Introduction, Context, and Motivations of a Circular Economy for Composite Materials

Marcello Colledani, Stefano Turri, Marco Diani, and Volker Mathes

Abstract Circular Economy is an emerging production-consumption paradigm showing the potential to recover and re-use functions and materials from post-use, end-of-life, products. Even if several barriers still exist at different levels, from legislation to customer acceptance, the transition to this sustainable industrial model has been demonstrated to potentially bring economic, environmental, and social benefits, at large scale. Composite materials, which usage is constantly increasing, are composed by a fiber reinforcement in a resin matrix. Among them, the most widely adopted are Glass Fiber Reinforced Plastics (GFRP) and Carbon Fiber Reinforced Plastics (CFRP). Their applications range from wind blades to automotive, construction, sporting equipment and furniture. The post-use treatment of composite-made products is still an open challenge. Today, they are either sent to landfill, where not banned, or incinerated. The application of Circular Economy principles may lead to the creation of new circular value-chains aiming at re-using functions and materials from post-use composite-made products in high value-added applications, thus increasing the sustainability of the composite industry as a whole.

Keywords Composites · Circular economy · GFRP · CFRP · Waste · Legislation · Circular value-chains

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1 Introduction

Glass fiber (GF) and carbon fiber (CF) reinforced polymer composites have revolutionized important manufacturing sectors, such as transport (automotive, aircraft, boats) and construction (building and infrastructures, plants, wind turbines), due to their lighter weight and intrinsically better corrosion resistance with respect to metals. The composite market by volume is dominated by glass fiber reinforced plastics (GFRP), with around one third of the whole production volume manufactured for the transport sector (cars, commercial vehicles, boats and to a lesser extent, aircraft), and another third for the construction industry (buildings, infrastructures, and wind turbines) [1]. Carbon fiber reinforced plastics (CFRP) are mainly adopted within the aerospace industry, with 32% of the total CFRP global demand, followed by the automotive industry which currently makes up around 21.8% of the total demand [1]. In the light of the above, the relevance of composite materials in many strategic industrial sectors appears significant. Therefore, the development of sustainable Circular Economy solutions to the post-use management of composite materials represents one of the key challenges of the modern manufacturing industry.

This chapter introduces the issues and the possibilities of the implementation of Circular Economy principles in this sector, starting with a general discussion about circular economy, introducing composite material features, the current market situation and the current composite waste treatments, with reference to existing legislation.

2 Circular Economy

In 1989 Pearce and Turner, introduced the concept of Circular Economy (CE) [2]. In their book they underlined the limitations of the traditional "take, make, dispose" approach, in view of the exploitation raw materials and the generation of increasingly relevant quantities of waste. They proposed the idea of a new closed-loop concept in which fractions from End-of-Life products can be reinserted as raw materials for new productions. The final goals are waste reduction and improvement of environmentally friendly processes, creating new adaptable and resilient systems.

This idea has been further enforced in the last years by the Ellen MacArthur Foundation [3], outlining the opportunities of Circular Economy: "*A circular economy is restorative and regenerative by design, and aims to keep products, components, and materials at their highest utility and value at all times. The concept distinguishes between technical and biological cycles. As envisioned by the originators, a circular economy is a continuous positive development cycle that preserves and enhances natural capital, optimizes resource yields, and minimizes system risks by managing finite stocks and renewable flows. It works effectively at every scale*" [3]. Value-chains should ensure not only environmental but also economic sustainability. The three fundamental pillars of Circular Economy have been defined as

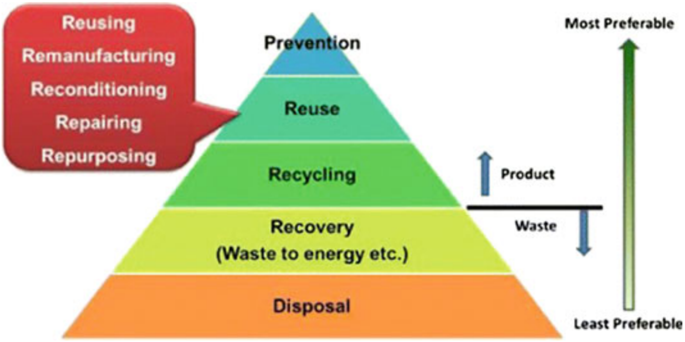

Fig. 1 Waste reduction strategies [4]

(i) preserve and enhance natural capital; (ii) optimize resource yields; (iii) foster systems effectiveness.

In practice, different Circular Economy options have been introduced to regain and re-use material fractions and component functions from post-use products, i.e. reaching the end of the first use cycles. The European Waste Framework Directive 2008/98/EC, introduced these options as the five-step hierarchy shown in Fig. 1 [4]. Solutions in the upper parts of the pyramid, which consider End-of-Life products as a resource, are preferable than those in the lower part. Prevention is considered as the most preferrable option, while disposal should be avoided whenever possible. The study identifies the role of legislation and policies in promoting and prioritizing the adoption of the most attractive Circular Economy options.

3 Current Circular Economy Solutions for Composite Materials

3.1 The Composite Material Market

Composite materials are a wide family of heterogeneous materials composed by two or more phases, with different physical properties. The resulting properties of composite materials are better than those of the constituting phases. The reinforcements could be in the form of short fibers, long fibers, filler, particulate and flakes, leading to different behaviors depending on the shape (see Fig. 2 [5]). Some examples of composite materials could be seen in nature. Wood is composed by cellulose fibers in a lignin matrix. The result is a flexible material with high elastic module. In the same way, bones are composed by collagen fibers in an apatite phase, leading to a material with low specific gravity and high mechanical properties.

Focusing on artificial composites, also called fiber-reinforced plastics, it is a wide group of materials mainly composed by fibers, resin matrix, additives and fillers.

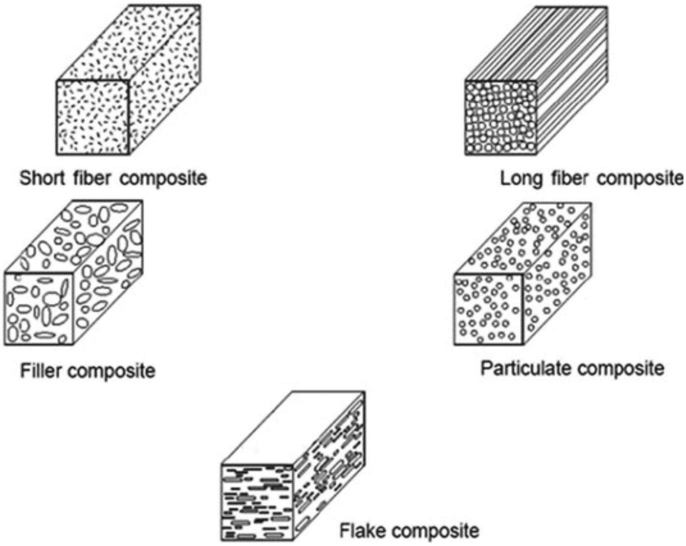

Fig. 2 Different fibers/matrix configuration in composite materials [5]

The two-phase composition results in high mechanical properties together with low weight, high corrosion resistance and, as a consequence, low maintenance needs. Different types of fibers are used in composite materials. More than 99% of the products currently on the market are composed by glass fibers, carbon fibers, natural fibers, aramid fibers and basalt fibers. Among them, glass fibers show the largest adoption rate, with 95% share of the worldwide production volume of composites [1]. This is due to the good mechanical properties that they bring, combined with their reasonable market price. For these reasons Glass Fibers Reinforced Plastics use is widespread in several sectors as wind blades, sports equipment, construction, marine and automotive (and transportation in general) and electronics (for E-glass fibers), as reported in Fig. 3 [1].

The second largest share of composite materials includes Carbon Fiber Reinforced Plastics. Carbon fibers present higher mechanical properties with respect to glass fibers, with higher cost (from 4 to 40 times the price of glass fibers depending on the specific application [6]). Due to their remarkable properties, CFRPs find application in high added-value sectors such as wind energy, sports equipment, sports car, construction, aircraft and aerospace [1].

In addition to the constituent material, fibers could be classified by their length. In particular, they could be short fibers, long fibers and endless fibers. Short fibers have a maximum length of 2 mm, long fibers are between 2 and 50 mm, while endless fibers are longer than 50 mm. The GFRPs market share shows a predominance of short fibers with respect to long and endless ones. Indeed, the European production volume of short fibers in 2021 has been of 1.51 million tons, while that of long and endless fibers has been of 1.1 million tons [1]. Even more, fibers are not only used

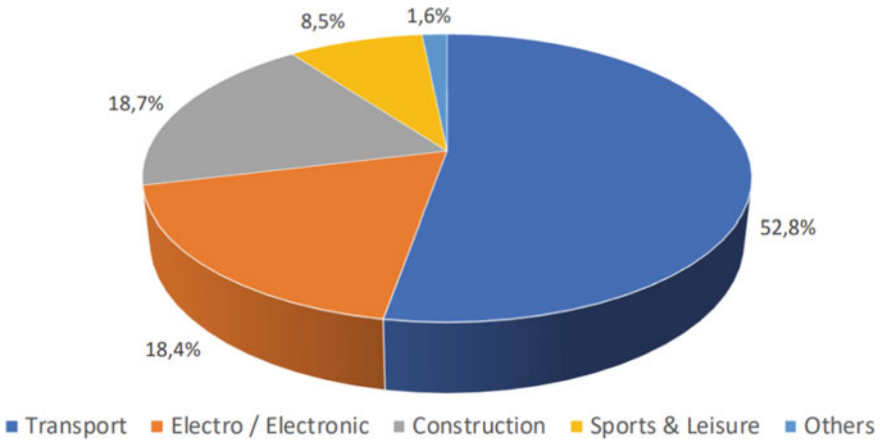

Fig. 3 GFRPs production in Europe by application industry [1]

as single chopped fiber or strand but also as semi-finished product as mats, textiles fabrics, knitted fabrics, non-woven and fiber pre-forms depending on the specific application.

Regarding the matrix, a spread variety of resins could be used. In the first analysis, they can be divided in thermosets, that could not be melted and reshaped once hardened, and thermoplastics, that can be reshaped several times through temperature. In particular, 86% of composite products made with long and endless fibers have a thermoset matrix [1]. Among thermosets plastics, the most used are unsaturated polyester, vinylester, epoxy and polyurethane, while regarding thermoplastics the most used in composites are polypropylene, polyamide, polystyrene, polyphenylene sulfide and polyetherether-ketone (also known as PEEK).

Finally, several fillers could be used to give specific characteristics to the final composite products. They are inert materials of mineral nature as calcium carbonate, frequently used as coating, aluminum trioxide and silica, as in piping and in polymer concrete.

In 2021 the production volume of CFRPs was approximately 52.000 tons, natural fibers reinforced plastics had a volume of more than 92.000 tons, while the volume of GFRPs was 2.910.000 tons. Aramid fibers and basalt fibers, with other types of fibers, have very low production volumes due to their very specialized applications. While the worldwide composite growth rate has been of about 8% in the last years, European composites production volume increased by 18.3%, returning to the pre-pandemic level [1]. In Fig. 4, the situation of composite production shares and volumes in Europe is reported.

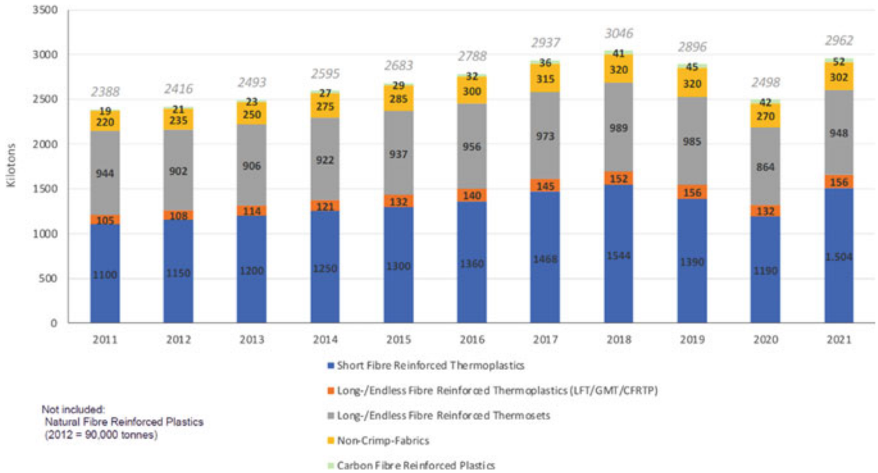

Fig. 4 Composites production volume in Europe in ktons excluding natural fibers [1]

3.2 Composite Waste Current Situation

Composite waste streams can be mainly divided into three groups. The first is composed by small components as consumables, sport and leisure equipment and design products, which are not systematically separately collected on a national or European level. Sorting of composite materials from mixed municipal waste streams is not performed due to technical and economic issues. This results in a high loss of composite materials and, as a consequence, of economic value.

The second group is composed by large infrastructures as wind blades, aircrafts, boats, construction structures and vehicles. The current situation and related opportunities of these groups will be better detailed in Chap. 2. As an example, the amount of End-of-Life wind blades, composed by GFRP, CFRP, steel and aluminum (in addition to other minor materials) is predicted to reach 30.000 tons per year in Europe by 2026, while it will increase up to 50.000 tons by 2030, as shown in Fig. 5 [7]. EoL leisure boats were also identified as significant GFRP waste stream with an estimated volume of up to 10.000 tons per year [8].

The third group of composite waste streams is formed by composite production waste. The collection and sorting of these materials is less problematic with respect to End-of-Life products. Moreover, in this case, the materials composition and the location are known. However, challenges are represented by the classification of these streams as "waste" and the related management challenges, as discussed in Sect. 4.

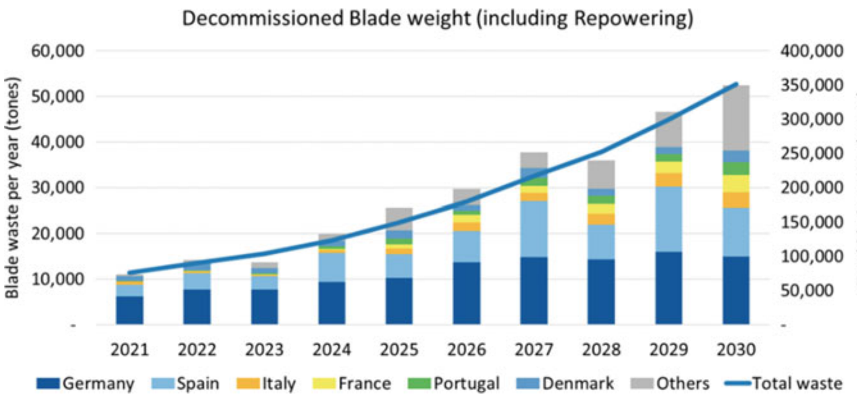

Fig. 5 Expected amount of End-of-Life wind blades in tons per year in different European Countries [7]

3.3 Recycling of Composite Materials

Recycling is the most widely investigated solution for the sustainable treatment of post-use composite products. Existing technologies and recycling methods are of three main types, namely mechanical, thermal or chemical [9]. In this section, a brief overview of the currently investigated solutions is reported.

Mechanical recycling: Mechanical recycling typically consists in one or more size-reduction stages (or shredding stages, also preceded by a coarse cutting step, if needed) to obtain particles, that remain composed of mixed fibers and resin, featuring desired dimensional distributions. The objective is to directly reuse the obtained fraction into new composite material formulations. To facilitate downstream reprocessing stages, the material flow in output from the size-reduction process can be sieved and divided into two or more fractions, with controlled characteristics [9]. The composite shredding powder residue instead can be used both as filler and as reinforcement. While the use of this powder as filler is possible in terms of physical and chemical properties, the costs are generally higher if compared to the costs of alternative virgin fractions, such as calcium carbonate or silica. In addition, in most applications, the weight fraction of composite powders that can be incorporated as a filler is limited, typically less than 10% [10]. This is mainly due to an increase in the viscosity of the compound, resulting in extended processing problems. For these reasons, powder fractions are usually incinerated to obtain energy due to their high content in resin. The possibility to use these materials for additive manufacturing has also been discussed and preliminarily demonstrated [11].

Concerning the reuse of granular fractions composed by fibers embedded within the resin, different studies have been performed both for thermosets and thermoplastics composite materials. As an example, up to 50% in weight of carbon fiber particles obtained through shredding have been successfully incorporated into new products with virgin PEEK resin through injection molding [12]. In addition, thermoplastic polymers could be subjected directly to reforming processes. Fiber-rich fractions of mechanically recycled thermoset composites are usually more complex to be reused as reinforcement, since the mechanical properties of the resulting materials are reduced due to poor bonding between recycled particles and the new resin [10].

Several works dealt with these issues finding innovative and promising solutions to use increasingly large fractions of recyclates, in particular focusing on Sheet Molding Compound (SMC) and Bulk Molding Compound (BMC) production techniques. As an example, it has been proven that longer mixing time of the paste with the recycled particles results in improved interface between recycled material and new resin. This leads to increased mechanical properties that are similar to those of the material obtained with virgin fibers [13].

In addition to the classical mechanical recycling processes, the possibility and potentiality to use an innovative process based on high-voltage fragmentation has been shown. The product is placed in a dielectric ambient as de-ionized water and a high-intensity and fast growing voltage (80–200 kV, <500 ns) is induced. As a consequence, a high electric field is generated, resulting in a dielectric breakdown and in the generation of a spark plasma channel with high pressure (10^{10} Pa) and temperature (10^4 K). After up to 1.000 discharge cycles the product is disintegrated. Working directly on the interfaces between different materials, this technology is able to clean the fibers from resin. Some works showed that applying this method to GFRPs it is possible to obtain lower amount of residual resins in comparison with comminution and a wider fibers length distribution [14]. The main disadvantages of this process remain the high cost and the low throughput [15].

Thermal recycling: Thermal processes used for composite materials recycling are pyrolysis, fluidized-bed pyrolysis and microwaves-assisted pyrolysis. These techniques work between 450 and 700 °C (depending on the resin), eliminating the matrix in controlled atmosphere. They are optimized to recover fibers and fillers but the resin is volatilized into lower-weight molecules, producing waste gases as carbon dioxide, hydrogen and methane and possible combustion residues on the fibers [9]. Exceeding the specific resin temperature, the process will concern combustion with energy recovery. This is typical in cement kilns in which the composite waste is converted into energy and fillers and fibers for the concrete.

Pyrolysis is a well-known process in which the temperature is higher than 450 °C in absence or in presence of oxygen, in a controlled atmosphere [16], to degrade the resin matrix. The output will be composed by solid products (in particular fibers, fillers and char), oil and gases. The char is stuck on the fibers, that need a post-treatment in a furnace at 450 °C, leading to more degraded fibers [17]. This effect is visible in particular for glass fibers. As an example, at the minimum temperature of 450 °C, mechanical properties of glass fibers are reduced by at least 50% of the original ones. In addition, as pyrolysis is an energy intensive and expensive process, the cost to recover glass fibers is higher than the market price of the virgin ones. For these reasons, pyrolysis process is mainly used to treat CFRPs. Furthermore, they are less sensitive to high temperatures, while char is still present on the surface of the fibers. To eliminate it in standard conditions, a post-treatment process with temperature up to 1300 °C is needed, resulting in significantly loss of strength of the fibers [18]. As a consequence, a compromise between resulting mechanical properties and the amount of remaining resin residues is needed. As an example, high tenacity carbon fibers after a two-steps process, the first one at 550 °C in nitrogen the second one at the same temperature in oxidant atmosphere, retained more than 95% of their tensile strength without resin residue on the surface [18]. A pyrolysis temperature between 500 and 550 °C seems to be the upper limit of the process in order to maintain acceptable strength of carbon fibers.

Fluidized-bed process pyrolysis uses a bed (e.g. composed by silica sand) made fluid by hot air, obtaining in this way oxidant conditions. In this way, it is possible to rapidly heat the materials and to release the fibers from the resin by friction. The organic fraction of the resin is then degraded through a second step at 1.000 °C for energy recovery [10]. Glass fibers treated with this process showed a reduction in tensile strength up to 50%, while carbon fibers of about 25% [19], probably due to damages from the fluidized sand. The added value of this process is the possibility to treat mixed and contaminated materials as products with painted surfaces or foam cores.

Microwave-assisted pyrolysis heats composite materials in an inert atmosphere, resulting in degradation of the matrix into gases and oil as for traditional pyrolysis. The main advantage is the energy savings due to the fast thermal transfer as the composites are heated directly in their core through microwaves [20]. This process has been used to treat both GFRPs and CFRPs. On the other hand, this process needs small particles in input. This results in an output composed by a wide length distribution of the fibers and in a reduction of resulting mechanical properties of the final product, with a limited percentage of recycled material that can be used (less than 25%) [21].

Chemical recycling: Chemical recycling of composite materials is performed through solvolysis, a chemical treatment to degrade a resin using a solvent. This technique has been widely used in the last 20 years, starting from the degradation of polyurethane into its monomers, carboxylic acids and glycols, and a styrene-fumaric acid copolymer [22]. Since this first positive attempt, several processes based on different solvents and techniques have been tried to recycle both thermoplastics and thermosets composite materials [9].

Solvolysis factors include the solvent, the working temperature, the pressure and the presence of a catalyst, depending on the resin to degrade. The reactive solvent diffuses into the composite and breaks specific bonds. In this way, it is possible to recover monomers from the resin, avoiding the formation of char residues.

To start the process, the activation energy of the polymer has to be reached. This could be achieved increasing the temperature or using a catalyst. The main advantage is that lower temperatures are required with respect to thermal treatment. On the other hand, reactors could be expensive, in particular for solvolysis in supercritical conditions (that is the process with best results), as they have to withstand high temperatures and pressures and corrosion due to modified properties of the solvents

[23]. As an example, polyester resins are easier to degrade through solvolysis than epoxy resins, leading to lower required temperatures.

The most used solvent is water, both alone and with co-solvent (e.g. alcoholic, phenolic, amine). Typical catalysts are alkaline catalysts as sodium hydroxide and potassium hydroxide or, less frequently, acidic catalysts (used in case of more resistant resins or lower temperatures). Other solvents are alcohols as methanol, ethanol, propanol and acetone or even glycols [9].

3.4 Existing Barriers

Despite the mature development phase of some of the reported technologies, key barriers exist bounding the systematic transition of the composite industry to a Circular Economy approach, including:

- *Governance*: significant fragmentation of stakeholders in different sectors, leading to poor-communication and inter-sectorial alignment of policies, thereby creating barriers to innovation; low priority of composite waste in the political agenda, thereby limiting stakeholder engagement; incomplete understanding of barriers to innovation, thereby limiting leadership and formation of an effective integrated innovation strategy;
- *End-users perception*: industry perception of composite waste as a cost to be minimized rather than a resource to be valorized;
- *Commercial uptake*: negative consumer and industry perception of products derived from recyclates and safety concerns;
- *Finance and regulation*: poor mobilization and alignment of finance across the sector; poor alignment of pricing methodologies and legislation, lack of suitable business models that support EoL composite valorization;
- *Technology development*: limited synergistic use of available inspection, repair and reprocessing technologies due to the different specialization areas.

4 Current Composite Waste Management Practices and Legislation

The following paragraphs provide a brief overview of the EU legislations concerning composite waste treatment and waste management.

End-of-Life GFRP and CFRP products are usually not systematically collected separately on a national or European level. Furthermore, sorting of composites from commercial or municipal mixed waste streams is not practiced due to technical challenges and economic considerations. Compared to other materials, the proportion of composites in residual waste as well as the commercial value of mixed GFRP and

CFRP waste are both relatively low, making manual or automated sorting unprofitable in practice.

As a result, once composites have entered a mixed or residual waste stream, they are not sent to recycling processes, regardless of the further treatment of the entire waste stream (as landfill, incineration or co-processing). This applies, in particular, to small scale consumption products, as sports equipment, consumer electronics or design objects, which are generally disposed by consumers via municipal residual waste.

Compared to small consumption products, EoL composite waste streams derived from large-scale consumption products, as leisure boats and vehicles, or large installations, as wind energy systems and construction waste, are significantly more attractive for centralized recycling approaches.

Despite the large amount of potential EoL-waste from these sectors, an estimation of the annually generated amount of waste is quite difficult and data about presence and treatment in different European countries are rare. Additionally, vehicles, as airplanes, cars, trucks and boats, as well as wind energy systems are assumed to have a relatively long lifecycle. Although the actual production capacities of these sectors are known quite well, the exact time of decommissioning is hard to predict. Furthermore, some products as vehicles underlie a massive exportation after their standard life-time, losing the possibility to be recycled and reused.

Contrary to composites EoL products, production waste appears as more reliable source of high-quality materials suitable for reuse and recycling processes. Their collection produces non-mixed waste fractions with well-known material properties and compositions. In addition, the amount of waste is constant or predictable and in a limited number of locations. Finally, databases elaborated for the estimation of current and future waste streams are easily accessible and precise.

Today, approximately 30.000 tons of post-consumer GFRP and CFRP-waste are produced annually in Europe. Additionally, 40–50.000 tons of commercial production waste are generated [24]. Due to an increasing use of composites in various applications, a steady increase of these amounts is expected.

Compared to other waste streams, the total amount of composite waste produced within Europe is relatively small. Therefore, this could lead to the assumption that an international collection and a centralized recycling could be economically beneficial. However, for the transportation of waste across borders—also within the European Union—some legal aspects must be considered. According to the European Waste Framework directive, establishments and undertakings which carry out waste treatment must obtain a permission from their corresponding competent authority. Within the EU, the import, export and transit of waste to, from or through EU Countries are subjected to approval procedures (called notifications), which are regulated in detail in the European Waste Shipment Regulation [25]. Transboundary movements of waste require not only a notification by the Country of waste origin, but also by the Country of destination and all others the transport will travelling through.

An exemption from notification can apply for non-mixed and non-hazardous waste. A final decision about an obligation for notification concerning a specific waste stream is up to each single national authority.

A single waste management company which intends to perform centralized GFRP and CFRP waste treatment using waste from different European countries, would have to arrange transport notifications with each single waste generator, independently from the type of the intended waste treatment (fiber/polymer recycling or thermal recovery in cement kilns). Since every notification procedure is time consuming and accompanied by considerable financial efforts, such a procedure for international waste collection does not seem practical. A more realistic approach could be a cooperation of waste management companies, which collect GFRP and CFRP waste on a national level and coordinate notifications for transboundary shipment to a centralized large-scale recycling plant.

In contrast to externally collected composite waste, an in-house reuse of residual materials from production processes is legally possible. Materials obtained from treatment of scraps (as recycled glass or carbon fibers), could be considered as product and not as waste, and would not be affected by the scope of waste management laws. However, also in this case a final decision about the classification of recovered materials as product, by-product or waste rests with the corresponding competent authority.

The already presented European Waste Framework Directive 2008/98/EC [4] sets the basic concepts and definitions related to waste management. It explains, through the so called "end-of-waste criteria", in which cases waste ceases to be waste and becomes a secondary raw material, and how to distinguish between waste and by-products. Although the Waste Framework Directive gives a lot of hints, explanations and definitions, a high degree of uncertainty is still present and different approaches concerning the handling of GFRP and CFRP waste in the EU Countries can be highlighted, mainly due to the absence of a clear classification of GFRP and CFRP.

Some examples from the different Countries are exemplified described in the following list.

- In the Netherlands LAP3 is the formal document of the Dutch Authorities regarding management of waste in general [26]. The document describes the waste stream plans for 85 sectors in Netherlands, including Plastics and Rubbers (considered in the Sector Plan 11). In this plan, for the first time, composites are mentioned, even if through a remark that specifies the difficulties to recycle fiber reinforced plastics. LAP 3 indicates for thermoplastics (including composites) that the minimum standard for waste management is reuse (or recycling). For thermosets plastics (including composites) the minimum standard for waste management is thermal recycling. Any further recycling technology "above" thermal recycling is acceptable, including the use in cement production. As in Netherlands most of the waste incinerators are qualified as thermal recycling installations, this means that landfill is not accepted anymore.
- In Belgium there is no specific rule for GFRP or CFRP. There are some companies collecting composite waste from the industry after waste sorting but there are no specific disposal rules.

- An interim solution concerning landfill can be found in Finland. There is a controversial situation in Finland regarding GFRP and CFRP. The landfill ban is in force as no more than 10% of organic content is allowed to be disposed. In the meanwhile, few landfills maintained a special exemption for GFRP to support the transition to a complete landfill ban. On the other hand, as there are not many real recycling processes for GFRP and CFRP, waste is processed by incineration (with energy recovery).
- Considering Europe (and not only EU), the situation is even more complex. As an example, in the UK landfill is still an opportunity. Landfill tax now stands at £98.60/ton (2022 rate), making the cost of landfill, including gate fees and transport, typically £140 to £150 per ton [27]. On the other hand, while sharp increases in landfill taxes are not expected, Germany and several other European countries have already largely banned landfill.

This small list shows the heterogeneous European situation that can affect the implementation of robust recycling and reuse of composites and the related circular value-chains. A common legislation at EU level is needed to coordinate actions and to promote best practices all around Europe.

5 Conclusions

Circular economy is showing its full potentiality only in the last years. The possibility to reduce environmental impacts while creating new circular value-chains acts as a game changer, not only at local but at global level. On the other hand, composite materials are more and more used in a widespread number of applications thanks to their peculiar characteristics of lightweight, high mechanical properties and corrosion resistance. Despite this, a systematic reuse of materials and components based on recycling and remanufacturing has not yet been implemented, even if different technologies are available. This is due for several reasons, from customer perception to the absence of a unique legislation at European level for EoL composites treatment. To solve these problems, and to transform them from an issue to an opportunity, a change at systemic level is needed, based on Circular Economy principles.

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