combusting composite scrap with other materials, any calcium carbonate filler in the composites will absorb sulphur oxides from the combustion and so reduce the sulphur emissions. This is similar to the addition of powdered limestone, which is commercially used for scrubbing combustion flue gases (Pickering, 2005). Finally, as stated before, the process recovers clean fibres and the energy from burning the organic composite resin can be retrieved as a source of energy.

Disadvantages of combustion are that the retrieved fibres are short and of a fluffy nature, and suffer from degradation in mechanical properties (Pickering, 2005; Yang et al., 2012), which makes it hard to compete with the virgin materials. Other barriers include that no matrix material is recovered, and that there is difficulty in replicating lab results (Bax et al., 2015).

WMH & TRL

<u>Waste Management Hierarchy level: Recycling</u> (Bax et al., 2015; Devic et al., 2018; Rybicka et al., 2016)
Combustion produces clean fibres that are fit for reuse, even though they are of less quality than virgin fibres. The matrix however is only recovered as energy, which means it does not fully recycle all material constituents.

<u>Technological Readiness Level: 4 – Lab scale</u> (Bax et al., 2015; Rybicka et al., 2016)
As there is difficulty in replicating lab results, combustion of composites has yet to move beyond lab scale. For the last two decades, the University of Nottingham has been researching the fluidised bed process, with professor Stephen Pickering as a very important figure in the field (Bax et al., 2015). Pickering states that although lab scale studies suggest that fluidised bed combustion has a potential to process a wide range of thermoset composites scrap (Pickering et al., 2000), further development work is needed to identify ways in which the material can be reprocessed into cost-effective new products to offset the lower fibre quality (Pickering, 2005).

5.2.3 Co-processing in a cement kiln

General

Co-processing is the use of waste as a raw material and/or energy source to replace natural mineral resources and fossil fuels in industrial processes. It is mainly used in energy intensive industries such as cement, lime, steel, glass, and power generation (Vijay et al., 2016). When co-combusting composite waste in a cement kiln, the fibres and fillers in the material act as a feedstock material for the cement (Job et al., 2016). This method is only discussed for glass fibre composites, as the carbon fibres are too valuable to be used for this sort of application.

Process

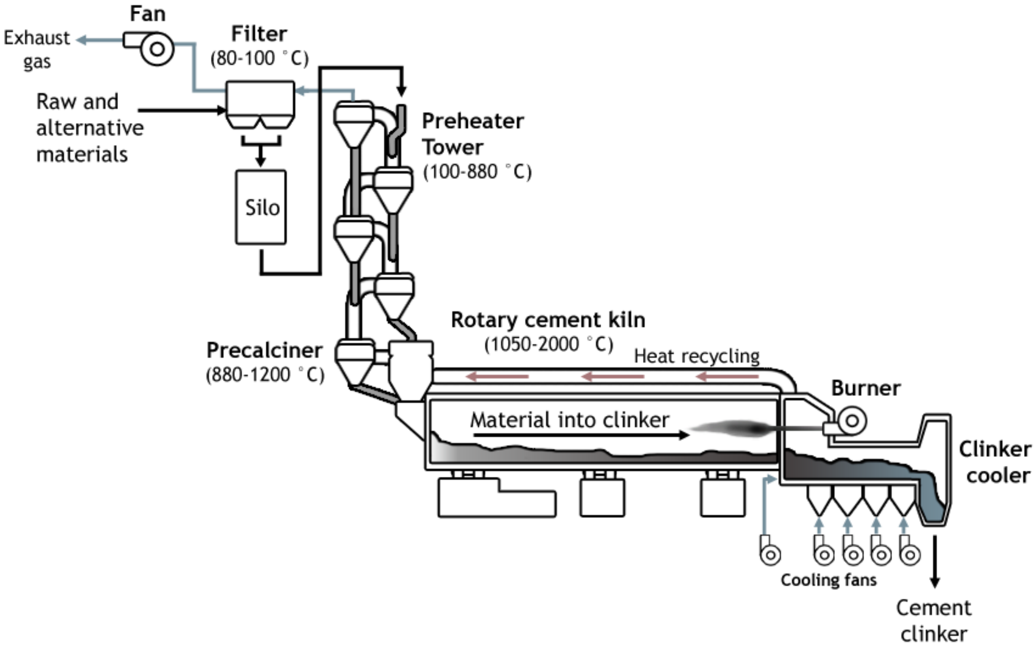

Figure 21: Co-processing of composite waste (alternative materials) in a cement kiln
Image constructed using Vijay et al., 2016; Ziegler et al., 2006

Figure 21 shows the basic cement kiln process system, which consists of the kiln and a suspension preheater. The raw materials (e.g. limestone and shale) are ground finely and blended to produce the raw meal. This raw meal is then fed in at the top of the preheater tower and passes through the series of cyclones in the tower to preheat and decarbonate it. More recently, an additional heater (precalciner) was added to further decarbonate the meal before it enters the kiln. This means a kiln can be smaller for the same output, which reduces the capital costs of a new cement plant (WHD Microanalysis Consultants Ltd., 2018). In the kiln, the materials are turned to clinker, which is then ground to form cement (Job et al., 2016).

When co-processing composite materials in a cement kiln, the parts are size-reduced and mixed with other solid recovered fuel to feed into the kilns (Job et al., 2016). These typically fed to the kiln system in the same way as traditional raw materials, but there are also other access points for lump fuel such as at the transition chamber at the rotary kiln inlet end (Ziegler et al., 2006). Within the cement kiln, the temperature is between 1050-2000 °C. With this temperature, the fibres are converted into ashes and mixes with clinkers, whereas the resin is converted into organic matter which provides energy for the burning process (Vijay et al., 2016).

Output

Co-processing is an effective route to recover some value from the incombustible material, as the glass fibres and fillers used contain minerals that can be incorporated in cement (Pickering, 2005). Any calcium carbonate calcines are converted to calcium oxide, the primary component of Portland cement, and alumina and silica also have cementitious properties in an alkaline environment (Job et al., 2016). The only mineral that has a negative effect on the cement production process is boron, which is found in E glass fibres. Too much boron (>0.2% boron oxide) can increase the cement setting time, although the ultimate strength is not affected. Therefore, glass fibre composite materials can substitute no more than about 10% of the cement kiln fuel input (Pickering, 2005). E-glass from European manufacturers now contains much less boron due to emissions regulations at manufacturing plants, though quantities are still significant in E-glass from China (Job et al., 2016).

(Dis)advantages

The main advantages of co-processing is that it reduces global environmental impact as it has lower carbon emissions, and it decreases the costs of waste management. Also, no ash residues are left that need to be landfilled afterwards as is the case with incineration (Vijay et al., 2016), and it saves on virgin materials.

A problem with co-processing composite materials is that it can generate dust containing glass fibres, which is harmful (see section 5.2.1 Incineration). Therefore, composite waste fed into cement kilns must not generate dust and must contain low concentrations of toxic materials and heavy metals, and no foreign material (such as metal inserts). Also, the waste must be of a designated size. Trials by Nomaguchi et al. in Japan discovered that shredding leads to a lower calorific value of the composite material as some of the resin was lost during the shredding process. This means that additional material must be mixed with the waste to reach the necessary calorific value for use as fuel by the cement industry. Moreover, it is still uncertain whether unburnt filler and fibres incorporated in the cement change the specifications of the product. (Halliwell, 2006). Finally, although the method does produce recyclate with a feasible application, there is no recovery of material that can be used for creating new composites as the recyclate is directly incorporated into the cement clinker or recovered as energy.

WMH & TRL

Waste Management Hierarchy level: Recovery (Bax et al., 2015; Devic et al., 2018)
Co-firing in a cement kiln mainly recovers the energy of the resin, and also has some recycling value for the minerals and fibres. However, as these materials are directly incorporated into the cement, it is not a recycling technology that brings the material back to its original state. This raises questions on the circularity of the process.

Technological Readiness Level: 9 – Industrial scale (Bax et al., 2015)
Recycling through co-processing has been technically and commercially demonstrated. It is increasingly used for managing composite regrind because of its technological potential, environmental benefits and cost effectiveness (Vijay et al., 2016). An example of co-processing on industrial scale is the CompoCycle collaboration: the first company, Fiberline, sends their waste to the second company, Zajon, who grinds and processes it into a recycling mix, which is then used by the third company, Holcim, to make the finished cement (Fiberline Composites, n.d.).

5.2.4 Pyrolysis

General

The most studied thermal process is pyrolysis, the heating of a combustible material with or without oxygen (Oliveux et al., 2015; Pickering, 2005). As mentioned before, pyrolysis is generally characterised by the absence of oxygen, but in practice pyrolysis requires a small amount of oxygen to minimise char formation (Job et al., 2016; Oliveux et al., 2015).
Pyrolysis allows the recovery of fibres and eventual fillers and inserts. The resin breaks down into lower-weight molecules, which produces mainly gases, an oil fraction, and char on the fibres. The gases that evolve can be used as fuel to provide heat for the process (Pickering, 2005).

Process

Pyrolysis processes operate between 450 °C and 700 °C, depending on the type of resin. Polyester resins decompose fully at a temperature of 400-450 °C, whereas epoxides or thermoplastics such as PEEK require higher temperatures of 500-550 °C (Cunliffe, Jones, & Williams, 2003; Job et al., 2016; Oliveux et al., 2015). Research at the School of Engineering in Bilbao, Spain (Torres et al., 2000) reported that pyrolysis products formed in the temperature range of 400-700 °C were remarkably consistent.
Regarding the fibres, a pyrolysis temperature of around 500-550 °C seems to be the limit in order to maintain acceptable strength for carbon fibres (Job et al., 2016). For glass fibres, less than 50% of their

mechanical properties are retained at the minimal temperature of 450 °C (Pickering et al., 2000). Therefore, this type of technique is better suited for recovering carbon fibres (Oliveux et al., 2015). To recover glass fibres in good conditions, a low temperature pyrolysis process was also investigated (Pickering, 2005).

The most conventional types of pyrolysis include classic conveyor pyrolysis and fluidised-bed pyrolysis (Job et al., 2016). Other pyrolysis-based methods are microwave-assisted pyrolysis, superheated steam pyrolysis, catalytic pyrolysis, and (reverse) gasification. For more a more elaborate overview on different pyrolysis types and its applications, there can be referred to Chen et al. (2015), Raja (2013) and Naqvi et al. (2018).

Job et al. (2016) found that for conventional pyrolysis the average energy demand was reported to be within the range of 23-30MJ/kg; no information on processing scale was included however. The energy demand could (partly) be covered using the recovered oil fraction, which would have a gross calorific value sufficient to heat the process. However, this requires additional separation and purification steps, which may escalate the process energy demand even further. Optimisation and upscaling the process will enable higher processing rate and lower specific energy demand (Job et al., 2016).
Microwave pyrolysis is reported as more energy efficient with an estimated energy demand of about 5-10MJ/kg. However, a side note is that classic pyrolysis can use gas as a direct energy source, whereas for microwave pyrolysis electric energy must first be generated. If this energy is generated using non-renewable sources, combined with the energy conversion losses it might not be that much more energy effective after all. To answer this, more research into the embedded energy of the methods is needed.

Classic pyrolysis

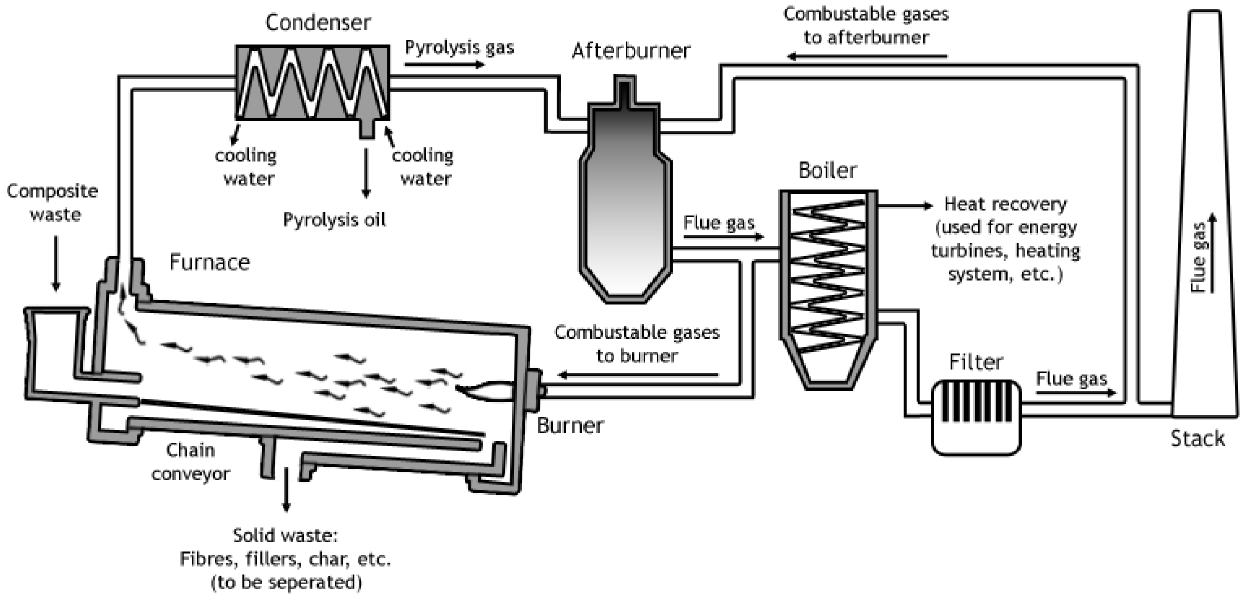

Figure 22: Classic pyrolysis of composite waste (here with chain conveyor)
Image constructed using Dirk Gerlach Engineer, 2009; Li et al., 2004; Pickering, 2005; ReFiber ApS, 2004; Sprenkels, 2018; WRS USA LLC., 2014

Classic pyrolysis transports composite waste through a furnace with controlled temperature and atmosphere. Commercial pyrolysis is typically undertaken in a chain conveyor (Job et al., 2016), but rotary kilns are widely used as well (Chen et al., 2015). In the process, waste is first moved through a furnace where the temperature is around 500-800 °C. The solid waste (fibres, fillers, char) is collected at the end of the furnace for further separation, whereas the flue gases move to the condenser to be cooled at a temperature of 50 °C. The precipitated gases form the pyrolysis oil, which can then be collected. The remaining gases are transported to the afterburner, where they are burnt at 1100-1200 °C. The hot gases from the afterburner are then used for heat recovery in a boiler (operating temperature of 900 °C) or direct combustion in the furnace. After passing through a filter, the remaining clean gases are emitted into the atmosphere (Dirk Gerlach Engineer, 2009; ReFiber ApS, 2004;).

Fluidised bed pyrolysis

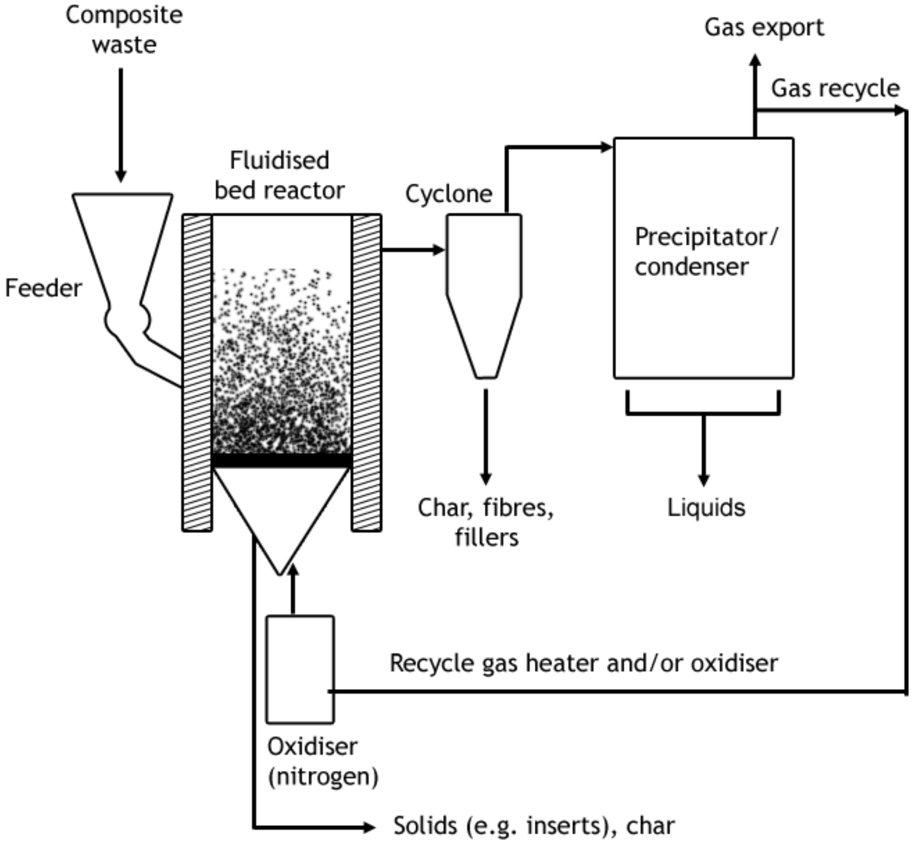

Figure 23: Fluidised bed pyrolysis (bubbling/stationary bed)
Image constructed using (Bridgwater, 2012, p. 5; Morin et al., 2012, p. 235; Oliveux et al., 2015; Pickering, 2005, p. 1210)

In order to achieve more rapid heating of materials, a fluidised-bed pyrolysis process has been developed. This involves passing the size-reduced waste material through a bed of silica sand, fluidised by a stream of hot air. This releases the fibres by attrition and thermal degradation of the resin (Job et al., 2016; Oliveux et al., 2015). Within the fluidized bed, there are many types such as stationary or bubbling beds, circulating beds, and vibratory fluidized beds. There are also several flow regimes that can be used, which include slugging, boiling, channelling, and spouting (Raja, 2013).

In the process, there are three output streams. Firstly, there are inorganic solids such as metal inserts, which sink in the fluidised bed when released and can be removed when regrading the bed.
Secondly, there are the inorganic solids such as the fibres and fillers, which all exist as airborne particles. The high value fibres can be separated from the low value fillers using a sieve separator. This removes the long fibres from the fluidising gas stream, while allowing fillers and short fibres to pass, which are then removed from the gas stream using a cyclone.
Thirdly, there are the polymer matrix and other organic constituents (e.g. mineral oils and facing paints) which are volatilised in the process.

Additional methods

In addition to the classic and fluidized bed methods, there are also other thermal processes that (roughly) belong to the pyrolysis category. Discussed below are microwave pyrolysis, superheated steam pyrolysis, catalytic pyrolysis, and (reverse) gasification.

Microwave pyrolysis heats the material in its core, which makes the thermal transfer very fast, potentially enabling energy savings (Job et al., 2016). In an experiment by Lester et al. (2004), prepreg material was suspended in a bed of quartz sand and powered for 8 s at 3 kW. To prevent the combustion of the fibres during heating, a stream of nitrogen gas was used to create an inert atmosphere. In the experiment, a gas trap was used to reduce the amount of polymer in the exhaust gases, and glass wool insulating was used to prevent solids leaving the microwave cavity through the glassware.

Superheated steam pyrolysis uses the presence of steam, which increases the polymer degradation rate and makes it easier to separate the fibres from the solid pyrolysis products (Pickering, 2005). Also, steam is a readily available material and enables uniform heating (Kim et al., 2017). In their study, Kim et al. (2017) heated carbon fibre composites to 550 °C in a SiC furnace for 60 minutes, first using a liquid flow of water before switching to air. Using air in the second oxidation step reduces the oxidation of carbon fibres and enables intensive decomposition of the resin.

Catalytic pyrolysis process has been developed by Adherent Technologies, Inc. (USA) for carbon fibre composites based on epoxy resins. This process, performed in a continuous pyrolysis reactor, combines low temperatures (typically around 200 °C) with the use of a catalyst at a processing time of 5 minutes. The polymer is completely degraded into low molecular weight hydrocarbons in liquid or gaseous form, and the remaining carbon fibres are substantially free from resin (Allred & Salas, 1994; Pickering, 2005).

Gasification is a process in-between combustion and pyrolysis (see Figure 17). This method has also been developed for carbon fibre composites. In this process, scrap is heated at 600 °C in a controlled flow of oxygen. The polymer is converted to shorter chain hydrocarbons and gases (H_2 and CO), whereas the carbon fibres can be recovered for reuse. There is some char residue on the fibres, but this is generally less than 10%. Alternatively, (an excess amount of) air can be fed into the furnace towards the end of the heating cycle to oxidise any char (Pickering, 2005).

In the reverse gasification process, scrap is heated in the presence of a limited amount of oxygen. The polymer is converted to lower chain hydrocarbon oils and gases and, in the best trials, only 2% of the resin remained on the fibres (Lester et al., 2004).

Output

The pyrolysis process produces a fuel gas, liquid oil products, and a solid residue consisting out of the inorganic fibres and fillers and a char residue. The ratios of these products depend on the composite type and resin content. In general, it can be said that solids account for the highest proportion (50 wt.% till more than 2/3) followed by liquid products (10–50 wt.%) and gas products (5–15 wt.%; Yang et al., 2012). Research by Cunliffe et al. (2003) reported more extreme numbers, with ranges of 30-90.2% for solid products, 8.8-59.4% for liquid products, and 1-12.6% for gas products at temperatures of 500–550 °C.

Resin

The pyrolysis process breaks the resin down into lower-weight molecules. It produces mainly gases and an oil fraction (Job et al., 2016), which contains a mixture of different classes of organic materials (Cunliffe et al., 2003). The pyrolysis gases are mainly CO_2 and CO, with a hydrocarbon gas content of less than 10%. The calorific value is generally low, about 14 MJ/Nm^3 (Torres et al., 2000) or less than 18 MJ/kg. The gases from epoxy and polypropylene however are rich in methane and propene respectively, and therefore have a higher calorific value of between 42-44 MJ/kg (Pickering, 2005).

The pyrolysis oil is reported to have a composition similar to heavy crude oil (Cucuras et al., 1991) or fuel oil, with calorific value of about 37 MJ/kg. It contains a complex mixture of organic compounds, typically containing 66% aromatic compounds and about 25% oxygenated compounds (e.g. ketones, carboxylic acids, alkylbenzenes; Pickering, 2005).

The resin products have been mainly considered as energy source (Oliveux et al., 2015). The pyrolysis oil has a typical calorific value in the region of 30 MJ/kg, giving it a high potential for use as fuel (Cunliffe et al., 2003). Another option is to use the liquid and solid condensable resin products as chemical feedstock. Especially the products from polyester composite are interesting: the condensable products include styrene (26% of liquid product) and phthalic anhydride (96% of solid product), which are potentially valuable products for recycling into new polyester/styrene resins (Cunliffe & Williams, 2003). In practice however, the resin products are typically burnt, in some cases with energy recovery (Job et al., 2016).

Fibres

Properties

The treatment conditions of the pyrolysis process play a great role on the resulting properties, as do the different fibre types. As the minimal process temperature is 450 °C, the especially glass fibres suffer from the high temperatures (Cunliffe & Williams, 2003) and lose between 52 and 64% of their tensile strength (Oliveux et al., 2015). Carbon fibres are less sensitive to temperature, although depending on the selected temperature for the process and post-processing, their tensile strength can be reduced anywhere between 4 and 85% (Oliveux et al., 2015). With skilled control however, carbon fibre mechanical properties can be maintained at 90% or virgin properties for classic pyrolysis (chain conveyor; Job et al., 2016). In industrial processes, this usually involves the blending of fibres retrieved from a diverse feedstock with different types of carbon fibres with varying properties in order to minimise property variation (Oliveux et al., 2015).

The length of the fibres is mainly limited by the pre-processing shredding and post-process chopping. During reclamation, some breakage of fibres occurs, but fibre length is mostly retained (Pimenta & Pinho, 2011). For example, carbon fibres recovered by ELGCF pyrolysis for the Fibre Cycle project reached lengths of over 500 mm (Oliveux et al., 2015). In industrial practice, chopping of the fibres is common to produce more even fibre length grades that can be blended to reach the desired results. ELGCF offers a chopped carbon fibre product (CARBISO™ C) in different lengths: 3 – 10 mm, 10 – 30 mm, 30 – 60 mm, and 60 -90 mm fibres (ELGCF, 2017).

Fluidised-bed pyrolysis fibres seem to be more damaged than with classic pyrolysis. For carbon fibres, the strength is typically reduced by about 25%, though stiffness is retained (Job et al., 2016). For glass fibres, the tensile strength was reduced by 50% at 450 °C and up to 80% at 550 °C (Pickering et al., 2000). In addition to the high temperature, attrition by the fluidised sand might also damage the fibres. (Job et al., 2016). However, there was only a small reduction in the fibre surface oxygen content, which indicates that the fibres have good potential for polymer matrix bonding when reused in a composite (Pickering, 2005)

For microwave pyrolysis, fibre strength retention was encouraging and slightly better than fluidised bed recovery (3.26 GPa against 3.05 GPa) as there is no attrition from sand (Lester et al., 2004). During the experiment by Lester et al., the microwaved fibres had a tensile strength reduction of around 20% and a stiffness reduction of 13%, compared to the tested virgin fibres. Furthermore, the fibres appear to be clean and homogenous. In the experiment by Åkesson et al. (2012), the tensile strength reduced by 25 % for 360 °C and 27 % for 440 °C. The original scrap size was 7-30 mm, and the retrieved fibres varied between 0.1-31 mm in length with roughly 52wt% of the retrieved fibres having a length of less than 15.7 mm.

When using superheated steam, (Kim et al., 2017) reported that decomposing carbon fibre composite waste at 550 °C for 30 min led to a 10-15% decrease in fibre tensile strength and a mild decrease in tensile modulus. Decomposing at 550 °C for 60 minutes or more resulted in similar decreasing behaviour, indicating that when the pyrolysis time is increased, the tensile properties begin to decrease. The decrease tensile strength can be because of two reasons: heat treatment damage, or surface oxidation caused by superheated steam. The surface of the superheated steam pyrolysis fibres was smooth, with little adhered resin. When the pyrolysis reaction times were 60 minutes or longer, epoxy char was practically eliminated. Additionally, the regenerated carbon fibres can naturally reform the surface during the process without any additional surface treatment, which improves the interfacial property (Kim et al., 2017).

For the catalytic process, it was found by Adherent Technologies, Inc. that the strength degradation of the recycled carbon fibres varied between 1 and 17%. When analysing the surface quality, the surface oxygen content varied from a reduction of 18% up to an improvement of 83% compared to virgin fibres. In both cases, the oxygen bonds were similar to those of virgin fibres. Therefore, the recycled carbon fibres would be suitable for bonding to a polymer matrix in a composite (Pickering, 2005).

For gasification, the loss of fibre properties is consistent with the effects of heat treatment of fibres. In the process of ReFiber, glass fibres have lost more than 50 % of their initial strength after heating to 500 °C, whereas the stiffness remains practically unaffected (ReFiber ApS, 2004). López et al. (2013) combined pyrolysis with gasification for carbon fibres, which gave a 28% reduction of tensile strength and about 10% reduction of stiffness with respect to virgin fibres after 30 min of gasification at 500 °C.

Surface quality

In addition to the mechanical strength, there is the surface quality to consider as well: any thermal or chemical process strips the sizing off the fibres (Job et al., 2016). Sizing is a surface treatment on the fibre (Pickering, 2005), used for example to protect the fibre surface, to bind fibres together for easier processing, to reduce static electricity, and to improve chemical bonding to the matrix. For the latter reason, they may also be referred to as coupling agents (Park & Seo, 2011). In the case of glass fibre, stripping the sizing results in dramatic loss of strength and processability, which makes the process more suitable for carbon fibres (Job et al., 2016). Also, there is a char-like substance from the resin degradation covering the fibres after pyrolysis (Oliveux et al., 2015). This leads to poor fibre-matrix adhesion when using the recycled fibres in new composite materials (Job et al., 2016).

To remove the substance and yield clean fibres, secondary combustion can be used. This significantly reduces the fibre strength however (Meyer, Schulte, & Grove-Nielsen, 2009); oxidising glass fibres at 450 °C for example leads to a 50% degradation in mechanical strength (Pickering, 2005). Different post-treatments have been tested in order to improve the fibre surface quality, but more research is needed on the relation between the surface quality of the recycled fibres and the mechanical behaviour of materials reusing these fibres (Job et al., 2016). Another option is to change the process conditions. In oxidant conditions, resins are more easily degraded than in inert conditions, leaving less residue behind. At temperatures of 500–600 °C, it is even possible to completely remove resin residues (Meyer et al., 2009). However, this has a negative effect on the fibre properties. This could be due to the fact that the fibres do not have a protective layer of resin anymore, which leads to more severe oxidation and thus increased deterioration of the fibres (Pickering, 2005). Therefore, a compromise is needed between the mechanical properties of the fibres and the amount of resin residue (Oliveux et al., 2015).

Application

The solid pyrolysis residue can be processed as a whole, such as grinding it down to a powder and using it as a filler. Cucuras et al. (1991) found in their study for SMC Automotive Alliance that up to 30% of the ground solid residue could be incorporated into a sheet moulded compound without affecting the processing or the mechanical properties of the moulded parts. As stated earlier however, conventional fillers are cheap which might make the economic viability of this application a challenge.

The solid residue can also be separated to yield recycled fibres and use them in applications that enable the use of chopped fibres with random orientation.

For carbon fibres, potential use could be in complex shape parts and panel shapes, as the discontinuous and shorter length fibres seem to produce high quality 3D complex and lightweight structure (Naqvi et al., 2018). Another option is to integrate them into sheet or bulk moulding compounds[4], or in non-woven textiles using methods such as wet papermaking, compressed air dispersion, needlepunching, and carding. Additionally, the recycled carbon fibre can be blended with thermoplastic fibres to create a semi-finished material for thermoplastic composite processing (Job et al., 2016).

Reusing glass fibres is more difficult due to the reduced fibre quality. Åkesson et al. (2012) for example used recycled glass fibres from the pyrolysis process to produce a non-woven mat. These mats had poorer mechanical properties compared to virgin fibre mats due to non-optimal matrix adhesion and the coarser recycled fibres that could potentially act as stress raisers. Therefore, the amount of recovered fibres in the laminate composite had to be limited to 25 wt.%[5]. To recover glass fibre properties, a post-treatment would be needed, such as the treatment proposed by the University of Strathclyde. If this can be scaled up to produce glass fibres that can compete with virgin glass fibres, the fibres could be incorporated into applications for chopped glass fibres, such as automotive thermoplastic composites. However, this may be economically challenging, given the low cost of virgin glass fibres (Job et al., 2016).

(Dis)advantages

Like fluidized bed combustion, pyrolysis methods are generally very flexible and able to treat mixed and contaminated waste, which means waste pre-treatment is less important; this makes the method very suitable for end-of-life waste (Job et al., 2016).
The main benefit of pyrolysis is that it is possible to recover both fibres as well as chemical feedstock from the polymer resin. However, It is not always possible to retrieve valuable products from the resin (i.e. monomers that could be reused to produce resins; Oliveux et al., 2015; Pickering, 2005). Refining these products from the pyrolysis oil may be difficult, and it seems likely that this would only be cost effective on a large scale. However, if a pyrolysis process only produces chemicals suitable for use as fuels, an energy recovery process that recovers high quality fibres may be more acceptable (Pickering 2005).

When considering the specific pyrolysis methods, classic pyrolysis takes quite a long time due to inefficient heating (Chen et al., 2015). This is why alternative processes such as fluidised bed and microwave pyrolysis have been developed, which provide faster heating. Fibres recovered from fluidized bed however seem more damaged compared to classic pyrolysis due to high temperatures and possible attrition by the fluidised sand (Job et al., 2016). Microwave pyrolysis does not have this drawback, and the emission of carbon dioxide is also considerably less than that of conventional processes (Kim et al., 2017).

WMH & TRL

Waste Management Hierarchy level: recycling (Bax et al., 2015; Devic et al., 2018; Rybicka et al., 2016)
Pyrolysis of glass fibres is not really a desirable recycling solution due to the reduced recyclate quality, but pyrolysis of carbon fibres rates highly as recycling method. It recovers both fibres and feedstock materials from the polymer, which could be used to create new resins. According to Rybicka et al. (2016), pyrolysis fits both in recycling and reuse, depending on how the recyclate is used.

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[4] The resulting materials showed only slightly decreased mechanical properties compared to compounds with virgin carbon fibres (Job et al., 2016)

[5] Åkesson et al. (2012) also state that it would probably be possible to produce non-woven fibre mats of higher quality when operating on an industrial scale

Technological Readiness Level: 4-8, depending on composite and pyrolysis types (Bax et al., 2015; Rybicka et al., 2016)
The Technological readiness level of pyrolysis depends on the specific type of pyrolysis and composite. Classic pyrolysis has been well-developed (Job et al., 2016) for carbon composites in particular (8 – commercial scale); the method is less developed for glass fibres (6-7 – pilot/commercial scale; Rybicka et al., 2016).
Fluidised-bed pyrolysis has been developed to pilot scale (Job et al., 2016). According to Naqvi et al. (2018), there is still a research gap on the performance of a pyrolysis process for mixed and contaminated wastes in a suitable experimental facility. They state that most research uses a muffle furnace or a fixed-bed reactor, whereas a fluidized-bed or spouted bed reactor (or any other reactor with the ability to process mixed and contaminated wastes) are not addressed yet.
For microwave pyrolysis, several universities have done trials but so far it has not been successfully commercialised (Job et al., 2016). Further research is still needed in order to characterize the extracted polymer and to verify the properties of the recovered fibres. Also, the reaction of how different polymer coatings respond to microwave heating need to be investigated. Still, from the work done, it appears technically feasible to recycle carbon fibres using microwaves (Lester et al., 2004).
Superheated steam pyrolysis is a very novel method (Kim et al., 2017), which still is being researched on at lab scale. Catalytic pyrolysis is still being further developed in a collaboration between Adherent Technologies, Inc. and Boeing (Giulvezan & Carberry, 2003), who were looking into moving to commercial operation in 2007 (Rush, 2007). Gasification is more mature, and often combined with pyrolysis on commercial scale, such as for example by the company ReFiber in Denmark (ReFiber ApS, 2004).

Although pyrolysis methods are more complex than fluidised bed combustion (Pickering, 2005), most industrial methods are based on pyrolysis (Kim et al., 2017). Industrial operations using (classic) pyrolysis for composite waste recycling mainly focus on recycling carbon fibres. Carbon fibre recycling using pyrolysis processes was first established in UK by Milled Carbon (now known as ELG Carbon Fibre; Job et al., 2016). Oliveux et al. (2015) list in Table 2 in their paper numerous companies that use pyrolysis as a recycling company, both located within and outside Europe. The technology maturity suggests that further development of this technology is a viable opportunity, although recyclate application is still limited (Rybicka et al., 2016). An example of recyclate use is by ELG Carbon Fibre, who has industrial scale capability to produce non-woven mats and veils, with or without thermoplastics from thermally reclaimed fibres (Job et al., 2016), or Karborec Spa in Italy that produces milled and chopped CF's and a multi-layered felt containing 95% recycled fibres (Oliveux et al., 2015).

5.3 Chemical recycling

General

Chemical recycling, often referred to as solvolysis, uses chemical depolymerisation of the matrix by using heated solvents or solvent mixtures. The fibres are not dissolved and are released from the resin to be collected (Job et al., 2016; Yang et al., 2012).

Solvolysis offers a large number of possibilities thanks to a wide range of solvents, temperatures, pressures, and catalysts (Oliveux et al., 2015). Depending on the solvent, it can be further classified as hydrolysis (using water), glycolysis (using glycols), and acid digestion (using acid). It can be used for both carbon and glass fibres (Yang et al., 2012), but these processes are more suited to carbon fibres. Glass fibres have low commercial value and suffer degradation from the process due to their fragility when exposed to thermal, acidic and alkaline conditions (Job et al., 2016; Oliveux et al., 2015). Therefore, to make chemical treatment applicable to glass fibre composites, improvements such as milder solvolysis conditions (lower temperature and pressure) are needed (Oliveux et al., 2015).

For the energy demand of the process, Job et al. (2016) report a range from 21 MJ/kg up to 63-91MJ/kg. This range is based on two projects: one at pilot-scale at Hitachi Chemical Japan processing tennis rackets, and one at lab-scale at the University of Birmingham. Optimising and upscaling the process will enable higher processing rates (compared to lab scale) and a lower specific energy demand (Job et al., 2016).

Process

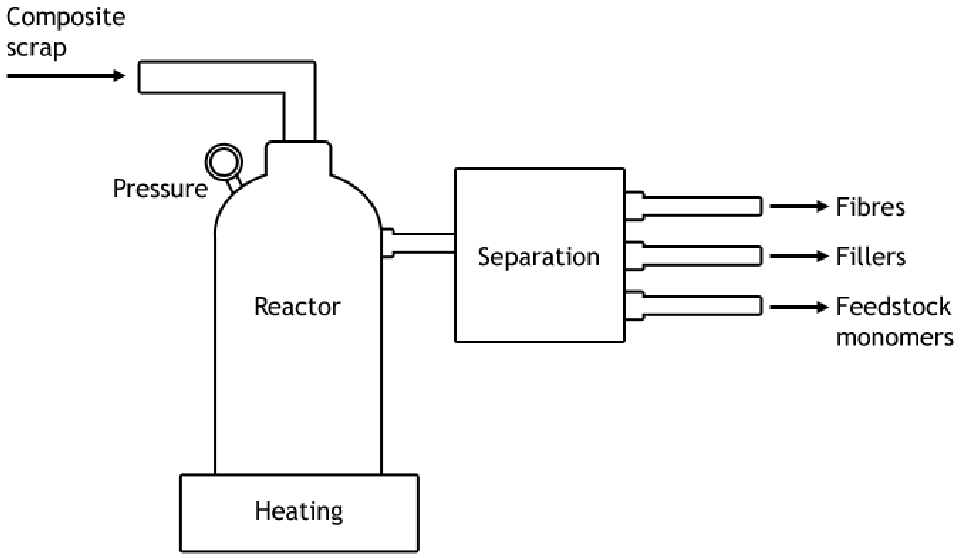

Figure 24: The Solvolysis process set-up. Image constructed using (Carbon TCG Co., Ltd., n.d.; Jacob, 2009)

Figure 24 shows an overview of the solvolysis process. A reactive solvent, sometimes mixed with co-solvent or co-reactive solvent, diffuses into the composite and breaks specific bonds in the resin. This makes it possible to recover not only the fibres, but monomers from the resin as well. Also, it avoids the formation of char residues on the fibres. When testing solvents, water appears to be used the most, but alcohols have been used as well (Oliveux et al., 2015). The benefits of using water or alcohol is that it is environmentally relatively clean, and that it could be separated from the dissolved solution using evaporation (for water) or distillation (for alcohol; Yang et al., 2012). The fluid solvent can be vapour, liquid, biphasic or supercritical[6], depending on the amount of solvent and on temperature.

Relatively high temperatures and pressures (HTP) are needed to degrade the resin, depending on the resin type. Polyester resins are generally easier to solvolyse than epoxy resins and can therefore be degraded at lower temperatures (Oliveux et al., 2015). There are also epoxy resins which have been developed to be easier to recycle (Job et al., 2016), such as by Adesso Advanced Materials (Japan) and Connora Technologies (USA; Oliveux et al., 2015). Adesso’s innovative approach includes the use of a degradable, recyclable curing agent (Cleavamine®), and recyclable epoxies (Recycloset®) that can be reused as toughening agents in adhesives and moulding compounds (Adesso Advanced Materials, 2013). Connora has developed a recycling technology (Recyclamine®) that enables the development and manufacture of high performance 100% recyclable thermoset composites. This includes a recyclable curing agent for existing epoxy systems (Connora Technologies, n.d.-a), and a low energy, solution-based process. The recycling process produces near-virgin fibres in woven mat form, and nearly 100% of valuable epoxy polymer in the form of epoxy thermoplastics that can be reused in conventional processes (Connora Technologies, n.d.-b).

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[6] In a vapour or gas-like supercritical phase, the process characterizes as more of a thermal process than as solvolysis (Oliveux et al., 2015)

To moderate the operating conditions, additives and catalysts can be used (Oliveux et al., 2015), but these can be very detrimental to the mechanical properties of the fibre, and to the environment and worker health (Job et al., 2016). Also, separation of the catalyst salt from the dissolves solution, a highly viscous oil, is still a challenge (Piñero-Hernanz, Dodds, et al., 2008). The catalyst molecules that remain are attached to the surface of the recovered fibres, which gives poor matrix adhesion in future applications (Job et al., 2016; Yang et al., 2012). In addition to (moderated) HTP solvolysis, there is also low temperature and pressure (LTP) solvolysis. This process uses more aggressive solvents to be able to operate at lower temperature (Oliveux et al., 2015). Both HTP and LTP solvolysis are discussed in more detail below.

HTP Solvolysis

Supercritical conditions have gained interest for solvolysis as the tuneable solvent properties change significantly from subcritical to supercritical conditions. The properties of supercritical fluids (SCFs) are in between liquid and gas phases, and benefit from low viscosities, high mass transport coefficients, and high diffusivities (Hyde et al., 2006). This means that SCFs are better able to penetrate into the material and could therefore be used to break down larger pieces of composite material, which is beneficial when looking to upscale the process. Through pressure manipulations, the solvent properties can be controlled, as well as the reaction rates and selectivities of the process (Wu, Klein, & Sandler, 1991). This makes the process very adaptable.

Water in particular has been considered for supercritical use (temperature >374 °C and pressure >221 bar), but the intense hydrolysis conditions require specific and expensive reactors (Oliveux et al., 2015). Only catalysts and semi-continuous conditions enable a significant reduction in temperature (Piñero-Hernanz, García-Serna, et al., 2008). With HTP solvolysis, an alkaline catalyst (e.g. NaOH or KOH) is normally used to achieve a higher dissolution efficiency and a faster dissolution rate (Yang et al., 2012). Acidic catalysts are used less often, mainly to degrade more resistant resins such as PEEK, or to degrade epoxy resins at low temperatures (see LTP section below; Oliveux et al., 2015).
Alternative solvents with lower critical temperature and pressure, such as alcohols or acetone, have been considered as well. These usually require equally high temperatures compared to water for sufficient resin elimination, but the pressure is much lower: about 255 bar at 450 °C for 1-propanol compared to about 630 bar with the same amount of water (Oliveux et al., 2015; Piñero-Hernanz, Dodds, et al., 2008; Piñero-Hernanz, García-Serna, et al., 2008).

The type and amount of resin is also important when choosing efficient temperature and pressure levels. If a resin concentration is too high, the liquid medium becomes saturated and the reaction is slowed down. Using semi-continuous conditions enhances the diffusion process, and it may also avoid resin residue on the fibres and degradation of valuable resin products (Oliveux et al., 2015; Piñero-Hernanz, García-Serna, et al., 2008). When testing supercritical fluids, used temperatures of 250–400 ◦C and a pressure of 4–27 MPa for water, and temperatures of 300–450 ◦C and a pressure of 4–17 MPa for alcohols to recycle carbon fibre reinforced composites. (Piñero-Hernanz, Dodds, et al., 2008; Piñero-Hernanz, García-Serna, et al., 2008)

LTP Solvolysis

LTP solvolysis is generally carried out using temperatures below 200 °C and at atmospheric pressure. As the temperature is very low, catalysts and additives are necessary to degrade the resin, and stirring can also be necessary. Acid mediums (e.g. nitric, sulphuric and acetic acids) have been used the most with LTP, compared to HTP which mainly uses alkaline conditions (Oliveux et al., 2015).

Output

The solvolysis process can regenerate clean fibres and fillers, as well as depolymerised matrix in the form of monomers or petrochemical feedstock (Yang et al., 2012). Gases are also produced but were not always mentioned nor analysed during research (Oliveux et al., 2015).

Resin

For all epoxy type resins, the main degradation product was phenol. Depending on the conditions and epoxy type, other phenolic compounds such as isopropyl phenol and cresol were identified, as well as anilines when the curing agent was an amine (Oliveux et al., 2015). Adding alcohols and amines with a long alkyl chain shows a significant effect on the process yield, as it enhances the degradation of crosslinked (thermoset) resins (Oliveux et al., 2015; Suyama et al., 2009).

The matrix resin feedstock could be used to make new resin again (Yang et al., 2012), but other applications include new polymers, monomers, fuels or other chemicals (Rush, 2007). According to Job et al. (2016), downgraded resins are also suitable for use as thermoplastics or adhesives.

Fibres

Properties

The effects of LTP solvolysis on fibre properties are comparable to HTP solvolysis due to the strong acid or oxidant conditions (Oliveux et al., 2015).

When recycling glass fibre composites only the resin degradation was studied in most cases, due to the low commercial value and fragility of glass fibres (Job et al., 2016; Oliveux et al., 2015). For carbon fibres, the regenerated fibre retains above 90% or virgin mechanical properties (Job et al., 2016; Yang et al., 2012), depending on the conditions. Using an alkaline catalyst (e.g. KOH) with supercritical water gives a resin elimination efficiency of over 90% and only 2-10% mechanical strength degradation for carbon fibres (Piñero-Hernanz, Dodds, et al., 2008). For supercritical alcohol, processing at 350 °C gave resin eliminations of 98% with the recovered carbon fibres retaining 85–99% of the strength of the virgin fibres (Piñero-Hernanz, García-Serna, et al., 2008).

Surface quality

The surface quality appeared to be the weakness of this recycling process. After solvolysis in a batch reactor, the recovered fibres are coated with an organic residue which might consist of material from the fibre/matrix interface. This residue compromises the sizing initially present on the fibre surface, and causes failures by fibre pull-out due to a weak fibre-to-matrix adhesion in new composites (Oliveux et al., 2015). A long and expensive rinsing is required to remove the residue afterwards, and even then some residue (3%) still remains (Oliveux, Bailleul, & Salle, 2012). For glass fibres, impairing the sizing results in dramatic loss of strength and handling/processability. This makes chemical treatments unsuitable for glass fibres, unless the fibres are post-treated to recover the fibre properties; such a treatment has been proposed by the University of Strathclyde. For carbon fibres it is possible to use oxidant process conditions to completely remove the resin. This results in fibres without residue and with an oxygen content higher than virgin fibres, but it significantly decreases the fibre tensile strength (Bai, Wang, & Feng, 2010; Oliveux et al., 2015).

Application

If the post-treatment process could be scaled up to produce glass fibres that can compete with virgin fibres, then the recycled fibres could be used in applications for chopped glass fibres, such as automotive thermoplastic composites. However, economic feasibility may still be challenging, given the relatively low value of glass fibre. (Job et al., 2016).

The application of carbon fibres recovered from chemical processing can be compared to those for thermal processing (Job et al., 2016).

(Dis)advantages

Job et al., (2016) report that, compared to pyrolysis, lower temperatures are generally necessary for solvolysis to degrade the polymers: circa 350 °C for solvolysis compared to 500-550 °C for pyrolysis. Also, the potential to reclaim chemicals from the resin is higher, and it can lead to cleaner fibres with no char formation.

The benefit of LTP solvolysis is that it allows better control of the occurring reactions compared to HTP solvolysis. Also, secondary reactions do not seem to occur as the temperature is low. This enables a higher recovery of epoxy monomers.

Disadvantages of solvolysis include that the process efficiency depends on the resin types, which means that pre-separation of composite types is critical. This makes the process suitable for processing production scraps for which the material characteristics are well-known, but it can be very difficult to use when treating mixed post-consumer composite scrap (Yang et al., 2012).
Also, reactors can become expensive, as they need to withstand high temperatures and pressures, as well as corrosion due to the modified aggressive solvents (Job et al., 2016; Oliveux et al., 2015). This cannot easily be covered by the retrieved value of the fibres, as a cost analysis of the subcritical hydrolysis process developed for glass fibres also showed that the recycled glass fibre would be more expensive than virgin fibre due to the treatment costs (Eurecomp, 2013).
Furthermore, especially for LTP solvolysis, there is the use of alkaline/acidic solvents and catalysts and the generation of toxic effluents. These can be very strong, and have a negative environmental impact and associated disposal costs and potential health risks (Job et al., 2016; Oliveux et al., 2015; Yang et al., 2012).
Finally, the high energy consumption of the process meant that the environmental efficiency of the process was lower than mechanical recycling but comparable to pyrolysis (Eurecomp, 2013).

WMH Level & TRL

Waste Management Hierarchy level: recycling (Bax et al., 2015; Devic et al., 2018; Rybicka et al., 2016)
Solvolysis recovers fibres and fillers, as well as valuable feedstocks from the resin that can be used to generate new matrix material. Yang et al. (2012) believe that only chemical recycling method can provide true materials recycling for both reinforcement and matrix. However, the environmental impact should be reduced and the recycling cost must be well compensated by the market value of the regenerated materials. According to Job et al. (2016), further development of chemical processing could lead to obtaining higher value from resin chemicals, although commercial viability has not yet been demonstrated.

Technological Readiness Level: 3-4 – Pilot scale (Bax et al., 2015; Rybicka et al., 2016)
According to Rybicka et al. (2016), there is a clear distinction in technological maturity between glass and carbon fibre research projects. Glass fibre projects are still residing on lab scale, whereas carbon fibre projects are moving towards pilot scale and demonstrators.
A study by Oliveux et al. (2015) concluded that numerous lab-scale have been carried out, but only a few studies have reached industrial or semi-industrial scale. Among these, ATI and Innoveox have proposed to sell or licence their technology, whereas Panasonic Electric Works (Japan) have built a pilot plant for their hydrolysis process to recycle 200 tons of GRP manufacturing wastes annually (Job et al., 2016; Oliveux et al., 2015). For further commercialisation of the process, the process scale requires much more research and development. If the environmental and cost issues could be resolved, for example through the use of more environmentally friendly critical water, there will be great potential in commercial application of chemical recycling (Yang et al., 2012).

6. Recycling in practice

This section will compare the different recycling methods, discuss their current applications, and consider the feasibility of each method from an economic, sustainable and recyclate quality point of view. This will lead to the future outlook in the next section.

6.1 Current state

In Table 6 on page 43, an overview can be seen of the gathered data for all recycling methods discussed. From this table, there can be concluded that all methods have both their benefits and drawbacks, and that it depends on the situation and desired result what method is most suitable. Most methods are suitable for treating post-consumer waste, but have as drawback that the recovered fibres are just moderate quality and that the resin is only recovered as energy. At the moment, pyrolysis seems to best balance flexibility and quality; this is also the method which is currently commercially exploited the most.

6.2 Method application

In Table 5, an overview of the availability and main application of each method (type) can be seen. This shows that the lower-end recycling methods in terms of recyclate quality are mainly used for glass fibre composites, whereas the higher-end methods are used for carbon fibre composites.

Table 5: The availability and application of the discussed methods

	Availability (Mativenga et al., 2016)	**Application**	
Mechanical grinding	Operating at commercial scale with wide range of potential applications available	Mechanical recycling is mostly used for glass fibre composites due to the relatively low values of the recycled products. It is suitable more for the waste sheet moulding or bulk moulding compounds, but may be difficult for the laminated compounds such as in wind turbine blades (Papadakis, Ramírez, & Reynolds, 2009; Yang et al., 2012)	
High voltage fragmentation	Laboratory and pilot scale machine available, e.g. SELFRAG		
Incineration and co-processing	Commercially available, several companies perform incineration of composite scrap (Bax et al., 2015)	Mainly glass fibres composites are incinerated/ co-processed. It is used for production of energy and recycling of ash in significantly lower value applications (Bax et al., 2015)	Thermal recycling processes have the advantage of being able to tolerate more contaminated scrap materials, making it suitable for processing mixed post-consumer waste (Pickering, 2005)
Combustion	Fluidised bed process investigated at pilot scale	Thermal and chemical processes are mostly used for carbon fibre composites. Any thermal or chemical process strips the sizing off the fibres, which makes these type of processes unsuitable for glass fibres unless post-treated (Job et al., 2016).	
Conventional pyrolysis	Industry scale process available at commercial level, currently operated by companies (e.g. ELG, UK and CFK Valley Stade, Germany)		
Alternative pyrolysis	Microwave pyrolysis has limited availability, even at pilot scale		
Solvolysis	Research done on lab scale, focus on carbon fibre composites		No (commercial) application yet

Table 6: Data overview of all composite recycling methods previously discussed

	Energy	Output	Recyclate application	(Dis)advantages	WMH level & TRL
Mechanical					
Mechanical grinding	Energy costs: 0.1-4.8 MJ/kg [1]	- Resin-rich powders [2] - Fibrous fractions [2]	Resin powders: filler [3] Fibres: filler, moulding compounds, injection moulding [4]	+ Suitable for post-consumer waste + No atmospheric/water pollution [5] (Ribeiro et al., 2016) + Low investment, fast processing, large volumes [5] - Highly inferior recyclate, low market appeal [5,6] - Continuous waste flow required waste [7]	WMH: Recycling [8,9,10] TRL: 8-9 for GF [8,10] 6-7 for CF [8,10]
High voltage fragmentation	Energy costs: 17.1-89.1 MJ/kg [4]	- Resin pieces [8] - Fibres [8]	Resin: not mentioned Fibres: filler, moulding compounds, injection moulding [4]	+ Suitable for post-consumer waste + Quite good matrix and fibre separation [8] + Relatively low-cost [8] + Fibre quality better compared to mechanical grinding [4,9] - Quite energy-intensive [4] - Moderate fibre quality compared to virgin fibres [8] - Resin residue on fibres [8]	WMH: Recycling [8,9] TRL: 5-6 [8]
Thermal					
Incineration	Exotherm Energy gain: approx. 30 MJ/kg [11]	- Energy from resin matrix [12] - Ash and solid residue (fibres and fillers) [13]	Ash: aggregates, construction applications, landfill [14]	+ Suitable for post-consumer waste + Energy recovery of matrix [12] - Near-worthless output [12,15] - Potential health, safety and performance issues due to roaming fibre fragments [16,17]	WMH: Recovery [8,9,10] TRL: 9 [8,10]
Combustion	Exotherm Energy gain: approx. 30 MJ/kg gain [11]	- Energy from resin matrix [6] - Fibres [11]	Fibres: moulding compounds [18], non-woven fibre fabrics [11]	+ Suitable for post-consumer waste + Very tolerant of mixed and contaminated materials [11] + No need to remove metal inserts [11] - Reduced fibre quality and fibres are short and fluffy [6,11] - No resin recovered, difficulty in replicating lab results [8]	WMH: Recycling [8,9,10] TRL: 4 [8,10]
Co-processing	Exotherm Energy gain: Approx. 30 MJ/kg gain [11]	- Energy from resin matrix [7] - Solid residue (fibre ashes, mineral fillers) [7]	Solid residue: incorporated in cement clinker [1,11]	+ Suitable for post-consumer waste + Treats mixed waste [1] + Lower carbon emissions of cement processing [7] + Decreases costs of waste management [7] + No residues left behind [7] + Saves on virgin materials [7]	WMH: Recovery [8,9] TRL: 9 [8]

[1] (Job et al., 2016), [2] (Pickering, 2005), [3] (Oliveux et al., 2015), [4] (Mativenga et al., 2016), [5] (Ribeiro et al., 2016), [6] (Yang et al., 2012), [7] (Vijay et al., 2016), [8] (Bax et al., 2015), [9] (Devic et al., 2018), [10] (Rybicka et al., 2016), [11] (Pickering, 2005), [12] (Larsen, 2009), [13] (Crowder & Richards, 2003), [14] (Williams, 2005), [15] (Halliwell, 2006), [16] (Hedlund-Åström, 2005), [17] (Limburg & Quicker, 2016)

				- Loss of all composite material value - Potential production of hazardous dust [16,17] - Effects on cement product uncertain [15]	
Pyrolysis ***Types:*** ***- classic*** ***- fluidised bed*** ***- microwave*** ***- superheated steam*** ***- catalytic*** ***- gasification***	Endotherm Energy costs: Classic: 23-30 MJ/kg [1] Microwave: 5-10 MJ/kg [1] Other: N/A *operating scale too small*	- Char, oil and gas from resin matrix [2,3] - Fibres, fillers, inserts [2,3]	Resin products: energy source [3], chemical feedstock[18] Solid residue: filler [19] Fibres: moulding compounds, non-woven fibre fabrics [1]	General + Suitable for post-consumer waste + Flexible, able to treat mixed and contaminated waste [1] + Recovers both fibre and resin feedstock [2] - Refining of resin products difficult, only feasible on large scale [2] Classic: - Takes a long time due to inefficient heating [20] Fluidized bed: + More effective heating [1] - More fibre damage [1] Microwave: + More effective heating [21] + Lower CO_2 emission [21]	WMH: Recycling [8,9,10] TRL: 4-8 depending on type [8,10] *Classic:* *8 for CF [1]* *6-7 for GF [10]*
Chemical					
Solvolysis ***Types:*** ***- HTP*** ***- LTP***	Energy costs: 21-91 MJ/kg[1]	- Monomers or petro-chemical feedstock and gases from depolymerised matrix [6] - Fibres [1,6]	Feedstock: new resin [6], fuels [22], polymers [22], thermo-plastics[1], adhesives[1], etc. Fibres: thermoplastic composites, moulding compounds, non-woven fibre fabrics [1]	General: + Lower temperatures compared to pyrolysis [1] + Higher reclaim of chemicals from resin [1] + Cleaner fibres with no char [1] + Suitable to treat waste with known characteristics (e.g. production waste) [6] - Difficult to treat mixed/ post-consumer scrap [6] - Reactors can become expensive [1,3] - Recycled fibre can be more expensive than virgin [23] - Environmental and health issues through solvents & toxics[1,6] - High energy consumption [23] LTP: + Better control of reactions [3] + No secondary reactions, leading to higher recovery [3] - The strong (mostly) acid conditions can be dangerous [3]	WMH: Recycling [8,9,10] TRL: 3-4 [8,10]

[1] (Job et al., 2016), [2] (Pickering, 2005), [3] (Oliveux et al., 2015), [6] (Yang et al., 2012), [8] (Bax et al., 2015), [9] (Devic et al., 2018), [10] (Rybicka et al., 2016), [11] (Pickering, 2005), [15] (Halliwell, 2006), [16] (Hedlund-Åström, 2005), [17] (Limburg & Quicker, 2016), [18] (Cunliffe & Williams, 2003), [19] (Cucuras et al., 1991), [20] (Chen et al., 2015), [21] (Kim et al., 2017), [22] (Rush, 2007), [23] (Eurecomp, 2013)

Table 7: Data overview of the recyclate of all composite recycling methods previously discussed (N/A = method generally not applied for this type)

Fibres					Resin	
	Carbon fibre		Glass fibre			
	Fibre length	Mechanical properties	Fibre length	Mechanical properties	Residual on fibre	Products
Virgin	Any length	*High strength fibre:* Tensile strength: 4.65e3 MPa [1] Stiffness: 225 - 245 GPa [1] *High modulus fibre:* Tensile strength: 2.4e3 MPa [1] Stiffness: 370 - 390 GPa [1]	Any length	*E-glass fibre:* Tensile strength: 2.00e3 MPa [1] Stiffness: 75 - 85 GPa [1] *C-glass fibre:* Tensile strength: 3.25e3 MPa [1] Stiffness: 60 - 70 GPa [1] *S-glass fibre:* Tensile strength: 4.75e3 MPa [1] Stiffness: 86 - 93 GPa [1]	0% resin content Clean surface	Virgin resin material
Mechanical						
Grinding	N/A	N/A	0.8-3.1 mm [2] mean: 2 mm [2]	*Not measurable*	49-59 % resin content [2]	Resin-rich powder [3]
High voltage fragmentation	N/A	N/A	0.5-9 mm [2] mean: 2 mm [2]	*Not measurable*	32-37 % resin content [2]	Resin chunks [4]
Thermal						
Incineration	*No fibres are retrieved*					Energy, ash [5,6]
Combustion	6-10 mm [3,7]	Tensile strength: *550 °C: –20 %* [3,7] Stiffness: retained [3,7]	6-10 mm [3,7]	Tensile strength: *450 °C: –50 %* [3,7] *650 °C: –90 %* [3,7] Stiffness: retained [3,7]	Very little [3]	Energy [7]
Co-processing	*No fibres are retrieved*					Energy [8]
Conventional pyrolysis ***Types:*** ***- classic*** ***- fluidised bed***	Fibre length can be retained [9] Commercially available: 3 –90 mm in various length categories [10]	*Classic* Tensile strength: –4 % to –85 % [11] *Possible to achieve 90% to near-virgin properties using blending* [12] *Fluidised bed* Tensile strength: –25 % [12] Stiffness: retained [12]	**No data found**	*Classic* Tensile strength: –52 % to –64 % [11] *Fluidised bed* Tensile strength: *450 °C: –50 %* [13] *550 °C: –80 %* [13]	None, but there is a char on the fibres [11,12] and the sizing is stripped off the fibres [12]	Oil, gas, char [11,12]

[1] CES Edupack (Granta Design Limited, 2018), [2] (Mativenga et al., 2016), [3] (Pickering, 2005), [4] (Bax et al., 2015), [5] (Larsen, 2009), [6] (Crowder & Richards, 2003), [7] (Yang et al., 2012), [8] (Vijay et al., 2016), [9] (Pimenta & Pinho, 2011), [10] (ELGCF, 2017), [11] (Oliveux et al., 2015), [12] (Job et al., 2016), [13] (Pickering et al., 2000)

Alternative pyrolysis ***Types:*** ***- microwave*** ***- superheated steam*** ***- catalytic*** ***- gasification***	**No data found**	*Microwave* Tensile strength: ca. –20% [14] Stiffness: –13 % [14] *Superheated steam* Tensile strength: *550 °C:* –10 % to –15 % [15] Stiffness: mild decrease [15] *Catalytic* Tensile strength: –1 % to –17 % [3] *Gasification (comb. w. pyrolysis)* Tensile strength: –28 % [16] Stiffness: –10 % [16]	*Microwave* Length varied between 0.1 mm to original scrap size length [17] Using scrap sized 7-30 mm lead to 0.1-31 mm fibre length, with roughly 52 wt% of the fibres being <15.7 mm in length [17]	*Microwave* Tensile strength: *360 °C:* –25 % [17] *440 °C:* –27 % [17] *Gasification* Tensile strength: *500 °C:* > –50 % [18] (similar to effects of heat treatment)	None, but there is a char on the fibres [11,12] and the sizing is stripped off the fibres [12]	Oil, gas, char [11,12]
Chemical						
Solvolysis ***Types:*** ***- HTP*** ***- LTP***	**No data found**	Generally 90% or near-virgin properties [11,12] *Supercritical water w. alkaline catalyst* Tensile strength: –2 % to –10 % [19] *Supercritical alcohol* Tensile strength: *350 °C:* –1 % to –15 % [20]	N/A	N/A	*SC water with alkaline catalyst* <10 % resin content [19] *SC alcohol (350 °C)* 2% resin content [20]	Gases [11], chemical feedstock elements; mainly phenol as well as other phenolic compounds [7]

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[3] (Pickering, 2005), [7] (Yang et al., 2012), [11] (Oliveux et al., 2015), [12] (Job et al., 2016), [13] (Pickering et al., 2000), [14] (Lester et al., 2004), [15] (Kim et al., 2017), [16] (López et al., 2013), [17] (Åkesson et al., 2012), [18] (ReFiber ApS, 2004), [19] (Piñero-Hernanz, Dodds, et al., 2008), [20] (Piñero-Hernanz, García-Serna, et al., 2008)

Considering the application of methods from the composite perspective is shown in Table 8 (Oliveux et al., 2015). When recycling glass fibre thermoset composites, the EuPC, EuCIA and ECRC[7] consider cement kiln co-processing to be the most sustainable solution (Oliveux et al., 2015). For carbon fibre composites, pyrolysis and solvolysis are the most preferred techniques: both have proven to enable the recovery of carbon fibres largely maintaining their reinforcement capability, although both are quite energy-intensive. Pyrolysis is the most widespread technique of the two as it is a proven and heavily used process in the chemical industry, but solvolytic processes have attracted increasing interest as it has lesser fibre degradation and avoids the formation of char contamination on the fibre surface. Also, the process can potentially be optimised to lower energy usage and increase recyclate quality (Oliveux et al., 2015). However, as stated before, it is more difficult to treat mixed post-consumer waste with solvolysis than with pyrolysis. According to Adherent Technologies, as both techniques have their advantages and drawbacks they should not be considered competing but complementary; the best treatment would be a combination of wet chemical processing followed by pyrolysis to remove the organic residues on the fibre surface (Oliveux et al., 2015).

Table 8: Summary of the recycling solutions according to the type of processed composite waste
Constructed using Oliveux et al. (2015)

Fibre type	Resin type	Direct reprocessing	Mechanical recycling	Cement kiln	Thermal treatment	Chemical treatment
Glass	Thermoset		x	x		
	Thermoplastic	x	x			
Carbon	Thermoset		x		x	x
	Thermoplastic	x	x		x	x

6.3 Recyclate quality

In Table 7 on page 45, an overview of the properties of the recyclate from each recycling method previously discussed can be seen. In this table can be seen that pyrolysis, although able to treat mixed waste and yield clean fibres, still has room for development with regard to the mechanical properties of the recyclate. The reduced mechanical strength and length, as well as the fluffy and tangled nature of the fibres, makes it hard to find market applications for the recycled fibres. Solvolysis shows promising results in regard to quality, but it still needs further development before this method will be able to treat mixed waste on an industrial scale.

Important to note is that the fibre length distribution depends not only on the degradation from the process, but also on the length distribution of the fibres in the waste feed (Pickering, 2009). For each recycling method, it is generally required to break down the composite into smaller fragments for easier processing, but this means that the fibre length of the recyclate is automatically limited by the fragment size. However, the smaller size might have a beneficial effect on the recyclate quality. For example, a fluidised bed process requires the composite feed to be shredded to fragments of 10 mm or smaller due to the high operating temperature of the bed. Larger fragments required longer residence times (which inhibits the processing speed) and were likely to increase fibre strength degradation (Pickering et al., 2000). For solvolysis, diffusion of limiting effects might occur with larger pieces of composites, which might leave concentrations of resin residues in the middle of bigger pieces (Oliveux et al., 2015). This leads to the assumption that there is a balance between the quality and length of recycled fibres, and that it is difficult to optimise both aspects. Therefore, when developing and optimizing processing methods, it is not only important to look at the ends results, but also at what input (sizes) can be processed.
Not much research yet focusses on determining exact retrieved fibre lengths. This could be because it is hard to say something about this due to the small size of the test samples being used, or because fibres are in

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[7] Respectively the European Plastics Converters (EuPC), the European Composites Industry Association (EuCIA) and the European Composites Recycling Service Company (ECRC)

different orientations such as woven mat platelets or all in a tangle. Some researches do present fibre length distributions of the fibres recovered in their experiments, but the results of this highly depends on the starting size of the samples used. This may skew the view on recovered fibre lengths presented in Table 7, as it is constructed using different researches and data types, and it also accounts for the data gaps.

6.4 Recyclate economy

6.4.1 Cost validation

When looking at the commercial viability of recycling, the value is mainly in the retrieved fibres. Therefore, the quality and price of the recycled fibres will dictate the commercialisation process, which is the main barrier for most methods at the moment (Yang et al., 2012). It is easier to achieve economic feasibility for carbon fibre composite and methods that produce better fibre length, as these have higher monetary and performance values and can therefore be processed on a smaller scale (Hedlund-Åström, 2005; Yang et al., 2012).

As stated before, thermal recycling methods are often combined to produce better quality fibres with less resin residue, which has not only performance but also monetary benefits. Combining pyrolysis with gasification for example also produces moderate-calorific-valued fuel gas and will be a competitive choice in the future. However, combining technologies makes the process more expensive; it may not always be affordable to do so when pyrolysis is required (Chen et al., 2015). To improve the economic viability of the more costly (thermochemical) recycling methods, it would be necessary to find a way to recover valuable products from the resins. Also, resin prices follow the evolution of petroleum prices, so it would also be a valuable low-cost source of monomers needed for resin production (Oliveux et al., 2015).

6.4.2 Recyclate commercialization

The key issue of commercializing recycled fibres and other recyclate materials is the costs of recycling, regardless of the method used. Current recyclates are too expensive to be able to compete with existing materials (Pickering, 2005). Due to low quality, these materials only compete with other lower value materials, such as aluminium or cheaper virgin material composites (Job et al., 2016). Furthermore, quantity is an important matter as well, as commercially viable operations require large throughputs. To be able to consume these quantities of recyclates, market development is needed. This means that either the cost of the recyclate must reduce to allow the recyclates to penetrate further into existing markets, or that the recyclates must find higher value end markets. The latter may mean developing new higher grade recyclates (Pickering, 2005).

Commercialising recycled glass fibres is difficult due to its low cost: generally €1-3/kg for the most common E-glass fibre type[8] (Cripps, 2017; Granta Design Limited, 2018; Gutierrez & Bono, 2013). This leaves little margin for recycled fibres, and companies struggle to find sufficient recyclate markets to operate at commercially viable levels (Pickering, 2005). Pickering estimated that when processing glass fibres using fluidised bed combustion, a throughput of 9,000 tonnes/year would be required in order to break even. This exceeds the existing industrial capacity (at the time), and is thus unlikely to be sourced within a feasible radius of the recycling plant (Pickering et al., 2000). A more recent analysis by (Cousins et al., 2018) concluded that, when recycling windmill blades, 50% of the glass fibre must be recovered and resold for approximately €0.25/kg, as well as 90% of the resin at a resell price of around €2.25/kg.

When considering which recycling method is most economically viable for glass fibres, Oliveux et al. (2015) state that no reuse of higher value is possible for thermochemical processing compared to mechanical recycling. From this there can be concluded that using thermochemical methods for glass might not be worth the increased cost. It could be feasible for use with long high value glass fibres, but this still needs to

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[8] Prices for the more specialized glass fibre types (R, S or T glass) can increase to around €13-28/kg (Cripps, 2017; Granta Design Limited, 2018)

be proven (Oliveux et al., 2015). This currently leaves mechanical recycling and co-processing in the cement kiln are the only economically feasible options. It is meaningless to compare the price of the recyclate price to that of virgin glass fibres, as the characteristics and applications are too different.

Commercial recycling of carbon fibres has progressed further, since it is an order of magnitude more valuable than glass: recent virgin carbon fibre prices are estimated at €30-60/kg (Granta Design Limited, 2018; Meng, McKechnie, & Pickering, 2018). Additionally, carbon fibre recycling initiatives have received support by large aerospace primes, while equivalent support for glass has been lacking (Job et al., 2016). Cost savings for recycled fibres are generally around 20-40% (Gardiner, 2014; Zoltek, 2019). When using fluidised bed for carbon fibres, a price of around 2-14 €/kg would be required to break even, depending on the operating scale of the recycling plant (Meng et al., 2018). Of course, the options for recycled carbon fibre are more limited in terms of fibre strength, length, alignment, and type (e.g. pellets and non-woven fabrics instead of long continuous fibres), but not all applications require the properties of expensive virgin fibres.

6.4.3 Market appeal

According to Oliveux et al. (2015) industrial applications using recycled fibres or resin are rare. They state this is partly because of a lack of confidence in performance of recycled fibres, but also because a batch of recycled composites originates from different manufacturers and often consists of different fibre grades. This makes it hard to control recycled fibres in terms of length, length distribution, surface quality (adhesion to new matrix) and origin. This doesn’t mean however that there are no feasible applications for the recyclate. Naqvi et al. (2018) state that there are many potential approaches to recycled fibres besides competition with virgin materials, such as penetrating new markets or forming new materials.

The value of a recyclate can be enhanced by exploiting its unique properties. For example, the coarse recyclates of glass fibre composites can be used as a core material as the higher permeability works as a flow channel (Pickering, 2005). Another example is that fluffy recycled fibres lean themselves quite well for wet paper making technology for example, as the fluffiness of the fibres provide extra grip (e.g. the Optiveil and Optimat products from Technical Fibre Products Ltd; Job et al., 2016). The 3D orientation of discontinuous and uniform fibre may also be used to give enhanced drape properties for automated forming (Naqvi et al., 2018). These markets should be explored and developed further to achieve the right market appeal to be able to further progress economically viable fibre recycling.

6.5 Sustainability

Previously in this report, the waste management hierarchy level of each method has been indicated. However, as almost all methods are placed under recycling, WMH levels don’t say that much. For example, even though they are all labelled as recycling, mechanical recycling produces still-mixed recyclate whereas pyrolysis and solvolysis recyclates are almost clean. This may give a skewed view when discussing the sustainable feasibility of the methods and the application of the methods in a circular economy. Co-firing composites in a cement kiln for example might seem favourable as it produces no composite waste for landfill, but it does not deliver any feedstock for creating new composite materials which means that it cannot be considered a feasible circular method. This section tries to give a more in-depth view on the sustainability aspects of the composite recycling methods, discussing both material and energy use.

6.5.1 Material

The sustainable benefit of recycling composite materials is that it avoids the use and production of (high embodied energy) virgin materials. This can be achieved both by replacing virgin composite constituents with recycled ones, but also by (cross sector) applications of the recyclate to replace other materials. For example, using glass fibre thermoset waste in railway applications can avoid the production of virgin or new concrete (Job et al., 2016), a material which is also difficult to recycle.

In a circular economy, it is desirable to keep the resources in use for as long as possible with maximum recovery, and with reuse for new composite and other viable high-end applications at the end of service life (Naqvi et al., 2018). Still, it is hard to create a closed loop in terms of resource efficiency for fibres, as recycled fibres cannot be reused in similar applications as the original fibre, especially those from high-end critical or structural parts (Oliveux et al., 2015). Therefore, it is important to use and develop methods which produce the highest recyclate quality and yield for both resin, fibre and filler. Also, feasible and qualitative applications for recycled composite materials should be developed in order to make optimum use of the remaining material value.

Another conclusion however could be that there is no place for composite materials in a circular economy, despite their weight (and thus energy) saving properties, as no sufficient circular use can be produced. Instead, in a circular economy context the composite is comparable to the monstrous hybrid: “a mixture of materials both technical and biological, neither of which can be salvaged after their current lives” (McDonough, 2015).

6.5.2 Energy

The energy demand for recycling fibres is relatively lower (10 to 20 times) compared to the energy demand for producing virgin glass and carbon fibres (13-32MJ/kg and 183-286MJ/kg respectively; Job et al., 2016). When placing the energy savings in context, it is nice to cite the example given by ELG Carbon Fibre managing director Barnes. He states that a car using virgin carbon fibres would need to be driven 155,000 km before the fuel savings offset the manufacturing energy, whereas for recycled fibres this balance is achieved in less than 15,000 km (Bakewell, James, 2017).

When comparing the different methods, there can be seen that the energy demand for chemical process is typically higher than others. However, as stated in the previous section, solvolysis is a favourable development in terms of recyclate quality and yield. Therefore, it pays to offset the energy demand of recycling methods and virgin fibre production to the material quality, as done in Figure 25. To score the recyclate quality of each method, a score was estimated keeping the reduction of strength and stiffness as well as the residual resin in mind. Also, the following categories were used to aid scoring: virgin quality (score = 10) – excellent (9-8) – good (7-6) – moderate (5-4) – poor (3-1) – no recyclate (0).

Figure 25 shows that methods with a higher recyclate quality generally have a higher energy demand as well, and vice versa. This means that although mechanical grinding might not be that good of a recycling method in terms of recyclate quality, only a small amount of energy is needed to reach this result. This is even more the case for the recycling methods with a negative energy demand, which actually produce energy as well as recycled material. This illustrates the balance between (energy) investment and resulting recyclate quality.

In Figure 25 there can also be seen that the methods most commonly used for glass fibre composite recycling score low on recyclate quality, whereas the methods used for carbon fibre composites score higher. This shows that a bigger effort is made for the higher residual value in the carbon fibres, whereas the glass fibres are considered much earlier not to be worth the effort.

In terms of development, the older more developed methods (e.g. incineration, mechanical grinding) are currently on the low end of recyclate scores, whereas the newer methods (e.g. solvolysis) score higher. Pyrolysis is a method benefitted from both previous development in the chemical sector as well as considerable recyclate quality, which explains why this is the main method used in commercial applications. Still, if solvolysis can be developed further, it should be able to compete.

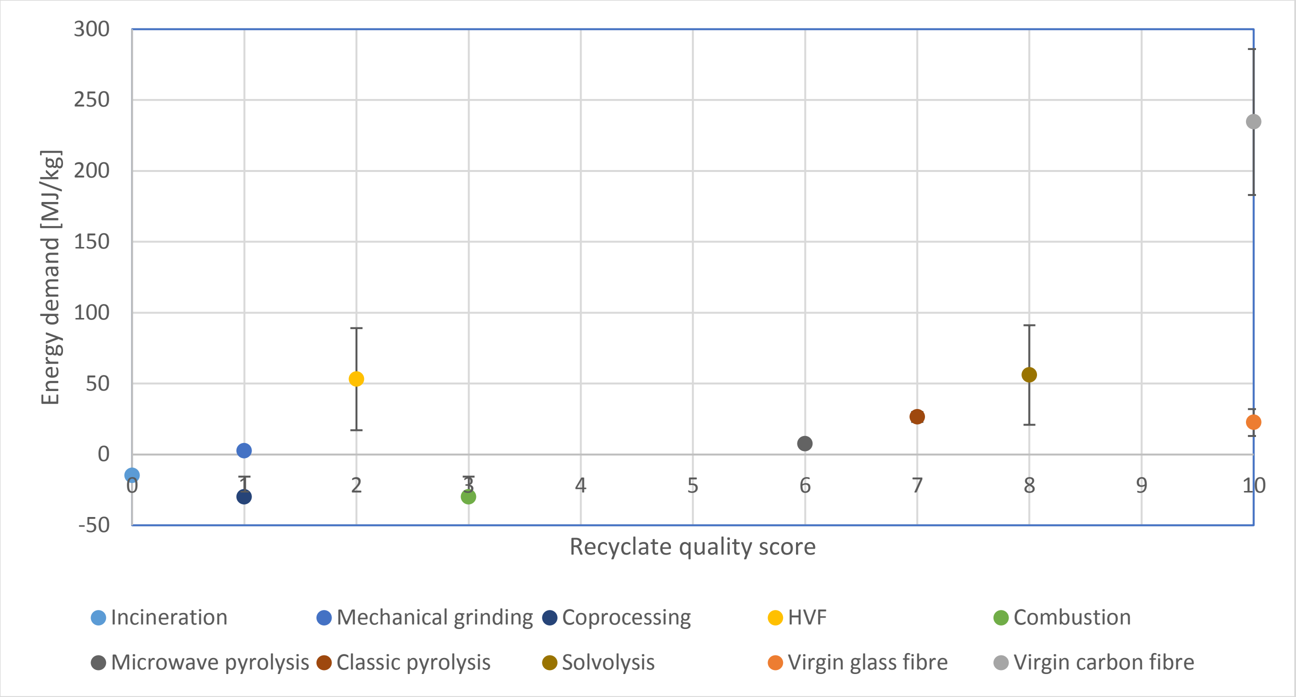

Figure 25: Offset of energy costs and material quality for the discussed recycling methods and the production of virgin fibres Negative energy demand means an energy gain (e.g. exotherm process). The recyclate quality score was estimated using the following categories: virgin (10) – excellent (9-8) – good (7-6) – moderate (5-4) – poor (3-1) – no recyclate (0)

6.6 Feasability

Considering the previous paragraphs, there can be concluded that there is currently no feasible way to recycle glass fibre composites on a near-virgin value level at industrial scale. Any commercial recycling of glass fibres currently undertaken involves downcycling methods and low-value applications.

For carbon fibres however, good process is being made: pyrolysis is exploited commercially and two solvolysis processes are available for commercial exploitation. Both techniques are able to provide clean and high quality carbon fibres, although they consume more energy than other methods due to the high processing temperatures. These are hard to lower for pyrolysis, whereas solvolysis could still be optimised (Oliveux et al., 2015).

The biggest barriers in composite recycling are the reduced quality of the recyclate which complicates circular usage of the material, and finding market applications for the recyclate products due to the lower quality and lack of confidence in recycled feedstock materials.

7. Future outlook

From the previous chapter there can be concluded that there is still plenty of room for development to be made on the subject of composite recycling. This section will review what developments are needed and what developments are being made, as well as what other options there are.

7.1 Current approach

Below, the developments around the current recycling practice is reviewed from different approaches, as derived from the biggest barriers within the current recycling practice: the reduced quality of the recyclate and finding market applications for the recyclate products (Pickering, 2005). Additionally, there are developments needed on the subject of implementation of these developments and changes, which will be discussed as well.

7.1.1 Recyclate quality approach

According to (Yang et al., 2012), recycling technology will become much more mature. This will either involve separating the reinforcement from the matrix, or recycling the composite as a whole by processing the (remeltable) matrix together with reinforcements. There is a high demand for the former approach, as there is sought to offer high efficiency and high quality of liberated and recycled products, without compromising environmental and economic constrains. (Yang et al., 2012).

To improve the market opportunities for recyclate material, either the recyclate needs to be improved or the manufacturing process tolerances should be increased process by using more recycled fibres, matrix or both. (Yang et al., 2012). In order to produce valuable chemicals from the polymer, research is being undertaken on scaling up chemical processes to commercial scale (Job et al., 2016). Meanwhile, there is also scope for improving pyrolysis processes to minimise fibre degradation (Job et al., 2016), but scaling up proves to be a challenge here as well. It was reported that laboratory-scale or pilot-plant pyrolysis led to better results in terms of fibre surface quality and mechanical properties than industrial-scale processes (Oliveux et al., 2015).

7.1.2 Market approach

Recovering high grade fibres only solves part of the problem: finding markets of sufficient size capable of using recovered fibre is also a major challenge (Job et al., 2016).
As glass fibres cannot yet be retrieved in any feasible manner, product specific development is needed to incorporate regrind as a reinforcing filler, for example in infrastructure products. (Job et al., 2016).
Recovered (carbon) fibres are in a physical form unlike any virgin fibre material, which limits the markets in which it can be sold. In order to make composites that compete with high value virgin carbon fibre composites, high carbon fibre volume fractions of up to 60% are needed. In order to achieve these fibre volume fractions, near unidirectional fibre alignment is needed. This requires the development of viable techniques for alignment of the recovered fibres (Job et al., 2016).

Instead of looking what development is required to make bringing the composite back to its constituents economically feasible and marketable, there can also be looked at how to make composite scrap marketable as a whole. A recent study by Conroy, Halliwell, & Reynolds (2006) on recycling construction industry composites reported that significant incomes could be earned if recyclates were used as filler materials replacing woodchips or recycled plastic. However, using recyclate in processes that are already optimised for existing products can be difficult, which can act as a implementation barrier . The future therefore lies in developing markets into which the recycled products can be sold at profitable prices, which is currently being studied (Pickering, 2005). In their paper, Conroy et al. (2006) list the valuable properties of recyclate material which can act as an opportunity, as well as requirements for using the recyclate material: the application should be beneficial, and not just for the sake of doing something with the recyclate.

7.1.3 Implentation development

The recycling of composites will play a vital role in the future, especially for sectors that depend on composites such as aerospace and automotive. These industries will require different recycling options for their products that will fit with current legislation and business models (Rybicka et al., 2016). To commercialise what is technically proven, appropriate business models need to be developed that integrate existing waste management supply chains using associated capital investment (Job et al., 2016). To this end, the research and development will be mainly centred about these sectors. Moreover, a very strong university–industry cooperation is important and will facilitate the success to reach the final goals (Yang et al., 2012).

Most (carbon fibre) composite products have a long service life, which means that it will take a long time before the end of life components will be available for recycling. This means that the growth of the recycling sector will lag significantly behind the current growth in the (carbon) fibre manufacturing (Job et al., 2016). To minimize the lag, it is important to already start further development of recycling methods in anticipation of the future composite waste.

Currently, the main driver for development is European legislation, which requires that recycling routes are available for composites (Pickering, 2005). Through legislation, the high cost of composite recycling will be compensated by forbidding landfill and incineration of composites, and by increasing production cost of virgin composite constituents (Yang et al., 2012). In order to further stimulate and financial assist composite recycling activities however, additional measures will be required.

It is also important to note that zero waste developments are cannibalising the area of composite recycling by diverting the waste as valuable materials. Therefore, widening the scope into considering system level transformation could be researched as well, as well as assessing not only technologies but strategies as well (Rybicka et al., 2016)

7.2 Alternative approaches

7.2.1 Alternative recycling methods

Instead of working on improving the recyclability of composite materials through improving the current recycling methods, there could instead be argued that different recycling methods are required that are more tailored to the specific needs of composite materials. In order to find new methods, research is being undertaken in multiple new directions.

One example could be microbial degradation of composite materials. This method gained interest quite a while ago now; in his 2003 paper, Gu reported two main research groups that explored microbial degradation for polymer composite materials, dating back to before 2000.

More recently in Europe, the Hohenstein Institute in Germany ran trials using microbiological systems to break down the synthetic matrix in a controlled way. This means that the plastic matrix can be broken down into molecules to be returned to the materials cycle, whereas the carbon fibres can be extracted without damage for use in new products (Composites Today, 2015). However, when searching the Hohenstein Institute on any word on the trials, nothing can be found. It is also hard to find any more recent research on the subject from other research groups. This raises questions on the success of the trials, as well as the method's flexibility for different matrix types and the feasibility of upscaling.

7.2.2 Material approach

Another approach is not designing new recycling methods, but instead the composite material itself. Yang et al. (2012) state that the link between composite design and manufacturing, and the reusability and recyclability of the materials is a very important research topic. This research area is the least developed, even though it has considerable importance and can be highly beneficial.

Enhancing composite recycling through material design could be a composite in which the matrix and fibre have a similar chemical nature. However, Yang et al. (2012) argue that this is almost always detrimental to the (end-use) properties of the material. They conclude that new and innovative concepts are needed in order to balance end-use properties and recyclability.

There is research being undertaken in order to develop the resins to be more easily recyclable, such as by Connora as cited in paragraph 5.3 Chemical recycling.

Another development is that of "green composites" or bio-composites: composites that use natural or biologically derived fibres and/or matrix materials, which benefit from bio-renewability and increased recyclability potential. The mechanical properties of natural fibres can easily compete with glass fibres, whereas improved weaving processes could for mechanical high-performance composites (Perry et al., 2010). According to Mitra (2014), biocomposites can supplement and eventually replace petroleum-based composite materials in several applications. They also expect bio-nanocomposites to have very strong future prospects, though the present low level of production, some deficiency in technology and high cost restrict them from a wide range of applications.

However, it will take a while before these materials are used in significant quantities, and even longer before these composites enter the recycling chain. This illustrates the delay between material development and resulting benefits, whereas method development takes effect immediately. This could explain why the material approach has not been favoured so far.

7.2.3 Alternatives to recycling

In this paper, recycling and recovery of composites is the main focus, but this only one step up from direct disposal. To retain as much value from composites as possible, there should also be looked at what other options there are to process composite products as close to its original shape and material as possible for maximum value retainment.

Perry et al. (2012) argue that main efficient recycling use of composite parts should be its full re-use, and that therefore the initial design must already consider recycling. They also state that the second best end of life solution is re-use as semi-products, where the initial part becomes a primary part for a new structure or product. For example, aircraft girder, plates or tubes can serve as basic assembly component, whereas thick parts could serve as raw material for screws, rings or all small elements. They conclude that in order to realize this, innovation combined with a perfect knowledge of the initial composite source is needed. Windesheim agrees, and states that composite recycling should not try to recover the base materials, but instead the end of life composite should be processed into big parts and used as reinforcement in new products. Their demonstrator products are riverbank protection profiles and construction chipboard, made from respectively strips and chips of recycled composite material.

In addition to salvaging composite material from parts and products, there are also opportunities to be found in repurposing products as a whole. For example, the master thesis of Speksnijder (2018) looked into repurposing windmill blades into a bridge constructions. Other examples that have already been realised are repurposing windmill blades as a playground (Superuse Studios, 2008) or constructional elements such as a roof (Bank et al., 2018).
These kind of solutions are possible with the current limited waste volume, but it would be difficult to find sufficient applications in order to meet large waste streams. Also, these products will eventually meet end of life as well, which means that there is still a need for feasible and qualitative recycling. However, it would be nice to be able to retain material value for as long as possible, and these projects could at least serve as inspiration or awareness raisers.
Ideally, a composite material (products) is used for multiple cycles on corresponding value levels to retain as much material value for as long as possible. Chapter 8. Implications sketches out what such an extended lifecycle might look like.

7.3 Future vision

In this paragraph, I will outline my expectations of the future, based on the data and insights gathered in this report.

In the near future, I expect that the practices described in the current approach will be used to optimize current recycling methods and bring them to a higher technology readiness level. Maybe the pre-processing shredding stage will also gain interest, as the size of the composite scrap that gets fed into the recycling process is the main limiting factor of the retrieved fibre length, and not as much the recycling process itself. This could lead to “smart shredding”, in which composites are broken down keeping the fibre orientation and location in mind in order to optimize the fibre length. This could be done for example by using manufacturing data, scanning techniques, etching break lines, and laser/CNC cutting in order to control how a composite part is broken down into chunks that are sized to optimally balance retrieved fibre length and process performance.

Then, as the bulk of composite materials start coming, the parties involved (companies, research, government, etc.) start realizing that this approach is not sufficient and that a lot of material value still gets lost. They then start searching for alternative methods and approaches. In the meantime, the use of composite materials will be limited and only applied it where it is strictly necessary or where it yields considerable sustainable benefit, for example through weight savings. Also, so-called green composites get more of a foothold due to their higher (perceived) sustainability.

When exploring possibilities for development in the far future, the use of nanotechnology could be an opportunity. So far, there has not yet been anything published on applying this technique for fibre reinforced plastics, but it has been researched in combination with recycling of plastics and creating higher value recyclates (Khan et al., 2014; Zare, 2013). If nanotechnology matures, maybe there is a possibility in using this technique to break down composites on nanolevel, improve recyclate quality, or find applications for (low-quality) recyclate in the creation of nanomaterials. However, for now this is still a long way off. Another far-fetched idea using the material approach could be to create a ‘smart composite’. Research has been performed on the subject of creating composite materials with embedded sensing networks to create information-aware materials (Schaaf & Leigh, 2008). Maybe, such a functionality could be extended to create materials which sense that they are being recycled, and change their structure accordingly to facilitate easier recycling, or even self-destruct, although then you would have to worry that no-one hacks your material and destroys it prematurely/while still in use.

Additionally, as oil and material prices start to rise, the manufacturing worlds starts to look more into using recycled materials. Manufacturing regulations are adapted to the use of recycled materials in face of the material shortage, and successful cases prove the potential and safety of recycled materials. I think the future of using recycled materials starts in changing the mindset of manufacturers and producers. Instead of perceiving it as a lower version of the virgin material, it needs to become a material in itself with unique properties and profitable price points. It should not compete with virgin materials, but instead a cheaper option for applications that do not require the properties of virgin materials.

Besides the development of improving the development and use of better recyclate constituents, I think there will also be more development in the direction of repurposing composite parts and materials as a “halfway step” before complete recycling as more of the material value is retained. In order to facilitate this, the share of thermoplastic resins will grow, as thermoplastic composites are far easier to reshape and repurpose than thermoset composites. To this end, multiple research programs and company-driven initiatives will be set up to look into the best way to realize this, and to match end-of-life parts and materials to new applications using the material and part-specific properties.

As there are so many types of composites, both in material type, structural build, and application, there is not a one-fits-all solution that will solve all problems. Instead, the solution space should be segregated into multiple sectors that focus on specific composite types. This calls for a good and well-structured overview that shows all involved stakeholders what is out there. A good start could be the scheme in Figure 26. This can be expanded to include for example research initiatives, materials available, developments and projects being undertaken, calls for partnerships, recycling methods available, etc. In such an overview, manufacturers could search what material needs there are and how to facilitate easier recycling with their materials to gain a competitive advantage. Producers on composite parts could find what material best fits their product with minimal costs and sustainable impact, and if instead of using virgin materials, it might be worth considering recycled constituents or even repurposed materials, parts or products. Also, they can search what recycling possibilities are there for the products at the end of life. I also think that responsibility for recycling should remain with the manufacturer and/or supplier (extended producer responsibility), as this creates more controlled waste streams and thus easier recycling and more qualitative recyclate. Products that use composite material are mostly more expensive and specialist products such as cars and airplanes, which should facilitate the reverse logistics, but they are also used in more low-end products such as sports equipment. Additionally, concepts such as lease models or return fees and educating the end user on remaining value and benefit of the product could be used to encourage them to hand their products back in. The costs of return logistics will eventually be recovered through the higher recyclate value. Meanwhile, corporate and/or government funding may be required to set up the system.

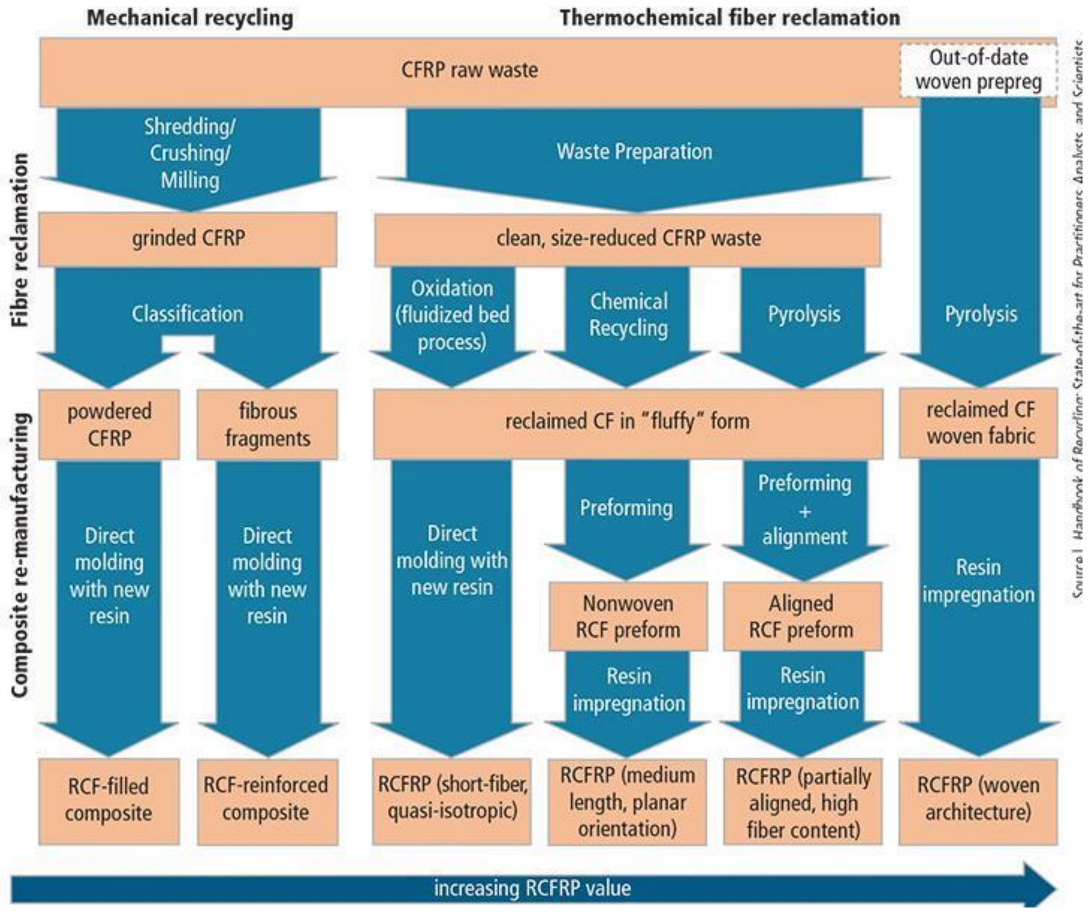

Figure 26: Overview of recycling techniques and products (Pimenta & Pinho, 2014)

Concluding, in order to realize a circular economy in which composite materials have a place, it is paramount that all stakeholders and parties start working together. As there is not yet a large bulk of composite material available, so now is the time to join forces and explore all possibilities to come to the best possible solution. When the composite waste starts coming in, we should be ready to start expanding our industrial practices in order to meet this growing waste volume. Meanwhile, new applications of recyclate material should be developed by regarding the recyclate not as an inferior material but instead as a new material type with its own unique properties and benefits, including the lower price point. To realize this all, an overview of all composite (recycling) activities should be made and maintained, as well as a timeline that plans out all the needed developments together with the required time management. This will facilitate the linking of input and output streams to finally come to a circular economy.

8. Implications

From the findings earlier discussed, there can be concluded that just upgrading and improving current practice will be sufficient to tackle the issue of composite recycling, especially at the expected waste scales in the coming years. There will be a need for new approaches and changed perspectives throughout the whole composite lifecycle, from initial design to end-of-life practice. All stakeholders involved should take up responsibility, and consider not only the end-of-life practice, but also additional lifecycles of composite materials and products. End of use does not necessarily mean end of life, and a lot of material value gets lost in making this mistake. From composites that get shredded up prematurely to airplanes that get abandoned in the desert: just because they are perceived to have lost all commercial operating value does not mean there is no value left to retrieve through other measures.

A new approach should start at changing these perspectives, and then work its way up into system and practice changes. Below in Figure 27, a first step is made in this approach by discussing the complete composite material lifecycle using multiple use and value cycles. Comparable lifecycle approaches have been made such as by Perry et al. (2012, 2010), but these either don't represent all available options and value levels, or give insufficient insights in what state the composite material

In the lifecycle in Figure 27, constructed together with fellow MSc student Julia Koelega, the different possible cycles for composite materials are shown, together with the routes that can be taken to arrive at each stage. The position of each element or stage illustrates its current value and the width of opportunities left available, with the upper left corner having the highest value and opportunity and the bottom right corner the lowest value and opportunity. In this diagram, there are three main value levels, represented by three product categories, as described in Table 9.

Table 9: Product categories for different product value levels in the composite lifecycle diagram

A-product	**B-product**	**C-product**
Definition: A high-grade product using long and high-quality long fibres for a highly-technical application.	*Definition*: A product using lesser quality short and/or randomly aligned fibres for more standardized applications, or a product using repurposed A-product (parts).	*Definition:* A low-quality product that uses but does not necessarily need composite material and integrates composite waste/shredded material as reinforcement.
Example: 1. A wind turbine blade 2. Aerospace airplane parts	*Example:* 1. A car dashboard using recycled short fibres 2. A wind turbine repurposed as bridge/playground	*Example:* 1. A tile or dam-wall using shredded composite waste 2. Recycled fibre non-woven veil product

When travelling from one stage to another, there are multiple routes that can be taken. Each route has its own effects on both value and opportunity, as can be seen. Within the diagram, it is possible to cycle multiple times within a value level or to move down a level, but it is not possible to move upwards in the diagram. Once a composite material is deteriorated in value for example by shredding the fibres short, it becomes very hard to obtain a higher value product again.

The thickness of the lines representing how common that specific route currently is. There can be seen that trashing is most common, as well as shredding and recycling to lower value stages.

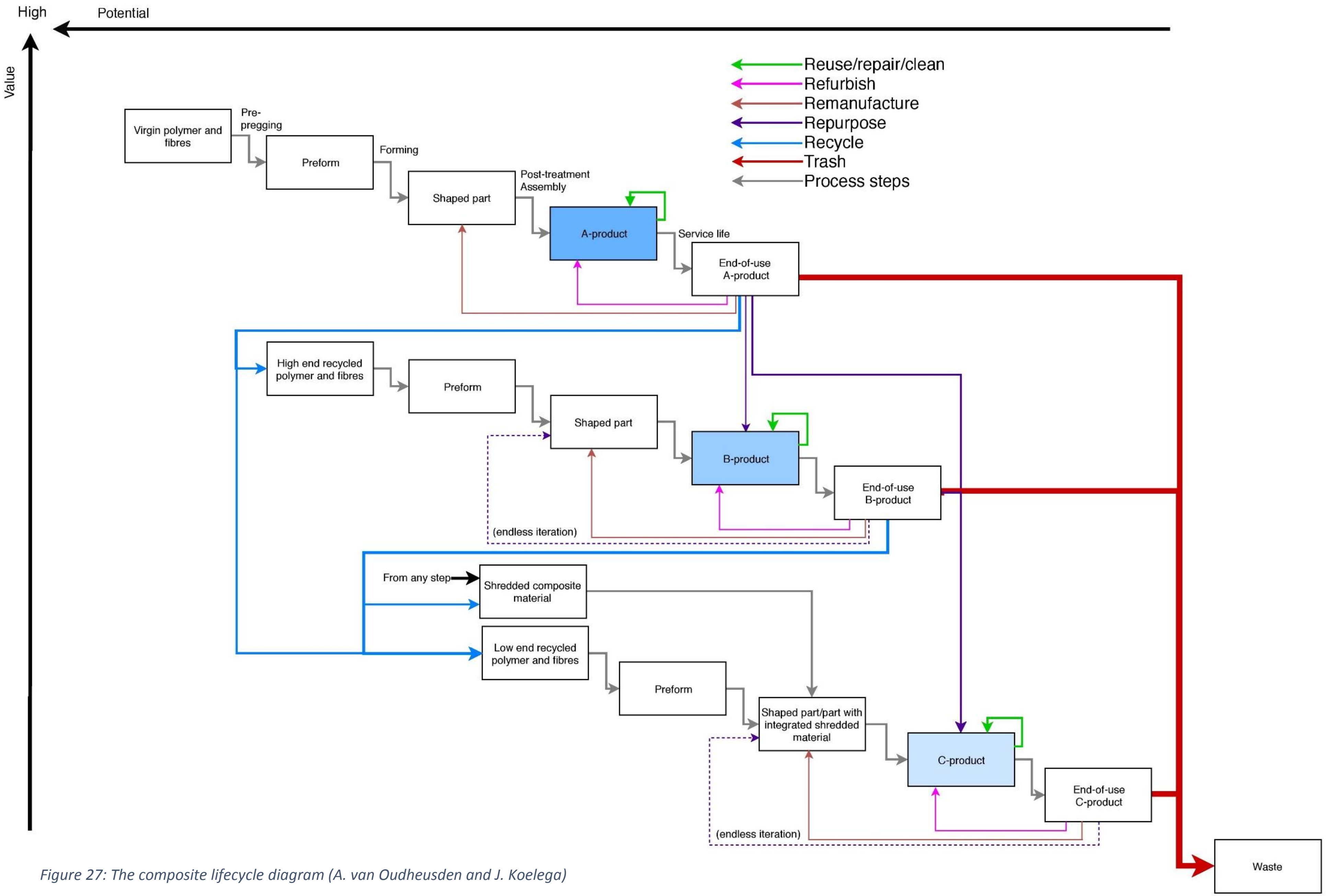

Figure 27: The composite lifecycle diagram (A. van Oudheusden and J. Koelega)

9. Conclusion

In this research paper, there was sought to provide an overview of the current recycling options for composite materials and the technical readiness level of these methods. This is realized through an overview table in chapter 6, as well as more detailed descriptions and visualisations in chapter 5. This data was used to draw conclusions on the feasibility of recycling composites. It turned out that although composite recycling has moved beyond landfill and incineration, there is still room for improvement in terms of process efficiency and recyclate quality, as well as scaling up from laboratory and pilot scale to industrial scale.

Other important findings are that although there is still room for development in the current methods, a different approach could be taken as well in order to create a more extensive lifecycle for composite materials, consisting of multiple use cycles on different value levels. When adhering to the circular economy philosophy, the material value of the composite and composite constituents should be kept as high as possible for as long as possible when processing through its use cycles.

Recommendations for future research are to create a more complete overview of composite economics and markets, as this is not yet a topic on which a lot of public research is available together with relevant conclusions. The topic of retrieved fibre length and properties is also interesting to look into, as well as feasible (market) applications for the recyclate. Finally, future research should move beyond the topic of recycling processes, and into the design phase. As extended producer responsibility is expected to be realized in order to make recycling easier, it is important that the designers and producers already consider secondary applications and recycling when creating their product. To aid this process, it might help to update and extend the terminology list, as well to determine what practices are common, and what practices should be made common. Creating a better future will only succeed when working together, which means that we should all be on the same page talking the same language.

All in all, I hope this paper this report will be a valuable aid for all those who search to gain more information on the topic of composite recycling. It should introduce those who are unfamiliar with the subject to all the possible opportunities for composite recycling and leads on where to start, as well as presenting a complete and extensive reference work for those who are more familiar with the subject.

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