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Recycling of composite materials

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ABSTRACT

Composite materials are used in a wide range of applications such as automotive, aerospace and renewable energy industries. But they have not been properly recycled, due to their inherent nature of heterogeneity, in particular for the thermoset-based polymer composites. The current and future waste management and environmental legislations require all engineering materials to be properly recovered and recycled, from end-of-life (EOL) products such as automobiles, wind turbines and aircrafts. Recycling will ultimately lead to resource and energy saving. Various technologies, mostly focusing on reinforcement fibres and yet to be commercialized, have been developed: mechanical recycling, thermal recycling, and chemical recycling. However, lack of adequate markets, high recycling cost, and lower quality of the recyclates are the major commercialization barriers. To promote composites recycling, extensive R&D efforts are still needed on development of ground-breaking better recyclable composites and much more efficient separation technologies. It is believed that through the joint efforts from design, manufacturing, and end-of-life management, new separation and recycling technologies for the composite materials recycling will be available and more easily recyclable composite materials will be developed in the future.

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1. Introduction

Composite materials provide design engineers with superior quality and long life span. Higher strength, lower weight and less maintenance have led to many engineering applications, in particular in the transport sector for significantly reduced energy consumption and impact to the environment (CO₂). Generally speaking, three types of composite materials are developed and widely used in numerous kinds of engineering applications: polymer–matrix composites (PMC), metal–matrix composites (MMC), and ceramic–matrix composites (CMC). According to the reinforcement types, composite materials can be classified into particulate composites, fibre-reinforced composites, and structural composites. Two types of classifications are illustrated in Fig. 1. Although it is hard to find statistics of the total global composites production, it was estimated with an output of 7 million tonnes in 2000 and could have reached 10 millions in 2006 [1]. For all types of composite materials, polymer–matrix is dominating the market, among which thermosets composites account for more than two

thirds, however the thermoplastics composites are growing more rapidly in recent years.

Two major application sectors (based on value) are automotive industry (over 30%) and aerospace industry (over 20%). Fig. 2 shows the application areas of composite materials according to the estimate for the year 2000. Defence & aerospace industry pioneered the use of composite materials: most defence aircrafts today have greater than 50% weight from composites. Composites have recently become a primary material for the new generation of commercial aircrafts such as the Boeing 787 “Dreamliner” (50%) and the Airbus A380 (25%) and the future A350 (53%). Weight saving technology in automobiles is crucial for improved fuel efficiency. As the largest application sector, use of composite materials in automotive industry is increasing very rapidly (construction of body, interiors, chassis, hoods and electrical components). Furthermore, composite materials are also used in sports and recreation facilities, boat and shipbuilding, in wind energy generation for wind turbines blades, as well as in oil and gas offshore exploration. Break-down and market share of composite materials used in Europe in the year 2000 is shown in Fig. 3. As can be seen, Germany takes the largest share, followed by Italy and France. These 3 nations account for more than 60% of the total. This can also be connected to the major automotive and aerospace industries in these 3 countries.

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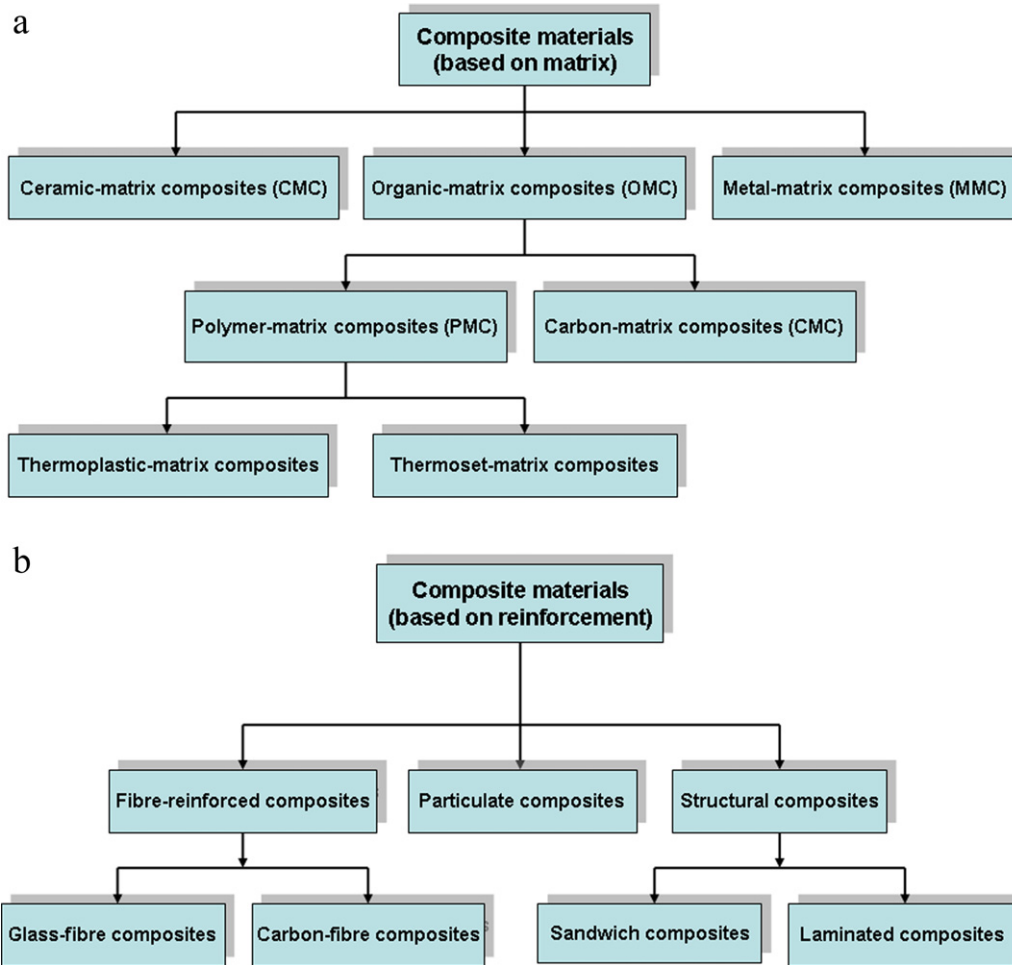


Fig. 1. Classification of composite materials. (a) Based on matrix materials and (b) based on reinforcement materials.

Recycling of engineering materials will contribute to the sustainability and sustainable development of industrial processes. Nowadays, metals, glass, thermal plastics and many other engineering materials are recycled to a great extent. However, composite materials, as a special category of engineering materials have not yet been properly recycled (both for the matrix and for the reinforcement materials). This is mainly due to their inherent heterogeneous nature of the matrix and the reinforcement,

leading to poor materials recyclability, in particular the thermoset-based composites. The current and future waste management and environmental legislations require all engineering materials to be properly recovered and recycled, from end-of-life (EOL) products such as end-of-life Vehicles (ELVs). Recycling will eventually lead to resource and energy savings for production of re-enforcement and matrix materials.

At present there are very limited commercial recycling operations for main stream composite materials, due to technological

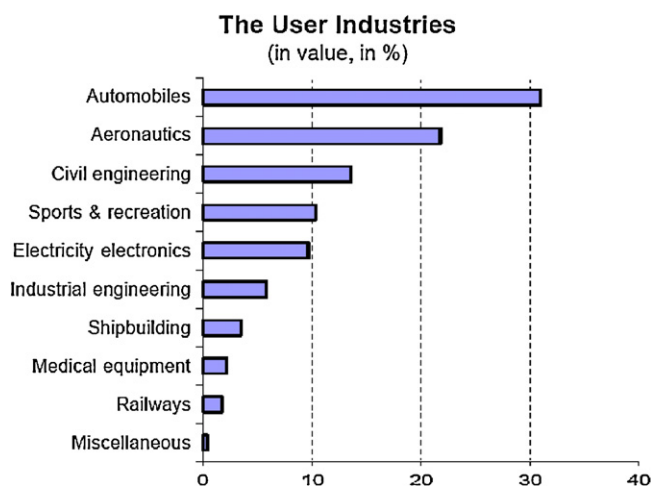


Fig. 2. Application of composite materials [1].

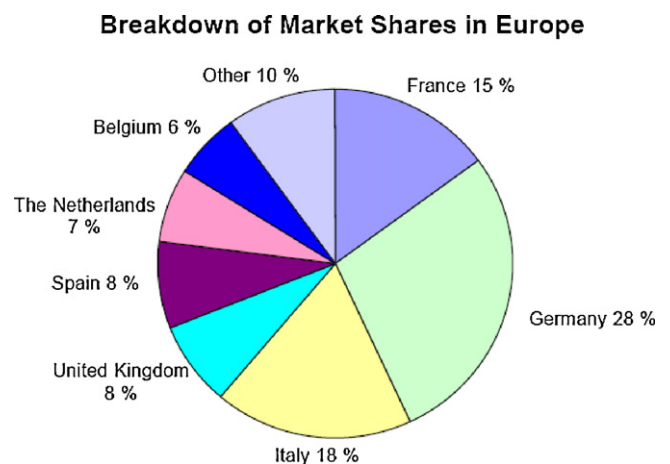


Fig. 3. Market of composite materials in Europe [1].

and economic constraints. Basic problem is the difficulty to liberate homogeneous particles from the composite material. Composite recycling is hindered both by the fibre and other types of reinforcement, and by matrix or binders in particular the thermoset type. Because of these challenges, most of the recycling activities for composite materials are limited to the down recycling such as energy or fuel recovery with little materials recovery such as reinforcement fibres. Relatively recent environmental legislation like the EU-directive for end-of-life vehicles [2] and the directive for waste electric and electronic equipment [3] causes increasing demand for recycling techniques that realize true material recycling.

Extensive R&D activities have been conducted, and various technologies, yet to be commercialized, have been developed basically in three categories: mechanical recycling, thermal recycling, and chemical recycling. *Mechanical recycling* involves shredding and grinding followed by screening to separate fibre-rich and resin-rich fractions for re-use. The method is very energy-intensive and the recyclates have relatively low quality. *Thermal processing* uses high temperature (between 300 and 1000 °C) to decompose the resin and separate the reinforcement fibres and fillers. Clean fibres or inorganic fillers are re-generated, and secondary fuel or thermal energy can be produced through pyrolysis, gasification or combustion. However, the quality of the recovered fibres or filler materials degrades to a varying extent during thermal processing. *Chemical recycling* aims at chemical depolymerisation or removal of the matrix and liberation of fibres for further recycling by using organic or inorganic solvent. Lack of flexibility and generation of waste chemicals with environmental concerns in chemical recycling caused the situation in which there is no active development at the moment. However, a cleaner process based on near- and super critical fluid (in particular water) technology has gained more attention in the research world and shown an interesting potential [4–6].

Lack of markets, high recycling cost, and lower quality of the recyclates versus virgin materials are major commercialization barriers, and will hinder further use of recycled composite materials in automotive, aerospace and other engineering and consumer products. Environmental legislation will help to promote recycling, but long-term technological developments are needed. Groundbreaking innovations are necessary in the following three areas:

- (1) Materials development for new and easy recyclable composite materials.
- (2) Materials recycling for more efficient and intensified separation and purification technologies.
- (3) Production techniques that can at least partially use the recycled fibres instead of only new fibres.

It is hoped that future innovative research and development, and new breakthrough separation and recycling technologies for the composite materials recycling will be available and more easily recyclable composite materials will be developed for the industry. As another long-term goal, we could even imagine a car could be built with completely recycled materials, and a dream of a car manufactured out of waste could be true.

2. Overview of recycling technologies for composite materials

Due to the technological, economical and environmental constraints, very limited industrial operations can be found for recycling of composite materials. Along with the increased future needs and stronger environmental legislations, a number of recycling technologies have been developed and proposed for composite

materials in the past decades. A comprehensive overview on recycling issues and technologies was given by Henshaw et al. [7,8]. An excellent technology review and market outlook was given by Pimenta et al. [9] on recycling carbon fibre reinforced polymers for structural applications. A recently published monograph edited by Goodship [10] updates and summarises most aspects regarding the recycling of composite materials. The recycling issues have already been addressed in the ASM handbook on composites [11]. A good overview on recycling technologies and developments for thermoset composites is given by Pickering [12] and Job [13]. Because of the great majority of polymer–matrix composites in the market, development of recycling technologies have focused more on this type of composite materials. It is also because of the special technical difficulties to separate thermoset matrix from the reinforcement materials, more research and development have been devoted to the recycling of thermoset type of composites. However, recycling technologies have also been developed for other types of composite materials such as thermoplastic matrix and metal–matrix based composites [14]. Table 1 summarises the different technologies mentioned earlier for recycling various types of composites.

2.1. General recycling system

As a general rule for recycling of any engineering materials, each recycling process involves a chain of operations which depends on one another. A failure in any step of this recycling chain implies that the recycling process cannot be completed. These chain operations are illustrated in Fig. 4 and explained as follows.

- (1) The availability of the composite scrap: this is the source of composites for recycling, which can originate from EOL post-consumer products – “old scrap”, or can be the production waste during manufacturing process – “new scrap” or “prompt scrap”.

Composites are volume-wise small in production and in the postconsumer products compared to metals and non-composite type of polymers. The production amount of different types of composites, MMC and CMC are even much smaller. This will lead to the limited availability of the waste composites (the scrap) for an economically viable recycling. Composites used in automobiles or

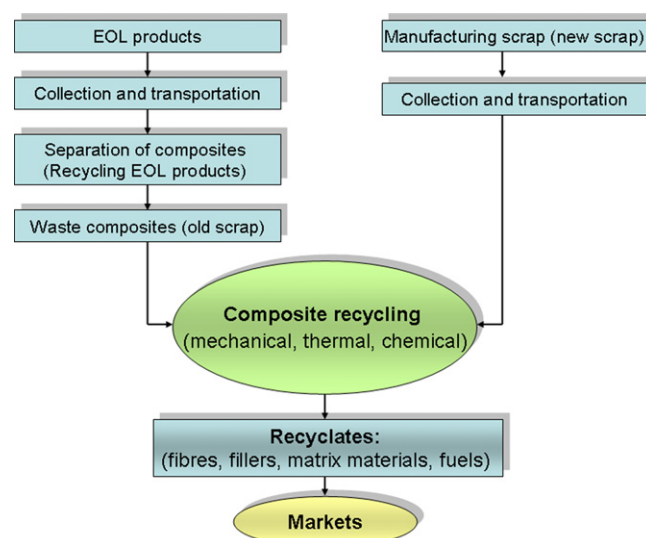


Fig. 4. Structure of recycling system for composite materials.

Table 1
Overview of recycling technologies for different types of composites.

Type of composites	Recycling methods	Technology features	Status of the technology
Thermoplastic–matrix composites	Remelting and remoulding	•No separation of matrix from the fibre •Regrinding – compression or injection moulding/extrusion – compression moulding •Product as pellets or flakes for moulding •Fibre breakage – property degradation	More studied for the manufacturing or process scrap. Commercial operation? unknown
	Chemical recycling	•Dissolution of matrix •Fibre breakage – property degradation	Not much studied
	Thermal processing	•Combustion or incineration for energy recovery (option for old scrap)	Not much studied or published
Thermoset–matrix composites	Mechanical recycling	•Comminution – grinding – milling •Products: fibres and fillers •Degradation of fibre properties	Commercial operation ERCOM (Germany) Phoenix Fibreglass (Canada) Promising technology
	Thermal recycling	•Combustion/incineration with energy recovery •Fluidised-bed thermal process for fibre recovery •Pyrolysis for fibre and matrix recovery	
	Chemical recycling	•Chemical dissolution of matrix •Solvolysis (supercritical organic solvent)/hydrolysis (supercritical water) •Product of high quality fibres, potential recovery of resin •Inflexibility of solvent and potential pollution	Hindered by the market for recycled fibres Only laboratory studies Promising
Metal–matrix composites	Re-melting – casting	•Dir-cast scrap: direct remelting – casting	MMC is much more expensive than the alloys or reinforcements Aiming at reuse of MMC
		•Foundry scrap: direct remelting with (dry Ar) cleaning •Dirty scrap: remelting – fluxing – degassing cleaning •Very dirty scrap: metal recovery only – remelting and refining to separate reinforcement from Al (alloy)	

aircrafts will live many years (10–50 years) before they are returned for recycling.

dictate the whole recycling process. The present challenge is, among others, the lack of market for the recyclates.

- (2) Collection and transport: proper and efficient collection system for “old” and “new scrap”. Collection and transport of EOL consumer products are essential steps in the whole recycling system. Nowadays, the collection of end-of-life vehicles (ELVs) and EOL aircrafts is very well organised. Transportation of these EOL products to the processing facilities may differ depending on the size of the product. ELVs are much more easily transported to the dismantling firms and then further to the shredding plant. However, EOL aircrafts due to their huge size need to be dismantled and cut into smaller sizes for ease of transportation. Incomplete and low collection rates may still be a challenging task for small electronic products as well as for sports and recreation facilities.
- (3) Reprocessing – recycling: these can be the application of physical (mechanical), thermal, or chemical technology, depending on the type of the composite materials. This step is the “core” of the chain operation of the recycling system. Most developments have taken place for proper recycling technologies. Unfortunately, the current available recycling technologies for composites all have difficulties to meet the requirements from product quality, environmental regulations and the operation economics (processing cost). There is great demand for more efficient separation technologies to meet all above criteria.
- (4) Market of the recycled products – recyclates. Market requirements and demand on the quality and price competitiveness compared to the virgin composites are crucial factors, which

2.2. Recycling of thermoplastic matrix composites

Although the market share of thermoplastic–matrix composites is much lower compared to the thermoset–matrix composites, the former has several potential advantages over the latter such as toughness and damage resistance to chemical attack, a more rapid processing cycle and better recyclability. Because of their fundamental ability to be re-shaped upon heating, thermoplastic matrix composites can be recycled directly by remelting and remoulding high value materials [8].

Mechanical breakdown into granules for use in the original processing stream is the most obvious technique for recycling fibre reinforced thermoplastics. However, fibre breakage induced by grinding and subsequent processing leads to devaluation of materials properties [15]. Study based on reprocessing of thermoplastics matrix composites shows certain reduction of tensile strength and Young's modulus, with poor surface appearance but increased failure strain and better moisture resistance. Recycling of thermoplastic matrix composites is more dealt with in the recycling of (non-composite) thermoplastics and polymers. It is thus not going to be discussed further in the current paper.

The main technical difficulty for the thermoplastic–matrix composites is its high viscosity of their melts (500–1000 times), which needs high pressure for the impregnation of reinforcement fibres. This leads to expensive product tooling and significant energy input in heating and cooling the tooling. In many application areas the disadvantages have outweighed their advantages and become

the obstacles for further market development. However, there are new developments in thermoplastic–matrix composites by using new generation of thermoplastics which can be processed in a water-like low viscous state. Thus much lower pressure and less expensive tooling and lower energy are required [16]. The possible liquid moulding of thermoplastics in the composites formation will bring new momentum for more commercial applications and market development. Increased future use of thermoplastic–matrix composites will definitely enhance the recyclability of composites materials.

Otheguy et al. [17] have demonstrated the recyclability of thermoplastic composite boat. It was shown that the hull of an experimental rigid inflatable boat (RIB), composed of glass/polypropylene laminate along with balsa core material and paint, can be recycled by melt processing into injection mouldable granules which have acceptable properties when processed. Although both balsa and paint have a deleterious effect on moulded strength, elongation-to-break and impact strength, the presence of balsa does have a small positive effect on modulus, and also on impact strength in the case of low wood content. In general, the achieved properties in the compounded granules are well within the region of commercial interest for reinforced polypropylene moulding materials. These materials could be used in non-appearance automotive applications, where talc and glass reinforced polypropylene is currently used. Alternatively, they could be used in decking and wood imitation applications where wood reinforced composites are currently being considered.

2.3. Recycling thermoset matrix composites

All 3 types of recycling methods have been widely investigated for thermoset matrix composites and to certain extent are available for future commercial use in industrial operations, and summarised below.

2.3.1. Mechanical recycling

Mechanical recycling process starts with the size reduction of the composite scrap by low speed cutting or crushing (to 50–100 mm). The size is then further reduced down to 10 mm to 50 μm through a hammer mill or other high speed millings for fine grinding. Afterwards the fine particles of the waste composites are classified with cyclones and sieves to fibre-rich (coarser) and matrix-rich (finer) fractions [12].

A recent research was published [18,19] for investigation of the potential use of recycled glass fibre composite materials as a replacement for virgin reinforcing materials in new thermoset composites. Specifically the closed-loop mechanical recycling of composites used heavily in the automotive sector known as dough and sheet moulding composites were studied. The mechanical recycling process and the collection of useful fibrous grades of recycled materials, recycle, by a novel air separation technique were developed. The properties of these recycle fibres were characterised and compared directly with the properties of virgin glass fibres. Single fibre tensile tests were employed to compare the strengths of the fibres and single fibre pull-out tests were used to investigate the strength of the interface between the fibres and a polyester matrix. These tests showed the recycle fibres to be weaker and have a poorer interface with the polyester matrix than the virgin glass fibres. Virgin glass fibres have successfully been replaced by recycle materials without disrupting standard production techniques and with minimal reduction of the mechanical properties of the resulting composites. As the loadings of recycle materials used were greatly increased both the flexural and impact strengths were significantly degraded.

Most of the mechanical treatment through crushing and milling is relatively simple, but it can be energy intensive and is only able to

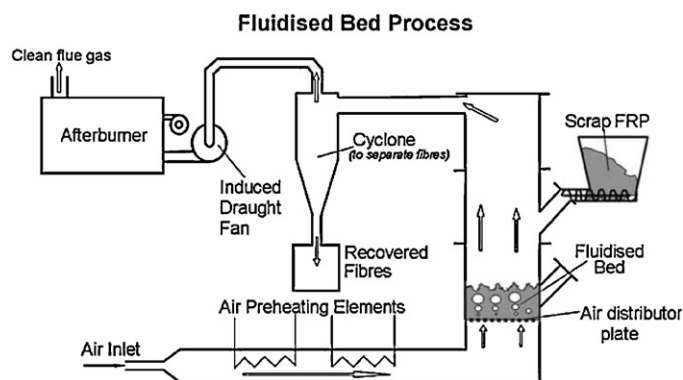


Fig. 5. Fluidised-bed process for fibre and energy recovery (combustion) [12].

produce short milled fibres with poor mechanical properties used as filler reinforcement materials. Two industrial scale development examples are ERCOM (Germany) and Phoenix Fibreglass (Canada) [12] which utilise mechanical recycling technology, and this will be discussed in detail in Section 4.2.1 later.

2.3.2. Thermal recycling

Thermal recycling of composites involves the processing at high temperatures. Thermal processing of the composite waste can include 3 types of operations:

- (1) Incineration or combustion for energy recovery only.
- (2) Combustion for fibre and filler recycling with energy recovery.
- (3) Pyrolysis with both fibre and fuel recovery.

Since incineration and combustion for energy recovery do not involve materials recovery, it is not classified as a recycling technology although the inorganic residues after combustion could be potentially used in the cement industry. However, Municipal Solid Waste Incinerators with a certain thermal efficiency are classified as 'recovery' installations. This distinction between 'recycling' and 'recovery' is also made in some of the European recycling directives. Thus there are only two types of thermal recycling methods, where the fluidised-bed recycling process has been mostly studied for both combustion and pyrolysis with promising perspectives.

2.3.2.1. Fluidised-bed combustion recycling process. Fluidised-bed recycling process developed at the University of Nottingham [12,20] is used to combust the resin matrix as energy and to recover the glass or carbon fibres. At the University of Hamburg a fluidised-bed pyrolysis process is used to recover both reinforcement fibres and secondary fuels from the depolymerisation process [21], which will be discussed separately.

Fluidised-bed technology was investigated to recover the glass or carbon fibres, and the organic resins are used as energy source and the combustion heat is recovered through waste-heat recovery system [12,20]. Fig. 5 illustrates the fluidised-bed recycling process. The composite scrap is firstly broken to 25 mm size before feeding into the fluidised-bed reactor operated with a sand-bed and preheated air. The reactor is operated at 450 °C for polyester resin composites and up to 550 °C for epoxy resin composites. The recovered fibres are clean and have a mean length of 6–10 mm. It was found that the recovered glass fibres suffer from 50% tensile strength reduction at 450 °C, while the carbon fibre has less degradation after the thermal treatment at 550 °C (with 20% loss in stiffness). Pickering [20] has described in detail the properties of the recycled fibres such as physical form, fibre length, mechanical properties for both glass and carbon fibres.

The fibre recycled through the fluidised-bed process is in fluffy form of individual short glass or carbon filaments, as compared to the virgin commercially produced continuous fibres [20,22]. It was found that Recycled fibres of up to 10 mm mean length were recovered and they retained ~75% of their tensile strength, while the Young's modulus remained unchanged and the surface condition was similar to the virgin fibre [22]. Together with the degradation in mechanical properties, this will limit the use of the recycled fibres to the applications with short fibres such as moulding compounds. Commercial viabilities have also been evaluated by Pickering [20], and it could only be economical if a processing capacity of above 10,000 tonnes/year can be reached for recycling glass fibre composite scrap. For carbon fibres the plant scale can be smaller due to the higher market value of the carbon fibres. The recycled fibres will bring the commercial value for the processing, and thus the quality and price of the recycled fibres will dictate the commercialisation process which is also the main barrier today.

2.3.2.2. Pyrolysis recycling process. Pyrolysis is a thermal decomposition of polymers or depolymerisation at high temperatures of 300–800 °C in the absence of oxygen, allowing for the recovery of long, high modulus fibres. A higher temperature of 1000 °C can be applied but the resulting fibre products will be more seriously degraded. It can be used for the treatment of polymers and polymer matrix composites. In the case of polymer–matrix composites, both the reinforcement fibre and the matrix materials (in the form of smaller molecules as oil, gas or solid char) are recovered in the pyrolysis process. Control of temperature and residence time in the pyrolysis reactor is important for the complete depolymerisation and cleanness of the recovered fibres. The process is well described by Pickering [12], Kaminsky [21] and Blazsó [23].

Compared to the combustion process where the polymer resins are oxidised to CO₂ and water vapour with energy release, the pyrolysis process will break down networked resins into lower molecular weight organic compounds in the form of liquid, gas and solid carbon char product. It generates the products with potential use as feedstock for further chemical processing [12]. This brings advantages over the combustion process with true materials recycling for the matrix polymers. Both glass and carbon fibre reinforced composites can be recycled through pyrolysis. Because of the much higher market value of carbon fibres, pyrolysis recycling of carbon fibre reinforced composites is more economically feasible, which is also the case for other types of recycling processes.

The pyrolysis process can be arranged in different types of reactors such as a fixed bed reactor, screw pyrolyser, rotary kiln or fluidised-bed reactor [23]. Out of these the fluidised-bed and rotary kilns are most suitable as pyrolysis reactors [7]. The multi-forms of pyrolysis products may be one of the difficulties in industrial operation. The solid product is normally a mixture of fibre glass or carbon fibre, filler materials, and solid carbon. Separation is needed in order to re-use the reinforcement fibres, or fillers. The condensed liquid product is often a mixture of complex organic compounds with relatively high calorific value similar to fuel oil (30–40 MJ/kg), depending on the type of matrix resins. The gaseous products are often a mixture of CO, CO₂ and hydrocarbons with relatively low calorific value (15–20 MJ/kg), and can be used as heat source to sustain the pyrolysis process (endothermic) through combustion.

The distribution of 3 types of pyrolysis products varies widely depending on the type of the composite scrap, and the pyrolysis temperature [12]. In general cases, the solid product accounts for the highest proportion (50% till more than 2/3) followed by liquid products (10–50%). The gas product falls in a range of 5–15% by weight.

In practice, the pyrolysis is combined with a combustion process in order to obtain clean fibres. This is in fact a kind of combination of pyrolysis and gasification. However, the high temperature and

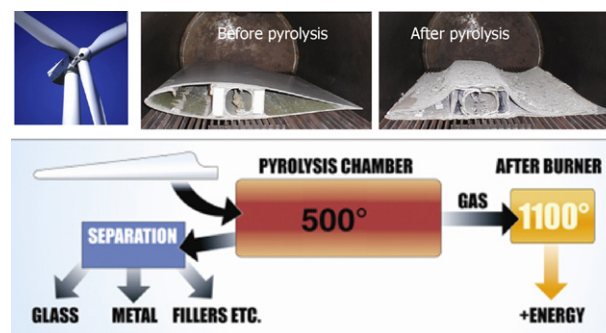


Fig. 6. Illustration of ReFibre process for recycling of wind turbine blades [24].

the oxidation may cause the degradation of the fibre strength. A pyrolysis–gasification process –ReFibre – is developed in Denmark to recycle the glass fibre and recover the thermal energy from end-of-life wind turbine blades [24]. In the process, the wind turbine blades are cut on site to ‘container’ size pieces with a hydraulic shear or similar tools. Once at the plant, the parts are shredded to hand-sized chunks. The material is fed continuously into an oxygen-free rotating furnace at a temperature of 500 °C, where the resin in the blades is pyrolysed to a synthetic gas. The gas is used for electricity production as well as for heating the rotation furnace. At the end of the rotating furnace or in a second rotating furnace the glass fibre material is ‘cleaned’ in the presence of air. Ferrous metals are removed by magnets for recycling. The dust is removed from the clean glass material remaining. Fig. 6 illustrates the ReFibre pyrolysis – gasification process.

The recovered glass fibres with strength degradation can be used in making thermal resistance insulation materials, and use in making new turbine blades is not recommended. However, the process has not been commercialized mainly due to economic reasons, since the landfill of the EOL turbine blades is the cheaper option and is still permitted.

2.3.3. Chemical recycling

Chemical recycling involves the process for chemical depolymerisation or removal of the matrix by using chemical dissolution reagents for liberation of fibres. The chemical recycling process can re-generate both the clean fibres and fillers as well as depolymerised matrix in the form of monomers or petrochemical feedstock. The dissolution process is often called solvolysis, and depending on the solvent can be further classified as: hydrolysis (using water), glycolysis (glycols), and acid digestion (using acid). When using alcohol or water, high temperature and high pressure are normally used under either sub- or supercritical conditions to gain a faster dissolution and a higher efficiency. For using acid digestion, atmospheric conditions are normally applied but the reaction rate could be very slow [25].

The solvolytic processes such as glycolysis can decompose the epoxy resin into its original monomers to produce a potential chemical feedstock. Supercritical fluids (SCFs), and especially supercritical water (SCW) and supercritical alcohols are also potential media for the recycling of fibres and resin [4,5]. Using water or alcohol is environmentally relatively clean, and both could be separated from the dissolved solution by using evaporation (for water) and distillation (for alcohol). The process could be used for different types of reinforcement materials (carbon and glass fibres). The re-generated fibre retains most of its mechanical properties. For reaching a higher dissolution efficiency and a faster dissolution rate, an alkaline base is normally used as catalyst (e.g. NaOH and KOH). However, separation of the used catalyst salt from the recovered oil and purification of the depolymerised products, highly viscous oil, is still a challenge [4].

According to Henshaw et al. [7], the use of hydrolysis to dissolve polyurethane foam has been extensively investigated by General Motors in the 1970s. The foam could be hydrolysed to diamine, polyol and CO₂ under high pressure steam and high temperatures (232–316 °C). In the more recent research reported by Piñero-Hernanz et al. [4,5], supercritical fluids were tested at temperatures of 250–400 °C and a pressure of 4–27 MPa for water, and at temperatures of 300–450 °C and a pressure of 5–17 MPa for alcohols (methanol, ethanol, 1-propanol and acetone) to recycle carbon fibre reinforced composites. It was found that the use of an alkaline catalyst (e.g. KOH) gives a resin elimination efficiency of over 90% in supercritical water, and the recovered carbon fibre has only 2–10% degradation in the mechanical strength [4]. In the case of supercritical alcohol, a resin elimination efficiency of 98% was reported at 350 °C and the recovered carbon fibres retain 85–99% of the strength of the virgin fibres [5].

However, these recent tests were conducted on a very small scale in the labs, and the reactor was a 10 ml stainless autoclave. Much more research and development is needed to scale up the reactor system. Furthermore, the efficiency of the chemical dissolution process (solvolysis) depends on the types of the organic resins, and pre-separation of the types of composites is critical. Thus the process could be applicable to the production scrap composites where the characteristics of the scrap is well known, but can be very difficult to treat the post-consumer composite scrap where mixture of various composites prevails and mechanical separation could be inefficient.

2.4. Recycling of other composites

Although the dominant composite market is the polymer–matrix composites (in particular the thermoset type), other types of composite materials deserve also certain attention when recycling is considered. Due to the specific nature, ceramic–matrix composites in principle cannot be recycled unless the ceramic matrix is remelted at extremely high temperatures, while metal matrix composites and fibre–metal laminates can be well recycled. Below the recycling technology and issues of these two types of composite materials are discussed.

2.4.1. Recycling of metal–matrix composites

Metal–matrix composites (MMC), in particular aluminium alloy matrix composites as the dominating MMC, are used increasingly in higher volume applications, for example in the automotive industry as engine parts. Most of the commercial MMC products are reinforced with short fibres, whiskers, or particulates such as SiC, Al₂O₃, graphite, boron, boron carbide, and titanium carbide. The volume fraction of the reinforcement is usually less than 30% [26,27]. The alloys can be discontinuously reinforced with particles and whiskers, or continuously reinforced with fibres and filaments [14]. For more information about MMCs, please refer to Miracle [28], where all aspects of MMC are summarised from science to technological significance.

MMC materials normally have much higher economic values compared to the base alloys used, and thus recycling for direct reuse of MMC as its original form is the main cost driver and should be considered first (primary recycling). If that is not possible, in particular for continuous reinforced aluminium MMCs, recycling MMC back into aluminium or its alloy is performed and the separated reinforcement materials are usually disposed in landfill [14]. In most cases, discontinuous MMCs, e.g. SiC reinforced aluminium MMCs in the form of die-cast and foundry MMC scraps are remelted back to the new MMC for direct reuse, and the quality of the recycled MMC is only marginally degraded for new manufacturing scrap. Only a slight change in the tensile properties of composite was observed after several recycling steps. Repeated remelting will

lead to quality degradation of the MMC, and sometimes dilution with virgin MMC can solve the problem. When dirty or old scrap is used, it requires fluxing and degassing for cleaning, or only the matrix metal is recovered through melting similar to the recycling of continuous reinforced MMCs [14].

For continuously reinforced MMCs, normally only aluminium or its alloys are recovered through re-melting, and leaving the reinforcement materials as wastes for landfill. Effective separation of the matrix alloy from the reinforcement fibres or filaments is important [14]. To separate aluminium or the alloy properly from the reinforcements, a salt fluxing technique with mixture of NaCl+KCl with additional fluorides such as Na₂SiF₆ and NaF is commonly used, because of effective de-wetting of ceramic particles from aluminium matrix using molten salts [27].

Metal matrix composites are recycled by melting composite scrap in various types of furnaces such as induction furnaces, reverberatory melters, hearth furnaces, and rotary barrel furnaces, and cast into ingots [29]. The remelting and refining technique is very similar to the recycling of aluminium metal and aluminium alloys. Nishida [26,27] classifies the separation of aluminium metal from the reinforcement into two types of methods: mechanical method and chemical method. In mechanical method, the matrix metal in molten state can be squeezed out from the composite, or removing the reinforcement particulate by filtration. In chemical method, a molten flux as mentioned above is used to absorb and wet the reinforcement particles and separate them from the molten metal.

A new separation method is recently reported by Kamavaram et al. [29] to recover aluminium metal from the MMCs using electrorefining in ionic liquid. The electrolytic melt comprised of 1-butyl-3-methylimidazolium chloride (BMIC) and anhydrous AlCl₃. Aluminium metal matrix composite (Duralcan®, Al-380, 20 vol.% SiC) was electrochemically dissolved at the anode, and pure aluminium (>98%) was deposited on a copper cathode at 103 °C. Current densities were in the range of 200–500 A/m² and current efficiencies in the range of 70–90%. Energy consumption was in the range of 3.2–6.7 kWh/kg-Al, which is still too high compared to the remelting with flux or filtration, or to a conventional aluminium recycling process.

2.4.2. Recycling of fibre–metal laminates

Fibre metal laminates (FMLs) are multicomponent materials utilising metals, fibres and matrix resins. Typical FMLs are prepared by stacking alternating layers of metal foils and fibre/matrix resin prepreg followed by consolidation in a press or autoclave. FMLs consisting of aluminium sheets and aramid fibre/epoxy prepreg were first developed by Vogelsang et al. at Delft University of Technology together with ALCOA in the 1980s and are known as ARALL (Aramid Reinforced Aluminium Laminate). GLARE, GLASS REinforced FML, which replaces aramid fibres with glass fibres, was introduced in 1991 [30]. Fibre metal laminates combine the best properties of the metal and the composite making them suitable for aerospace applications. GLARE is already used in the fuselage of the Airbus A380, and is expected to find broader use in aerospace industry.

FMLs can be viewed as structural composite materials. Although the volume of production is limited and they are mainly applied in the aerospace industry, recycling of both the manufacturing scrap and EOL scrap becomes a relevant issue and should be seriously considered. There has been arguments whether GLARE should be recycled or not [31] regarding the relative low production value and high recycling cost. Landfill would still be an option, but will be most probably prohibited in the future. There will be no options to dispose in the future and recycling solutions must be investigated and should be available now. The challenge and criteria are that the recycling technology and process should bring much less

environmental impact compared with landfill, in particular on a relatively small scale.

2.4.2.1. Mechanical separation – cryogenic liberation and eddy current separation. Tempelman [32] was the first to study the recycling options for GLARE. Because of the very limited market value of epoxy resin and glass fibre, only the aluminium alloy was the objective to recover. Since generation of the EOL GLARE scrap is not earlier expected than around 2030, only the manufacturing scrap of GLARE was investigated. To recover the aluminium alloy, delamination is the first step. Tempelman developed a low temperature cryogenic liberation process to separate the aluminium foils from the epoxy resin and glass fibres, using the large difference of thermal expansion coefficients between aluminium ($2.4 \times 10^{-5}/\text{K}$) and glass fibres ($0.8 \times 10^{-5}/\text{K}$). In the process, the GLARE scrap will be first reduced into small pieces of typical 10 mm size in a granulator in the presence of liquid nitrogen at -196°C . Inside the granulator cryogenic liberation takes place. The mixture of liberated aluminium pieces and un-separated GLARE is then processed in an Eddy Current separator, where the liberated aluminium particles ($\sim 10\text{ mm}$ size) are separated from the resin, fibres and the un-liberated composites according to their differences in density and electrical conductivity. Reasonable liberation and separation efficiencies were obtained; however, different particle size requirements between cryogenic separation (granulation at maximum 10 mm) and eddy current separation (minimum 10 mm) have negative effects on the final separation and the scrap quality. From a commercialisation point of view, the cost of low temperature cryogenic liberation is high compared to the market value of the recovered aluminium scrap.

2.4.2.2. Thermal delamination. In contrast to low temperature cryogenic liberation, a high temperature liberation process has been studied at Delft University of Technology. Templeman [32] tried to delaminate the GLARE at 220°C to destroy the epoxy resin in an open furnace, but the delaminated aluminium was still covered with solid residues which required mechanical treatment such as sand blasting. Delamination in a fluidised bed reactor is another possibility. A recent investigation by using thermal delamination was conducted at the authors group at Delft University of Technology. It is proven that new GLARE scrap can be thermally delaminated at a temperature of 500°C in the presence of air, in a process that is very flexible with respect to the scrap size. After thermal delamination, relatively clean glass fibres and aluminium plates are generated. The glass fibre could be used for a lower level of applications, and the aluminium is remelted and refined in a secondary aluminium smelter in the presence of chloride salt (NaCl–KCl–cryolite mixture) and cast as ingot. Ideally, the aluminium alloy could be refined back to its original quality and could be used to make new GLARE. Research is under way with focus on refining of the aluminium alloy at Delft University of Technology.

The combustion of the epoxy resin ($\sim 10\text{ wt.}\%$, or $\sim 32\text{ vol.}\%$) provides more heat than required for the separation at the delamination temperature, and the excessive thermal energy could be used to supply heat to the melting and refining of the separated aluminium scrap in the case of an integrated recycling plant. The combustion products of epoxy resin (only CO_2 and H_2O) are considered to be non-hazardous if complete combustion is achieved.

3. Composite recycling in aerospace, automotive and wind energy industry

3.1. Composite materials in aerospace industry and recycling status

Composite structures have been developed and applied for military aircraft for over 50 years. Current commercial aircraft

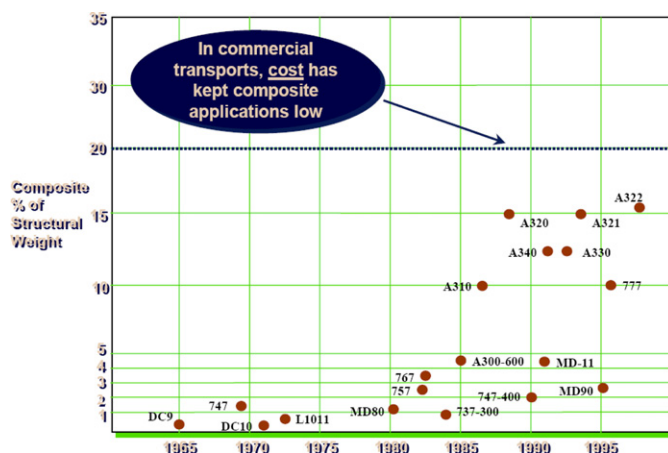


Fig. 7. Composite usage trends in commercial transports and general aviation aircrafts [33].

structures and applications are rapidly progressing from metallic parts and structures to composite parts and structures. The next generation aircraft is already entering the market by utilising large percentages of composite materials. Historically reserved for control surfaces and secondary structures, composites are now being employed for primary structures by the two largest and most famous commercial aircraft builders the Europe-based Airbus and the Boeing Company from the U.S.

The use of composite applications to replace metal alloys is mainly driven by the desire of airline operators to reduce operational costs by improving fuel economy. The drive to lower operational costs has led to the use of lighter and stronger composites. As an illustration GLARE offers between 15% and 30% weight savings over aluminium alloys. Fig. 7 shows an increased trend of using composite materials in commercial aircrafts [33]. Now the 20% cost limitation has been well broken by Airbus A380 and future A350 as well as Boeing 787. Table 2 summarises the increased use of composite materials in both Airbus and Boeing commercial aircrafts.

In general, carbon fibre manufactured waste composites are being disposed of through landfill. They are considered to be unrecyclable largely based on the inability of the industry to utilise the individual components that make up the composite materials, which places a burden on the substantial amounts of manufacturing waste produced each year. Also, retired aircraft are left parked in the desert mainly due to a variety of economic reasons, while the aircraft owners were largely unaware of the material value still represented in these airframes. Carbon fibres are targeted for reuse due to their economic value which can amount up to \$50 per pound. The financial incentive for the aircraft owner to leave a retired aircraft parked is due to the book value. When parked the aircraft still represents a book value of several millions even though the aircraft deteriorates over time making it un-airworthy. When the decision is made to scrap the aircraft the aircraft will lose as much as 75% of its value which makes it cheaper on an accounting basis to leave the aircraft parked in the desert than the option for scrapping and recycling. However, Boeing believed that aircraft could be recycled in a way that offered both economic advantages to operators and environmental benefits. Boeing's goal is to achieve 90–95% recyclability of the world's fleet by 2012 with the materials recovered in these recycled aircraft directed toward high-value commercial manufacturing applications [37]. Both commercial aircraft manufacturers Boeing and Airbus have been involved with research efforts into carbon fibre recycling over the past several years.

As a good reference, the EPSRC report [36] gives a comprehensive overview on The Aircraft at End of Life Sector, including the

Table 2
Overview on use of composite materials in Airbus and Boeing commercial aircrafts.

Aircraft model	Composites use (wt.%)	Main composite structures
Airbus [33–35] A300 A310 A320 A340 A380 A350	4.5 6 10 13 25 53	Rudder, radome and fairings Vertical tail fin, airbrakes, spoilers, elevators Entire tail structure, fairings, leading and trailing edges, bottom access panels etc Horizontal stabilizer, rear pressure bulkhead, keel beam, fixed leading edge on the wing GLARE in front fairing, upper fuselage shells, crown and side panels and the upper section of the forward and aft upper fuselage; carbon and glass fibre reinforced plastics in wings, fuselage sections, tail surfaces, and doors; composite honeycomb panels in the belly fairing Carbon composite wing, fuselage, skin, frame, keel beam and rear fuselage; complete horizontal and vertical tail plane etc.
Boeing [33,36,37] B777 B787	10 50	Full composite empennage, fairing, floor beams, wing trailing edge surfaces, gear doors, and the empennage-including the horizontal and vertical stabilizers, elevators, and rudder All composite fuselage and wing box, engine fan blades and casing

composite use and recycling issues. Both the efforts from AFRA and PAMELA are introduced, which will be discussed later in the paper.

3.1.1. Recycling efforts – Boeing

In 2006, Boeing [37,38] and 10 other aerospace companies formed the Aircraft Fleet Recycling Association (AFRA) with a common commitment to improve the way retired aircraft are managed [38]. AFRA's objectives include addressing the environmental concerns of retired aircraft and creating and sharing upgraded processes. AFRA is dedicated to the concept that end-of-service is not end-of-life. Its mission is to help airlines achieve the best return for their retired aircraft while promoting responsible recycling and developing safe and sustainable solutions for the reuse of aircraft parts and assemblies from older aircraft.

For the past several years, Boeing has been working with a number of third-party technology firms on the recycling of aerospace grade composites. Recent tests involved primary composite scrap material from the 777 and 787. This research indicated that the fibres could be recovered from the matrix, the recovered fibres are comparable to the virgin fibres in strength and bonding properties and are potentially suitable for high-end industrial applications. Research was also focused on using recycled 777 and 787 CFRP in high-end industrial manufacturing applications that include electronics casings using required radio frequency shielding and high-end automobile parts. Boeing also addressed another issue for recycling and disposal of carbon fibre composites coated with hexavalent chromium primer [39]. These composites are coated with hexavalent chromium and can be classified as hazardous waste and thus may/should not be disposed on land due to possible leaching of the chrome into the ground. This makes the recycling of such coated composite more challenging.

Boeing has started testing recycled carbon fibre in non-structural components of commercial airplanes and military

aircraft. The research has shown that the reclaimed fibres serve as a viable replacement for new fibre in many high-end industrial manufacturing processes, and offer a significant savings of money and carbon dioxide. Estimates by Boeing suggest that carbon fibre can be recycled at approximately 70% of the costs to produce virgin fibre. The costs for manufacturing are \$15–\$30 per pound (lb) with the amount of energy consumed during the process of 25–75 kWh/lb against \$8–\$12/lb and 1.3–4.5 kWh/lb for recycled carbon fibre [37].

Efforts by Materials Innovation Technology (MIT), Recycled Carbon Fibre Ltd (RCF) (<http://www.recycledcarbonfibre.com>) in conjunction with The Boeing Company and its many suppliers, have lead to the recovery and reuse of high value carbon fibres. Both utilise a pyrolytic process for fibre recovery from the matrix. This process strategy is enabled by the differential rates of oxidation of the rapid matrix and slow oxidation of the carbon fibres. The combination of recycled carbon fibres and virgin thermoplastic resins to manufacture injection moulded products was investigated. Aerospace carbon fibre prepreg material was used along with cured composites panels and thermoplastic resins used for compounding.

It has been shown that properties of the recycled carbon fibres recovered from the MIT process showed performance adequate for use in existing applications, given its equivalence or improvement to virgin fibre filled grades. The use of RCF recovered fibres in injection moulding compounds exhibited a marked decrease in nearly all properties except modulus. This is most likely due to the pyrolysis recovery process with damage occurring at the filament level and possibly during compound processing and moulding. The RCF made composites were still stronger than the native resins and offered improvements in stiffness over the virgin carbon fibres. The stiffness improvements for the pyrolysis recovered fibre over the virgin fibre is most likely due to the higher modulus of the aerospace-grade fibre which is largely unaffected by the recovery process [40].

It is the desire of the Boeing Company to promote the use of recovered carbon fibres in order to mitigate the disposal of carbon fibres and also to alleviate a market constraint on the carbon fibre market by making more fibre available for use in thermoplastic compounds.

3.1.2. Recycling efforts – Airbus

In 2005, Airbus established its process for advanced management of end-of-life aircraft (PAMELA) consortium with the objective to increase the amount of aircraft recycled material from the current 70–75% upwards to 90% in the coming years [41]. Similar to Boeing, Airbus used pyrolysis to extract carbon fibres from the composite material matrix in an effort to scale-up the process and to determine best practices for the recovery of the large volumes of composite materials being utilised on new generation aircraft which need to be treated in the future.

The practices as described above clearly indicate that current end-of-life (primary and secondary) waste material from aircraft is being treated through thermal treatment through the pyrolysis method. The aerospace industry is expected to generate a stable potential for 10–15 million pounds of carbon fibre recyclate, generated from both manufacturing and end-of-life in the short term. By 2029, it is estimated that there will be more than 50 million pounds of carbon fibre to be reclaimed [40].

The challenge in handling these volumes of waste material is being able to deal with the diverse nature of the feedstock such as prepreg, off cuts and end-of-life waste materials but also high modulus aerospace materials in combination with standard modulus scrap which all has to be treated in the same way. Furthermore there is the issue of consistency of the recyclates when dealing with different feedstock. Recycled Carbon Fibre Ltd processed carbon

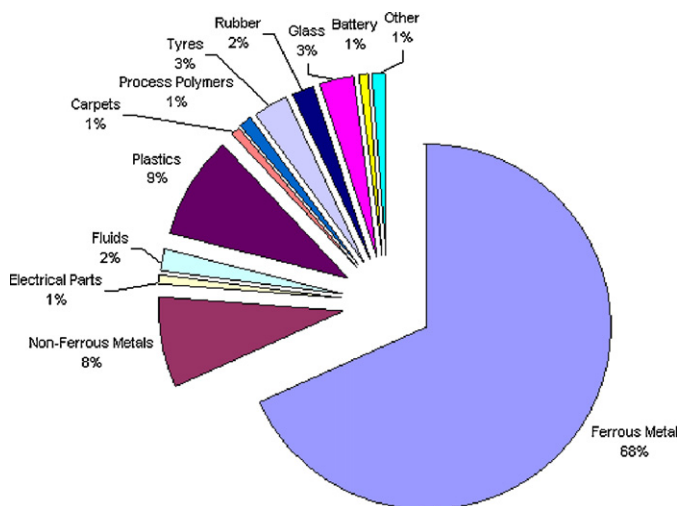


Fig. 8. Average material compositions of a vehicle produced in 2000. <http://www.wasteonline.org.uk/resources/InformationSheets/vehicle.htm>.

fibres from aerospace as well as the automotive racing (Formula One) industry both being high modulus fibres. The stiffness properties of these recycled products can, however, be too large to be used in industrial applications, which force the recycler to blend the feedstock to achieve constant properties [39].

The major current challenge is the establishment of a market for recyclates. Life cycle analysis has to be developed in order to determine the environmental, economic and technical advantages of recycled carbon fibres over other materials and with respect to disposal. First the potential market must be identified and product pricing established. This requires the determination of the characteristics and properties of different recycled carbon fibres, the assessment of their processing times costs and the establishment of the value for the recyclate [42].

3.2. Composite materials in automotive industry and recycling status

The automotive industry is one of the largest users of composites today and use of composites in newer models is increasing steadily. Composites have contributed to lowering the weight of an average passenger car by more than 200 kg and the potential for further savings is enormous. This weight saving translates into fuel economy. This becomes a huge environmental saving when multiplied by the 17 million cars that are built each year in Europe. The average plastics weight content of a vehicle in Europe today is around 120 kg, of which about 20% is composite material [43].

The automotive industry's use of structural composite materials began in the 1950s. Since those early days, it has been demonstrated that composites are lightweight, fatigue resistant and easily moulded to shape, a seemingly attractive alternative to metals. However, there has been no widespread switch from metals to composites in the automotive sector yet. This is because there are a number of technical issues relating to the use of composite materials that still need to be resolved including accurate material characterisation, manufacturing and joining [44].

Nowadays the polymer-matrix composites (PMC) are in competition with the existing metal components in the automobiles. Fig. 8 shows that the majority of materials in the personal cars are metallic (steel, aluminium, magnesium, copper), and that more than 3/4 of the car weight consists of metals [45]. Approximately 9% of the mass is plastics, but the use of the composites in the volume produced cars is very limited. Although the automotive industry has the highest share in using composite materials, they are usually low

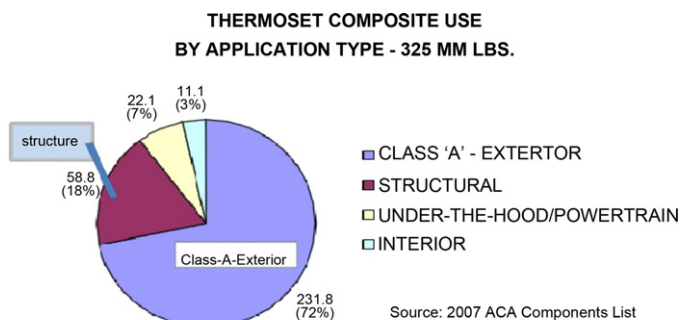


Fig. 9. 2007 Composites use by applications [46].

value composites in contrast to the high performance applications in aerospace industries. Furthermore, the proportion of composites applied in each individual vehicle is small.

The structural parts commonly made of composite materials in automobiles include composite modular front end, tail doors, side doors, and seating [46]. Fig. 9 and Fig. 10 show the use of thermoset composites in a standard automobile and the use of thermoset composites per OEM in 2007 [46].

Recycling of plastics from end-of-life vehicles (ELVs) turned to be difficult. The majority of plastic material from an ELV is reported to the auto shredder residue (ASR), from which it is very difficult to extract the plastics for recycling. Removal of plastic components from ELVs prior to shredding is labour-intensive and costly. Therefore only a small portion of plastics used in the cars is recycled such as bumpers, dashboard and battery casing. As can be deduced, recycling of composite materials from ELVs is much more difficult than plastics, and all composite materials used in the cars will end up with ASRs and goes to landfills or at most are incinerated.

Today, the biggest obstacle to the recycling of composite components is again the lack of end-users for the recycled material. The overall cost of recycled composite materials (e.g. reinforcements, or fillers produced by grinding) is considerably higher than their virgin equivalents. The quality and technical performance of the recycled reinforcement or filler are still inferior to the virgin materials. Consequently, there are currently virtually no automotive products that are manufactured predominantly from recycled composites [44].

To overcome the problems of recycling composites, new 'self-reinforced' materials have been developed. Natural fibre reinforcements (e.g. flax, hemp, coconut, abaca, basalt, animal hair, bird feathers, etc.) are also applied at an industrial level for cosmetic and semi-structural applications. Although a lot of development work is needed in this area, especially for applications where long fibre

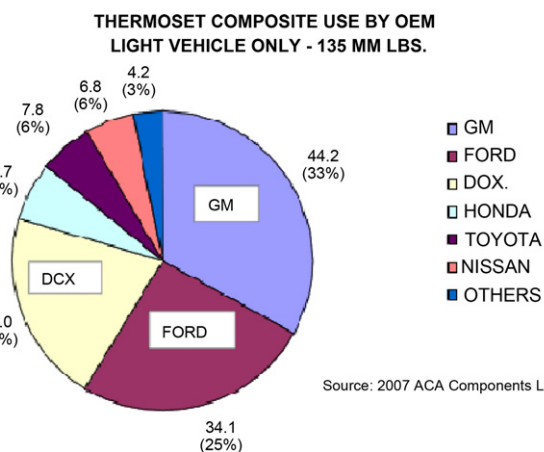


Fig. 10. 2007 composites use by OEM [46].

reinforcements are required, these materials seem to be promising from a recycling perspective. Although laboratory tests demonstrated that it is possible to grind and remelt short-fibre reinforced thermoplastics many times with little loss of structural performance. However 'on the road' degradation of these composites during the years of service life will cause the recycled composites much different from their virgin counterparts. For this reason, only a small amount of recycled material (10–20%) is added to virgin material for new components [44].

This again implies that the recycled composites or its fibre constituents may be more easily to find a new life in a less critical application. It is very important not to forget that both the reinforcement and the polymer matrix (both thermoplastics and thermosets) will experience degradation during the recycling process and during their previous service life for ELVs. Without using sufficiently large proportions of the virgin materials, neither the recycled composites self (such as fibre reinforced thermoplastics) or recycled fibre from thermoset matrix composites could be adequately re-used in making the new parts of automobiles.

Mangino et al. [44] emphasized the importance of 'design for recycling' practice. Careful consideration needs to be given to material selection and design for separation. Materials and components need to be classified in terms of re-use, energy recovery and recycling. Procedures and processes for dismantling and recycling need to be developed.

The increasing presence of multi-material hybrid components is a challenge for recycling that has not yet been resolved by car manufacturers. Currently, there are two trends – to shred the component or to dismantle it. It is essential that research is undertaken at a European level to investigate the management and recycling of hybrid material structures and components. It would also be worthwhile to combine the development of new recycling technologies and strategies with other industrial sectors with similar constraints [44].

There is a non-debatable fact that the increased use of strong and light-weight composite materials in the automobiles for higher fuel efficiency will face another problematic issue of recyclability demanded by the legal recycling targets, if no efficient and more appropriate recycling technology is developed in a short term. Proper processing of ASR for materials recovery and the development of reasonable market for the recyclates compatible with the recycling cost level are two practical solution perspectives for the automotive industry. Replacing gradually more thermoset matrix composites with thermoplastics matrix composites will ease greatly the recycling challenges.

3.3. Recycling of composite materials from wind turbine blades

3.3.1. Composite material in wind turbines and the recycling issue

Wind energy is one of the most direct accessible global green energy supply. Over the past ten years, global wind power capacity has continued to grow at an average cumulative rate of over 30%. In 2008 more than 27 GW new capacity has been added and the total installed capacity has reached 120 GW by end of 2008 [47].

The wind turbines are generally based on the Danish 3-blades rotor design, where the rotor blades represent approximately 4% of the total turbine construction weight in large-scale power generation wind mills of 1–3 MW. For a 1.65 MW Vestas V82 wind turbine, the rotor weighs 42.2 tonnes compared to the total construction weight of 1061.2 tonnes in which 16.8 tonnes consist of glass-epoxy composites, nearly 40% of the total blade mass [48]. Almost all turbine blades are made of glass fibre reinforced epoxy composites, with a fibre-matrix ratio of 60%. During manufacturing up to 10% of the prepreg materials turn into waste due to cutting, generating large amount of production waste composites. This would lead to a global generation of 1200 tonnes

non-recyclable manufacturing waste per year, according to the estimates by Papadakis [48].

Furthermore, the problem scale of recycling EOL wind turbines is even greater. In the case of a 1.65 MW turbine with three 40 m composite blades, a total of 18.6 tonnes of composite materials are used. With an average life time of 20 years, the total amount of waste composite materials arising from the wind energy industry will exceeds 1 million tonnes over the next 20 years [48]. Based on the moderate wind energy growth rate, an annual generation of EOL wind turbine composites could reach 300,000 tonnes in c.a. 20 years time (2008–2028) [48].

As the wind turbine manufacturing industry relies heavily on thermosetting composite materials for key turbine components – the rotor, recycling and disposal of new and EOL composite turbine scrap is becoming a pressing global issue. Until now, there are still no commercial operations to recycle the new and EOL composite materials for wind turbines [48].

3.3.2. Current industrial practice

At the moment, there are three possible routes for the manufacturing waste and dismantled wind turbine blades: landfill, incineration, and recycling [24]. The first option is largely on its way out in European Union countries because of the EU Landfill Directive (99/31/EC) [49]. Germany, for example, already introduced a landfill disposal ban on glass fibre reinforced plastics (GRP) in June 2005. The most common and practical route is incineration. In the combined heat and power (CHP) plants, the heat from incineration is used to create electricity, and to feed a district heating system. However, 60% of the scrap is left behind as ash after incineration because of high inorganic content of the composites. The incineration ash may be pollutant, and must be either land filled or recycled as a substitute construction material. The inorganic loads can also lead to the emission of hazardous flue gases and the small glass fibres in the flue gases cause damages to the flue gas cleaning devices [24,48]. Due to the limited efficiency of electricity conversion and the low heating value of the composites (c.a. 15 MJ/kg) used in the turbine blades (40–60% of glass fibre as inorganic materials), incineration will have only a short future. However, there are further developments in using the incineration residue (glass) as insulation materials or raw materials for cement kilns.

The best alternative is recycling – either material recycling, or product recycling in the form of re-use (e.g. in the market of the used turbines to the developing countries). At the moment, however, there are few established methods for the recycling of wind turbine blades, in particular due to the relative low market value of the glass fibres and high processing cost. Mechanical recycling technology is suitable more for the waste sheet moulding or bulk moulding compounds, but may be difficult for the laminated compounds used in the wind turbine blades [48]. A recent research from an EU consortium (REACT) found that the mechanically recovered fibres are difficult to bind with the new resin and longer fibres would be needed than virgin fibres. Mostly the mechanically recovered fibres are more suitable for lower end applications. There is also doubt that mechanical grinding and milling may consume a lot of electrical power, which could become a economic barrier for a feasible recyclates in the market [24]. Pyrolysis is perhaps the most suitable technology, as is practiced in the ReFibre's recycling process, described previously in this paper. As has been proven, the quality of the recycled glass fibres is degrades and it is suggested to retire as a thermal insulation materials instead of using for new turbine blades. However, as Larsen explained the there are needs from the government regulations to ban the landfill or even incineration before a real economically feasible pyrolysis comes into commercial operation [24].

With the recycling pressure for a rapidly increased capacity of wind power and the involved use of GFRP composites, as well as the

lack of legislation and proper market for the recyclates, new materials for making wind turbines are being developed. Thermoplastics (e.g. PET foam) is one of the options regarding the recyclability, but it is limited to very small wind turbines. Even a natural material of bamboo has been tried, but the applicability to large scale wind turbines is doubtful and a system approach is needed to reconsider the replacement materials [24]. It is clear and important that recyclability of the current GFRP turbine blades is a pressing issue and a driving force for the development of the more recyclable replacement materials.

It is expected in the near future that a similar legislation will be established for prohibiting the incineration of the composite materials from wind energy and transport sectors. All the parked retired aircraft, ELVs and its auto shredder residues (ASRs), as well as retired windmills, will have to find a real solution and more environmentally friendly disposal. However, as a message to the government bodies, the forced recovery and recycling should not bring more environmental burdens than simple landfill or incineration. A complete life cycle assessment (LCA) must be conducted and be used as criteria to implement any new technology.

4. Challenges for better recyclable composite materials

4.1. Challenges and opportunities

The concept of composite materials itself has already defies its recyclability by virtue. This is a paradox in our real life: superior mechanical properties can only be gained through complex structures such as coatings and composites, and this will in great conflict with the demands in good separability and recyclability! Problems must be addressed in two different levels for the whole materials society: recycling of existing composite materials, and development of new and better recyclable composite materials.

For the existing composite materials, how to find a low cost, efficient recycling technology to separate and recycle the current in-use composite materials. To reach this goal industrially and commercially, there are a number of boundary conditions and constraints.

- The available technologies to recycle fibres and fillers, and/or matrix materials.
- Availability of composite scrap versus economy of scale of recycling operation.
- The compatible quality of the recyclates with the existing composites markets.
- The environmental regulations on landfill and incineration of the composite materials.
- The overall cost and new environmental burdens in the recycling technology.
- Profitability and sustainable operation of recycling business.

At present, barriers exist at almost all above aspects, and the main issues to overcome as summarised by Pimenta et al. [9] are: *global strategy* with organised networks for all involved parties of scrap generators, composite users, recyclers and researchers; *incentives for recycling* supported by the government with penalties for scrap generators in landfill and credits for recyclers; *implementation of suitable legislation* for recycling technologies and reasonable quota for materials and energy recoveries similar to EU ELV Directive [2]; *logistics and cooperation for a continuous and steady scrap supply chain* with proper identification and pre-separation of the composite scrap; *market identification and product pricing*; *life cycle analysis* of the recycling process and the recycled products; and most importantly *the market establishment* which is the greatest challenge and requires all above issues to be solved simultaneously.

Development of new and better recyclable composite materials, which is the challenge for the materials scientists and manufacturing and application industry. Further development of thermoplastic–matrix composites should have a promising future. Furthermore, composite materials based on same or similar materials for both matrix and reinforcements will promote recyclability. Polymer–polymer composites could be a good example, but there is a long way to go to develop this composite family and find wide applications in the established composites markets.

Polymer–polymer composites are a new family of composite materials, where surface-modification of polymer particles enables them to be combined with and to bond to polymer systems with which they are normally incompatible [50]. By combining surface-modified polymer particles with various polymers, novel polymer–polymer composites are formed. Because of the several degrees of freedom that one has in creating these composites, this is a powerful approach for custom tailoring the properties of materials. Polymer–polymer composites have been developed to achieve combinations of physical properties that make them ideal for specific applications.

Through discussions within this paper, it is evident that recycling of composite materials designed and produced to date is very difficult and challenging, due to the inherent heterogeneous nature of composite materials. Recycling and reuse of the composite materials in its original form are only limited to certain clean manufacturing scrap of thermoplastic–matrix and metal–matrix composites, because of remelting and reshaping capabilities. For the majority of the composite materials based on thermosetting, recycling back to composite materials is impossible, and thus only constituent materials either reinforcement fibres and fillers, or the matrix resins could be recovered and used as raw materials for making new composite materials. In most cases, due to quality degradation of the recycled reinforcement fibres it is restricted to use them for making the same type of composite and applications and a lower quality composites could be made for less critical applications.

Practically speaking, the current developed technologies cannot deliver the same quality reinforcement fibres as the virgin fibres except for the chemical de-polymerisation process. Although carbon fibres experience less degradation compared to glass fibres through thermal recycling process (e.g. pyrolysis), reuse of the recycled carbon fibre for making the same type of composites as with virgin fibres is still a challenging task. Chemical recycling method is still in a research stage, far from commercialisation, but it is a recycling of composites back to their constituents or feed-stock materials. It is the authors' belief that only chemical recycling method can provide true materials recycling of composite, however care must be taken that much less environmental impacts are generated and the recycling cost must be well compensated by the market value of the re-generated reinforcement and matrix raw materials.

4.2. Industrial perspectives

In spite of many technological, social and environmental, and economical challenges, commercialisation efforts have been made constantly to recycle various types of composite materials by both independent recyclers and by the composite users such as automotive and aerospace industries. Due to the special technological difficulties for thermoset composites, the discussion will focus more on the commercial operations of recycling thermoset composite materials.

As has been mentioned a number of times in this paper, there has been no commercial operation in recycling of thermoset composite materials. However, this does not imply that commercial technologies are not available. The main barriers for commercialisation are

the lack of markets for the recyclates and the profitability. A historical overview and future prospects for commercial operation have been well described in recent articles by Pickering [12] and Pimenta et al. [9], and a short summary is given in this position paper.

4.2.1. Mechanical recycling technology

Two industrial scale development examples are ERCOM (Germany) and Phoenix Fibreglass (Canada) [12], utilising mechanical recycling technology of shredding – milling and classification. The ERCOM process uses a mobile shredder and a hammer mill. The Phoenix Fibreglass process uses two stage shredding and pulverising followed by products classification. However, ERCOM terminated and Phoenix Fibreglass has stopped operation in 1996 due to economic problems and lack of suitable markets for recyclates. Due to the relatively low values of the recycled products, mechanical recycling is mostly used for glass fibre reinforced polymers. The recycled short fibres are not sufficiently clean, and could be used mainly as fillers or reinforcement in new low-end composite manufacturing.

The European Composites Recycling Services Company (ECRC) developed mechanical recycling technology for automotive sector. ECRC was founded in 2003 by a number of key players in the composites industry, with its initial focus on solutions to meet the demands of the European Union (EU) Directive on end-of-life vehicles (ELV) [43]. Today, ECRC is on track to provide its members with a closed circuit of cost effective logistics, shredding and grinding (the mechanical recycling approach), and second generation outlets for the products they are now being required by original equipment manufacturers (OEMs) to take back after their useful life has ended. There are indications that there is interest from cement and gypsum users for the recovered fibres in the building industry for applications in floor and wall coverings, as a result of a high alkaline resistance resulting from their exposure to polyester resins. The reground aggregate is also proving to be competitive fillers for thermoplastic resins and could bring additional benefits in processing. The first commercial automotive application incorporating recycled SMC materials would be in production for a structural component in a reinforced polypropylene (PP) compound predicted at the report time (July 2008), but the status is not known now.

4.2.2. Pyrolysis recycling technology

Compared to other recycling methods, pyrolysis is the most realistic and practical recycling technology for both carbon and glass fibre reinforced polymer composites. Although matrix resin can also be recovered as secondary fuels or feedstock polymers (with smaller molecules), fibre recovery is the main driver and more practical goal to reach. The matrix resin is most practically decomposed as secondary fuel for use within the process or combusted further for generating electricity.

With pyrolysis recycling technology, 3 commercial operations were reported [9]: RFCL, JCMA, and MIT-RCF. The world's first commercial scale continuous recycled carbon fibre operation is made by Recycled Carbon Fibre Ltd. (RCFL) in the UK. According to Pimenta et al. [9], the process is implemented as a semi-open continuous belt furnace with controlled atmosphere to avoid char formation; it complies with all legislation on the treatment (post-combustion) of off-gases, and the resin's calorific value is recovered and fed back in the process *since material-recovery from the polymer is not economically viable*. The company has successfully reclaimed fibres from virtually all types of waste; the large dimensions and continuity of the furnace belt allow for entire out-of-date prepreg rolls to be recycled while maintaining the architecture of the reinforcement. The group recently launched Green Carbon Fibre Ltd. (GCF) for commercialisation of recycled products (e.g. milled and chopped fibres or pellets).

According to the News from REINFORCED Plastics.com [51] on 30 March 2010, The RCF Group or RCFL operates the world's first commercial scale continuous carbon fibre recycling plant from its site at Coseley in the West Midlands, UK. The site has the capacity to process 2000 tonnes of scrap carbon fibre composite each year. The Group's second plant will be based in the USA and it is expected to be operational in 2010/11. Until then, feedstock materials will be collected in the USA and processed at the UK facility.

4.2.3. Chemical recycling technology

Compared to mechanical and thermal processing technologies, chemical recycling technology has not yet been developed. Different chemical dissolution systems using various solvent have been recently studied in lab scale. Potential environmental issues (generation of toxic effluents, and use and disposal of alkaline catalysts) need to be resolved. Process scale up should be continued, and use of supercritical water shows a promising future due to its more environmental friendly nature. If the environmental and cost issue could be resolved, there will be a great potential in commercial application of the chemical recycling technology, since the recycled fibre will not encounter any degradation problems, and the de-polymerised resin could be made to new resin again. A real materials recycling for both reinforcement and matrix constituents could become true. Of course for the matrix resins it falls into the "Tertiary" recycling as feedstock material.

4.3. Multidisciplinary demand in knowledge and future need in R&D

To reach these ambitions and goals, multi-disciplinary knowledge is highly needed, and the joint efforts in further research and developments from materials design, material production, product design and recycling are indispensable!

For composite materials development, more easily recyclable composites are strongly needed. Although this is in contradiction with the required properties and performance by industrial applications, groundbreaking innovations are highly wanted. The manufacturing process for composites components and parts in various applications requires also drastic adaptations, aiming at reduced scrap rate and increased tolerance of more used of recycled reinforcement fibres and fillers. As far as recycling technology is concerned, there are high demands in developments of both mechanical separation technologies and thermal and/or chemical recycling processes, which offer high efficiency and high quality of liberated and recycled products without compromising environmental and economic constraints.

The following proposed topics cover 3 essential R&D challenges, and will be the key to the success for the development of truly recyclable composite materials and their true recycling from all aspects.

- (1) Recycling of composite materials and their constituents.
- (2) Product design and engineering for end-use properties including recyclability.
- (3) Maximization of product quality of each material use.

The first research topic focuses on the development of new and more efficient separation technologies (physical, chemical and thermal) for the recycling and recovery of the existing composite materials from EOL consumer products. The research and development will be centred about important industrial application sectors: automotive, aircraft, and wind turbines, where composites materials have been broadly used. A very strong university – industry cooperation is important and will facilitate the success to reach the final goals.

The second research topic focuses on the link between the composites design and manufacturing with the reusability and recyclability of the materials. Though the importance and benefits are obvious, this is the least developed area. This will be future important task for the material development and product manufacturing. For instance, a fibre re-enforced thermal plastics with similar chemical nature between the matrix and fibre will enhance greatly the recyclability of such composites. However, this is almost always a contradicting factor between the (end-use) properties and the recyclability. New and innovative concept is needed for the future development in order to meet simultaneously the end-use properties and the recyclability.

The third topic relates to the product quality of the recycled materials. The quality degradation, in particular the re-enforcement fibres, has been a common problem now from different types of recycling technologies. The lack of practical markets for the recyclates hinders the composites recycling seriously. This problem could be solved in both directions: (1) improving the quality of the recycled materials, (2) increasing the tolerance of the manufacturing process by using more recycled fibres, matrix or both.

5. Future perspectives

For the materials recyclability for automobiles, the bottleneck can be identified in the plastics and composite materials, and to a large extent this goes to the ASR. Without proper solutions to the recycling issue for the plastics and composite materials, more use of strong and light weight composites will be strongly limited. Another example is the increased use of composite materials in aerospace industry, as illustrated in future Airbus (A350) and Boeing (787 Dreamliner) commercial aircrafts. All these industrial development trends will give more incentives and raise higher demand for better and true materials recycling of composite materials. The first generation wind turbines are reaching their end of life. The turbine blades made of glass fibre reinforced plastics need immediate recycling. Materials recycling (at least for the glass fibres) issue deserves great attention and immediate action from both governmental (legislation and incentives) and industrial parties (manufactures and users or owners).

5.1. Vision 2015

Year 2015 is just 5 years ahead and we will not expect drastic changes in industrial operations. But the EU Directives for ELVs require 85% materials reuse and recycling or 95% reuse and recovery including a maximum 10% energy recovery, compared to 80% materials reuse and recycling since 2006. This implies that ASR which accounts for 25% of the car mass has to be recycled to a very large extent. Technology development and commercial operation in further processing ASR for materials recovery is under way, and an interesting example is the ASR processing plant operated by ARN in Tiel of Netherlands based on Volkswagen-SiCon process in the newly built PST (Post Shredder Technology) plant [52]. However, whether the 20% composites within the plastics fraction are recoverable is questionable within 5 years. Looking into the aerospace industry, there are no legislations yet for composites recycling like the automotive sector of EU ELV Directive [2]. The same is true for the EOL wind turbines.

The technologies are waiting for the legislation and better economics of the recycling process. At the same time the technologies are going through further developments, in particular for the increased quality of the recycled products and cost reduction of the recycling operation. The increased accumulation of EOL wind turbines and increased use of composites in automotive and

aeronautic industries will further draw attention from the society and government bodies to promote commercialisation of the composite recycling process.

Can the Beacon developed during the Delft Skyline Debate in December 2009 “A car out of waste” become true by 2015? Based on the current technology and industry status, it is not yet possible. This will not only depend on the recycling technologies of various engineering materials, but will also depend on the quality requirements and tolerance of a car design. Fig. 11 below illustrates the status of the recycling technology and industry and the challenging sides of the recycling.

Current recycling of metals and glass is far ahead the recycling of polymers and rubbers. Even now it should be possible to build a car with all recycled steel and non-ferrous metals (copper and aluminium), as well as the glass windows and lights. Since metals and glasses do not degrade in quality and properties during their service life, approximately 45% of steel is already produced with the recycled steel scrap. Almost no single steelmaking process works only with virgin raw ferrous materials (hot metal produced in the blast furnace with iron ore concentrates). Electric Arc Furnace (EAF) steelmaking even uses 100% recycled steel scrap to make new steel. Aluminium parts of 100 kg or more per car in Europe can all be made out of recycled aluminium or its alloys. Recycled copper and brass alloy can meet all quality requirements by the automobiles (10–35 kg/vehicle). As far as the glass is concerned, the glass window of a car in about 3% of the car weight can be made out of recycled glass without technical difficulty.

The situation is quite different for plastics and rubbers, and even in 5 years time there would be no dramatic change for the car parts made from the recycled plastics and rubbers. This will be greatly limited by the quality degradation of polymeric materials and rubbers, and hindered by the difficulty in general polymer recycling. However, there will be no doubt that new scrap of these types of materials could be used in making new car parts. Even recycling from bumper to bumper could be possible if the quality of the EOL plastic bumpers is still sufficiently good so that it can sustain and serve another life of a new car. Therefore, mixed use with virgin polymers would be much more realistic. Degradation of rubber is perhaps much more severe, and it is generally not recommended to used recycled rubber for the same function of car tires. In 5 years from now, there would be no big change of composite recycling technology, and any commercially recycled fibres in the near future will be most likely used in lower levels of non-structural applications with less critical quality requirements.

What will be the situation in 2030, 2050 and beyond?

5.2. Vision 2030 and beyond

What will happen after another 15 years time from 2015 then? We have now 20 years to develop technology, infrastructure and legislation. Simply from resource availability point of view, it will force us to recycle and recover not only the valuable reinforcement materials (carbon and glass fibres and a like) but also increasingly scarcer organic polymer matrix materials. It will be hard to imagine if the EOL wind turbine blades are still buried or burnt in incinerators, and composites car parts are being mixed with general plastics and discarded as the rest stream of waste.

By year 2030, the manufacturing process will become so efficient that much less manufacturing composites wastes will be generated. The product design can tolerate much more use of the recycled fibres. Legislation will promote contribution of all involved parties to the recycling efforts and cost so that the high recycling cost based on state of the art technology will be more than compensated by the market value of the recyclates. A much more healthy market tolerated by the manufacturing industry would be ready based on groundbreaking innovative design. A proper balance for

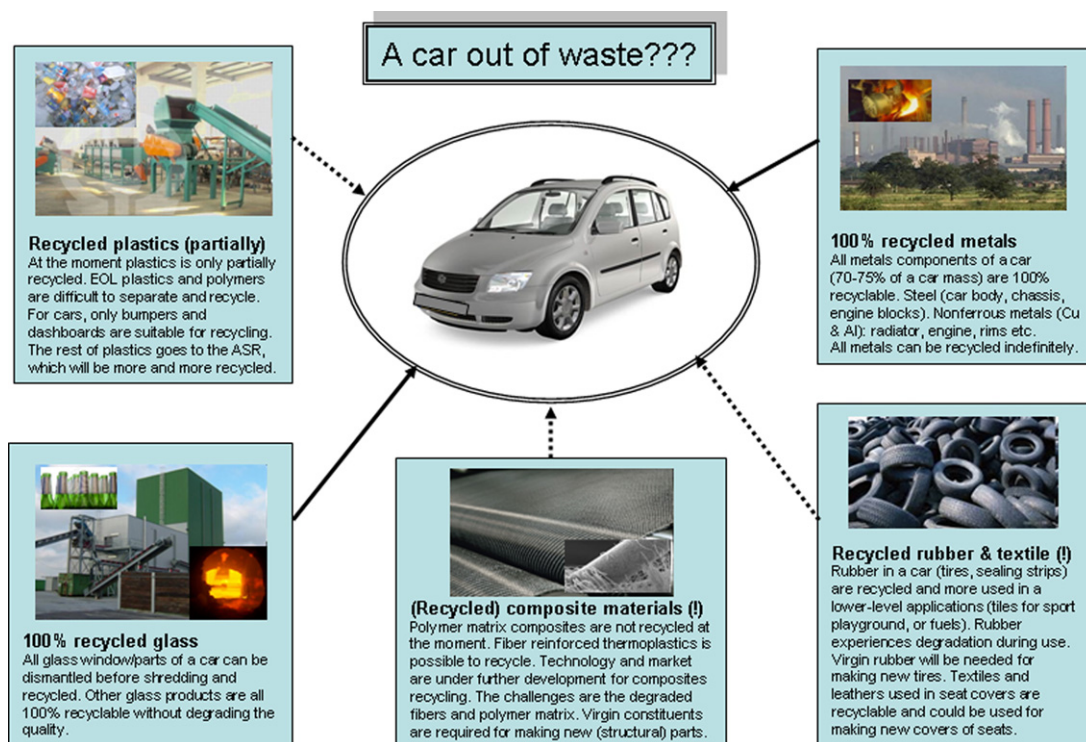


Fig. 11. Is a car made out of 100% recycled materials possible in the future: 2015, 2030, or 2050?

the production and use between virgin and recycled constituents of composite materials would have been established. The consequence of the success in the recycling of composite materials will be the direct benefits of increased use of this type of stronger and light weight materials in transport (automotive and aerospace) and other civil sectors.

Looking into a much longer future, by year 2050 the fundamental contradiction between the heterogeneity and recyclability would become less critical, by using newly developed reinforcement materials which will have much more similarity with the matrix materials in their chemical nature. The use of non-remelttable thermoset matrix will be significantly replaced by thermoplastics. Recycling technology will become much more mature to either separate the reinforcements from the matrix materials most likely based on clean chemical recycling technology such as super critical water, or recycle the (re-melttable) matrix together with reinforcements. The high cost of composite recycling will be compensated by legislation for forbidding the landfill and incineration of composites waste and EOL products, and by increasing production cost of virgin composite constituents (reinforcement fibres and polymer matrix).

With the constantly developed new expertise and knowledge and the joint efforts from all involved parties, we can turn many today's dreams in composite world to reality in the coming 30–40 years!

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