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Grand Challenge Paper

# Grand Challenges in Resource Recovery from Polymer Composites

Angela Daniela La Rosa*[1]

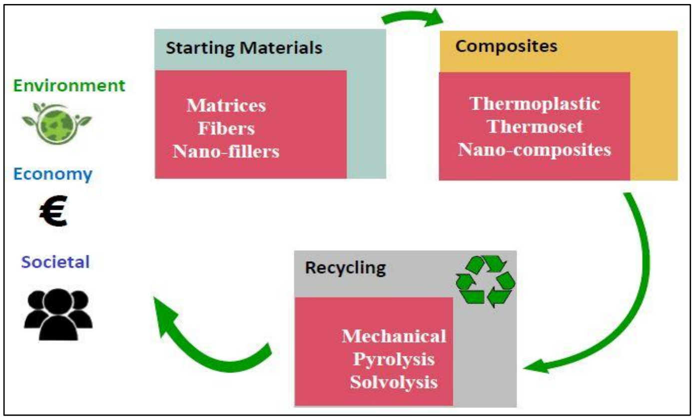
[1] *Department of Manufacturing and Civil Engineering, Norwegian University of Science and Technology, Teknologivegen 22, 2815, Gjøvik, Norway.*

## ABSTRACT

Due to their unique properties of light weight and high durability, composite materials have been widely used and their application is expanding into established sectors as well as new ones. As a result, PMC waste generation is on the rise, making the application of more sustainable end-of-life solutions urgent. Composites are tough, durable and inhomogeneous. For these reasons they are difficult to recycle and the commercialization of recycled waste materials is challenging. In the future, new strategies are needed to reduce waste, recycle as much as possible and reuse recovered materials, minimizing the fraction of waste that needs to be incinerated for energy recovery and completely avoiding disposal. The application of eco-design and circular economy principles and the use of tools such as life cycle assessment (LCA) and life cycle costing (LCC) will contribute to more environmentally friendly products and economically feasible.

**KEYWORDS**: Sustainable composites; recyclability, eco-design; circular economy; LCA.

## GRAPHICAL ABSTRACT



## HIGHLIGHTS

- Circularity of composites from raw materials demand to end of life management
- Development of green composites and nanocomposites
- LCA of composites for environmental impact assessment and sustainability

## 1. Introduction

### *1.1. About polymer composites*

Polymer matrix composites (PMCs) are an important class of structural materials used in many sectors (e.g.: aerospace, automotive, industrial applications, etc.) often competing with other structural materials, such as steel, aluminium alloys, titanium alloys, etc. Typically, PMCs are combinations of a polymer matrix with high-strength fibres that provides a product with a much higher modulus and strength than the polymer matrix itself, and therefore, it is often called a fibre-reinforced polymer (FRP). Figure 1 is a diagram of possible combinations of matrix and different types of reinforcements. The matrix can be either a thermoplastic polymer or a thermoset polymer, such as an epoxy and a polyester. Fibres commonly used as reinforcement are glass, carbon, or aramid.

PMCs have found much greater use than either metal matrix composites or ceramic matrix composites in aerospace, automotive, and other industries,

Correspondence to: Angela Daniela La Rosa.
*E-mail address: angela.d.l.rosa@ntnu.no*

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primarily because of their durability, relative ease of processing and lower density (depending on the fibre type and volume fraction, the density of PMCs is between 1.2 and 2 g/cm3). Because of such a low density, the strength-to-density ratios and, in many cases, modulus-to-density ratios of PMCs are comparatively higher than those of metals (Mallik, 2018).

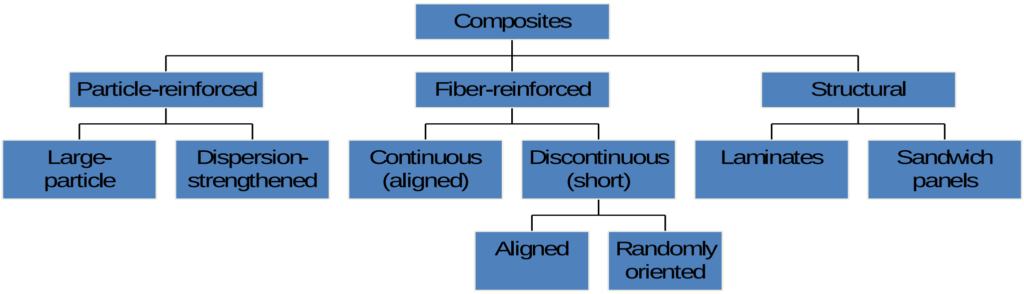

**Figure 1.** Types of reinforcement in polymer matrix composites.

### 1.2. Challenges

Due to their unique properties of lightweight and longer duration, composite materials have been widely used and their application is expanding in consolidated sectors as well as in new sectors. As results, PMC waste production is increasing and hence the need of more sustainable solutions for their end-of-life management. Composites are tough, durable, and non-homogeneous, which makes them intrinsically difficult to recycle and makes the commercialization of recycled waste materials a challenge. The main environmental issue related to polymer composites wastes is the increase of the impact on marine ecosystems and other categories e.g.: the depletion of fossil resources and the climate change. To contrast this huge environmental concern actions are to be taken especially in two main aspects:

1) Reduce the use of the raw materials that are at elevated risk of depletion (e.g., crude oil, copper, etc.) or prepare alternative for them.

2) All product parts should be designed and manufactured in a way that they can be recycled or reused, following the inspiration of the Ricoh's Comet Circle (Figure 2), shown in the figure below. Ricoh's objectives are to reduce the input of new resources by 87.5% by 2050 from the level of 2007.

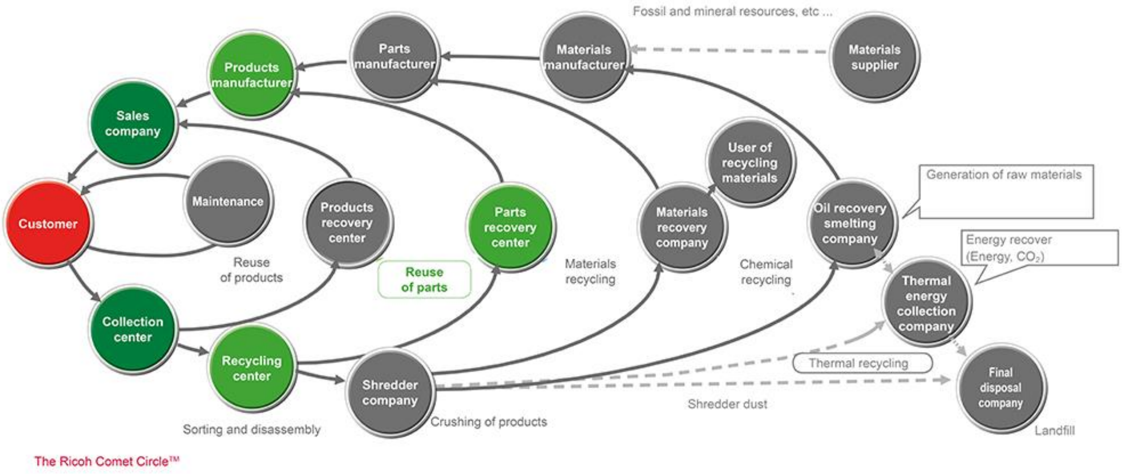

**Figure 2.** The Ricoh's Comet Circle (The comet circle, 2008).

Many challenges currently prevent a higher degree of circularity in the composites sector, and they are related to all the phases of PMCs life cycle: raw materials needed, manufacturing, use and end of life.

## 2. Polymer composites life cycle

### 2.1. Raw material demand

In Europe, it is estimated that 683,000 tons of composite waste will be generated in 2025 while the global annual FRP recycling capacity is estimated to be less than 100,000 tons (Mordor Intelligence, 2021). This constitutes a big challenge, but also a good business opportunity especially for FRP end-users, remanufacturers, and recyclers. The demand for new materials could be covered, in part, by secondary (reused, remanufactured, recycled) components and materials in agreement with the circularity goal that aims to minimize the extraction and use of virgin raw materials and energy resources. Another initiative that has been investigated in the last 2 decades is the development of alternative sustainable reinforcing fibers and matrices. Raw materials for composites are currently mostly based on finite fossil fuels. Most CFs are made of PAN (some small percentage is made of rayon or the petroleum pitch process), while about 80% of the polymers used to fabricate composites are obtained from non-renewable sources (Andrew and Dhakal, 2022).

*a) Natural fibres*

As sustainable reinforcement alternative, natural fibres have been tested due to their advantages over synthetic fibers in many impact categories and above all for the lower energy needed in the production step, that contributes also to lower greenhouse gas (GHG) emissions during their entire life cycle (Joshi et al., 2004; La Rosa et al., 2013). The use of natural fiber reinforced polymer composites is increasing worldwide in particular flax fiber. The applications of flax fiber composites have been found in automotive, maritime, sports and leisure industries. Besides that, there is a large potential to employ flax fiber composites in musical instruments, aerospace components, satellite structures, short wind turbine blades, and other engineering applications. Table 1 highlights some of the applications of flax fiber reinforced composites in different fields (Das et al., 2022).

**Table 1**

Some applications of flax fiber reinforced composites in different fields. Taken from Das et al. (2022) pag.226.

| Area | Components or Parts |
|---|---|
| Automotive | Door panels, Side panels, Floor dashboard, Seat backs, Trunk elements, Car bonnet, Head rests, Parcel shelves, Roof upholstery, Middle consoles, and other interior parts |
| Maritime | Small to medium sized boats, Boat hulls and canoes, Sailing yachts, Kayaks, etc. |
| Sports and leisure industry | Surfing, hockey sticks, tennis rackets, skis, baseball bats, canoe, kayak, snowboards, surfboard, stand-up paddle boards, most kind of boards, bow, bike or bicycle frames, racing car components (e.g., Tesla Model SP 100D), etc. |
| Musical instruments | Musical speakers, guitars & ukuleles, soundboard, top sheet of the guitar body, soundboard of violin, backside, etc. |
| Aerospace | Secondary parts, interiors, trunk load floors, interior fitting elements (i.e., partition panels, tray tables, baggage compartments) |
| Space applications | Satellite structures (development project under BComp and European Space Agency) |
| Wind energy | Wind turbine rotor blades (small to medium size) |
| Others | Consumer goods, furniture, panels, tubes, sandwich plates, building materials, tank, sound insulating panels, concrete reinforcement, or concrete structure for bridge pier, etc. |

*b) Green matrix*

Bio-based composites from natural fibres and bio-resins are receiving increasing attention. Depending on their chemical structure, bio-resins can be thermoplastic or thermoset, biodegradable or not. Thermoset bio-derived resins commercially available in the market are mainly epoxy resins, unsaturated polyester resins and polyurethane. These resins have a bio-content (derived from biomass) ranging between 20 and 50% and are not biodegradable. Among the thermoplastics, we can find in the market non-biodegradable bio-resins like bio-polyolefin, bio-nylons, cellulose and lignin-based polymers, and bio-degradable bio-resins like poly (lactic acid) PLA, poly (butylene succinate) PBS and polyhydroxyalkanoates (PHA). An interesting new class of liquid thermoplastic that could replace the thermoset matrix is also available. The advantage of using a liquid thermoplastic to replace the thermoset matrix is that the derivatives thermoplastic composites are easily recyclable compared to the thermoset composites.

*c) Nanocomposites*

A growing class of composite materials consists of polymers reinforced with nano-fillers. The reinforcement of nano-fillers (e.g., carbon nanotube, graphene, or nano-TiO2) is significantly better than micro- and macro-fillers due to the large degree of contact between nanofiller and polymer, known as the “nano-effect”, that may enhance the polymer properties (electrical conductivity, dielectric properties, mechanical properties, fire resistance, etc.). Many future potential applications are expected for nanocomposite with high filler content (> 50%) and interesting research activity is currently focused on this topic (Harito et al., 2019). Environmentally friendly nano-fillers (nanocellulose) have attracted increasing attention in recent years due to low cost, renewable precursor, and biodegradability (Lee et al., 2014; Chinga-Carrasco et al. 2011).

### 2.2. End of Life and Circular Economy

In the future, fully recyclable composites should be produced. The highest priority of the Circular Economy principle is to reduce waste production and, at the same time, minimize the extraction of raw materials as schematized in figure 3 (Composite Materials Report, 2022). The key points are:

i. Reduce waste, if necessary, through technology innovation.

ii. Develop ways of reusing production by-products directly or in collaboration with the material suppliers.

iii. Recycle as much as possible and reuse the recovered fibers in the own process to close the material life cycle. Collaborate with other markets for the re-use of high-performance materials in other sectors.

iv. Upcycling: moving resources back up the supply chain.

v. Minimize waste fraction that must be incinerated for energy recovery.

vi. Avoid disposal routes completely. Even though landfilling of composites is still permitted in some countries, avoiding landfilling and comparable treatment routes is a key goal.

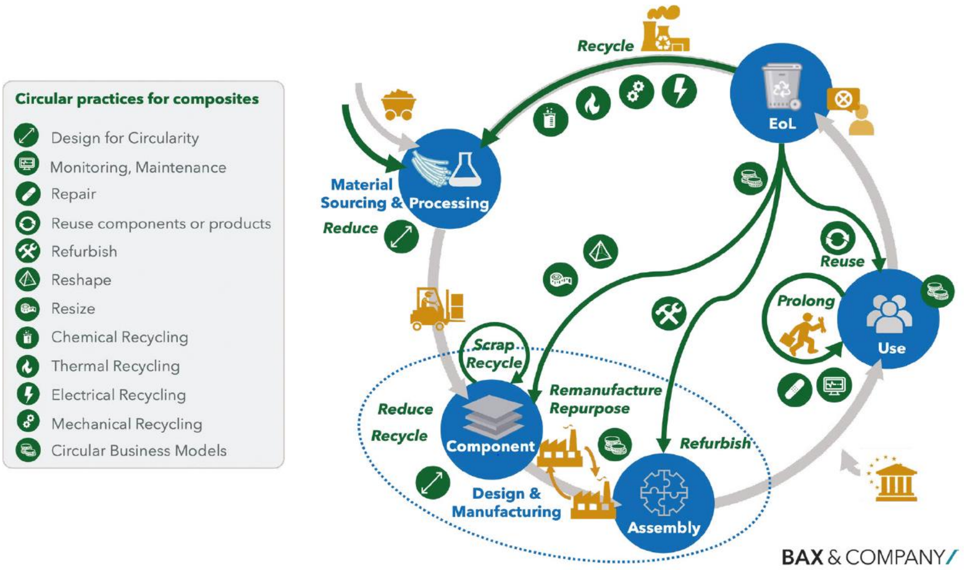

**Figure3.** Circular Economy principles in the FRP sector (source: Bax & Company).

*2.2.1. Recycling techniques for FRP waste*

Polymeric matrices are divided into thermoplastics and thermosets. Thermoplastics can be melted by heating and solidified by cooling. This property is of high interest for recycling because thermoplastics can be re-shaped. Thermoset materials instead, once polymerized cannot be re-melted or re-formed. The major limitation of thermoset matrices is their limited recyclability options. Once reacted, thermosets form a network which renders them un-processable. This limits the use of thermoset in those applications where recyclability is a must. In the automotive field, in EU nut also in USA, there is an increasing need for materials easy to recycle with lower impact on the environment. Pimenta and Pinho, (2011) discussed the different techniques for thermoset recycling providing some numbers on the process scale (Fig. 4).

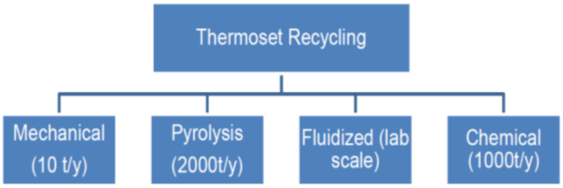

**Figure 4.** Thermoset recycling techniques with production rates accordingly to Pinho Pimenta and Pinho, 2011 and Cicala et al.2016.

Systems have been developed to recover carbon fibres from production waste and end-of-life components without significantly degrading the fibre properties. ne of recycling processes used to recover carbon fibre from composite waste is pyrolysis, where high heat burns off the resin. The high value of carbon fibre resulted in several recycling companies starting up around the world. An Italian example is Karborek, set up in 1999 with the aim of optimizing carbon recycling by means of a pyrolysis process. The fibres recovered by this process possess 70-90% of the mechanical properties of the original material and can therefore be reprocessed to produce composites like virgin carbon fibres. However, there are significant obstacles to the return of recycled fibre materials to the market (Composite Materials Report, 2022). The reason is that today's recycled products exist in forms suitable for use as compounds (ground and chopped fibres in powder or pellet form) and for layering (non-woven mats such as dry fabrics as well as prepregs and SMC materials), but the use of these products has been very limited so far. The recycling options for glass fibre composites are limited by the low value of glass fibre. The main reason is the insufficient knowledge of the mechanical properties and processing characteristics and the lack of large-scale applications that can demonstrate the economic, technical, and environmental justification for the use of these materials.

*2.2.2. Waste as resources*

The EC Waste Framework Directive (WFD), 2006, contains all the relevant points related to the waste issues in chapters1and 2: - The Waste Hierarchy (article 4) that applies the following priority order in waste prevention and management legislation and policy: (1) prevention; (2) preparing for re-use; (3) recycling; (4) other recovery (e.g., energy recovery); and finally (5) disposal.

- The By-products, article 5.

- The EoW (end of waste status) article 6.

- The ERP (extended producer responsibility) article 8.

These are the main crucial points for a definitive solution for the management of special waste so that it can be considered and used as a resource. They deserve to be adequately treated through a broader vision that focuses on environmental protection as a true incentive to develop its potential. A specialist vision in the environmental field, and a vision from entrepreneurs; a vision that creates new opportunities and that focuses as a priority on the protection of the territory (sea and dry land), because if we manage to save and even improve the environment, companies will gain advantages instead of further problems.

## 3. Polymer composites design for sustainability

To achieve a circular economic business model the product should be designed from the start for recyclability or reuse. Manufacturers should be encouraged to prioritize recyclability early in the product design process. A challenge in composite recycling is the transparency into the composites supply chain. It should be possible to know if companies are using virgin composite materials or composite recyclate (recycled composite materials), and what these materials contain to plan for how their scrap and products can be recycled or reused. A consistent raw material can produce a consistent product but if the raw material is a recyclate, that becomes more and more challenging because it may not be the same every time. It would be helpful if the industry developed a database where composite material suppliers could list their material contents. Then composite material users and their recycling partners would be better able to develop end-of-life solutions for composite parts and products.

### *3.1 Eco-design and Circular Economy by means of LCA and LCC*

Moving towards eco-design and circular economy, the life cycle assessment (LCA) and life cycle cost (LCC) have been recognized as the most useful tools for materials and processes selection, to obtain more environmentally friendly and economically feasible products, according to rigorous methodological framework described by ISO 14040 (2006) and by ISO 14044:2018 (2018) standards. Specifically, the LCA must include four main steps: 1) the study's goal and scope definition, 2) the inventory of all material and environmental flows among the life cycle of the product, 3) the environmental impacts calculation and 4) the interpretation of the results. Comparison of alternatives material choices can help decision makers to reduce environmental impacts and LCA is often employed to compare several product or design alternatives with the same function and/or to reveal the environmental "hot spots" of a product life cycle" and/or to achieve recyclability by design. An example of application of the LCA method to different end-of-life scenarios for composite materials is reported in figure 5 (Stergiou et al., 2022). The comparison of the impacts at end point categories shows that pyrolysis, fluidized bed and chemical recycling processes perform better due to the energy recovery, heat and electricity co-generation. This is also due to the greater extent of recovery of a possible reused product with a high content of embodied energy, compared to mechanical recycling routes.

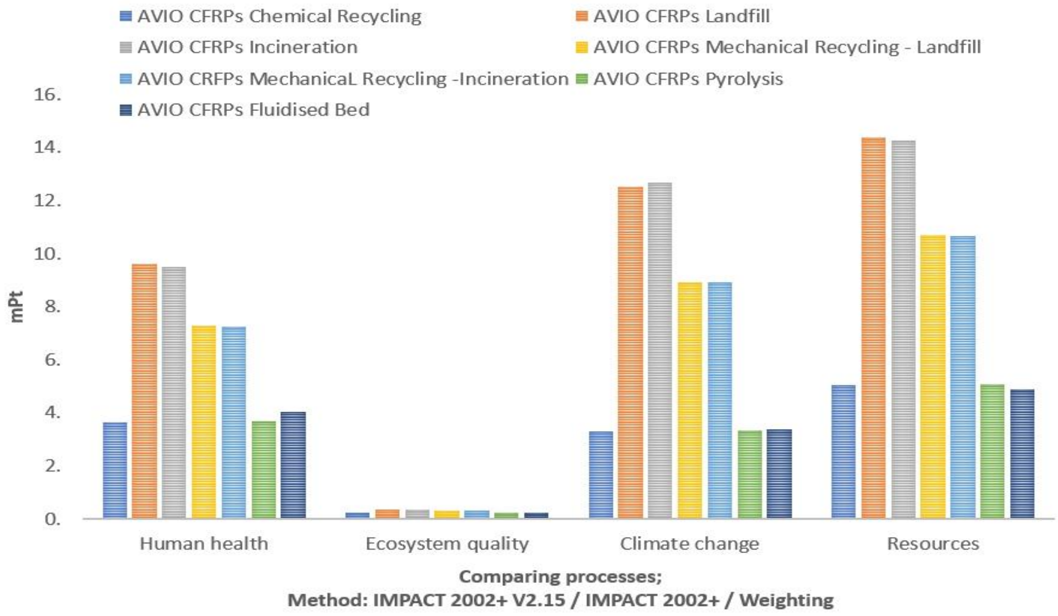

**Figure 5.** Results of overall comparison of recycling routes into endpoint categories. Taken from Stergiou et al., 2022.

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