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REVIEW

En route to realistic sustainable structural fiber-reinforced polymers in a circular economy

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Abstract

Fiber-reinforced polymers (FRP) have faced exponential growth for decades due to their exceptional strength-to-weight ratio, permitting previously unreachable performances. In particular, in the necessity of diminishing the human overall environmental footprint, they allow safer, lighter, and more performing structures with on-demand properties and infinite engineered possibilities. As a consequence, substantial materials and energy savings can be expected. Yet, the environmental footprint of these materials and structures remains poor. This is attributed to their sourcing (oil-based mainly), their highly energy-intensive production, the complexity of the material, and the challenging handling of their end-of-life. Also, their highly multidisciplinary nature, requiring organic and polymer chemistry, material, processing, and mechanical engineering, among others, complexifies the interactions between actors to embrace and solve these issues fully. To this date, FRP industries remain a fully linear economy that cannot be carried in a (more) sustainable future. This review provides a multidisciplinary and extensive overview of current market needs and research development over all aspects of FRP to guide both research and markets toward sustainable and circular solutions. Sourcing, production, performances, and end-of-life are discussed, and, whenever possible, the environmental, economic, societal, and industrial needs are considered. The work intends to provide a general overview and future perspective to, one day, reach truly sustainable and circular structures.

Keywords Fiber-reinforced polymer (FRP) composites · Sustainable materials · Covalent adaptable networks · Recycling · Circular economy · Green polymers

1 Introduction

The first principle of thermodynamic laws states that energy cannot be created or destroyed in an isolated system. From a material perspective, A. Lavoisier, one of the fathers of modern chemistry, was credited with the apocryphal maxim: "Nothing is lost, nothing is created, everything is transformed." If the former deals with energy and the latter matters, one must admit that our actual mode of production and consumption of materials, ruled out by these fundamental paradigms, does not follow these principles. The additional entropy augmentation due to the irreversibility of our system forecasts a limit to the economic growth [1]. The (actual) linear production of materials consists of a series of value-added steps, consuming (fossil) energy and resources towards a final point of discarding, following a "make, use, dispose of" motto. This leads to the (definitive) loss of energy and matters in the ecosystem [2]. To pursue a more sustainable approach, both academia and industry must consider sourcing materials from renewable resources, including waste. They should prioritize the principles of "reduce, reuse, and recycle" in that order, focusing on eco-design to facilitate the integration of materials into a circular economy [3].

The history of mankind has always been intimately linked to materials. From stone to bronze, later to steel, the advances

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in materials science and technologies intensely followed humanity's evolution, alongside the access to dense and accessible energy [4]. The significant metallurgy progress on steel in the eighteenth century, followed by aluminum in the nineteenth, allowed new infrastructures and advances, to the extent of structures "heavier than air" to fly. One could say that we entered the era of plastic in the middle of the twentieth century. Although natural polymeric materials have been used for thousands of years, as old as ancient Egypt [5], it was in 1907 that Leo Baekeland developed the first synthetic polymers. Twelve years later, Nobel laureate Hermann Staudinger emitted the concept of macromolecules [6], opening the doors to unlimited advances in that field since then. The idea of reinforcing polymers with fillers, i.e., composite materials, concomitates with Baekeland's first works, using asbestos to reinforce his bakelite. However, it is the developments of glass fibers (first patent in 1880) and carbon fibers in the '50s [7], boosted by the world wars and Cold War, that brought a whole new kind of materials, able to overpass metallic structures: structural fiber-reinforced polymers (FRP) [8]. While metallic materials are inherently recyclable and relevant in a circular economy, their higher density has pushed toward their replacement by FRPs, but doing such has led to a step back in recycling and circularity.

FRPs enabled exceptional strength-to-weight ratio materials with a wide range of properties at competitive costs. The developments in the last 50 years have seen substantial advances, and a deep understanding and mastery of knowledge and techniques have allowed significant gains in structural performance. Tremendous efforts have been made to extend polymer matrices to various applications and markets depending on the manufacturer and end-user requirements, costs, and used fiber. Among them, unsaturated polyesters (UPR), epoxides (EP), and polyurethanes (PU) represent the vast majority of matrices in FRP [9], while glass fibers largely dominate the market. Nowadays, FRPs are seen as essential assets in a more sustainable future to diminish fuel consumption in transportation through weight savings, high-pressure vessels, renewable energy production equipment, and more [10].

Ultimately, the exponential growth of FRP use poses a significant challenge in the framework of a sustainable and circular economy. The production of fibers requires access to high quantities of energy at competitive costs. It also causes the depletion of non-renewable resources (sand and oil mainly) while releasing large amounts of greenhouse gases (GHG) in the atmosphere [11]. The production of matrices also requires significant amounts of energy and fossil resources and yields hazardous waste. Additionally, thermoset polymers are usually considered non-recyclable and cause downgraded composite structures to be lost in the ecosystem at their end-of-life (EoL), either through landfill or incineration, making the recirculation of their constituents in the economy unreachable so far [12]. Moreover, manufacturing composite parts generates a vast amount of additional waste [13]. The high embodied energy and environmental footprint prevent the true environmental benefits of using FRPs instead of conventional metallic structures from being effective [14]. The problems of the EoL arise nowadays with the first generations of large composite structures now reaching replacement time and the continuous growth of the composite market [9]. Major efforts are now put into improving the eco-design of FRPs in the frame of a circular economy through better sourcing of the building blocks, process evolution, and EoL scenarios.

Exploring the sustainability of FRPs demands a wide range of scientific competencies, as there are numerous ways to tackle this subject. While new systems are quickly being developed in academic laboratories, including bio-based polymers and dynamic networks [15, 16], they often struggle to achieve a well-established and competitive maturity level. Implementing these innovations in an industry characterized by inertia and significant past investments is difficult, hindering the willingness to embrace disruptive changes [17, 18]. Moreover, significant efforts are still needed to develop streamlined and economically viable pathways tailored to the existing FRP structures, including the recycling of "old" chemistry, such as UPR or EP. Reaching recyclable FRP that could embrace a circular economy requires mobilizing competencies and knowledge to address the complexity of FRP across various fields: organic chemistry (building blocks synthesis), polymer chemistry (polymerization and depolymerization), processing, materials science, and mechanical engineering. Addressing all the aspects of FRPs, between fibers and polymer matrices, from the sourcing of raw materials to the EoL, including production, processing, and supply chains, is essential to ensure the development of global and relevant solutions [10, 19]. Circularity should be at the heart of eco-design for composites to ensure their seamless integration into a sustainable economy and a lasting future.

The future of the circular economy for FRP lies in optimizing these technologies to balance environmental impact, cost-effectiveness, and material recovery rates. Progress in these areas will be crucial for reducing the overall carbon footprint of industries heavily reliant on composite materials. Achieving net-zero goals will force the composite sector to tackle sustainability challenges within its supply chain (see Fig. 1). Until now, limited attention has been given to the role of chemistry in enhancing the sustainability of composites.

There are numerous reviews discussing sustainable polymers [20], thermosets [21], epoxy resins[15], covalent adaptable networks [22, 23], natural fibers [24], composites [25, 26], and to some extent recycling [13, 27–29]. However, to the author's best knowledge, none has provided a critical overview of the whole composite value chain, from sourcing

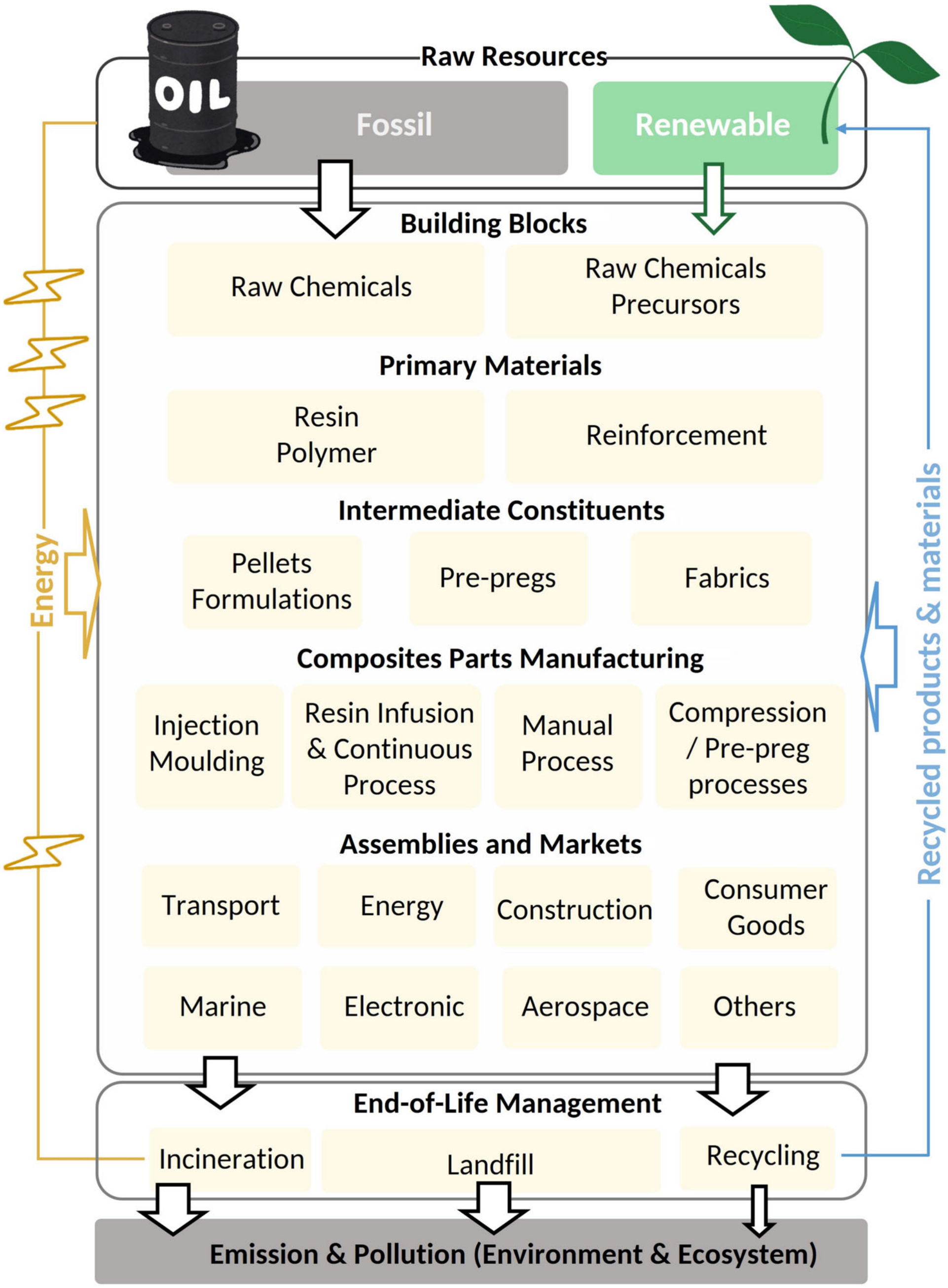


Fig. 1 Non-exhaustive schematic overview of the composite market value chain

to end-of-life, in a frame of a circular economy. In particular, the critical role of chemistry in composites, regarding its influence on processes, properties, and end-of-life considerations, must be discussed to a larger extent. Access to renewable feedstocks and monomers to design sustainable composites with advanced properties is critical.

This state-of-the-art aims to provide a multidisciplinary overview, with a priority on chemistry, about the recent advances in the realm of structural composites for a future circular economy, while considering market needs and research developments. After briefly introducing the current composite market, the concept of circular economy, as applied to polymer chemistry and FRP, is discussed. The role, properties, and limitations of fiber reinforcements, as well as the pivotal role of matrices in the eco-design of FRP, are addressed. In the case of matrices, the evolution of sustainability over time will be discussed in relation to each chemistry, ranging from established to emerging ones. This will include sourcing, scale-up, production, performance, end-of-life, and, whenever possible, the environmental, economic, societal, and industrial needs are also considered.

Our work aims to provide a comprehensive overview and future perspective, ultimately leading to the development of genuinely sustainable structures within a durable and circular economy. However, FRPs remain highly complex materials with complex behavior used in highly demanding applications. This leads to multidisciplinary aspects and complex value chains with numerous steps in the production process that cannot be treated in a single review. For this reason, some aspects have been deliberately overlooked in this work but should not be forgotten when considering the sustainability of these materials such as the (eco)-design of composite parts and the complex manufacturing associated (exhaustive analysis on manufacturing, lay-up design, assembly, painting, multimaterials...), or service-life aspects (aging, repairing, monitoring...). Although thermoplastic and short-fiber composites are mentioned due to their growing importance, they are not the primary focus. Similarly, although crucial in composite performances, the fiber/matrix interface is briefly discussed for a comprehensive overview, and the reader is referred to specific reviews [30–32].

2 Current composite market situation, trends and needs

In 2023, around 13 Mt of composite materials were produced worldwide, valued at 41 bnUS$ [9]. When extended to composite manufactured parts, the market represents 105 bnUS$. The COVID-19 pandemic caused a slowdown of the composite market in 2020–2021. However, it fully recovered and resumed its precedent growth trajectory in 2022, driven by various sectors such as construction, energy, and electronics. This growth was particularly notable in Asia (47% of the global volume), with China developing its renewable energy sector exponentially [9, 33, 34]. North America and Europe follow, with 26% and 19% of the market volume, respectively, focusing more on high-added-value applications like aerospace and transportation. The rising demand for renewable energy, electric vehicles, and hydrogen storage pushes forward the need for composite materials. Despite the ongoing global uncertainties that recently affected the market, including the COVID-19 economic crisis, Boeing 737 Max crashes, energy supply issues, and the wars in Ukraine and the Middle East, the market still forecasts a 5–10% compound annual growth rate (CAGR) up to 2030 [9]. For comparison, the global structural steel market represents 117 bnUS$ with a 5–6% CAGR, and the total aluminum market weighs 162 bnUS$ with a projected 3–4% CAGR in the same period. The overall polymer (including FRP, thermosets, and thermoplastic) turnover in 2023 was estimated to be 624.8 bnUS$ with a projected 4% CAGR; the total volume is around 416 Mt. The global plastic trade mainly depends on single-use plastics (disposable) and commodity thermoplastic polymers such as polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC), polystyrene (PS), poly(ethylene terephthalate) (PET), and polyurethane (PU, mostly thermosets) [35, 36]. More importantly, while FRPs represent only around 3% of the total plastic market volume, they account for around 16–20% of the total plastic market value. This makes FRPs a well-established key sector in structural materials and the global economy, yet a niche sector in the polymer industry.

The strict distinction in fibers for composites differentiates discontinuous and continuous fibers, with discontinuous being short (<1 mm) or long (>1 mm). However, we consider the similarity in behavior and applications for all types of discontinuous fibers compared to continuous fibers, particularly for structural applications. Short fibers refer to discontinuous fibers in this work, and long fibers are deemed to be continuous ones.

Construction (25%), transportation (24%), electronics (17%), and energy (13%) represent the largest segments of the market (see Fig. 2). However, the requirements for these sectors are vastly different in terms of matrices (thermoset vs thermoplastic), fiber type (glass or other), and length. While the transportation sub-segment largely uses short fibers and thermoplastics (glass/PA6, glass/PP) to match production rates and cost requirements, the energy, electrics, marine, and aerospace sectors predominantly use long (continuous) fibers. Due to costs, equipment size, and property requirements, the energy sector (mostly wind turbines) primarily uses unsaturated polyester resin and long fibers. In general, the FRP market remains mainly dominated by glass fibers, short and long [9], representing more than 90% of the global volume (for 77% of the value) thanks to their competitive strength-to-price ratio. Natural fibers represent the second largest reinforcement, with 7–8% of the total volume and about 4% of the value. They are especially used in the transportation segment as short fiber reinforcements due to their low cost, low density, damping properties, and environmental benefits. Conversely, the aerospace sector requires high-performance materials and prioritizes performance over cost, making the sector the first consumer of carbon fibers and epoxy. This translates into a 19% market share in value for only 1% of the market volume. Carbon fibers are approximately 14 times more expensive than glass fibers [37].

The long fiber-reinforced composite (LFRC) market is more nuanced and evenly shared than the global composite market. LFRC combines the strength of long fibers with the flexibility of polymer matrices, providing superior mechanical properties, reduced weight, and excellent corrosion resistance compared to traditional materials such as steel or aluminum [38]. The global LFRC market was valued at approximately 25–30 bnUS$ in 2023 and is projected to grow at a CAGR of 8–10%, potentially reaching USD 50–60 bnUS$ by 2030. Construction and energy (including electric and electronic) applications account for about 33% of the market each, with composites being used for infrastructure reinforcement, renovation, and building materials (rebars, beams...). Large-scale wind turbine blades are predominantly

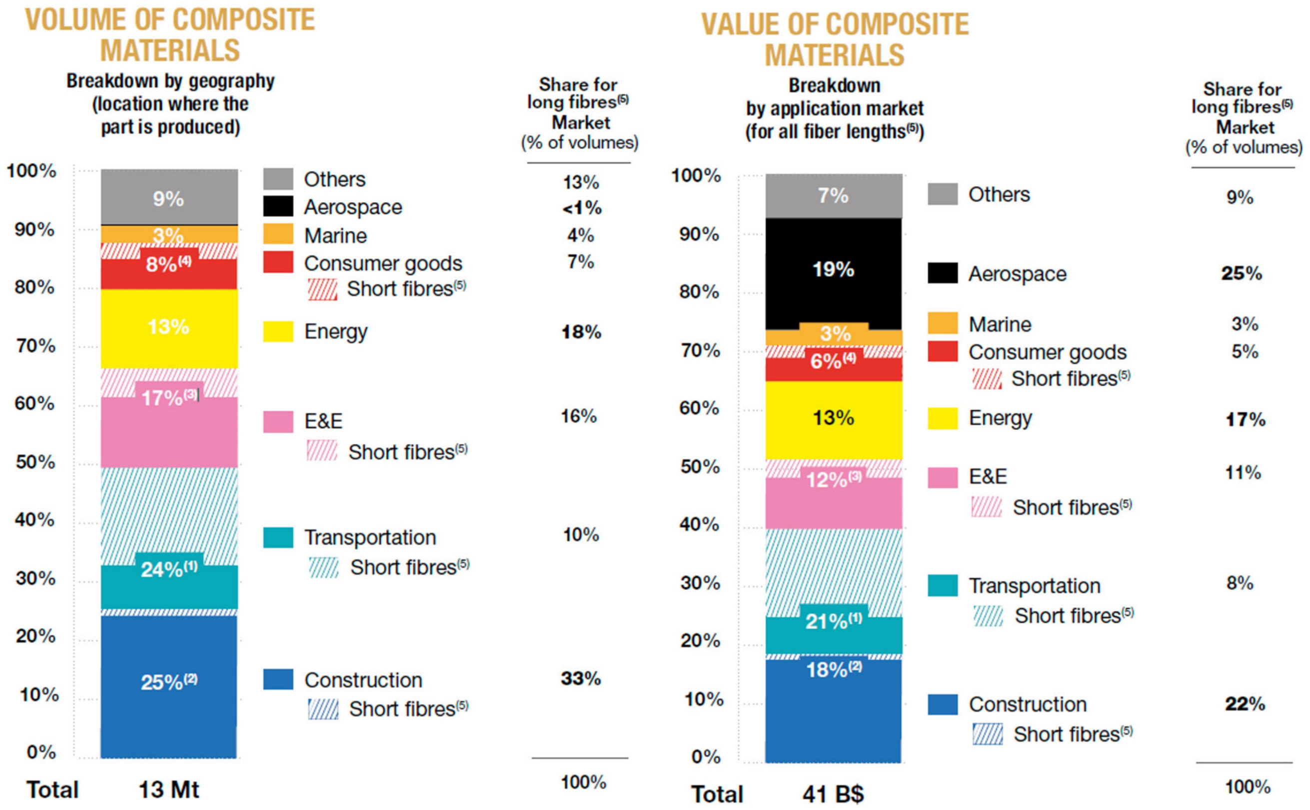


Fig. 2 Composite market breakdown in 2023 in volume and value per application [9]. Copyright 2024 JEC. Long fiber relates to continuous, and short fiber to discontinuous

made of glass fiber composites [33]. Aerospace, marine, and transportation follow with 15–20% of the market, driven by the need for materials with high strength-to-weight ratios to improve fuel efficiency and reduce emissions. Glass fibers still account for 50–55% of global fiber use in structural composites, again due to their cost-effectiveness and good mechanical properties. Carbon fibers make up 25–30% of the market and are mainly used in aerospace and high-performance automotive sectors, sports, and leisure. In these applications, strength-to-weight ratios are crucial, making the higher costs of carbon fibers secondary. Aramid fibers contribute 5–7% of this fiber market, mainly used in applications requiring impact resistance, such as defense and aerospace. Meanwhile, natural fibers such as flax, hemp, and jute are gaining ground, representing about 5–10% of the fiber consumption, particularly in automotive and construction, in which eco-friendly solutions are in growing demand.

The market is evolving with the increasing use of carbon fiber composites for larger, more efficient blades in wind turbines or hydrogen storage, boosted by price reductions. Many efforts are being made to decrease the cost of carbon fiber, such as the technology developed by the Canoe Technology Centre, allowing an outstanding 40% cost reduction on carbon fiber (8€/kg in 2018) [7, 39]. Moreover, the extra costs related to metal corrosion boost the demand for composites in construction (including gas and oil pipelines). Recently, traditional metallic and cement-based bridges have been replaced by FRP-based structures, decreasing the need for maintenance, structural health monitoring, and heavy equipment (Smart Circular Bridges project, Flax pedestrian bridge by FiberCore and TU/e, Almere, The Netherlands). Besides, natural fibers are quickly expanding as a sustainable alternative to glass fibers. Thermosetting resins like epoxy, polyester, and vinyl ester account for 80–85% of the LFRC market due to their higher affinity with the fibers and inherent stability. Thermoplastics, including polyamide, polypropylene, and PEEK, become increasingly popular due to their recyclability and toughness, representing 15–20% of the market, but face severe limitations related to their processability [40]. Higher process temperature and pressure are usually required to melt the thermoplastic and allow the shaping and welding of the laminate; such requirements can lead to additional environmental impacts and energy consumption, which should be taken into account. Additionally, the wetting of fibers with thermoplastic is challenging, limiting the exploitation of the full fiber potential [30].

Thermoset polymers remain the preferred matrices for composites, representing 60% of the market (see Fig. 3). They offer good stability over time, are required for long service-life, have easier tooling and lower energy than ther-

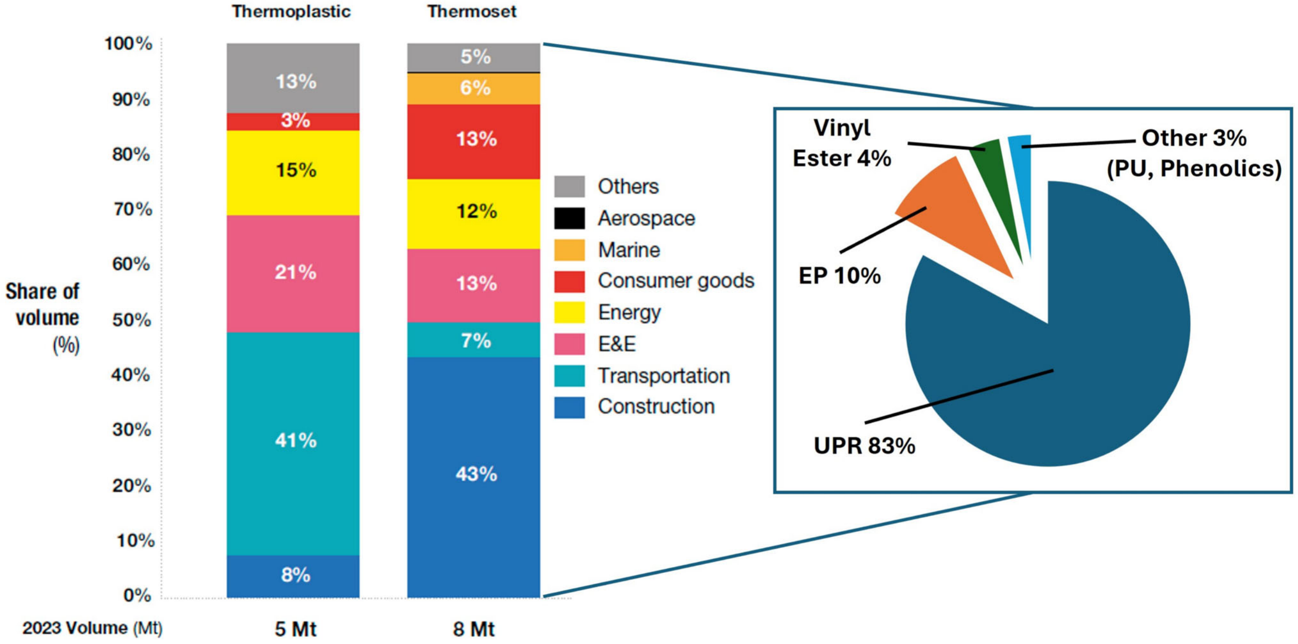


Fig. 3 Resin market breakdown for composite by applications [9, 41]. JEC Copyright

moplastic, and are more versatile to adjust properties and constituents in various markets [42]. Among them, unsaturated polyesters (UPR) possess the largest market share, with 83% of the total of thermosets used, followed by epoxy (10%), vinyl ester (4%), and others (polyurethanes, phenolics, bismaleides, polyimides...). The strong domination of UPR is due to its low cost, low viscosity (simplifying processes and giving access to large-scale structures), and versatility. However, their poor environmental stability, fire resistance, and mechanical properties make the epoxy more relevant in demanding applications [9, 43].

Thermoplastics (TP) matrices have seen significant growth over the last decades to stabilize at around 40% of the market volume [9]. Thermoplastics are mainly considered for their recyclability, flexibility, and cost-effectiveness, particularly in automotive (41%) and electronics (21%). As discussed earlier, the vast majority of these thermoplastics are in combination with short glass and natural fiber in order to improve their properties at a lower cost [40].

The choice of thermoset or thermoplastic matrices and the related chemistry remain strongly dependent on the targeted applications and, therefore, certain requirements. The most significant limiting parameters are mainly cost and properties. They influence all the steps of the industry, from sourcing to the manufacturing process and final application [44]. Thermosets are preferred for long fiber structural applications because of their effectiveness in wetting the fiber and their durability. Thermoplastic-based materials are ideal choices for low-cost, mass, and high-speed production.

The choice of composite constituents affects not only the final properties and total costs but also the manufacturing process [45]. The manufacturing process also plays a key role in the possible attainable properties, costs, and final applications. For instance, manual processes, such as hand lay-up, pre-preg, and thermo-compression, have lost their dominant position due to the high labor cost engaged, requiring numerous highly qualified workers. Their market share decreased from 52% in 2018 to 35% in 2023 [9]. Conversely, fast and automated processes have experienced rapid growth. Injection molding, which uses short fibers, now represents 40% of the composite market (Fig. 4) due to the extensive increase in thermoplastic use and continues to be a growing field. The share of injection molding was only 13% in 2018 [9]. For thermosets, out-of-autoclaves are the most promising processes [46] as they offer faster production rates, less energy, and less labor. Out-of-autoclave includes continuous processes (pultrusion, pull-winding, continuous sheet, filament

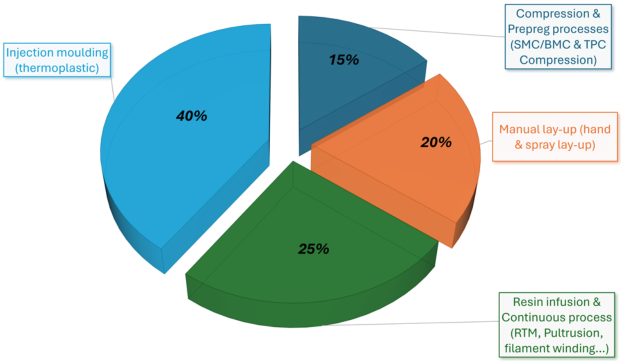


Fig. 4 Composite manufacturing processes—market breakdown 2023, data from [9]

winding...) and infusion processes (resin transfer molding, liquid infusion...). They represent 25% of the market, with a 5–10% predicted CAGR up to 2030 [9].

As previously seen, the main difficulty in obtaining sustainable material is its large portfolio of solutions that make each material distinct and, somehow, unique. This results in a highly diversified market with extremely complex EoL management. To date, there is no unique solution, and EoL remains the least developed part of the composite value chain [28]. Only a few actors have actively offered solutions to recover and valorize decommissioned composite structures. Although many SMEs and start-ups are flourishing in that segment, they remain underdeveloped and not fully suited to the current market needs. Currently, EoL primarily involves landfill incineration (with or without energy recovery) and grinding for downgraded use (cement fillers, clinker, composite filler, etc.). Progress in developing standards and integration of recycled materials will be essential for market growth. [10, 34]. Nowadays, only four countries worldwide (Germany, Finland, the Netherlands, and Austria) have legislated on composite EoL to ban landfills and encourage recycling [47], but still tolerate some exceptions due to the lack of reliable solutions.

In the coming years, the LFRC market is expected to continue its strong growth trajectory, fueled by technological advancements in material processing, increasing demand for lightweight and durable materials, and the push toward sustainability. Improvements in thermoplastic composites, recycling technologies, and automation in manufacturing are likely to further propel this market, making LFRCs more cost-effective and accessible across a wider range of industries.

3 Sustainable structural materials in the frame of a circular economy

3.1 About the concept of circular economy

In a linear economy, the current model, an economic value, is created during the extraction of resources and increases through the subsequent production/transformation steps into manufactured goods and their final consumption by end-users. The value of goods becomes negative once they are discarded as waste (see Fig. 5) [2].

Since the latter half of the twentieth century, many economists have identified some issues with the current economic model. The economist K.E. Boulding introduced the idea of an (economic) isolated system by analogy to thermodynamics. In what he called the "spaceship Earth," resources are not unlimited and should be considered capital stock. To ensure the spaceship's future, the main goal of the inhabitants should be to maintain this capital, i.e., preserving the resources [48]. The economist N. Georgescu, who pioneered the concept of "ecological economics" [49], more precisely formulated the deep connection between the economic model and thermodynamics. As the economy is deeply dependent on energy and matter transformation and consumption, Georgescu suggested visualizing the economic sphere as a system that will reach maximum entropy, and human activities accelerate this process. Accordingly, he proposed the terminology of "low entropy" for renewable resources and "high entropy" for waste and pollution. In other terms, for Georgescu, low entropy is transformed irreversibly into high entropy. Because such a pathway is irreversible, the resources are scarce (over humanity's time frame).

Later, the concept of circular economy was formally introduced by Pearce and Turner [50]. The limits of the continuous exploitation of finite resources and the increasing waste production were exposed. They proposed, to tackle that problem, to re-incorporate end-of-life products as raw resources for new materials production, referred to as cradle-to-cradle, creating a closed-loop economy. Importantly, they stated that the main goals were to reduce waste and build more resilient systems. Unlike the linear economy's "make, use, dispose," the circular economy first relies on the 3Rs "reduce, reuse, recycle," i.e., firstly by mitigating the consumption of goods, secondly by extending the service-life, and finally by processing materials to recover their components/constituents. The 3R concept finally grew to 10Rs, as summarized in Fig. 6. In a so-called circular economy, matter and energy are no longer deemed as an irreversible flow but rather as an isolated system with a finite amount of resources preventing

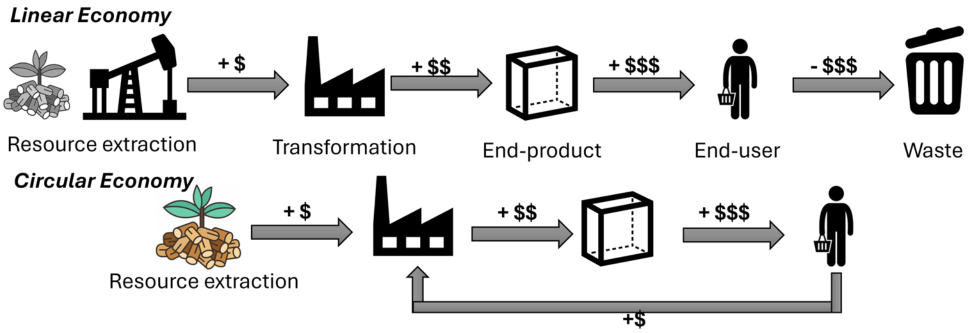
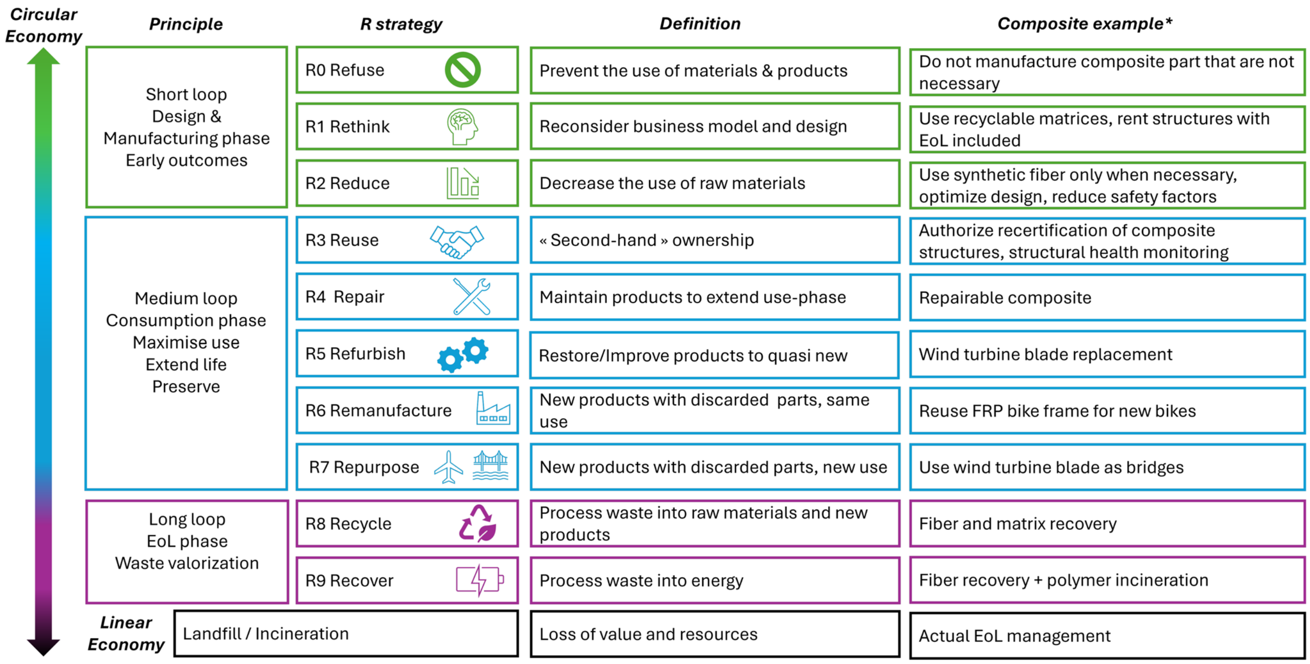


Fig. 5 Simplified overview of an idealized linear and circular economy value chain. The dollar icons represent cumulative values



Principle	R strategy	Definition	Composite example*
Short loop Design & Manufacturing phase Early outcomes	R0 Refuse	Prevent the use of materials & products	Do not manufacture composite part that are not necessary
	R1 Rethink	Reconsider business model and design	Use recyclable matrices, rent structures with EoL included
	R2 Reduce	Decrease the use of raw materials	Use synthetic fiber only when necessary, optimize design, reduce safety factors
Medium loop Consumption phase Maximise use Extend life Preserve	R3 Reuse	« Second-hand » ownership	Authorize recertification of composite structures, structural health monitoring
	R4 Repair	Maintain products to extend use-phase	Repairable composite
	R5 Refurbish	Restore/Improve products to quasi new	Wind turbine blade replacement
	R6 Remanufacture	New products with discarded parts, same use	Reuse FRP bike frame for new bikes
	R7 Repurpose	New products with discarded parts, new use	Use wind turbine blade as bridges
Long loop EoL phase Waste valorization	R8 Recycle	Process waste into raw materials and new products	Fiber and matrix recovery
	R9 Recover	Process waste into energy	Fiber recovery + polymer incineration
Linear Economy	Landfill / Incineration	Loss of value and resources	Actual EoL management

Fig. 6 10Rs strategies in the frame of an idealized circular economy. *Representative examples are given and might not be legally or economically viable

their depletion. Later, architect R.B. Fuller defended the concept that waste "is nothing but resources we are not harvesting because we have been ignorant of their value" [51], which is particularly relevant to the composite industry.

In 2017, Kirchherr et al. [52] identified 114 different definitions of circular economy and proposed a unified definition: "circular economy describes an economic system that is based on business models which replace the 'end-of-life' concept with reducing, alternatively reusing, recycling and recovering materials in production/distribution and consumption processes, thus operating at the micro level (products, companies, consumers), meso-level (eco-industrial parks) and macro-level (city, region, nation and beyond), with the aim to accomplish sustainable development, which implies creating environmental quality, economic prosperity, and social equity, to the benefit of current and future generations."

It is clear how important materials and energy streams are in this context and the role that chemistry and the chemical industry can play in the realm. More specifically, composite materials, particularly made of glass fibers and carbon fibers, should be designed to fit the requirements of a circular economy. This includes the design of polymeric matrices able to be repaired, recycled, and depolymerized, with the view to ideally retrieving both fibers and matrix or monomers. The main goal should be to transform large composite waste into resources and valuable compounds [2]. They should be conceived to fulfill this definition and help designers and engineers achieve the full recirculation of waste.

From the above definition, materials embedding the principle of the circular economy must fulfill the following:

- Sourcing from renewable resources: biomass, by-products, post-consumer waste...
- Limit energy consumption at all stages
- Be easily repairable to extend their service life
- Be able to be easily deconstructed and separated from the entire structure
- Be able to be repurposed to a new function
- Ultimately be able to be recycled
- Avoid the use or release of toxic and hazardous compounds

3.2 Materials in circular economy, from lab concept to market reality

The current global economic system is intricate, involving numerous actors, i.e., public institutions, governments, and industries at different scales (local, regional, and international). Therefore, to globalize a circular economy, it is essential to engage both states and governments as well as companies and industries.

The rising awareness of environmental challenges has significantly increased our insights about the recycling of materials, its economy, and the associated challenges [51]. The United Nations is pushing towards goals such as net-zero carbon. Stricter environmental regulations [17], along with treaties promoting and regulating trades, encourage the reuse and recycling of materials and wastes [53, 54]. Although the current efforts are still insufficient, there is a consensus on the need for cooperation toward a global circular economy [4]. Industries should adopt a business model where the economic system mimics Nature, with flourishing interdependent actors [3]. However, such an ecosystem must be encouraged and supported by strong policies and adapted contexts [51, 53].

Only a few countries have already engaged in programs to promote a circular economic system. China has invested widely and legislated to support the industrial recycling of by-products and waste. They created circular economy industrial parks that saved 14 million tons of GHG in 2016 [17, 53]. Economical simulation plans trillions of dollars of savings if these strategies are extended nationwide [17]. Pushed to an extreme, the Club of Rome projected in a multiple-case scenario [55] from 5 European countries (Finland, France, the Netherlands, Spain, and Sweden) that shifting to a circular economy through energy efficiency, material efficiency, and renewable sourcing would cut carbon emission up to 70% with 1.5–2% increase in the Gross National Income (GNP) and reduce unemployment by nearly a third. It is clear that the change from our current linear business model to a circular one is a monumental challenge with significant risks for the private sector. This would include the establishment of long-term common strategies among different actors, creating interdependence that can be beneficial but also present risks for investors [54, 56]. This is the reason why it requires strong support from the government through financial support, tax incentives, and regulations [53].

Implementing global circular strategies for materials would lead to substantial economic and environmental benefits but also secure supplies for industries [51], including high-demand sectors, such as aeronautics. As an example, aircraft engine manufacturer Rolls-Royce developed the Revert program [51]. Aircraft engines require high-quality and high-performance alloys (so-called superalloys), based on titanium and/or nickel, including some minor elements like vanadium, rhenium, and cadmium [57]. The Revert program allowed Rolls-Royce to secure the supply chain by recovering alloys from production waste, dismantling decommissioned turbines, and sending back sorted alloys to their supplier. By doing such, the supplier has easier and guaranteed access to low-cost "raw" materials, while Rolls-Royce secured aero-grade alloys at a lower cost. Around 60–70 jobs have been created while increasing the profits of Rolls-Royce and its business partners as well as lowering the environmental footprints of their products, by reducing the need for virgin ores from extraction to treatment [51]. Other successful examples include carpet manufacturer Desso, which recovers old carpets from customers to recycle [3] or tire manufacturer Michelin, which rents transporter truck tires by the mile [2].

In polymer chemistry, recirculating carbon offers significant environmental benefits [19] as well as economic gains through the creation of local employment [2] and the value maximization at each stage. Recycled plastics can be cheaper if competitive processes are set up in addition to favorable regulations [19], even though common recycling technologies (mainly thermo-mechanical) lead to a reduction of their properties and/or an increase in the cost of the recycled product [58]. Many studies have demonstrated the interest in sourcing chemicals from renewable feedstocks, in particular, C1-chemicals, to promote a circular carbon and plastic economy [19, 59]. Vidal et al. [19] proposed a bold system change to align with IPCC recommendations and limit CO_2eq emissions [60] by proposing the "full" recirculation of "carbon" atom. They highlighted that recycling alone cannot sufficiently reduce emissions but also estimated that renewable feedstocks as raw materials cannot be the sole source; carbon capture and utilization (CCU) [61] must be developed to provide new building blocks. Renewable and cheap energy remains a crucial parameter, along with population sensitivity to such issues. In addition, a smarter design of polymer-based materials is essential, particularly by maximizing service lifespan and recoverability. By doing so, they estimate 85–90% of CO_2 savings in construction sectors, a major consumer of composite materials. More importantly, one must keep in mind that GHG emissions are not the sole criterion of the environmental footprints, and other parameters such as water eutrophication, acidification, resources depletion, water and land use, or wildlife are as crucial as CO2 emissions and are (too) often overlooked [62]. Additionally, measures towards GHG savings should ensure that sustainable practices do not transfer impacts from one category to another.

3.3 Complexity and strength of the specialized composite market, can composite enter a circular economy?

Both energy and matter are vital to material sciences, even more so to composites. Up to now, the recovery of energy, or matter, from polymers and composites is still to be addressed. According to the current situation, the composite market is inherently linear and unsustainable. Efforts must be made to recirculate the large amounts of waste in this sector. In particular, these wastes still possess an important value, embodied energy, and potential that are highly relevant for a more circular economy.

Each year, an exponential number of composite structures are decommissioned with limited recycling possibilities, mostly ending up in landfills. Even though some interesting works have been performed in the academic literature to tackle this problem [13] (see Sect. 8), to the author's best knowledge, the full circular ecosystem of composite does not yet exist. Some start-up companies started to develop such strategies by developing the recycling of carbon or glass fibers and the valorization of these recovered fibers a few years ago such as Composite Recycling (Switzerland), Nova Carbon (France), Gen2Carbon (UK), Lineat (UK), and V-Carbon (UK), among others. Others focus on repairable composites to extend service-life (CompPair, Switzerland), new resins (Mallinda Inc, USA; Recyclamine, India), and sustainable fibers (EcoTechnilin, France; Bcomp, Switzerland; Isomatex, Belgium).

However, the potential for structural composites to lead to high-performance materials in a circular economy remains uncertain. Unlike commodity (single-use) plastics, structural composites are designed with dedicated processes to have long service-life (>20 years), and specialized actors that can gather and recycle more efficiently as aforementioned [63]. Composite materials are primarily used in high-performance technical applications, such as transportation (aeronautics, naval...), sports and leisure, or construction [9]. These sectors are characterized by the complexity of their products and their high-performance requirements, where materials must not only meet strict safety standards but also be optimized for weight, durability, and cost. The complexity of these industries has a direct influence on the sourcing and manufacturing of composites, challenging the recycling of these materials [33, 64]. The development of custom-tailored solutions, such as thermal, chemical, and mechanical recycling methods, to handle the different fibers and matrices used in composites is required. Such approaches must also integrate new design philosophies, such as modularity and disassembly, to enable easier repair, reuse, and recycling. However, their specificity limits the number of actors and ensures continuity between suppliers and clients, making the recovery of decommissioned structures easier than that of consumer goods, as exemplified by the Rolls-Royce initiative for aeronautics alloys.

There is still a challenge in conciliating performance, durability, and end-of-life. Composites' presence in aircraft, like those from Boeing or Airbus, has substantially increased over these years, and their eventual decommissioning constitutes a significant challenge. As a result, the circular economy in aerospace must focus on optimizing disassembly and reusability, which can only be achieved through highly specialized, market-driven recycling methods [13, 39].

Similarly, in the wind energy sector, the extensive use of fiber-reinforced composites in turbine blades and the need for wind turbines to operate efficiently over 20–25 years have spurred innovation in composite materials. Yet, the complexity of recycling these blades at their EoL, given their size and multi-material compositions, requires a tailored approach [33, 34]. Wind turbine blades are a prime example of the complexity of the demand for specialized composites, as decommissioned blades must be recycled in ways that ensure they remain profitable for reuse or repurposing in new applications [37]. Despite the challenge, 90% of wind turbine mass is already recyclable/recycled from the metallic components, making the process of disassembly and sorting already efficient. Several million tons of composite materials will need to be recycled from wind blades in the coming years [65]. In response, new recycling methods are being explored to more efficiently separate fibers and matrices, with processes like thermal and mechanical recycling being considered for different types of composites [34]. Recycling strategies are increasingly being integrated into digital platforms (B2B) that optimize the reuse of materials, reducing waste and improving resource efficiency [66].

Other market sectors for composites, such as construction and transportation, that represent a non-negligible part of the market volume could benefit from this high-value market, promoting sustainability and closed-loop recycling in composites. However, due to the specialized nature of these markets, circular economy strategies cannot adopt a one-size-fits-all approach. As already observed by Vidal et al. for all polymers [19], multiple parallel and complementary pathways must be developed to preserve and recirculate resource capital.

Efforts in the right direction are being observed in the industry. Recently (March 2025), the first industrial circular model was launched in the composite sector by Arkema, Groupe Beneteau, Veolia, Composite Recycling, Owens Corning, and Chomarat [67]. The alliance aims to provide a fully closed-loop model in the nautical sector. While the Elium® (Arkema) thermoplastic resin will be used along with glass fibers (Owens Corning, fiber production; Chomarat, textile reinforcement) to produce pleasure boats, the end-of-life and production waste will be collected by Veolia. Composite Recycling will treat the composite waste through pyrolysis (detailed in the dedicated section of this review), allowing for the recovery of the fiber that Owens Corning will remelt into virgin material. The pyrolysis oil, mainly composed of acrylates, will be purified by Arkema to produce new batches of their Elium resin. This example represents a significant step towards a more circular model and also proves the interest of the industry to build upon such initiatives.

The composite sector highlights the need for adaptable and complementary circular economy strategies. The materials must perform at peak levels during their service-life, while their end-of-life management requires market-driven solutions that address their unique characteristics and preserve their value (Fig. 7). In that sense, fibers, matrix, and their interactions should be advanced and selected to best suit the circularity concepts.

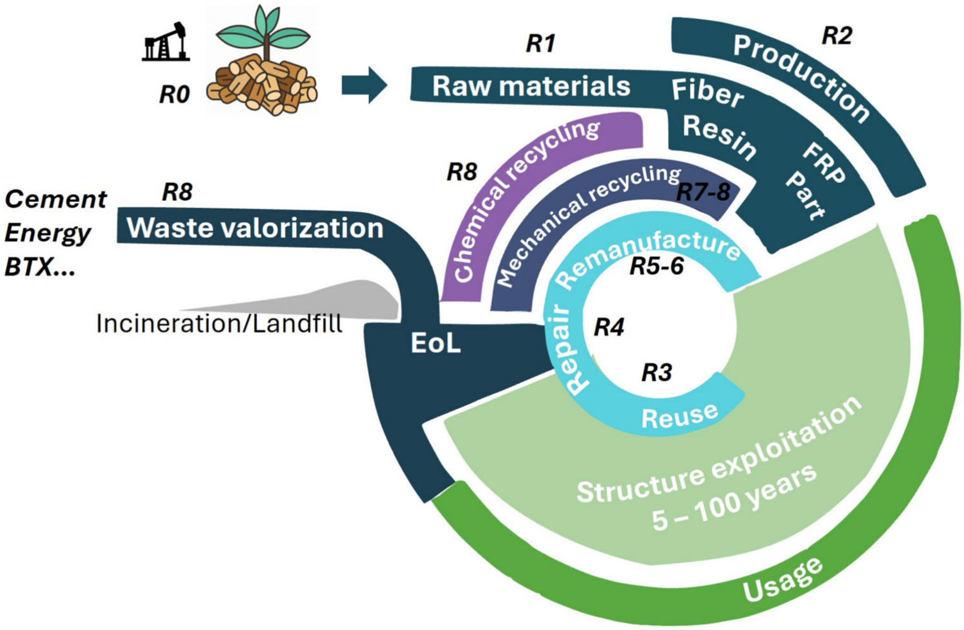


Fig. 7 Implementation of the R's into the composite economy, a figure inspired from [10]

4 Fibers and reinforcements—drivers for properties and environmental footprint

Structural composite properties are largely determined by the fibrous reinforcement, which commonly represents 60–80% of the total mass. The type of fibers imparts the main properties of these composites as well as their costs and environmental impacts.

4.1 Glass fibers—the unsustainable market volume

As previously seen, glass fibers widely dominate the FRP market. Their excellent strength-to-weight ratio, resistance to environmental degradation, and cost-effectiveness make them materials of choice in many applications. However, several challenges can arise with the use of glass fibers that must be addressed in the frame of a circular economy.

4.1.1 Sourcing and production

Glass fibers are silica-rich amorphous materials with several grades of glass fibers available depending on the final requirements (see next section). All grades possess a high silicon dioxide content (SiO_2), which constitutes 50–70% of the total composition and is responsible for the final product's mechanical strength and thermal properties. The composition is completed with other additives such as lime (calcium oxide, CaO_3), dolomite (calcium-magnesium carbonate $CaMg(CO_3)_2$), and alumina (kaolin, Al_2O_3) as network modifiers and alternate network formers to adjust the properties [68]. Minor oxides such as boron oxide (B_2O_3), magnesium oxide (MgO), or alkali oxide (Na, K) can also be included to enhance specific properties, such as thermal resistance or corrosion resistance, and facilitate fiberization. Although glass fiber grades meet certain content specifi-

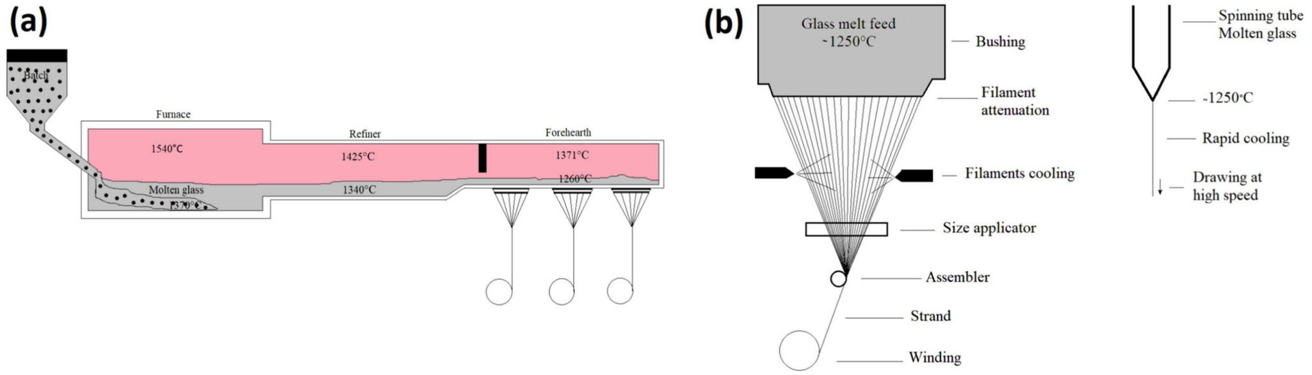


Fig. 8 Schematic representation of glass fiber production. **a** Vitrification of glass in a furnace and **b** GF continuous filament forming process. Reprinted from [26] Copyright 2024, with permission from Elsevier

cations, each manufacturer possesses its own (patented) recipes, making generalization and complete circularity complex, as compositions might vary from supplier to supplier.

Glass fibers are produced in an uninterrupted continuous process to limit costly pre-heating and maximize production rates (see Fig. 8). The raw materials are kept in separate batches before being mixed in the desired composition. The homogeneous batches are first melted and vitrified at about 1500–1800 °C to become glass. A second refinement step is performed at 1100–1300 °C. The molten glass is then extruded through a bushing into filaments [69]. This step is critical as it determines the diameter of the fibers, which ranges from 5 to 24 μm. The molten filaments are rapidly cooled and solidified by water spraying (or air jets), forming continuous glass filaments. At this stage, chemical sizing agents (generally water-based) are applied; this can also include a thin protective coating. The sizing agents reduce the abrasion of the equipment used to process the fibers (waiving, chopping...), protect the fibers, and enhance the adhesion with the future matrix. Typical sizings include silanes, epoxy-based compounds, and polyurethanes, which are tailored to the resin type (e.g., polyester, epoxy, or vinyl ester) used as the matrix in the final composite. A supplementary thermal step is commonly applied to cure and stabilize the sizing [69]. Finally, the continuous glass fibers are processed in desired forms (chopped, rovings, yarns, mat...). Rovings (continuous strands) are commonly used in structural composites, while mats and chopped strands are employed in applications where flexibility and moldability are important, such as in automotive or marine parts.

4.1.2 Properties, advantages and limits, applications

Various types of glass fibers can be produced, each with distinct compositions and mechanical properties. A-glass (Alkali-) is the oldest glass fiber type with a composition similar to glassware and lower properties than other types. They are mainly used in low-reinforcement and insulation applications [70]. E-glass (Electrical-) is the most widely used glass fiber type, known for its high tensile strength, cost-effectiveness, and suitability for general-purpose applications. E-glass is composed mainly of silicon oxide, alumina, and calcium oxide. S-glass (Structural-) fibers have higher tensile strength and modulus compared to E-glass, making them ideal for high-performance applications such as aerospace and defense. S-glass has fewer impurities (more controlled composition) than E-glass, with magnesium oxide used instead of calcium to bring more strength and stiffness (20–40 wt%). However, their compositions present more elevated melt viscosity, requiring higher energy inputs and more scarce and pure raw materials. This makes S-glass fibers more expensive and environmentally impactful [26, 71]. Other glass fiber grades such as C-glass (Chemical-), AR-glass (Alkali-Resistant-), and D-glass (Dielectric-) exist for specific applications but represent a small volume and will not be discussed further. All glass fibers possess a pure elastic-brittle behavior, with tensile strengths ranging from 2000 to 4500 MPa, modulus in the range 70–90 GPa, and 4–6% of elongation at break. Compared to other structural fibers, they have a relatively high density, between 2.4 and 2.6 g/cm^3, making them too heavy for some lightweight applications [72]. Except for specific grades, glass fibers are sensitive to strong oxidative, acids, alkalines, and salty environments and possess a large operating temperature range, between −50 and 300 °C [72].

4.1.3 Environmental footprint and values for recycling

The extraction of raw materials for glass fiber production raises environmental concerns, especially regarding silica and limestone mining. Sand mining is widely recognized as a major ecological issue [73]. Sand is required in massive quantities for construction, resulting in overconsumption that surpasses natural renewal rates [74] and augments the pressure on the mined ecosystems through water depletion, landscape destruction, wildlife habitat destruction, and pollutant emission [75]. Additionally, during the production process, high energy inputs are required to melt and extrude the glass fibers. The embodied energy of glass fibers is typically around 25-50 MJ/kg [76, 77], with associated CO_2 emissions at around 3-5 $kgCO_2eq/kg$. Typical furnaces use natural gas, which emits large quantities of greenhouse gases, including nitrogen oxides, carbon monoxide, carbon dioxide, methane, and traces of other gases and particulate matter [78]. Sustainable practices in raw material sourcing have become a priority for most industry leaders. Strategies include developing mining protocols that reduce landscape impact and actively sourcing from regions with stricter environmental regulations [75, 79] and higher quality ore deposits to reduce processing requirements. However, the lack of transparency in the protocols implemented by industries, along with economic interests, limits the ability to achieve truly sustainable practices.

Most efforts toward sustainable glass fiber production focus on reducing emissions, conserving raw materials, and improving energy efficiency throughout the production process. In particular, the advances in more efficient electrical furnaces, which can be alimented by renewable energies, allowed substantial energy savings and emissions reductions [76]. Moreover, the integration of digital monitoring and AI-driven control in furnace operations offers potential improvements in terms of yield and energy management [80]. Despite all the good practices that can be taken, the pressure on sand stocks may remain. In this regard, recirculating glass fibers can reduce the environmental burden of glass production [81] and secure feedstock supply to limit any eventual future costs. Techniques such as closed-loop recycling of cullet (broken or waste glass) back into the melting process could reduce the need for virgin materials and lower furnace energy requirements. Equally, recovering glass fibers from composites and re-melting could provide sustainable sourcing for glass fiber producers [82].

Additionally, research into bio-based, environmentally friendly sizings and low-emission binders might help the industry to shift towards more economically viable, sustainable pathways. The key challenge is to balance the demand for higher-performance fibers while aligning sustainability goals and maintaining competitive costs. With continued investment in sustainable practices, glass fiber production for composites is poised to meet the growing demand for lightweight, durable materials across diverse sectors, including automotive, aerospace, and renewable energy.

Given the increasing demand for glass fibers in the wind turbine and transportation sectors, it is essential to find more eco-friendly replacements and alternatives to alleviate market pressure. This includes developing advanced recycling methods and exploring new materials that can offer similar or superior performance with a lower environmental footprint.

4.2 Basalt fibers, a potential direct replacement of glass fibers

Basalt fibers (BF) are mineral fibers obtained from fused volcanic rocks. Some authors classify BFs as natural fibers (from minerals), as they require much less human intervention than glass fibers. Basalt is a major constituent of the oceanic and continental crusts [83]. It is widely available worldwide and is considered a largely accessible and renewable resource. Indeed, large quantities of basalt are generated each year due to volcanic activity. BFs were originally developed in the 50s in the Soviet Union (Moscow Research Institute of Glass and Plastic), mainly for military, spatial, and aerospace applications [84]. For this reason, the majority of the production and use remains nowadays concentrated in Russia, Ukraine, Germany, and China. However, new actors from the United States and Europe are now developing BFs as a potential alternative to glass fibers. While the glass fiber market represents nearly 30 bnUS$, the basalt fiber market is estimated to be 0.27 bnUS$.

4.2.1 Sourcing and production of basalt fibers

BFs are deemed a viable and greener alternative to glass fibers, with similar properties [71]. Most BFs are obtained from the direct melting and fiberization of basalt ores. Similar to glass fibers, basalt is mainly constituted of SiO_2 (40-55%), with high content of Al_2O_3, Fe_2O_3, MgO, and CaO [85]. The composition of BF depends on the sourcing of the basalt ores. In that sense, some ores and basalt grades are not suitable for continuous filament processing. In particular, some (crystalline) minerals such as olivine or biotite nucleate the crystallization of the cooling basalt melt, making the fiberization impossible due to fiber breaking during the spinning [86]. Specific care must be taken in the composition of the basalt ores. This could lead to the over-exploitation of basalt quarries, leading to environmental and landscape destruction. Some recent works and companies (as an example, FILAVA® from Isomatex S.A., Belgium) analyzed their basaltic ores to better control their compositions and thus their properties. This could enable the achievement of a more evenly distributed exploitation of basalt sources worldwide and the development of a new economy for structural materials [87].

Interestingly, the basalt fiber manufacturing process does not differ much from glass fibers, but the overall process is considered slightly cheaper and less energy-intensive [88]. Knowledge transfer, equipment adaptations, and market development could benefit from that with no major investment or detrimental modifications compared to existing glass fiber plants. Basalt rocks are crushed and washed before being loaded into furnaces. As the basalt already possesses terminal composition, a metering system is not required [83]. The raw materials are melted at 1400–1500 °C and extruded into 10–20 μm fibers through a continuous spinning process. The cooling must be performed adequately to avoid any crystal nucleation, being detrimental to the fiber properties [86]. Similar to glass fibers, sizing agents are required to protect the fibers, limit the abrasion of equipment, and improve the fiber/matrix adhesion.

4.2.2 Properties, advantages, and modifications to move from glass fiber production to basalt fibers

BFs are attractive for structural composite applications due to their high strength, corrosion resistance, and thermal stability. They are more expensive than E-glass, but the expected developments of the market and technologies, driven by high demand, are expected to lower basalt fiber costs to a similar level to glass fibers [84, 89]. They display properties similar to S-glass fibers with tensile strengths between 3000 and 4900 MPa and an elastic modulus ranging from 80 up to 100 GPa, but lower costs [90]. Interestingly, the elongation remains relatively high, ranging between 3 and 6%, which make them tougher than carbon fibers. BFs perform well under extreme temperatures and resist chemical degradation. They are more resistant to alkaline and acidic conditions [89] than glass fibers and possess a broader operative window starting from low temperatures (−200 °C) up to high temperatures (+700 °C). On the contrary, glass fibers are more susceptible to chemically aggressive media and cannot sustain temperatures higher than 300–400 °C before considerably losing their mechanical properties [85]. However, BFs are slightly denser than glass fibers, with a density between 2.5 and 2.8 g/cm^3 (compared to 2.4–2.6 g/cm^3 for GFs). In contrast to asbestos, another natural mineral fiber, BFs are completely inert and do not release any harmful compounds [71, 90], being compliant with REACH regulations. They have an expected lifetime of 50 years, longer than glass fibers, which could further enhance the durability of structures [91]. When properly manufactured and sized, BF exhibits excellent adhesion properties with various resins, improving composite matrix integration and leading to improved load-bearing capabilities and longevity of the composite structures [92, 93].

Such inherently excellent properties position BF as a strong alternative to traditional glass fibers and a cost-effective option compared to carbon fibers. They are suitable for high-temperature applications in aerospace, automotive, and fire-resistant construction materials. They can be exploited in corrosive environments, such as concrete reinforcement in construction and infrastructure applications. This combination of heat resistance, mechanical strength, and chemical durability makes basalt a more sustainable, reliable, and direct alternative to glass fibers. Despite the promising and growing interest, the development of the basalt fiber industry remains limited, with ongoing technical difficulties in ensuring a consistent supply. Future work should focus on better control of the properties through composition and processes and optimization of the fiber-matrix interface. Specific care should be taken to ensure benefits for a circular economy and environment through job creation while avoiding the replication of glass fiber issues, such as the impact of sand extraction.

4.2.3 Environmental footprint and values for recycling

The environmental footprint of basalt fiber composites has been poorly investigated [94]. This could be understood from the currently low usage volume, and this point must be more extensively investigated in the future. In particular, the sourcing of basalt ores should be considered in terms of environmental burdens along with transportation. Recent works [95] have highlighted that basalt fiber bars (epoxy resin matrix) could lead to a reduction of up to 44% of the carbon footprint compared to similar glass fiber bars and a 50 to 89% reduction compared to galvanized and stainless steel, respectively. The major contributor to the LCA of basalt fiber remains the energy required to melt and process the basalt ores [96]. The advances performed for glass fibers, using electrical furnaces with high efficiency and renewable energy, could also help reduce the environmental footprint of BFs further. Utilizing sun or wind-based electricity, available worldwide, basalt could be competitively produced on-site, reducing transportation needs and supporting local economies, contributing to the UN Sustainable Development Goals [75]. Importantly, the high thermal and chemical resistance of BF could be of high interest in pyrolysis and chemical recycling processes to retrieve the fiber, allowing it to separate from the matrix and reuse these fibers in a state similar to the virgin ones.

4.3 Carbon fibers, high structural performance but poor environmental footprint

Although carbon fibers comprise a minor share of the composite market, they represent non-negligible values and an even less negligible contribution to composites' environmental footprint. They are used for highly demanding applications where cost is secondary to performance.

4.3.1 Sourcing and production

Carbon fibers (CF) were initially developed out of the carbonization of cellulose fibers in the USA during the early 50s [7]. However, the most advanced materials were obtained from the development of acrylic-based fibers. In particular, the key stabilization of polyacrylonitrile (PAN) discovered by serendipity by Shindo and industrially developed by Toray from the end of the 50s in Japan led to continuous developments since then [7]. Today, PAN-based CFs now represent 90% of CFs, with the remainder being pitch-based and rayon (viscose). PAN is obtained from the free radical polymerization of acrylonitrile in an inert atmosphere with other monomers such as styrene, butadiene, or vinyl acetate. The thermoplastic is then transformed into a filament by wet spinning using dimethylformamide (DMF) as solvent. Acrylonitrile is obtained from propylene through the SOHIO's process (see Fig. 9). Pitch offers a more straightforward sourcing as it is a by-product of the coal industry and can also be bio-derived [97]. Mesophase pitch can be melt-spun to obtain primary fibers that can be later transformed into CF. However, pitch-based CFs do not match the PAN-based properties (see the section below) [98]. The final sourcing is rayon fibers (Lyocell and viscose fibers) obtained from regenerated cellulose [99].

In all cases, the overall process to yield CF remains consistent. The raw materials are first spun into fibers, typically using solvents (DMF) or mechanically through dry spinning. They are then stabilized, gradually oxidized to 350 °C, and finally carbonized. Carbonization is usually performed through step heating between 400 and 1600 °C. For high-modulus (HM) CF, an additional graphitization stage is performed at temperatures between 2500 and 3500 °C [101]. Surface treatment (plasma, oxidation) is generally performed to enhance the grafting of sizing agents (as for glass and basalt fibers). Finally, the continuous CFs are winded into tows, tapes, or any other reinforcement shape.

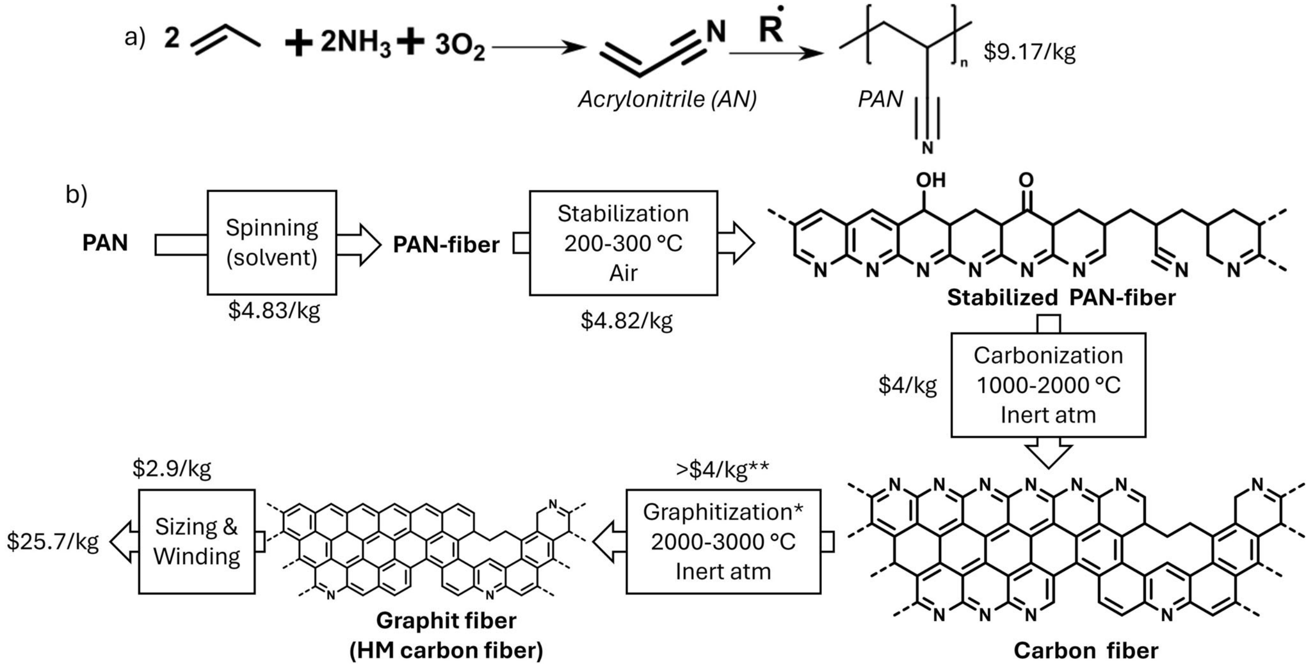


Fig. 9 Representation of the most common pathway to obtain carbon fiber. **a** Acrylonitrile synthesis by the SOHIO process and PAN polymerization. **b** Carbon fiber manufacturing process and representative macromolecular structures. Prices are taken from [100]. *Graphitization is optional, **no data available

4.3.2 Properties, advantages and limits, applications

Inherently, CFs are highly aromatic and crystalline carbon-based materials (carbon content > 92%). The high covalent energy of C=C bonds (145 kJ/mol) due to their aromatic nature results in outstanding properties. However, the process and the raw materials significantly influence the microstructure of the materials and, as a consequence, their properties. The understanding of the role of copolymerization in improving the oxidation/stabilization of PAN has led to the development of high-performance fibers after carbonization [102]. PAN-based CFs consist of highly ordered crystalline-oriented carbon layers, with amorphous regions structuring the networks and allowing efficient stress transfer between the regions, creating a “nano-carbon reinforced carbon” [103]. The crystalline domains are more refined, with less spatial separation between crystalline domains in PAN-CFs than in pitch-CFs, resulting in superior strength [104]. The defects at the surface act as crack nucleation sites, reducing the overall properties. In that sense, outstanding properties were achieved through the precise control of the microstructure together with the efforts in processing defect-free fibers, reaching 8 GPa of tensile strength for the Toray® T1200, with improved fracture toughness. Standard carbon fibers have a modulus ranging from 200 to 500 GPa, with tensile strengths between 3 and 6 GPa. However, CFs remain brittle with low elongation (0.5–1.5%), requiring high-security coefficients and over-dimensioning CF structures to prevent catastrophic

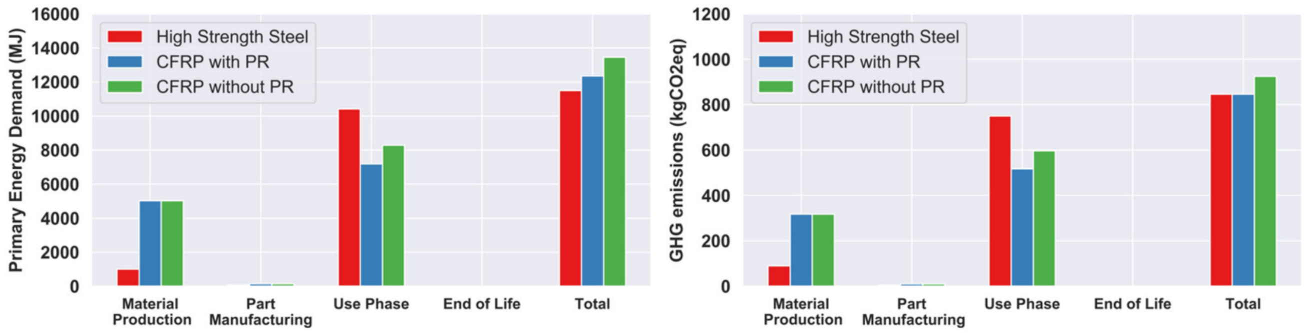


Fig. 10 Primary energy demand (left panel) and GWP emissions (right panel) for steel and CFRP components with and without powertrain resizing (PR) with 50% secondary mass savings. Reproduced with permission from [107]

failure. Interestingly, advances in molecular dynamics [105] along with experimental works [7], such as using carbon nanotubes to nucleate the fiber crystals, forecast continuous improvements. Perspectives towards reducing the fiber size, thus defect size, also predict future enhancement of CF properties [106]. These improvements could benefit other raw materials, such as pitch or lignin, to extend the portfolio of carbon fiber solutions through a better nanostructure control [103]. Along with the improvement in the engineering design of carbon laminates, CFs display a bright future for ultralightweight structures, but the sustainability of such structures must be considered (see Fig. 10).

4.3.3 Environmental footprint, sustainability and values for recycling

The general manufacturing process of CFs is not sustainable. The production of PAN and its monomers contributes to a substantial part of the CFs' footprints. In addition to their impactful fossil sourcing, PAN monomers are widely recognized as highly hazardous, posing problems in terms of waste, emissions, and workers' safety. Among structural materials, CFs possess the highest embodied energy and environmental footprint [108], as shown in Table 1. Even if the weight savings in the total life cycle demonstrate the benefits of using CFs over common structural materials such as steel or aluminum [11, 108], they yet do not align with net zero goals [109]. More efforts towards greener CFs are currently being made, from sourcing raw materials to processing the fibers. Producers predominantly focus on advances in process efficiency through higher energy efficiency, renewable energies, and carbon compensation. Main actors such as Toray, Hexcel, or Mitsubishi have set different goals. Hexcel targets a 30% global warming potential (GWP) reduction by 2030, while Toray plans to become carbon neutral by 2050. Mitsubishi had the most ambitious roadmap, with its KAITEKI program in 2021 aiming to achieve carbon neutrality by 2023 and carbon positivity by 2030 [110]. They reported a 26% reduction in GHG emissions in 2022. Since then, their carbon neutrality objectives have been reviewed, but no clear date is currently set. In general, producers do not share extensive data about the sustainability advances of carbon fibers.

Other exploration routes investigated by academic researchers are emerging, like the CO_2-sourcing of PAN [61], by utilizing algal carbon capture. In this process, algae are transformed into glycerol (and bio-diesel), methanol, propylene, and, ultimately, acrylonitrile. The authors performed a techno-economic study that showed the viability of the process, even at pilot scales. The environmental benefits were seemingly improved compared to traditional PAN synthesis routes.

Other works focus on new precursors for CFs, such as cellulose or lignins. Cellulose was one of the original precursors of CFs; now, a small quantity of CFs is produced from regenerated cellulose, which has a more controlled composition. However, the properties of these CFs do not compete with PAN-based CFs [7]. Regarding the regenerated cellulose, rayon fibers such as viscose or Lyocell® have a poor environmental footprint. Viscose requires carbon disulfide, a highly toxic gas, in addition to hazardous solvents and the generation of large amounts of wastewater [99].

The most promising route, so far, comes from lignin [111]. After cellulose, lignin is the second most abundant organic material. Vast amounts of waste lignins are produced each year as a by-product of the wood and paper industry, with limited exploitation of their value. This makes it a relevant material for the circular economy through the use and recirculation of waste streams. Up to date, several pioneering works have demonstrated the possibility of obtaining CFs from lignin, usually with a copolymer (such as polyurethanes) to facilitate the spinning and stabilization of lignin [112]. Although not competitive with PAN, satisfactory properties were obtained, with modulus ranging from 40 to 80 GPa, strengths up to 1000 MPa, and elongation between 0.5 and 1.4% [98]. Importantly, LCA demonstrated lower environmental footprints of lignin-based CFRP than PAN [108, 111, 113]. Depending on the process, GWP reduction between 20 and 60% was estimated, along with lower energy consumption.

Beyond their sourcing, the high embodied energy of carbon fibers makes them highly interesting for recycling to lower environmental impacts. Their high stability and mechanical properties forecast the potential of CFs for being reused in new applications while maintaining high performance. Although various recycling processes exist (as discussed later on), and should be adapted to the samples to be recycled (including the polymer matrix), the recovery and reuse/repurpose of carbon fibers seems to be consistently beneficial [28, 114, 115], and economically viable [116].

4.4 Natural fibers—the sole fully sustainable fiber for composites

Natural fibers are likely the oldest reinforcement in human history; for instance, straws were used to reinforce brick houses tens of thousands of years ago [117]. More recent examples date back to the first half of the 20th century, with the pioneering work of De Bruyne for aircraft structures [118]. During WWII, due to supply shortages, kraft paper/phenolic resins were used to replace aluminum in bombers. In 1941, Henry Ford, facing steel shortages, manufactured the conceptual "Soybean Car" made from natural fibers (Hemp and Ramie) and plastics partly made of soybean oil (among others) [119]. However, the project remains at the prototype stage. As noted in Section 2, NF currently represents 8-10% of the composite market. However, they are mostly employed as discontinuous (short) fibers and fillers. Since the early twenty-first century, driven by increasing environmental awareness, significant efforts have been deployed to master NF properties and extend their use in load-bearing applications.

4.4.1 Sourcing and production

Natural fibers come from a wide variety of sources, including various plants (such as flax, bamboo, kenaf) and different parts of these plants (bast, straw, seed, leaf, etc.) (see Fig. 11). Such diversity results in varying crop requirements, extraction methods, and main properties. In all cases, and unlike synthetic fibers, the length of NFs is limited by the finite size of the plants. One advantage of NF is the potential for locally sourcing natural fibers, depending on the crops available in a region (e.g., ramie in China, jute in India, and kenaf in sub-Saharan Africa...). For structural applications, bast fibers (flax, hemp, nettle, etc.) are typically the ideal candidates as they are rapidly grown and have well-known, established extraction pathways, including advantageous mechanical properties. So far, flax has been the most investigated fiber due to its broad (commercial) availability and superior properties, with hemp also emerging as a strong candidate. To simplify the readership, flax and, to a lesser extent, hemp will be primarily discussed, although other promising natural fiber exists [24].

Currently, Europe (France, Belgium, Netherlands) is the leading producer of flax fibers, representing two-thirds of the world's production [24]. A complete value chain that benefits textile know-how exists (specifically in Europe) and explains the rapid adoption of flax for composite over other fibers.

Significant work has been performed [120] to optimize fiber quality, properties, and economic outcomes through the careful selection of flax varieties, growth conditions, and extraction techniques. Natural fiber production can be divided into the following main stages: seeding, growth phase, harvesting (including retting), and finally fiber extraction [24]. At each stage, various parameters influence the future properties and economic values of the fibers. Before seeding, a variety selection is needed in accordance with the growing location and conditions. Interestingly, different varieties do not significantly change the properties of elementary fiber [121]. Despite height differences, oleaginous flax and textile flax displayed similar properties, allowing for the complete valorization of flax products and their integration into a circular economy. However, this should be carefully handled as lignification of the fiber occurs during the production of the seeds, complexifying the fiber extraction process. The seeding rate plays a crucial role in future properties [120]; a high seeding rate (>1800 plants/m^2) yields thinner flax stems with lower mechanical properties due to the competition between plants. On the other hand, a low seeding rate (<1200 plants/m^2) decreases the areal fiber production, thus reducing the economic returns.

The growth stage, from seeding to harvesting, also affects the fibers' properties. As the plant naturally grows, flax (and hemp) goes through several stages: germination, growth, flowering, seed formation, and aging [120]. Climate conditions (e.g., drought, precipitation, temperatures) strongly affect the development of the plants and, consequently, the fiber properties. Drought affects the morphology and chemical composition of the plants. Surprisingly, it has a negligible impact on the mechanical properties of the elementary fibers [122]. However, the yields were reduced, leading to variations in incomes for farmers and fluctuations in composite material prices. Hemp shows a similar growth stage to flax. However, the stem tends to be longer (about 1 m for flax, 2 m for hemp), with faster growth, greater versatility in soil types, and lower requirements in fertilizers [123].

Once the plants reach sufficiently advanced maturity, they are harvested. This includes uprooting, retting, and rolling. The most critical stage is retting, which involves fungi and microbial organisms degrading lignins and binding components by enzymes. Retting eases future fiber extraction

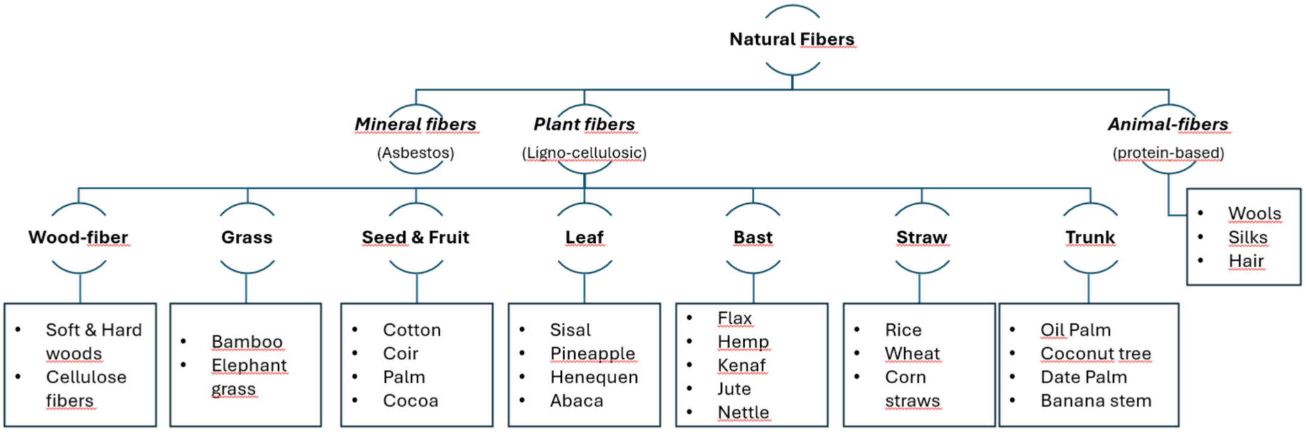


Fig. 11 Classification of natural fibers

by separating the wood from the fibers and increases the mechanical properties [125]. Retting can be performed in the open air, in the field of production (dew retting), or in a water bath with enzymes and chemicals (wet retting). Wet retting, the oldest technique, is more controlled than dew retting but was progressively banned (in Europe in particular) due to the large amount of wastewater and pollution involved [24]. Dew retting is now the preferred method to obtain flax and hemp fibers. It allows the easy separation of the technical fibers from the other components, but it must be perfectly controlled. Under-retting results in higher energy input and potential damage to the fiber during the extraction, while over-retting will downgrade the fiber's properties through the degradation of the fiber cell walls [125].

Finally, the flax stems are scutched to extract the structural fibers. Proper fiber separation is critical to obtain homogeneous fiber impregnation in subsequent composite manufacturing [126]. The scutching yields long fibers, short fibers, and shives, all of them being differently valorized. An additional step, hackling, can be performed to refine the flax fiber quality. Once scutched (or hackled) fibers are obtained, they can be processed to obtain reinforcement through spinning, weaving, or unidirectional tapes.

From seeding to harvesting, these stages are also critical in the environmental footprint of natural fiber in terms of land use, eutrophication, and water consumption and wastewater generation [127] (see Fig. 12). The energy required for seeding and harvesting is also accounted for. Scutching and hackling are major energy consumers in the production stages. Hackling is particularly energy-consuming but optional. Some works highlighted the low interest in hackle flax for composite applications, given its environmental costs [127].

4.4.2 Natural fibers as key materials in a circular economy

NFs, being inherently biobased, are widely regarded as sustainable materials. Up to now, the sustainability of NFs and their resulting composite materials can vary. As mentioned in the Section 4.4.1, the environmental footprint of NFs depends on the plant species, growing conditions, and treatment process. Flax and hemp are particularly promising as they require less water and fertilizers than other crops [117]. As a comparison, 600 to 1000 L of water is necessary to produce 1 kg of flax when the consumption for 1 kg of cotton is estimated to be 7000–29,000 L [127]. Using fertilizers and pesticides increases the eutrophication of water and should be limited to a minimum. Replacing GFRP with NFCs in case studies with similar mechanical performances demonstrated substantial gains in most sustainable metrics (except land use and eutrophication) [81, 127, 128]. However, it is somewhat unclear how the destruction of ecosystems and landscapes by sand extraction (glass fiber) is deemed, which could further favor NFs. The embodied energy of flax fibers is estimated to be around 10 times lower than glass fibers (4.4 vs 45 MJ/kg) [127]. From an economic viewpoint, flax benefits crop rotation and boosts farmers' revenues. However, the use of flax in composites is limited by competition with feed crops and textile use.

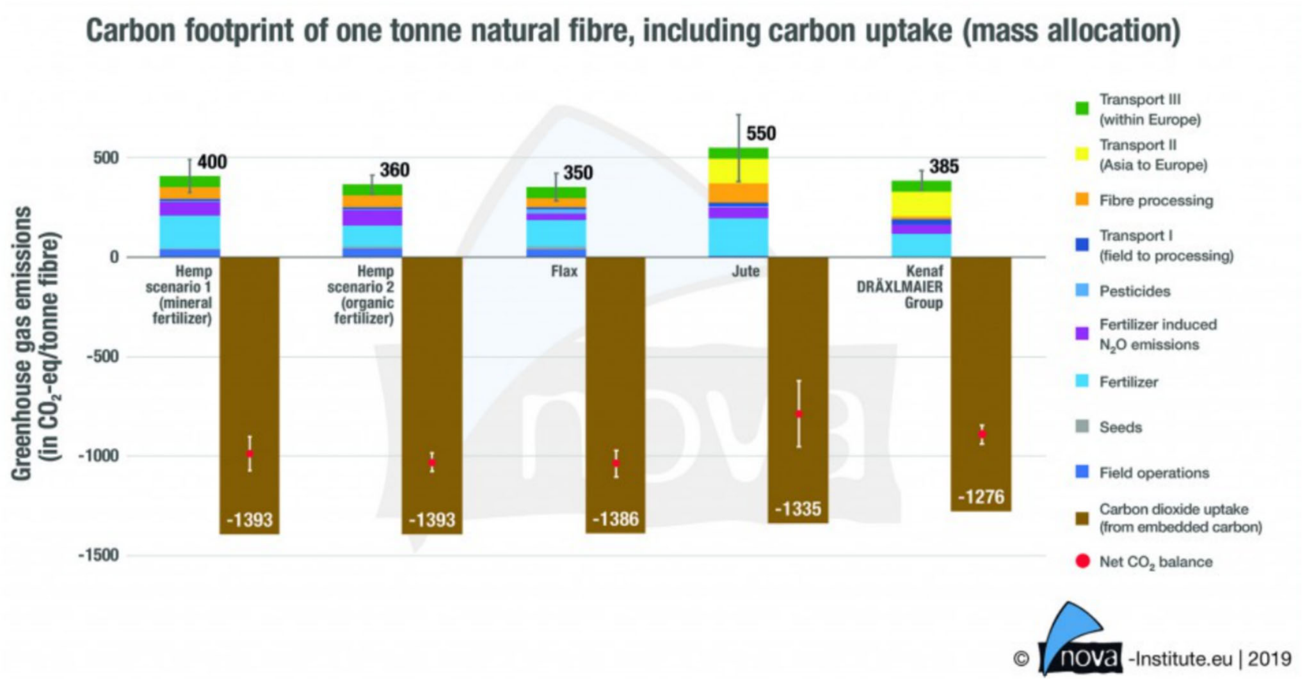


Fig. 12 Greenhouse gas emissions per tonne of natural fiber and carbon storage by photosynthesis. Copyright 2019 Nova-Institute.eu [124]

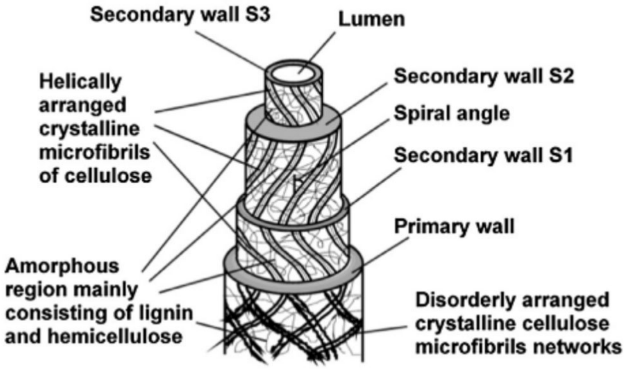


Fig. 13 Ultrastructure of a natural vegetable fiber cell, in which the secondary wall S2 makes up to 80% of the total thickness and thus acts as the main load-bearing component. Reproduced with permission from [131]

Hemp could further decrease the environmental burden of composites. Hemp is even considered more sustainable than flax as it requires less water and does not need fertilizers or pesticides [123]. The greater variety of hemp strains makes them adaptable to more climates, allowing hemp to be more accessible for global implementation. Hemp plants are taller than flax (2–4 m vs 1–2 m), resulting in higher fiber yield. In addition, different hemp parts can be valorized through multiple uses and markets (including pharmaceuticals) to ensure the full exploitation of their value and the competitive cost of their fibers [123]. However, hemp fibers are less developed than flax ones, and further developments are still needed. Other fibers, such as nettle [129], could also provide sustainable alternatives without competition for arable land.

4.4.3 Properties, advantages, and limits to their applications

Natural fibers are characterized by a complex ultrastructure that generates intricate behaviors and challenges in the design of natural fiber composites (NFC) [130]. They are highly anisotropic [24]. More importantly, the plant type greatly affects the chemical composition (lignin, cellulose contents), cellulose crystallinity fraction, and morphological structure (microfibrillar angle, cross-section shape), thus affecting the final mechanical behavior. The ultrastructure of plant fiber is shown in Fig. 13.

While cellulose crystallinity is crucial for providing strength and stiffness to the fiber, the microfibrillar angle (MFA) also plays a critical role. MFA represents the rela-

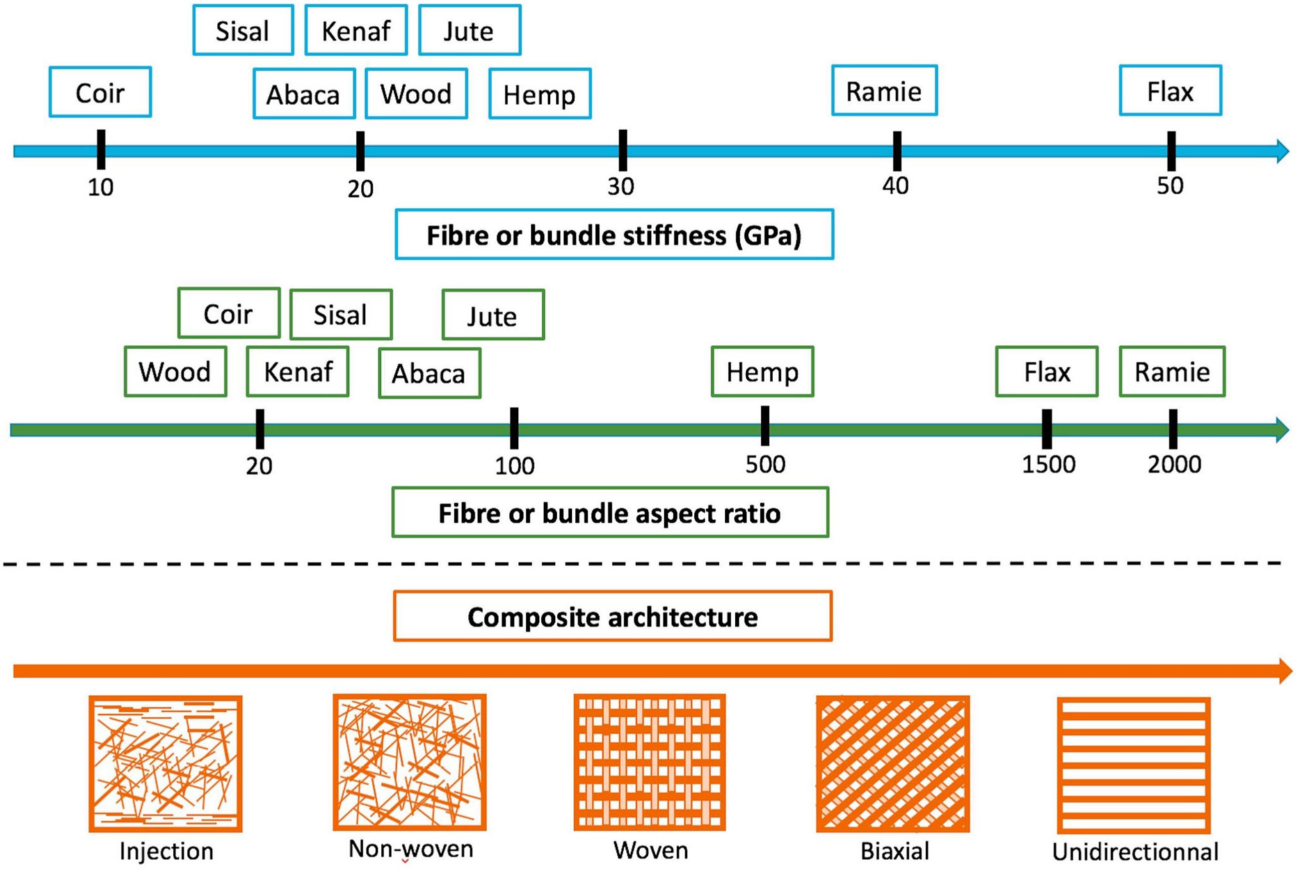


Fig. 14 Panorama of plant reinforcement's mechanical or morphological specificity and composite's architecture diversity. Reproduced with permission from [24]

tive angle between the constitutive cellulose microfibrils and the fiber on its own. To improve the mechanical properties of NFs, the MFA must be as close as possible to 0°, allowing the cellulose microfibrils to bear loads in their preferential directions. Typical MFA for flax range 2–7° [132] while hemp range 7–11° [24], with both 60–90% of cellulose (of which 60–70% crystalline). As a comparison, cotton possesses a higher content of cellulose (82–98%, including crystalline one) but a higher MFA (15–25°), resulting in a lower modulus (5–13 GPa).

As the mechanical properties of NF vary upon plant species, Baley et al. [24] proposed a hierarchization to simplify NF selection for composite design, as represented in Fig. 14. With a modulus ranging 30–70 GPa (average 53 GPa) and strength of around 1000 MPa, flax is usually considered to have the best mechanical properties, followed by ramie and alfa fibers [24, 121]. Hemp presents slightly lower mechanical properties (around 20–45 GPa in modulus and 800 MPa in strength), but the recent progress in the processing of hemp fibers, driven by European-funded programs (H2020 and CBE-JU, Ssuchy project), has shown promising results using hemp reinforcement [133]. While glass, basalt, and carbon fiber behave like pure linear elastic materials with a brittle behavior, natural fibers notably possess a non-linear visco-elasto-plastic behavior [134, 135]. This visco-elastic behavior gives NFC better damping properties than synthetic fiber composites and metallic materials [136], which is an advantage in some applications requiring insulation and vibration damping. A bilinear behavior (with two apparent moduli, E1 and E2) is typical in tensile loading. A stiffening effect, reported for flax [137] and hemp [134], exists when subjected to repeated load. Although well known, its pure origin is still debated in the scientific community but is mainly attributed to the MFA reorientation under load [135, 137, 138].

Although the mechanical properties are significantly lower than those of glass fibers, their specific properties (i.e., relative to density) are competitive [139]. Specific properties are commonly used metrics for designing sustainable structures as they directly relate to the strength (or stiffness)-to-weight ratio. In the frame of a circular economy, other metrics should be considered in the design of materials, in particular Global Warming Potential (GWP), such as the strength-to-GWP ratio or embodied energy/GWP-related metrics [76]. To help such design, Shah proposed an Ashby approach for composite, including specific properties (see Fig. 15, and eco-impact related to processes (see Sect. 7). The normalization of mechanical properties to eco-impact data is given in Table 1.

Some specific behaviors and properties of NFs further influence the design of structural natural fibers. The organic composition of natural fibers, primarily consisting of carbon

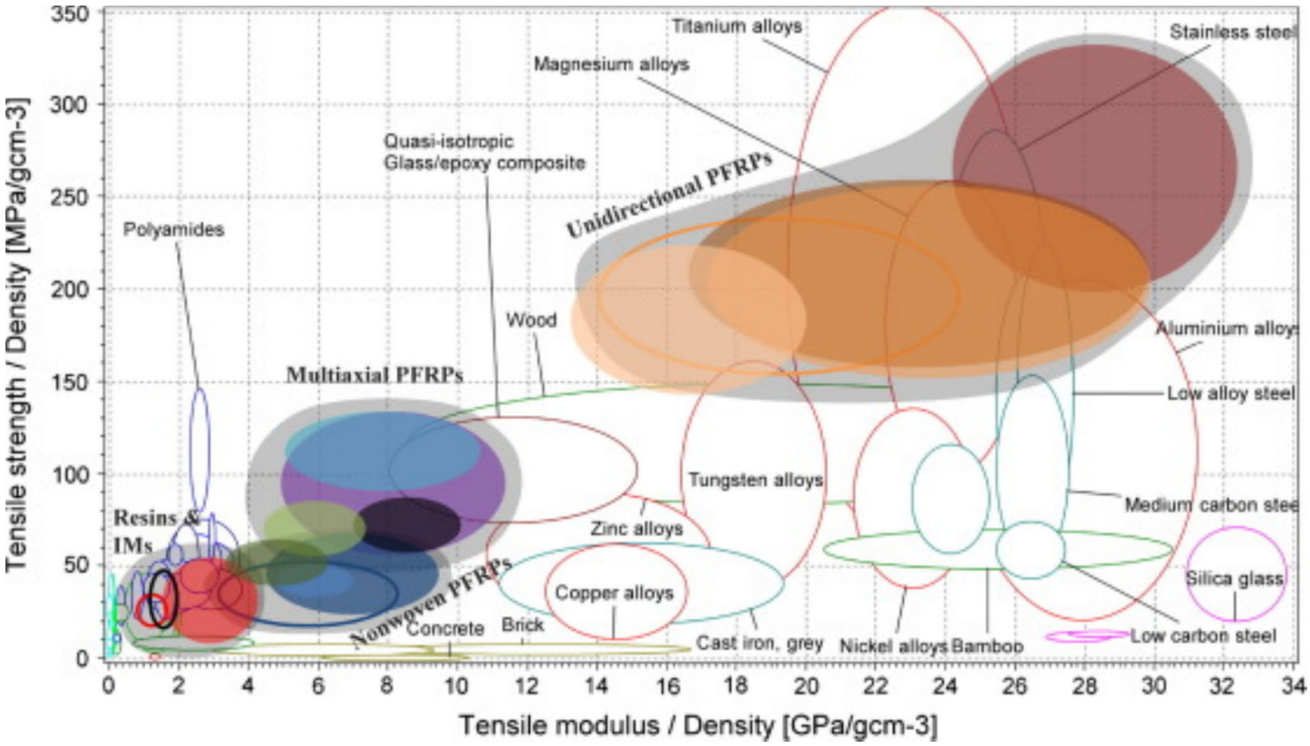


Fig. 15 Ashby plot comparing the specific tensile modulus and specific tensile strength of NFC (filled balloons) with various other engineering materials (unfilled balloons). Reproduced with permission from [140]

Table 1 Overview of reinforcement properties and eco-environmental impacts range (typical values; flax, elementary; hemp, bundle)

Material	Absolute properties range (typical value)						
	Density g.cm^{-3}	Tensile Modulus GPa	Tensile Strength MPa	Elongation (break) %	Embodied Energy MJ.kg^{-1}	GHG kgCO2eq. kg^{-1}	Cost €.kg^{-1}
Carbon fiber (HS)	1.8	200–300 (250)	4000–6000 (4800)	1.0–2.0 (1.8)	100–1500 (1170)	30–80 (72)	25–50 (35)
Carbon fiber (HM)	1.9	350–600 (500)	3000–5000 (4000)	0.4–1.5 (1.2)	500–1500 (1450*)	50–100 (85*)	50–130 (65*)
E-glass	2.6	70–76 (72)	3300–3700 (3400)	4.0–5.0 (4.8)	30–50 (44)	2.650	1.1–2.8 (1.8)
S-glass	2.5	80–90 (85)	4300–4900 (4500)	4.0–6.0 (5.2)	30–50 (54)	3.140	2.0–3.5 (3.3)
Basalt	2.6–2.8	90–100 (92)	3100–5000 (4800)	3.1–6.0 (4.5)	12–52 (24)	0.02–1.0 (0.955)	2.5–3.0 (3.0)
Flax	1.53–1.56	30–70 (53)	600–1500 (970)	1.6–3.6 (2.15)	3–10 (4.4)	–1.4 (0.5)	6–10 (9.08)
Hemp	1.4–1.6	15–40 (28)	300–1100 (800)	0.8–3.3 (2.3)	3–10* (4.4*)	–1.4* (0.5)	0.5–10* (9.08*)

Table 1 continued

Material	Specific properties		Properties per unit eco-impact		Properties per cost		Ref
	Specific modulus	Specific strength	Modulus/ (GHG * Density)	Strength/ (GHG * Density)	Modulus/ (Cost * Density)	Strength/ (Cost * Density)	
	GPa/ (g.cm^{-3})	MPa/ (g.cm^{-3})	GPa/ (kgCO2eq. kg^{-1}. g.m^{-3})	MPa/ (kgCO2eq. kg^{-1}. g.m^{-3})	GPa/ (€.kg^{-1})	MPa/ (€.kg^{-1})	
Carbon fiber (HS)	139	2667	2.0	37	4.0	76	[37, 108, 158, 159]
Carbon fiber (HM)	263	2105	3.1	24.8	4.0	32	[37, 108, 158, 159]
E-glass	28	13237	10.5	500	15.5	735	[68–70, 76, 90, 91]
S-glass	33.5	1779	11	566	10	539	[68–70, 76, 90, 91]
Basalt	35	1811	35	1897	11	604	[89–91, 94, 95]
Flax	35	634	69	1268	3.9	70	[24, 127, 160]
Hemp	20	571	40	1143	2.2	63	[24, 123, 127, 160]

and oxygen from sugar-based cellulose, hemicellulose, along with aromatic lignin, makes them susceptible to fire. [141]. Additionally, their thermal stability is lower than that of synthetic fibers, as their degradation starts at temperatures above 200 °C [142, 143]. NFs are generally considered for applications under temperatures below 100 °C [144]. Additionally, the high number of hydroxyl functions makes them sensitive to moisture, leading to undesirable weight gains, fiber swelling, and some plasticization effects [145]. However, in the case of flax, it was shown that moisture saturation did not reduce the fiber tensile strength or fatigue properties in the longitudinal direction [146] but decreased the stiffness. Moisture can also significantly reduce the transverse properties through the weakening of the interfacial strength [147]. Hydrothermal and hygrothermal aging, particularly cyclic processes, can significantly degrade both static and fatigue properties. This was attributed to composition changes within the fiber but, more importantly, to the cyclic swelling of the fibers that generate cracks within the matrix (whose swelling is much less pronounced than NF) and the debonding of the fiber-matrix interface (Fig. 16). These factors should be considered when designing natural fiber composites, including the importance of fiber-matrix adhesion in NFCs. Moreover, designing matrices able to withstand such swelling-shrinking cycles and maintain cohesion over time could benefit long-term behavior.

4.4.4 Natural fibers in structural composites—reaching their full environmental potential

The reinforcement generally represents 50–75% of the mass in structural composites (continuous). In this regard, it is evident that most work mainly focuses on the sourcing, design, and reinforcement process to improve the composites' environmental score. NFs appear to be one of the most promising reinforcements for most composite applications, offering the potential to efficiently replace glass fibers with promising ecological benefits.

Many works have explored fully or partially bio-based matrices with NFs to ensure sustainability when using NFCs. Both thermosets and thermoplastics have been investigated. Thermoplastics offer several advantages. By nature, thermoplastics are recyclable, which simplifies the EoL scenario through direct thermo-mechanical recycling and provides lower LCA over multiple cycles [149]. However, thermo-mechanical recycling involves the decrease of fiber length and the degradation of the properties between the original continuous fiber composite, meaning that the recycled one must be only used in non-structural applications [150]. Solvolyse could help separate the fibers from the thermoplastic, but it has been poorly investigated, and the use of (hazardous) solvents is debatable. Polylactic acid is biobased and bio-compostable, making it an ideal candidate for biobased composites [148, 151]. However, in all cases, the poor interfacial adhesion between non-polar thermoplastic polymer and polar NFs leads to unsatisfying properties compared to the potential of NFs [152]. In addition, thermoplastic processing requires high temperatures to reach a satisfying melt viscosity to impregnate the fibers, which complicates the processing. Starting from thermoplastic monomers such as lactide to PLA [153], or methyl methacrylate to PMMA (Elium® by Arkema, [150]) offers some processing advantages while keeping the recyclability aspect of thermoplastics. However, their properties remain less competitive compared to thermoset matrices.

Thermosets, and in particular epoxy resins, provide improved wetting of the fiber and, hence, better properties [154]. Several works have exploited partially bio-based epoxy [155–157], some of them being already commercially available such as the Sicomin GreenPoxy® [64, 154].

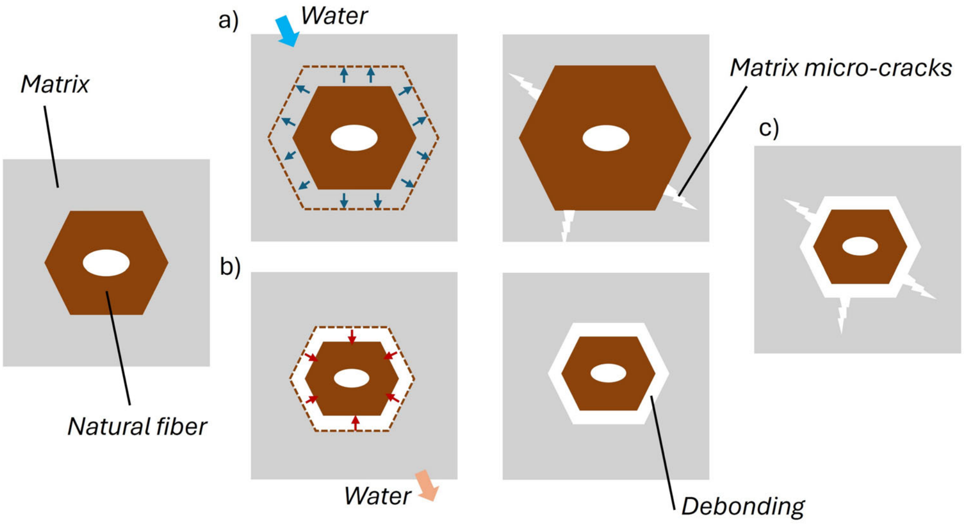


Fig. 16 Influence of NF hygroscopic behavior on the interface and composite integrity [148]. **a** Micro-cracks due to moisture swelling, **b** decohesion caused by moisture desorption and shrinkage, and **c** combined mechanism through aging and cyclic sorption–desorption

Compared to glass fibers, similar specific properties were obtained, with environmental benefits in cradle-to-gate scenarios. Yet the recyclability of thermosets is an issue, often resulting in the landfill or incineration of biobased composites once decommissioned.

Finally, it is important to note the wide diversity in sourcing and properties of fibers suitable for structural FRPs. This diversity allows the selection of different fibers adapted to the applications. To date, the most important criteria are either costs or performances, favoring the use of glass fiber in the former and carbon fiber in the latter. However, they possess the highest environmental impacts (Fig. 17). Other criteria such as embodied energy, resource depletion, and GWP should ideally be considered by engineers to design more sustainable structures. Regulations and tax incentives policies could trigger the designing criteria to foster the production and use of plant and basalt fiber, that display competitive properties when properly used. The far superior properties of carbon fiber make them, currently, challenging to replace in their main applications. In that sense, the first R's strategies (Refuse, Rethink, Reduce) and the last (Reuse, Repurpose, Recycle) should be prioritized, i.e., limit the use of carbon fibers as a last resort, maximize use, and ensure recycling. For most applications, natural fibers are the most sustainable pathways in Europe with negative GWP and could help reach carbon-neutral goals. However, efforts are still required to match the fiber advantage with the matrix footprint.

5 The complex case of fiber/matrix interface and the issue in natural fibers

The fiber/matrix interfacial adhesion is critical in FRPs, but it is probably the most complex aspect to apprehend [30].

Various physical and chemical phenomena influence the interface (Fig. 18). Covalent or ionic bonds [161, 162], intermolecular bonds such as hydrogen bonding, Van Der Waals interactions, or molecular entanglements, but also interdiffusion, electrostatic interactions, and mechanical interlocking affect the interfacial strength. [31, 163]. Good interfacial adhesion between the fiber and the matrix allows the efficient transfer of external loads to the fibers [164]. Through an efficient stress transfer, the fibers responsible for most of the high performance of FRPs can be exploited to their full

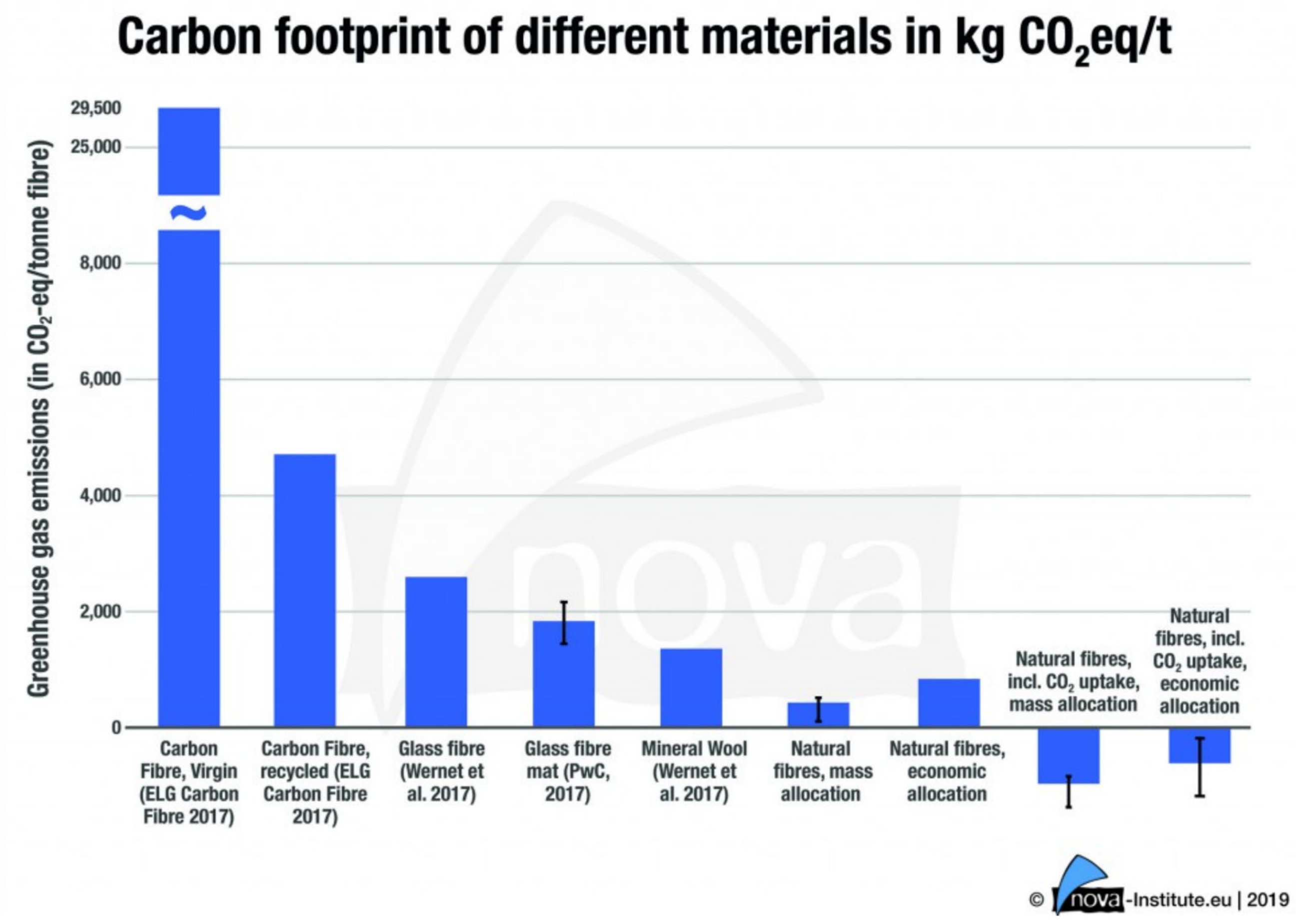


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Fig. 17 Greenhouse gas emissions per tonne of fiber used for composite application. Copyright 2019 Nova-Institute.eu [124]

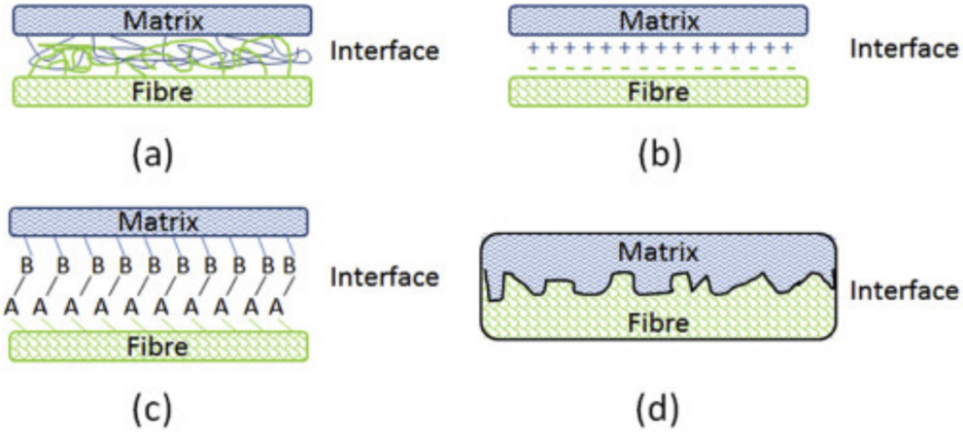


Fig. 18 Fiber-matrix interfacial bonding mechanisms: **a** molecular entanglement following interdiffusion, **b** electrostatic adhesion, **c** chemical bonding, and **d** mechanical interlocking. Reproduced with permission from [168]. Copyright 2016 Elsevier

potential, maximizing the composite properties. The interface, particularly its shear strength and toughness, governs failure mechanisms of composites [165]. Higher interfacial strength yields an increase in strength in all types of solicitations. The improvements are more pronounced in the off-axis directions, where the interface is directly solicited. High adhesion leads to a shift from interface-dominated to matrix-dominated failure modes [165]. In addition, poor interfacial adhesion can facilitate the nucleation and propagation of (micro)cracks that drastically degrade fatigue properties [26]. It is important to note that “too-strong” adhesion is not necessarily beneficial as it leads to brittle failure of the materials, while fiber pull-out and partial delamination can facilitate increasing the toughness of FRPs [88, 166]. Therefore, designing a high-strength interface with improved toughness is pivotal in composites. Through chemical sizing, physical modifications, and processing steps, the interface of synthetic fibers has been extensively optimized to reach the best properties [167]. In parallel, the matrix chemistries were adapted and modified to fully combine with fiber sizing and maximize the performance. The combination of fiber nature, sizing treatment, and matrix chemistry should be considered when developing new composite systems. Indeed, “high-performance” matrices, or new chemistries (for instance, among CANs), could not be suitable if the interface is of poor quality. More importantly, one must keep in mind that the sizing of synthetic fibers is a critical step and often an industrial secret, allowing competitors to differentiate themselves from each other, making the sharing of information on the chemical composition almost impossible. Therefore, new chemistries should, in an ideal case, be compatible with sized commercial fibers, thus close to UPR and EP chemistries in chemical affinities.

By comparison with synthetic FRPs, in which their structure is more controlled through mastered sizing processes, the interface properties in NFCs are even more problematic to investigate and tailor. The intricate geometry of natural fibers results in greater variability and less reliable outcomes. Moreover, the poor interfacial compatibility between NF, which is generally hydrophilic, and the polymer matrix, which is often hydrophobic, hampers the maximization of NF properties.

Most chemical modifications of NFs induce lignin or hemicellulose removal by alkaline or acidic treatments [169, 170]. Additionally, chemical grafting can enhance fiber and matrix compatibility. For instance, thanks to the reactivity of the -OH groups of cellulose, silane treatment can yield amino-terminated or epoxy-terminated functions on the fibers, making them more compatible with epoxy resins [171]. Carboxylic acids or anhydrides, acetylation, and esterification can enhance compatibility with polyester resins and polyolefin thermoplastics. It increases roughness and mechanical interlocking, decreases the water uptake of NFs, and increases the dimensional stability [170]. Other chemical treatments have also been explored by reacting through isocyanates, acrylonitriles, peroxides, and benzoylation [172, 173]. Improvements ranging 10–30% of strength increase and a diminution of up to 50% in water uptake are typically reported.

However, these results should be taken with care as most of the studies are performed on short fibers ($<20\,\text{mm}$) or non-woven randomly oriented reinforcements where even a slight increase in the adhesion strength can lead to improvements [174]. Additionally, the sustainability and relevance of such treatments from economic and environmental viewpoints have been poorly discussed [175] but were shown to severely affect the environmental footprint. The toxicity of the compounds involved and waste chemicals might hamper the obtained benefits. NFs are mainly investigated to reduce the environmental footprint of composites. To ensure competitiveness with GF, cost must be minimized [173]. Therefore, the interest in applying additional chemical steps should be investigated and avoided whenever possible.

6 Matrix—the key towards sustainable composites in a circular economy?

The need to shift from a linear to a circular economy requires thinking more globally about the design of composite materials. The macromolecular engineering of resins is critical for ensuring the implementation of composites in a circular economy. In an ideal case, resins must be sourced from renewable feedstocks (bio-CO_2-derived and/or post-consumer waste), scalable, and adaptable to existing composite manufacturing processes (drop-in), all while delivering the expected properties. In addition, integrating recyclable by-design systems would facilitate the recovery of valuable compounds at the EoL, ideally, all of them, i.e., fiber and polymer building blocks. This comprehensive approach would not only reduce the consumption of fossil resources but also enhance the durability of structures and create value at the end of their service life by transforming them into new resources for materials with properties similar to virgin materials.

6.1 Toward a complementary and sustainable value chain for polymer building block sourcing

Matrices for structural materials are mainly polymers containing heteroatoms (through the ether, amine, ester linkages). Some identical challenges can be identified, particularly in the sourcing of these matrices. Epoxy, polyesters (linear and unsaturated), polyamides, and polyurethanes require either (poly)amines, diols/polyols, or carboxylic acids.

For example, bisphenol A (BPA) is a typical component of epoxy, benzoxazines, phenol-formaldehyde, and vinyl ester resins and can also be used in polyurethanes. 4,4’-Methylenedianiline (MDA) is a typical hardener in epoxy resins and is used to produce methylene diphenyl diisocyanate (MDI). All these substances are petro-sourced and are currently under scrutiny due to their toxicity, carcinogenicity, and neurotoxicity. Identifying better sourcing for polyols, phenols, and amines is vital to achieve more sustainable building blocks. Their hazardousness should also be considered to avoid health accidents similar to those experienced in the past [184].

The importance of sourcing chemicals from renewable resources has already been mentioned. Two main strategies are essential. The first one aims to switch from petro-based sourcing to renewable sourcing of the same chemicals [18, 185]. This strategy focuses on efficient chemical pathways to obtain competitive biobased compounds. The customers can readily use them as they remain the same compounds as the petro-based version. This is, for example, the case of bio-aniline and bio-phenol to obtain MDA and BPA, among others. However, this does not address health considerations. Industries mainly deploy this strategy. Moreover, the processes can adversely affect the actual environmental benefits, which are often undisclosed by sectors.

The second strategy consists of finding biobased alternatives to existing compounds. Most of these researches are performed by academics and typically have a lower Technology Readiness level (TRL), making it more complex to implement in the industry as the value chain does not necessarily exist. However, it is often the most advantageous strategy as it contributes to the valorization of waste products such as lignin and can help reduce the toxicity of the chemicals used [186].

For both strategies, certain resources and pathways are of particular interest in the fields of polymers (and, subsequently, composites) as they might provide relevant and steady quantities through industrialization processes (keeping in mind the reduced consumption associated with a circular economy) [59, 187]. Namely, lignins, non-edible (or mass-produced) carbohydrates (cellulose and starch), and CO_2 are promising chemical platforms for sourcing monomers that are suitable to replace existing petro-based monomers. Additionally, terpenes, vegetable oils, and miscellaneous agricultural waste fermentation can provide valuable chemicals for the polymer industry [58].

6.1.1 Lignin, from waste to valuable phenols and amines

Lignin is a plentiful biopolymer produced in large quantities by the paper industry. It is the most abundant source of aromatic chemicals [188]. About 100 Mt are produced annually, with the most being used as a low-grade fuel [189]. The depolymerization of lignin into valuable compounds and their separation is challenging [190]. Different processes exist, such as kraft lignin or soda lignin, but discussing the different depolymerization processes, while critical, is beyond the scope of this work [189].

Several phenolic compounds have been obtained from lignin that can effectively serve as platform chemicals, as schemed in Fig. 19. Lignin is principally constructed of syringyl, guaiacyl, and hydroxyphenyl primary compounds. However, depending on the lignin source and the extraction process, there is a too large variety of compounds extracted [194]. Extraction yields are generally low and produce considerable quantities of by-products [195]. The catalytic fractionation of lignin is valuable through the efficient valorization of these products [196] while reducing GWP emissions. Innovative pathways have emerged to stabilize the depolymerization of lignin and promote high yields and well-defined chemicals through in-situ functionalization [190, 197]. This strategy produced biobased polyamides with a reduction of GWP and economic advantages compared to conventional PA-6,6 [198].

From lignin, guaiacol is an exciting compound [186, 192, 199]. Guaiacol is analogous to phenol substituted by a methyl

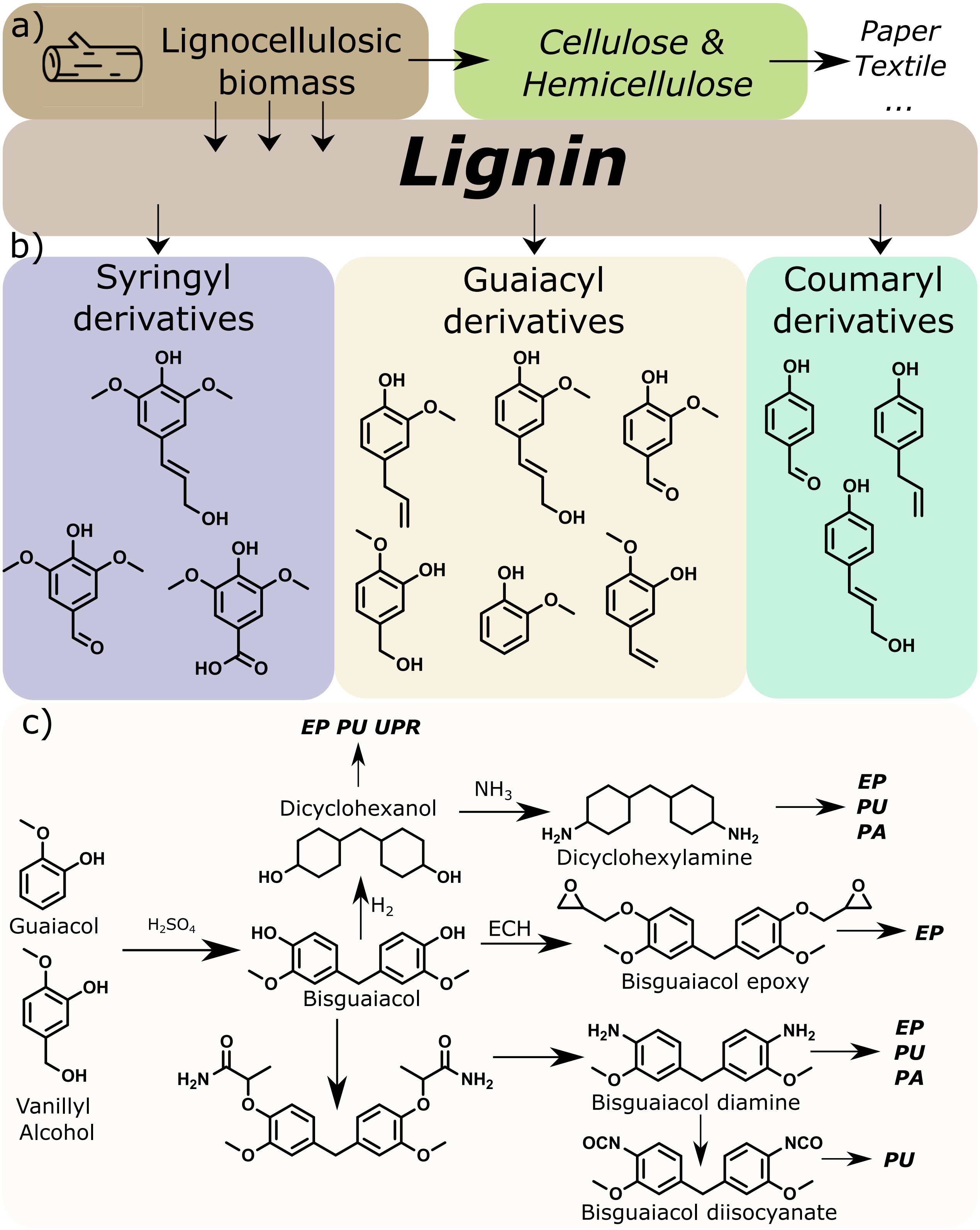


Fig. 19 Lignin valorization into key building blocks for composite applications. **a** Schematic representation of the lignocellulosic value chain, with lignin as a major by-product. **b** Typical lignin constituents and small phenolic compounds. **c** Catalytic valorization of guaiacol compounds into key monomers for composite and polymer applications. The bisguaiacol pathways are summarized from [191–193]

ether in the ortho position. Guaiacol can efficiently replace BPA [186, 200], but can also be aminated to replace MDA and MDI [192] (see Fig. 19). Other phenolic compounds, such as vanillin [201] or eugenol [202], demonstrate promising potential for producing sustainable materials.

In addition to phenols, amines can also be produced from lignin [199, 203]. As previously mentioned, phenolic can be aminated through Williamson etherification and Smiles rearrangement [192] to obtain aromatic amines. The group from Barta explored the use of Raney Nickel to catalyze both the hydrogenation of phenol into cycloaliphatic alcohols and their subsequent amination in the presence of ammonia [204, 205]. Despite being potentially biobased, no environmental assessment was found using ammonia, which might hamper benefits [206], due to the fact that ammonia production is one of the most polluting chemical processes [207]. Nevertheless, the direct fabrication of primary amines from alcohol is promising for polymers and composites, as they are the most challenging compounds to replace for epoxy resins.

6.1.2 Carbohydrates and polysaccharides, easy access to precious building blocks

Polysaccharides (cellulose, hemicellulose, starch, chitin) are the most abundant biopolymers on earth, accounting for 70–

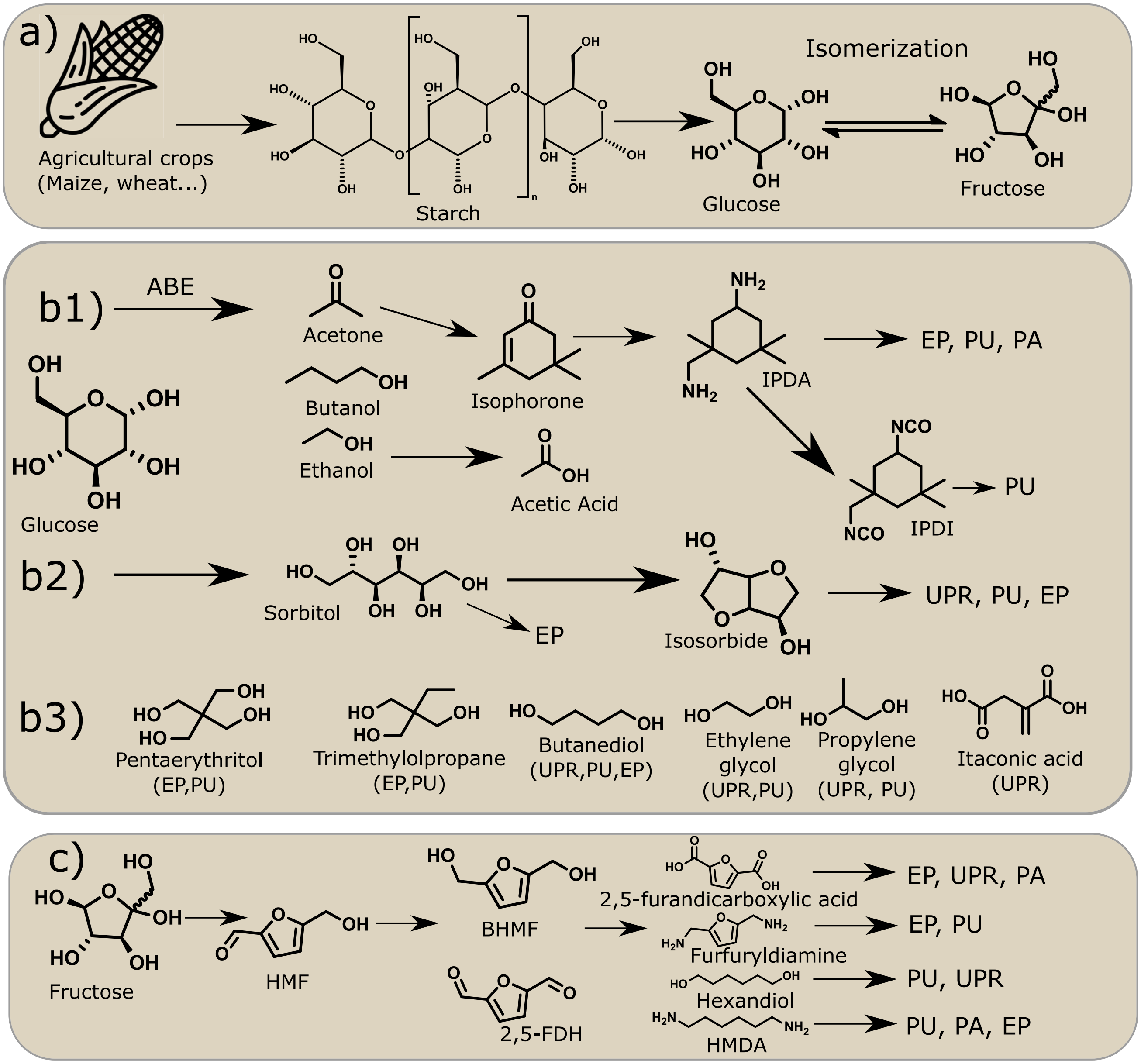


Fig. 20 From crops to valuable compounds. **a** Starch fermentation to glucose and fructose, **b** glucose route, **b1** acetone-butanol-ethanol (ABE) process to isophorone diamine, **b2** sorbitol route, **b3** various compounds from glucose, and **c** fructose route to furanic compounds

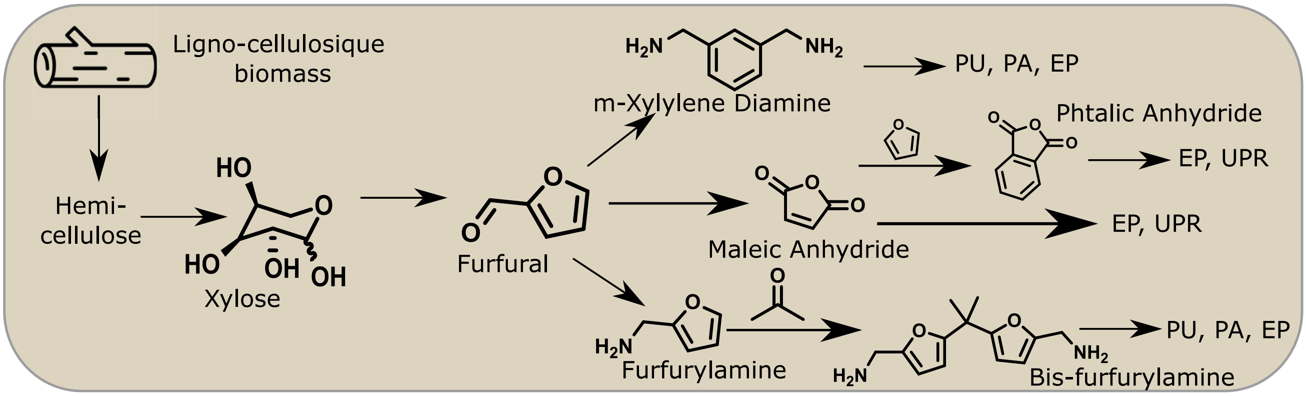


Fig. 21 Hemicellulose route to furanic compounds

80% of the total biomass. They are the primary source of energy for living organisms but also offer a renewable feedstock for the polymer industry.

Typically, the use of such resources begins with the depolymerization of polysaccharides into simple sugars such as glucose/fructose (from starch and cellulose, Fig. 20) or xylose (from hemicellulose, Fig. 21). Subsequently, these sugars are transformed into more valuable compounds for industries. All these transformations can be developed through bio-fermentation (in the presence of bacteria, yeast, etc.), enzymatic, or chemical pathways. Common challenges are related to scaling and managing by-product generation. The toxicity of produced compounds to fermenting organisms typically imposes highly dilute media that increase energy consumption and lower yields [208]. It is also crucial that competition with food production is avoided on a large-scale perspective. Moreover, many crops, such as maize, have detrimental environmental footprints due to the use of large quantities of water, fertilizers, and pesticides.

Over the last 20 years, numerous chemical compounds have been obtained from glucose, and some are industrially produced or at a pilot scale. The most famous example is the production of polylactide (PLA), from the fermentation of starch to lactic acid [209]. PLA is employed as a bio-compostable (and sometimes biodegradable) composite, often combined with flax [148].

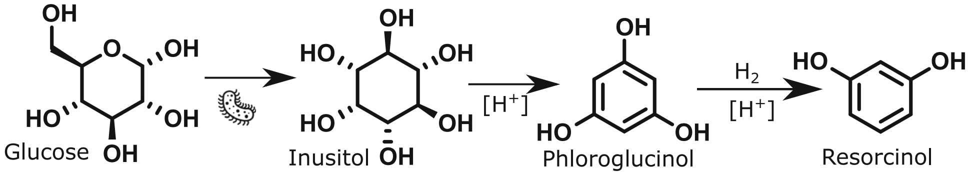
Sorbitol [210] and isosorbide [211] are also promising building blocks to provide polyol and amines used in epoxy, polyurethane, and polyester resins. Various polyols are obtained through the fermentation of starch by bacteria, leading to trimethylolpropane, pentaerythritol, and butanediol [208]. These are standard components for epoxy resin and polyurethanes. Biobased butanediol is produced on a large scale and represents an important fraction of partially biobased epoxy resins. The long-time known acetone-butanol-ethanol (ABE) fermentation process is also regaining industrial interest [212]. From bio-acetone, Evonik Industries AG is now producing bio-isophorone, a mass-produced precursor of isophorone diamine and diisocyanate, typical crosslinkers in epoxy and polyurethanes.

Phloroglucinol and resorcinol, which are potential aromatic alternatives to BPA, are also produced from glucose (see Fig. 22). They have already shown significant promise in the preparation of epoxy resins [157] and other high-performance resins [213, 214]. However, the carcinogenicity of these phenolic compounds remains a major concern [215].

Aniline, a multimillion-ton produced aromatic amine, mainly for epoxy and polyurethane resins, has been successfully produced and commercialized by Covestro AG from the fermentation of glucose [217] (see Fig. 23). The GWP was shown to be reduced when compared to traditional petro-based aniline [218].

Finally, furanic compounds are emerging as a versatile platform for various applications as they can provide numerous building blocks. Alternative pathways to petro-based existing compounds, such as m-Xylylene diamine (mXDA) [219], maleic and phthalic anhydride [220], and hexamethylenediamine (HMDA) [221] were demonstrated. However, these pathways may include multiple steps and may not be industrially or environmentally relevant. The

Fig. 22 Phloroglucinol and resorcinol production from glucose, adapted from [216]



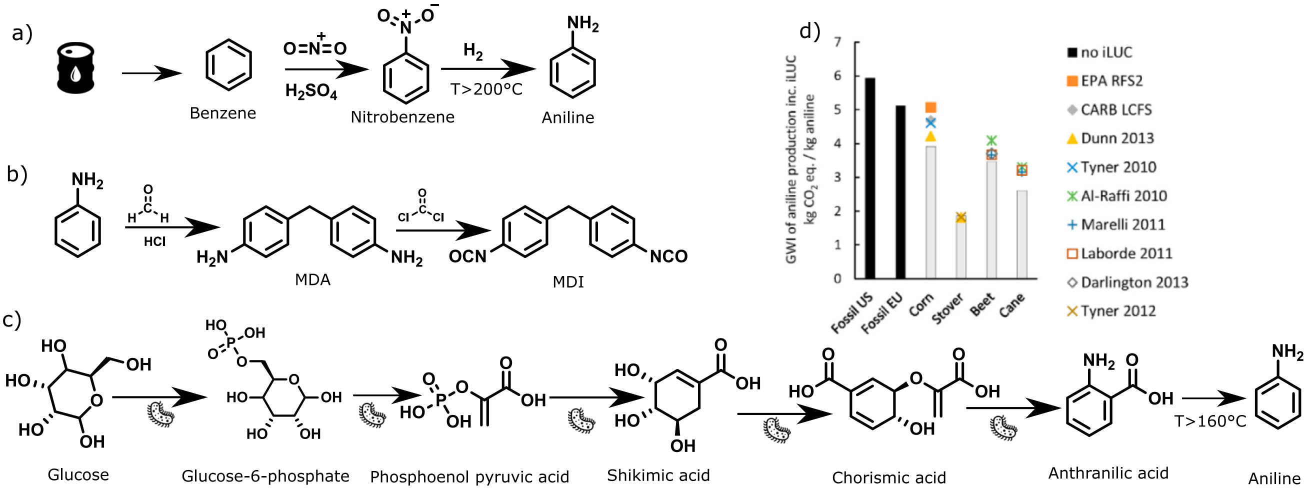


Fig. 23 **a** Conventional petro-based pathway for aniline, **b** transformation of aniline to typical polymer building blocks, **c** fermenting process from glucose to anthranilic acid (simplified from [217], and **d** GWP comparison of petro-based aniline and biobased one [218]

overall yields of mXDA and the 5-step process from furfural may hinder the practical applicability [219]. The synthesis of HMDA from fossil resources was demonstrated to be more economically and environmentally friendly than from biomass [221], and it is currently produced at pilot scales by Coverstro AG and Genometica.

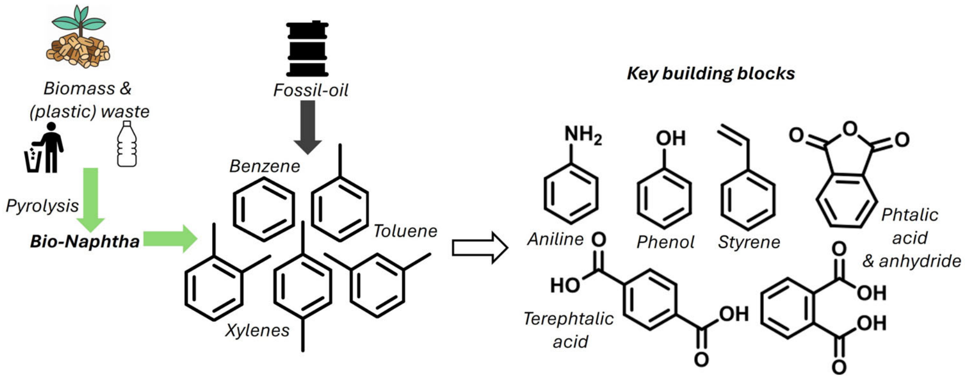
Furan compounds can also provide new chemical structures with a lot of promise. For instance, 2,5-furan dicarboxylic acid (FDA), an aromatic diacid that brings stiffness and thermal stability to polyester [222], has been shown to be advantageous compared to its petro-based counterparts, terephthalic acid [223]. FDA provides high thermal stability as a replacement to BPA in epoxy [224]. Amine crosslinkers can also be obtained to replace conventional aromatic diamines [225, 226].

6.1.3 Pyrolysis by-product valorization

Complete recirculation of organic compounds is not feasible. Vidal et al. [19] have already acknowledged that some polymers cannot be recycled. Moreover, certain waste materials might not be environmentally or economically viable to recirculate. In these cases, pyrolysis could be a realistic ultimate solution.

The thermal decomposition of biomass and organic waste in an inert atmosphere has emerged as a promising pathway for producing BTX (benzene, toluene, xylene) from renewable sources (see Fig 24). The technique effectively converts lignocellulosic biomass into aromatic hydrocarbons [185]. By exploiting pyrolysis to recover valuable aromatic compounds (and small molecules of gas), the process can complement chemical extraction and waste management efforts [19]. Notably, the production of bio-naphtha and bio-BTX from renewable feedstocks has been successfully developed by Neste Oyj, a Finnish oil refiner with several patents to control the composition, even in the presence of nitrogen and oxygen impurities [227, 228]. From this bio-BTX, bio-phenol was produced and applied by Covestro for BPA-derived plastics [229].

Fig. 24 Biomass and waste valorization by pyrolysis to bio-naphtha and key building blocks for composite matrices



6.1.4 Carbon capture and utilization, exploring CO_2 as a valuable chemical

The use of carbon dioxide in chemistry has a long history [230] and is used for a variety of compounds, some of them being commercialized (see Fig. 25). CO_2 is a key element in urea production from ammonia, representing nearly 75% of the 150 Mt produced annually [231]. Other chemicals such as salicylic acid (50%), methanol, dimethyl carbonate, and ethylene carbonate are industrially produced from CO_2, albeit on a smaller scale [231]. The detrimental overproduction of CO_2 has pushed policies to encourage industries and academics to increase efforts in efficient carbon capture and utilization (CCU). Significant progress has been made towards the capture of CO_2 either from industrial processes or from open-air, using biotechnologies, amines, and covalent organic frameworks among others [232, 233]. CO_2 is remarkably stable (−394 kJ/mol), which makes its activation complex and economically difficult without strong governmental policies [232]. Despite this, CO_2 remains a promising chemical that could help partially decarbonize the composite industry.

Although mainly limited to academia, CO_2 has been proven to be a valuable compound for the production of building blocks in advanced polymers [235]. High molar mass linear polycarbonates were produced from CO_2 and oxides, presenting advantageous properties when compared to BPA-phosgene derived polycarbonate [236, 237]. Acrylic acid and methacrylic acids, key components for the preparation of unsaturated polyester and vinyl ester, were also reported, but the results demonstrate discrepancies in the reproducibility [238–240].

One of the most promising strategies can be related to the synthesis of cyclic carbonates [235, 241]. Cyclic carbonates can be obtained from the cyclo-additions of CO_2 into epoxides [242]. The process can be quantitatively performed in solvent-free conditions using supercritical CO_2. These cyclic carbonates are valuable for producing Non-Isocyanate Polyurethanes (see Sect. 6.7). Other intermediates to provide more reactive cyclic carbonates (such as exovinylenecarbonate) were also developed [243] in order to obtain high-performance polymers such as polycarbonate [244], and polyurethane [245], that can even be recycled more efficiently than traditional ones. Recently, CO_2-derived oxazolidones (cyclic urethanes) have been demonstrated to be a versatile polymer with easy recyclability and suitable for manufacturing composites with natural fibers [246]. However, the development of cyclic urethanes remains at a very low TRL and is not yet suited for rigorous investigations aimed at composite applications. They remain potential future solutions if production is scaled to a few kilograms.

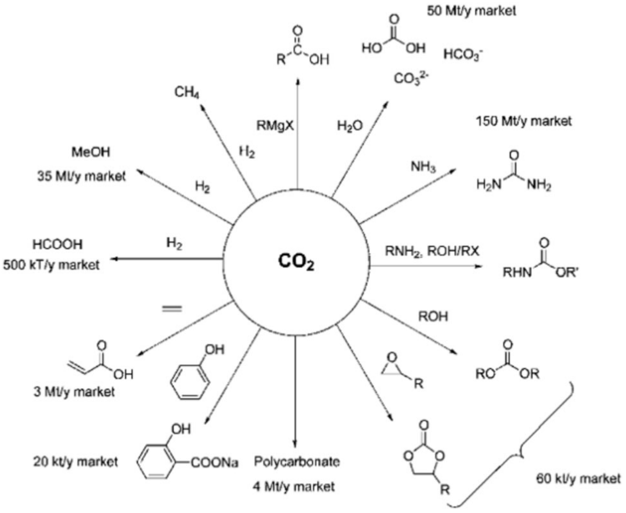


Fig. 25 Chemical transformation of CO_2 into commodity chemicals and market volumes. Reproduced with permission from [234]

6.2 Unsaturated polyesters—the low-cost large-scale resin

Unsaturated polyesters (UPR) are the most commonly used thermoset matrices for composites. They represent 87% of thermoset resins and 65% of the total composites production. Despite their widespread use in various composite applications, UPRs pose several issues arising from their sourcing and end-of-life perspectives.

6.2.1 Backgrounds of UPR, composition, polymerization, and properties

Unsaturated polyesters consist of polyester oligomers or prepolymers containing reactive unsaturations, a reactive diluent that often plays the role of crosslinker, and a free-radical initiator (see Fig. 26). The prepolymer is prepared from unsaturated dicarboxylic acids or their anhydride equivalents, polymerized with a diol. Maleic (cis) acid/anhydride or fumaric acid/anhydride (trans) as unsaturated dicarboxylic acids or their anhydride equivalent are preferred unsaturated diacids, with fumaric acid being more reactive for free-radical crosslinking[177]. Glycols such as ethylene glycol, propylene glycol, butanediol, and cyclohexyl dimethanol are used in various proportions to tailor the properties and prepare the prepolymer [177]. In addition, aromatic saturated diacids/anhydrides are used to improve chemical resistance, strength, or toughness, particularly with isophthalic acid/anhydride and adipic acid that are commonly used for such.

Maleate and fumarate cannot be homopolymerized efficiently. Due to their molecular weight, polyester prepolymers have relatively high viscosity. Therefore, reactive diluents, typically styrene or methyl methacrylate, are used to lower the viscosity and crosslink the network [247]. Typical UPR formulations have viscosities ranging from 0.5 to 2 Pa.s,

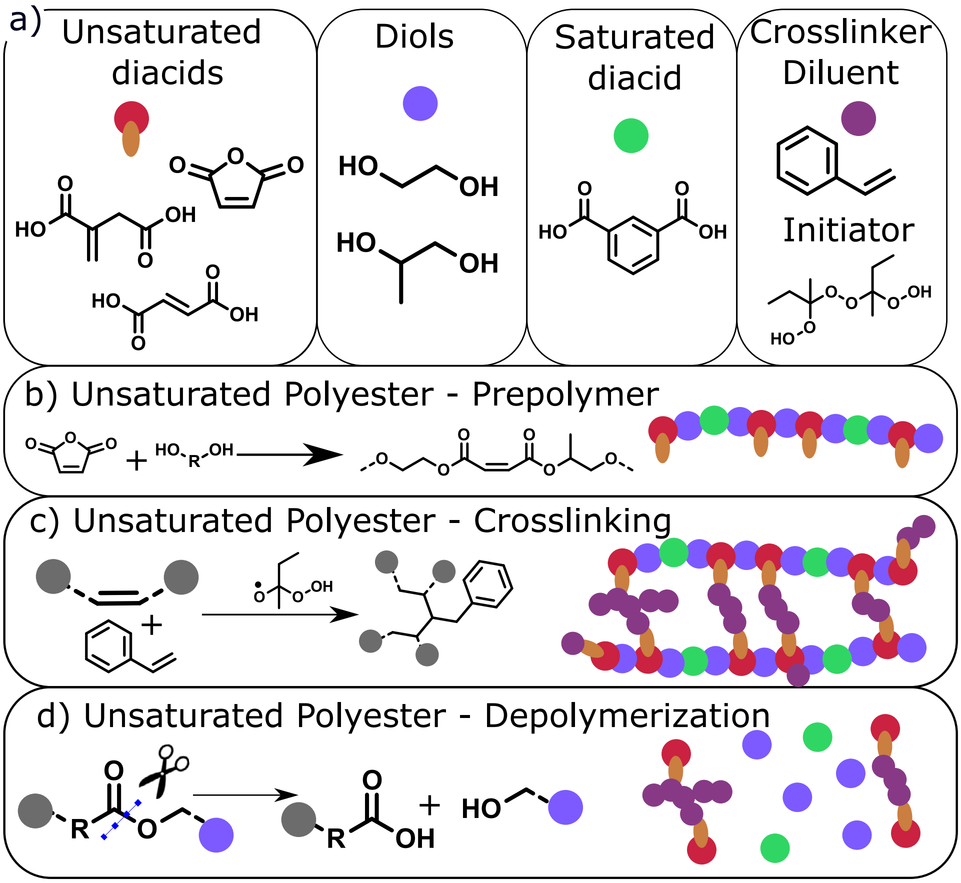


Fig. 26 Unsaturated polyesters. **a** Typical monomer precursors, **b** pre-polymerization from diacid and diols to unsaturated polyester, **c** crosslinking of UPR, and **d** representative hydrolysis of UPRs

making them ideal candidates for infusion processes at low temperatures.

UPRs undergo crosslinking through a free radical polymerization process. Typically, peroxides such as methyl ethyl ketone peroxide (MEKP) or benzoyl peroxide (BPO) are used to initiate the free radicals when heated. An accelerator (cobalt-based) can lower the free radical initiation temperature and accelerate the curing process [248]. Room temperature curing is feasible by employing MEKP and cobalt accelerator [248]. The initiator and catalyst composition play a significant part in the polymerization conditions as well as the future properties and stability of the network over time. During free radical polymerization, the unsaturated polyester bonds are consumed, forming new stable C-C bonds and crosslinking the polyester chains. Due to the nature of the bonds formed, UPRs are inherently stable and difficult to depolymerize. Particular attention must be brought to ensure the total consumption of the crosslinker (styrene) and the initiator to prevent the release of these (hazardous) compounds in the environment [176].

The low viscosity of UPR formulations, their versatility, and the ability to cure at room temperatures have fostered their use in large composite structures. Marine applications and infrastructures (construction and wind energy) are the most prominent consumers of UPRs. They are mostly combined with GF for producing low-cost composite materials [151] but are also investigated as matrices for natural fibers [249]. UPRs present moderate resistance to hydrolysis and solvent for general purposes but poor resistance to weathering and UV aging in outdoor applications. They tend to yellow and degrade under UV exposure. To avoid these undesirable effects, stabilizers and modifiers should be used. UPR is suitable for applications with service temperatures up to 80 °C, with glass transitions generally ranging from 70 to 110 °C. Typical mechanical performances of UPRs are reported in Table 2. UPRs are relatively brittle materials and are prone to fire [250]. Flame retardants, often containing toxic halogen compounds, are used to mitigate this issue[251]. Despite their prevalence in fiber-reinforced plastics (FRPs), there is ongoing research to enhance the sustainability of UPRs, aligning with circular economy principles.

6.2.2 Routes to greener UPRs in the frame of a circular economy

The sustainability of UPRs depends primarily on sourcing components and designing polymers that are easier to decon-

Table 2 Overview of matrices composition, properties and applications for composites

Resin chemistry	Part A	Part B	Catalyst	T_g (°C)	E (GPa)	σ (MPa)	ϵ (%)	Advantages	Drawbacks	Applications	Biobased potential	Recycling	Ref
Unsaturated polyesters	Unsaturated polyester prepolymer	Styrenic (styrene, vinyl toluene) and methacrylic diluent	Mandatory: radical initiator (peroxides), optional: accelerator (cobalt, amines)	80–250	2.5–4.0	60–100	2.0–8.0	Low cost, easy processing, moisture resistant, chemical resistance, fast cure	VOCs, brittle, high shrinkage, fire sensitive, limited performances, thermal stability	Marine, construction, energy	Everything except styrene	Hydrolysis, pyrolysis, chemolysis: polyols and acid. Styrene rich compounds are challenging	[176, 177]
Epoxy	Epoxy monomer	Amines, anhydrides, thiols, acids	Not necessary in the majority. Required for anhydrides, thiols…	60–150	2.0-3.5	40–80	1.0–5.0	High strength, low shrinkage, good adhesion, chemical and thermal resistance	Long curing time, expensive, brittle, hazardous constituents, burning behavior, bisphenol A	Aerospace, automotive, marine, energy	Potentially 100%. Biobased amines are the most challenging	Acidolysis, pyrolysis, solvolysis. Challenge to valorize products	[15, 43, 178]
Polyurethanes	Polyester polyols, Polyether polyols	Isocyanates	Preferably, Tin catalyst, strong base	50–120	0.5–3.0	30–70	5.0–20.0	Toughness, curing speed, versatility, adhesion	Hazardous constituents (isocyanates), UV sensitive, thermal stability, moisture sensitive process, burning behavior	Flexible composites, elastomers, automotive	Polyols. Isocyanates are challenging	Feasible, challenge in purifications. Hydrolysis, chemolysis, acidolysis, pyrolysis: polyol, amines or oils	[179, 180]
Vinyl esters	Vinyl ester prepolymer (Epoxy + Methacrylic acid, Polyol+MA)	Styrenic and methacrylic diluent	Mandatory: radical initiator (peroxides), optional: accelerator (cobalt, amines)	90–180	3.0–4.0	70–100	3.0–7.0	Chemical resistance, adhesion, mechanical performances	VOCs, cost (vs UPR), bisphenol A	Chemical tanks, marine structures	Epoxy and polyol precursor. Styrene and methacrylate difficult	Potentially, cleavage of ester groups, challenges in styrenic phase	[181, 182]
Phenolics	Phenolic prepolymer (Novolac, Resole…) (Phenol+ formaldehyde)	Formaldehydes, Hexamethylene tetramine	Acid or base	150–350	3.0–4.0	40–70	1.0–3.0	High thermal resistance, burning behavior, dimensional stability	Brittle, lower mechanical strength, curing time and temperature, hazardous constituents	Fireproof structures, electrical parts	Phenol and formaldehyde	Challenging, pyrolysis	[183]

struct through macromolecular engineering. While UPRs represent nearly 90% of the market, it is important to emphasize that they are not investigated upon the corresponding contribution. Since 1997, less than 173 papers have been published using “bio-based” and “unsaturated polyester” as keywords, and fewer than 300 on “recycling” (Scopus database). In contrast, bio-based epoxy has been present in nearly 2000 papers since 1996, with more than 100 papers published annually since 2017 and 227 papers in 2023 alone. More than 2000 papers on epoxy recycling have been published since 1996, with more than 300 papers in 2024. Most of the works on the sustainability of UPR-containing composites have focused on the type of natural fibers, the process parameters, and the modifications of the fibers to improve the interfacial adhesion [249].

Even though some independent transversal work on the synthesis of monomers could be relevant to UPR, it appears that the sourcing of UPR monomers from biobased and renewable sources has only been partially investigated. Much more extensive research and effort are needed in that area. Comprehensive solutions addressing both sourcing and recycling have yet to be addressed.

Still, some works have progressed in developing renewable alternatives to existing systems, but challenges remain. For instance, petro-based compounds such as isophthalic acid can be replaced with FDA [253]. Biobased diols, including ethylene glycols and polyols like isosorbide, have also shown promising results recently [253, 254]. However, sourcing biobased unsaturated dicarboxylic acid remains difficult due to limited options. Biobased routes to obtain maleic acid (MA), typically obtained from the catalytic oxidation of benzene [255], have been developed from butanol [256] or furfural [257]. LCA analyses highlight that the furfural route is more environmentally favorable than bio-butanol [258], including pilot-scale feasibility. However, valorizing by-products must be performed to diminish the footprint and E-factors, a common issue in biomass chemical transformations. Fumaric acid, the trans isomer of MA, is naturally occurring and can be easily obtained from fermentation [259]. From an economic perspective, most fumaric acid is still produced from the catalytic isomerization of MA [255].

Itaconic acid [176] and muconic acid [260] are other unsaturated dicarboxylic acids showcasing promising potential for UPRs whether their production can be scaled to industrial quantities. Notably, if the interest in exploiting bio-based UPR has been demonstrated in some cases [252], the pathway to obtain biobased compounds should be systematically discussed. Moreover, a recent LCA on bio-based UPR by Shahid et al. [252] demonstrated the dominant role of styrene in the environmental footprint (see Fig. 27). This reactive diluent represents the most problematic compound to urgently replace [261]. In this regard, current solutions are based on bio-based methacrylates such as isosorbide (di)methacrylate [250], bio-phenol grafted methacrylates [247, 261], and terpenes [261]. Yu et al. [247] investigated several sustainable aromatic analogs to styrene, such as veratrol and guaiacol derivatives. The reactivity and final properties are suited for considering them as alternatives. However, the viscosity of these alternatives was consistently too high to compete with styrene-based formulations, making them unsuitable for composite infusion. Moreover, the costs of lignin-derived aromatic vinyl were not discussed, but they are not competitive up to now.

6.2.3 About the recycling of unsaturated polyesters

As previously mentioned, the recycling of UPR has been poorly investigated. While linear polyesters are among the most advanced polymers in terms of recycling with existing pilot plants, exploiting enzymatic [262] or catalytic [263] pathways, strategies for crosslinked UPR are much less

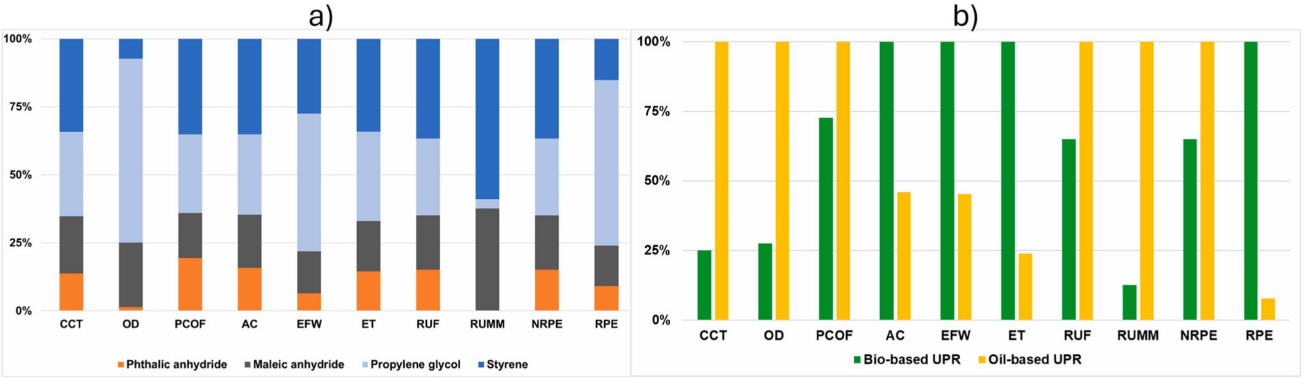


Fig. 27 **a** Environmental contribution of various components of oil-based UPR and **b** comparison of the environmental performance of bio-based and oil-based UPRs for each impact category (100% corresponds to the highest impact in each category). Reproduced with permission from [252]

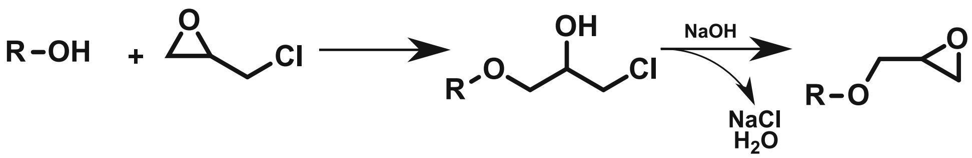


Fig. 28 Conventional pathway to obtain glycidyl ether

developed [264]. Some authors have investigated chemical recycling, primarily through hydrolysis of the ester bonds [265, 266], to recover some valuable diols and dicarboxylic acid (isophthalic acid). However, the MA/styrene adduct is difficult to valorize due to the stable nature of the C-C bond formed. Pyrolysis was also employed to retrieve carbon or glass fibers, by thermally degrading the matrix [267, 268]. In all reported cases, MA and styrene are wasted, further highlighting the urgency to find renewable alternatives to styrene and MA. Direct pyrolysis of UPR-based composites remains, to date, the most realistic pathway. Pyrolysis management and perspectives for composites are discussed later on in the dedicated Sect. 8.2.

6.3 Epoxy resins—versatile, competitive, and problematic

Epoxy resins (EP) account for about 10% of the composite market. They are often the preferred choice when high performance and stability are required. EPs are highly reactive and versatile and can be used in many applications. The curing process of epoxy can easily be tuned by playing with hardener chemistry, (macro)molecular structure, and catalyst. EPs are probably the most investigated thermoset resins for composites, particularly in NFCs.

6.3.1 Basic principles of epoxy resins, monomers, curing agents, and common properties for targeted applications

Epoxy resins are two-component formulations consisting of (a) low molecular weight epoxy precursors containing oxirane moieties and (b) a hardener. Depending on the hardener and some specific requirements, a catalyst might be necessary. Epoxy monomers are synthesized from precursors containing nucleophilic groups, such as hydroxyl or primary/secondary amines, that are reacted with epichlorohydrin (ECH) by nucleophilic substitution (Sn2, see Fig. 28), leading to glycidyl ethers. The production of epoxy monomers has high atom efficiency (generating only NaCl and water as by-products) and requires a base catalyst (typically NaOH) at a moderate temperature (below 120 °C). Although ECH is a volatile and highly toxic compound, it is extremely economical [184]. They can also be produced by the oxidation of unsaturation, such as unsaturated fatty acids, triglycerides [269], and terpenes [270]. Bisphenol A is the preferred precursor for epoxy resin. Diglycidyl ether of bisphenol A (DGEBA) is economical and widely available. The resulting resins derived from DGEBA have good properties (mechanical, thermal, stability to UV). Depending on the manufacturing process and applications, reactive diluents, such as aliphatic epoxies, and tougheners can be used

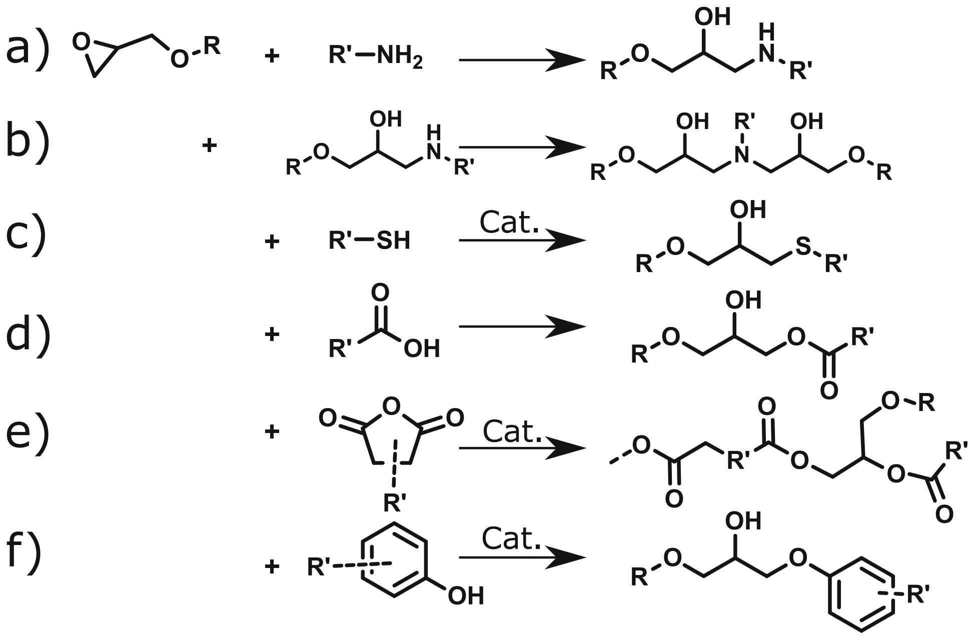


Fig. 29 Curing agent reactions for epoxy resin. **a**, **b** Aminolysis, **c** thiol-epoxy, **d** epoxy-acid, **e** epoxy-anhydride, and **f** epoxy-phenol

to adjust the formulations [271]. EPs generally have higher viscosities than UPR, but viscosities as low as 250 mPa.s can be achieved for infusion processes.

Epoxy resins are highly reactive compounds due to their strained three-membered ring structure. Substituting the oxirane ring with electron-withdrawing groups (e.g., carbonyl, aromatic...) or donating groups (ether, amines...) strongly influences EP's reactivity, creating a very versatile and convenient toolbox for adjusting EP formulations to the complex requirements of composite manufacturing. The ability to tune the reactivity of epoxy resins by choosing specific curing agents and adjusting the curing conditions allows them to be used in various applications, ranging from construction to electronics under diverse forms like adhesives, coatings, and composite materials. This tunable reactivity is the key factor explaining the widespread use of epoxy resins in industry. The oxirane ring is more susceptible to nucleophilic attack [272], making it very reactive with various curing agents like amines, thiols, anhydrides, phenols, and acids (see Fig. 29). The reactivity of epoxy resins depends on the type of hardener used, its structure, and reaction conditions, such as temperature and the presence of catalysts. Primary amines are the most commonly used hardeners due to their low viscosity, fast reactivity, and wide availability. Aromatic amines such as MDA are largely used as they possess low reactivity at low temperatures and provide high T_g, mechanical properties, and stability. However, high temperatures are required to fully cure the system since secondary aromatic amines are less reactive than primary amines. Aliphatic primary and secondary amines present similar reactivity [15]. Due to concerns about the toxicity and hazards associated with primary aromatic amines [273], there is a growing interest in alternatives such as (cyclo)aliphatic amines. The most commonly used EP precursors are displayed in Fig. 30, along with health hazards related to each one.

Anhydrides are the second most frequently used hardener for epoxy resins. They require an initiator, such as Lewis bases, to start a chain-step copolymerization [15]. Such polymerization requires higher temperatures than with amines and is considered a latent formulation. However, most anhydrides are solid and must be solubilized or melted with the epoxy to be processed. This can limit its exploitation in certain composite processes [274]. Different fiber sizing agents might be required for epoxy-anhydride and epoxy-amine formulations as well.

Catalysts like tertiary amines, imidazoles, or metal salts can modify the reaction rate and the properties of the final product. These catalysts enhance the epoxy group's reactivity, allowing them to cure faster and at lower temperatures. However, these catalysts can also cause the yellowing of

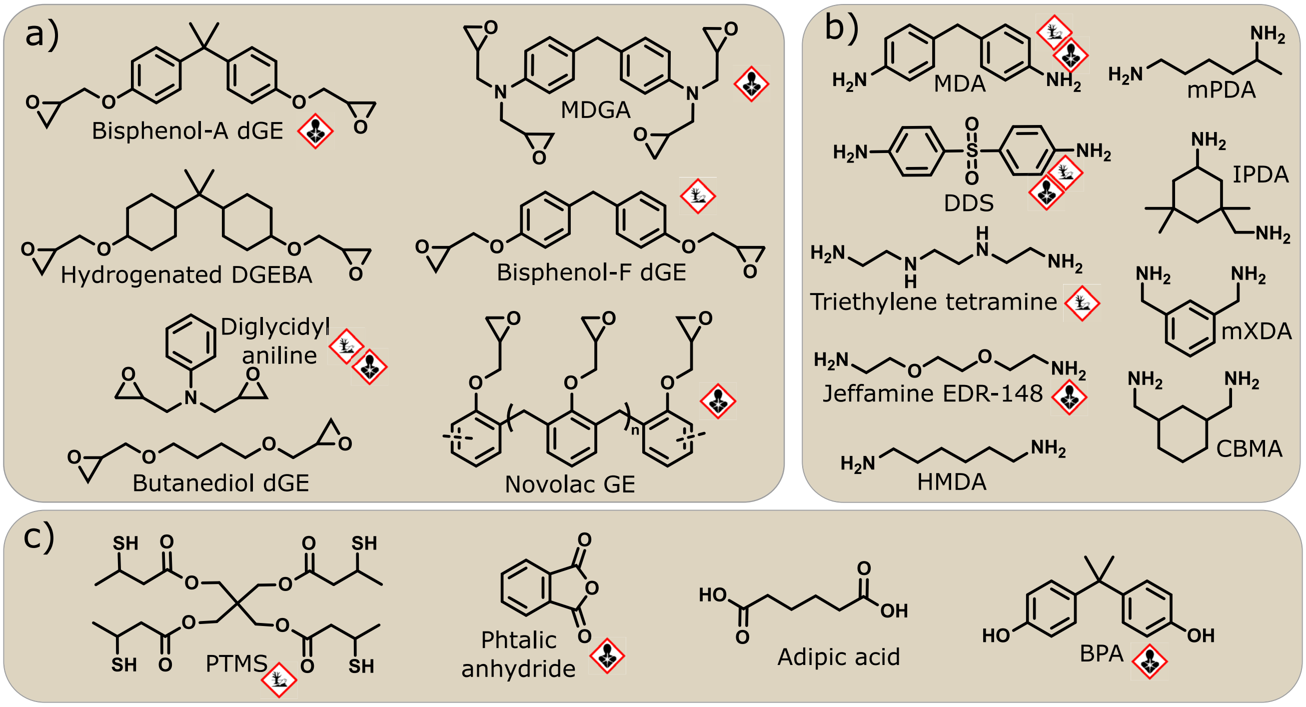


Fig. 30 Typical EP constituents and potential health hazards. **a** EP monomers, **b** typical amine hardener, and **c** other common hardener. Acronyms: GE, glycidyl ether; MDA, 4,4'-Methylenedianiline; MDGA, 4,4'-Methylenebis(N,N-diglycidylaniline); PTMS, Pentaerythritol tetrakis(3-mercaptopropionate); CBMA, cyclohexanebis(methylamine), DDS, diaminophenyl sulfone; mPDA, methylpentane diamine

cured resins, diminish the environment's stability, and are often hazardous, with a risk of leakage from the matrix. When possible, avoiding the catalyst is preferred [272].

EP resins possess excellent mechanical properties, chemical resistance, adhesion, and thermal stability, representing solid assets for structural composites. Upon curing, EPs form a hard, densely cross-linked structure, which enhances their rigidity and structural integrity. This network imparts high mechanical strength and durability, allowing epoxies to withstand significant stress and maintain good toughness [15]. Furthermore, epoxy resins exhibit good thermal stability, performing well under heat and even high temperatures, especially when cured with appropriate hardeners like anhydrides or aromatic amines. EPs also offer excellent insulation properties, making them suitable for use as encapsulants and insulators in electronics and electrical applications [275]. Additionally, epoxy resins offer excellent chemical resistance, particularly to alkalis and solvents, which is valuable for protective coatings in harsh environments. EP tends to swell with strong acids but can withstand common acidic conditions [276].

Polar functional moieties formed during curing play a crucial role in enhancing the interfacial interactions with fibers during the fabrication of FRP composites [277]. They particularly improve interfacial strength and toughness, leading to high performance. The initial low viscosity also helps improve fiber and surface wetting for all types of fibers. Significant efforts in the field of sizings aimed first to enhance the interface of epoxy using either amine or epoxy silane agents [278]. The polar functions at the surface of NF have made EP resins the best matrix for NF, with unidirectional laminates commonly reaching a modulus of 30 GPa, and tensile stress above 320 MPa [154, 156, 279].

The properties of epoxy resin are significantly influenced by its structure, molecular weight, and chain length. Additionally, the number of epoxy functional groups, or oxirane rings, is critical for the crosslinking reaction, which in turn affects the performance of the epoxy thermoset. Therefore, the clever design of the epoxy monomers through careful selection of starting compounds is paramount to perfectly control epoxy-derived compounds. Anhydride acids have seen a surge as hardeners for epoxy because they are not reactive at room temperature, allowing one-pot formulations with extended working times [210]. They provide hard epoxy resins with high weather stability; however, they are more brittle [274]. Alternatively, the curing process can be conducted via UV-triggered polymerizations by using thiols-epoxy, as a base catalyst can trigger fast curing, suitable for fast light-induced designs such as 3D printing. They are highly transparent, with strong adhesion and good water resistance. Their mechanical properties are lower than amine-hardened epoxies, and their commercial availability and diversity are much lower.

6.3.2 Pathways towards more sustainable epoxy—a not so impossible bio-sourcing?

Significant scientific and industrial efforts have been made to improve the sustainability of epoxy resins. These efforts focus on two main aspects: (i) finding bio-based and non-toxic alternatives to commonly used DGEBA and aromatic amines and (ii) developing recycling pathways to break down the matrix and recover fibers as well as valuable building blocks.

For the past 50 years, the main route for ECH production was through the propene chlorination to allyl chloride, further oxidated to dichloropropanol (DCP) (see Fig. 31a). DCP was then ring-closed using sodium hydroxide. This was a particularly competitive pathway because of the low cost of the reagents and process efficiency despite the high toxicity of all involved compounds (chlorine, allyl chloride) [280]. In the late 2000s, a more sustainable pathway was developed, starting from glycerol to epichlorohydrin (GTE process) [280, 281]. This process is more straightforward, with the chlori-

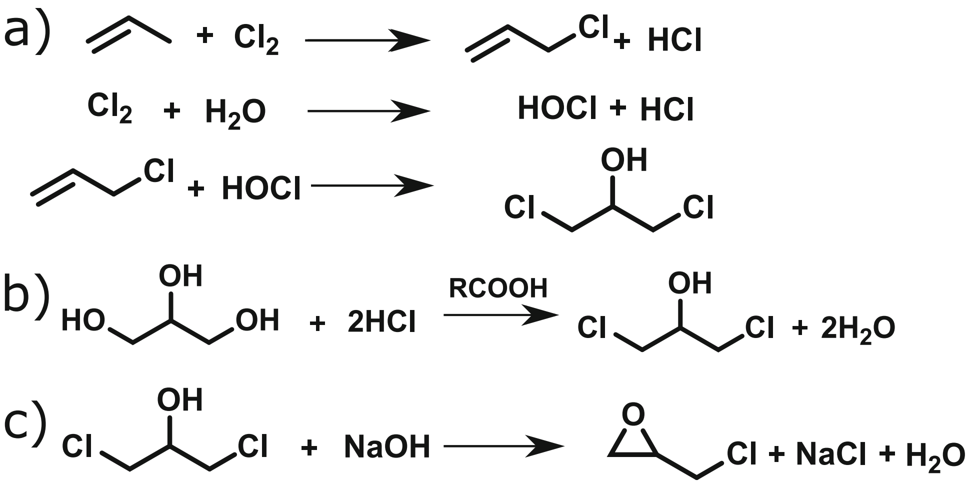


Fig. 31 Routes to synthesize epichlorohydrin. **a** petro-based pathway to dichloropropanol, **b** (bio-)glycerol route to dichloropropanol, and **c** final synthetic step from dichloropropanol to ECH

nation of glycerol by hydrochloric acid (see Fig. 31b) using a carboxylic acid catalyst (typically caproic acid) at moderate temperature (70–120 °C) to obtain the key DCP intermediate. DCP can then be closed to ECH in the same manner as starting from propene.

This process was patented and commercialized by Solvay under the brand name Epicerol® with a production capacity of about 100 kt/yr [282]. They claim a 60% GWP reduction compared to the traditional process, consistent with the available literature data [283, 284]. Moreover, the global production of glycerol has jumped since the beginning of the 21st century due to the increase in biofuel production and the generation of glycerol as a by-product. The valorization of this waste is a good example of creating complementary value chains in a circular economy and providing additional revenue streams for bio-fuel producers [282].

Current efforts now focus on finding alternatives to BPA, through renewables routes and reducing risk for health [285]. The possibilities of sourcing bio-derived phenols and amines have been discussed in Sect. 6.1.

To the author's best knowledge, there is no commercialized bio-derived phenol epoxy. Partially biobased epoxy is, however, commercialized, such as Greenpoxy® or Fairpoxy® with biobased contents ranging 20–56%. This bio-content only includes the epoxy part and not the hardener, that is, amines are still petro-sourced. The final biobased contents range from 14 to 45%. The most common formulations include bio-based butanediol diglycidyl ether as a reactive diluent, largely available with DGEBA. For the highest bio-content, bio-ECH is used to produce DGEBA, leading to 33% of bio-carbon in it.

Scientific literature has extensively explored alternatives to replace BPA. Indeed, BPA is an endocrine disruptor that mimics estrogen and has anti-androgen capacities, causing damage to different tissues and organs, including the reproductive system, immune system, and neuroendocrine system [285].

Recently, Witthayolankowit et al. [200] proposed a comprehensive study on developing a biobased epoxy derived from bis-guaiacol. The guaiacol was extracted from birch bark. The epoxy was cured with a bio-derivable anhydride and used as a matrix with hemp reinforcement, providing a fully biobased composite. They demonstrated no endocrine activity for the epoxy. They investigated the environmental footprint (see Fig. 32) and demonstrated the benefits of their strategy but highlighted the importance of valorizing all compounds of the lignin treatment to create an economical and environmentally beneficial value chain [195].

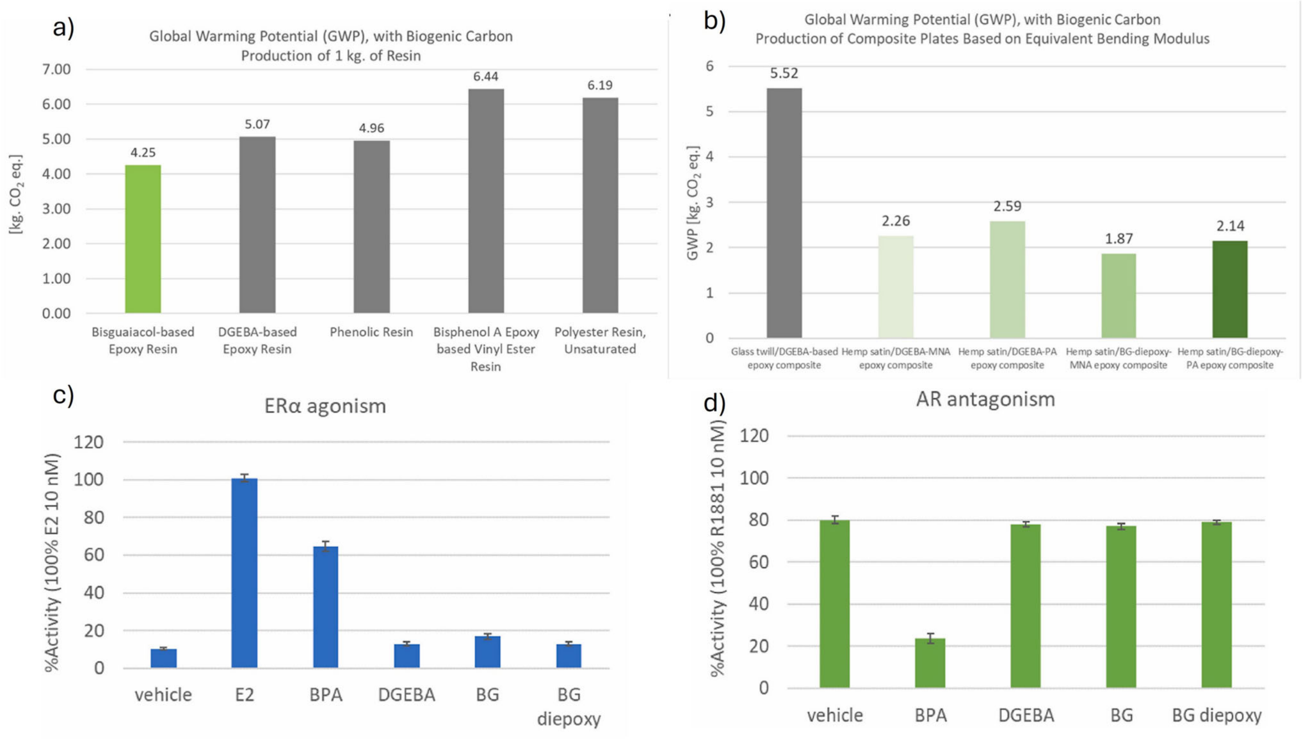


Fig. 32 GWP as determined by an LCA for **a** production of 1 kg of bis-ethylguaiacol epoxy (BG) and other benchmark resins, **b** production of one hemp fiber-reinforced composite plate and compared to a glass-fiber-reinforced epoxy composite plate, all with an equivalent bending stiffness. **c** Oestrogen receptor agonistic activity of chemicals and **d** androgen receptor antagonistic activity of chemicals. Vehicle is DMSO. Reproduced with permission from [200]

Other strategies have been deployed to develop bio-based epoxy resins that are easily depolymerized. Wu et al. [224] developed a system based on epoxidized FDA and a lignin-derived cycloaliphatic diamine [204], thus avoiding the use of BPA and MDA (see Fig. 33). By this approach, it remains a conventional epoxy-amine system with high T_g (around 170 °C), low sensitivity to water, and satisfying mechanical properties, suitable to manufacture glass fiber and flax fiber composites. More importantly, the incorporation of ester moieties in the presence of tertiary amines allowed the internally catalyzed depolymerization by methanolysis of esters at low temperatures (70 °C, 48 h). The process was selective and enabled the recirculation of all compounds after purification. No LCA was performed, but positive results might be expected as FDA in polyethylene furan dicarboxylate (PEF) was reported beneficial compared to petro-based compounds [223, 286], as well as lignin catalytic depolymerization [197]. However, these should be confirmed through dedicated cradle-to-cradle analysis, considering the recycling steps and yields.

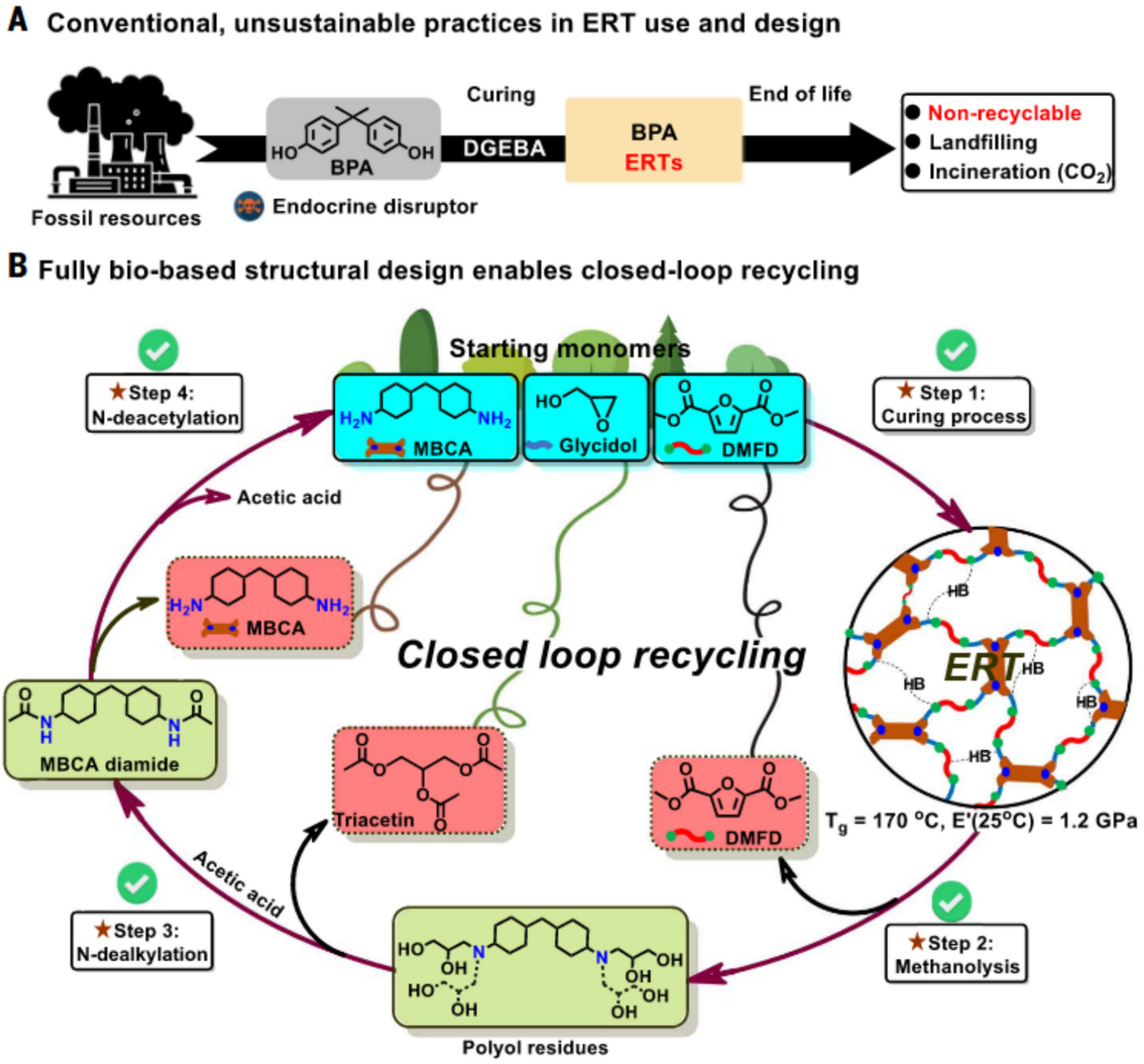


Fig. 33 Schematic representation of the closed-loop of epoxy materials developed by Wu et al. as compared to the conventional BPA route. **A** The unsustainable status quo of BPA-based thermosets sourced from fossil resources. **B** Synthesis and closed-loop recyclability of a fully biomass-derivable epoxy resin. From [224]. Reprinted with permission from AAAS

A parallel approach was developed by Rorrer teams, named PECAN (polyester covalently adaptable network) [210]. This interesting strategy is based on already commercially available building blocks. By reacting biobased epoxy derived from sorbitol and butanediol with methylhexahydrophthalic anhydride (MHHPA), a liquid anhydride, they

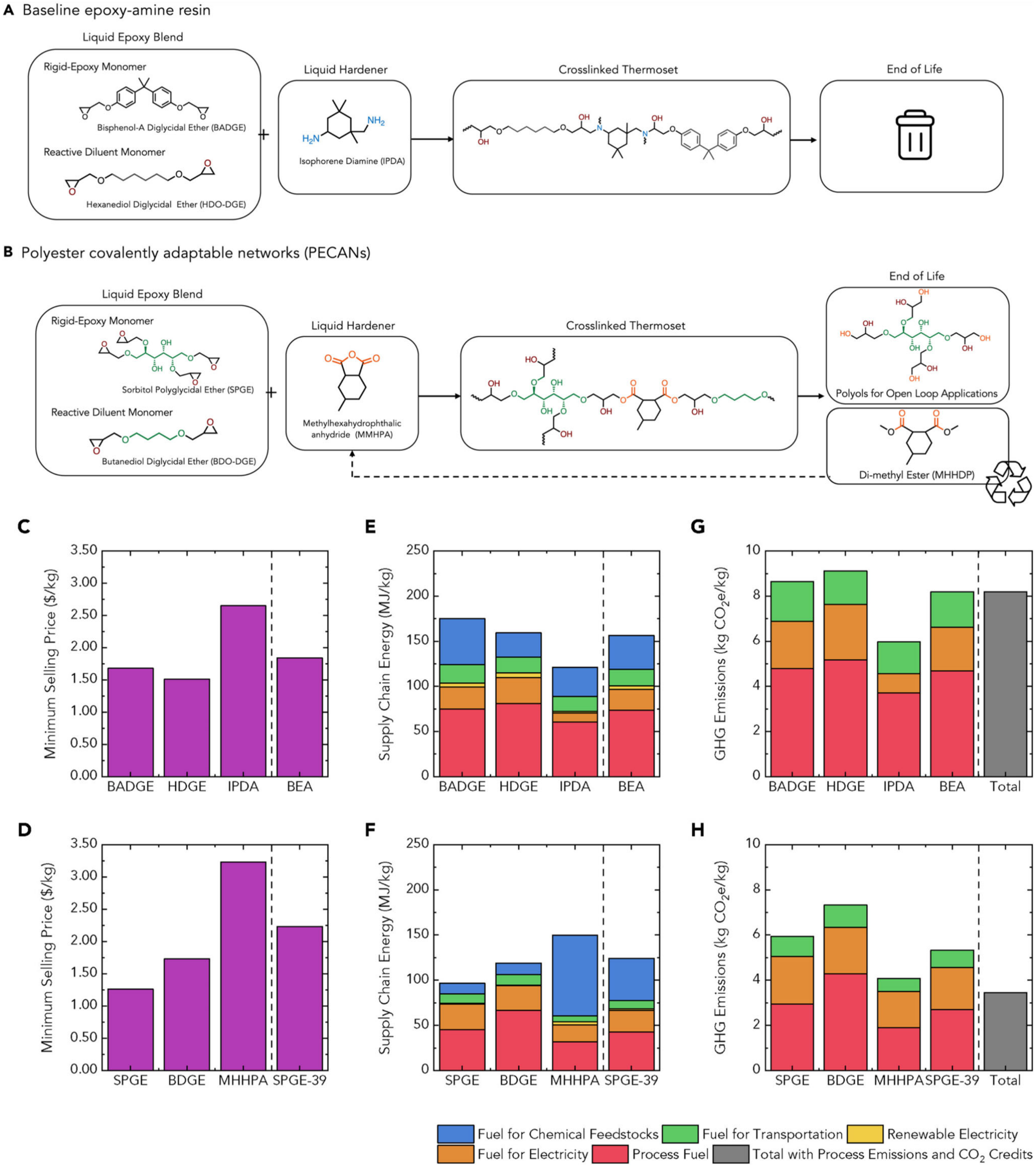


Fig. 34 Polyester Covalent Adaptable Network (PECAN) strategy to obtain cost-competitive biobased epoxy network with low environmental footprint. **a** Benchmark DGEBA-based epoxy resin, **b** PECAN constituents and depolymerization (methanolysis) route, **c**, **d** minimum selling price estimation for the constituents and resin, **e**, **f** cumulative energy demand for the resin production, and **g**, **h** estimated GHG emissions, including curing, for DGEBA-IPDA and PECAN resins, respectively. Reproduced and adapted with permission from [210]

obtained fully biobased epoxy-based polyesters. The properties are similar to benchmark epoxies with glass transitions of about 100 °C, modulus 1–3 GPa, and elongation of about 3–4%. The formulations have low viscosity, below 0.5 Pa.s, suitable for composite manufacturing. They demonstrated positive environmental results compared to epoxy anhydride with a GWP reduction of about 35% (see Fig. 34). Although an increase by 25% of costs was estimated, and could represent an economic burden for some industries, it remains commercially relevant. The technology was successfully implemented into a GFRP wind-blade [287]. They demonstrated creep resistance and advanced weathering resistance compared to DGEBA epoxy and the thermoplastic Elium®.

Using methanol and a base catalyst (K_2CO_3), they successfully depolymerized the network within 7 days at 50 °C or within 6 h at 225 °C by batches of 500 g. The recovered glass fibers were of high quality. The polymer matrix was degraded into a polyol that was subsequently used as a crosslinker for polyurethane (open-loop) and a methylated diester, which was reconverted to the initial hardener (closed-loop). Unfortunately, the LCA was not performed on the recycling of the composite nor the matrix, and the temperature and solvent proportion might hamper true benefits. Despite this, their work pioneered the exploitation of competitive bio-derived epoxy through a drop-in approach within existing manufacturing techniques. This opens the door to the recycling of large composite structures. Their approach should be extendable to many epoxy-anhydride systems with minimal adjustments.

Other bio-based epoxy and alternatives to BPA have been investigated over the last two decades [288], exploring bio-phenols [186], such as vanillin [289], resorcinol or eugenol [290], and cyclic polyols, such as isosorbide [291]. These results demonstrated similar properties to DGEBA, but characterizations were not thorough enough regarding the applicability of these materials. Parameters such as viscosity and curing behavior, weathering, and environmental durability were often missing. Despite their importance, LCA and techno-economic analyses are not often conducted due to the complexity of implementing such work alongside the chemical developments of new resins.

6.3.3 Towards recyclable epoxides—between cheap yet not selective and expensive yet full recovery

Aside from the few mentioned works that developed a comprehensive approach to tackle the issue of epoxy-based composites by designing the resin, the EoL of these stable thermosets (epoxy-amine) remains an issue. The Recyclamine® commercialized by Aditya Birla (previously sold by Connora) incorporates acetal linkages in the amine hardener. Under acidic conditions (acetic acid), these linkages are cleaved, allowing the recovery of the fibers on one side and a thermoplastic on the other side [292]. The LCA on the recycling process was shown to be positive [293], but the chemical synthesis of the Recyclamine® is not taken into account, while it can involve several steps and the use of toxic reagents [294, 295].

Epoxy-specific depolymerization protocols have been developed over the last decade towards two main strategies. The first one focuses on unselectively depolymerizing the EP under mild conditions to retrieve the fiber efficiently, often exploiting oxidative procedures. The second one is the selective catalytic cleavage of specific bonds to retrieve valuable compounds in addition to the fibers.

Unselective oxidative cleavage has been achieved with several strategies. In general, an oxidant such as hydrogen peroxide or tert-butyl hydroperoxide (TBHP) is used with a solvent and sometimes a catalyst. The pathways involved the formation of radicals that can cleave aryl-ethers and C-N bonds in the network [296]. A suitable solvent is required to swell the epoxy network, thus facilitating the penetration of the solution and the solubilization of the degraded products. Acetone [296], DMF [297], and acetic acid [298] were efficiently exploited with hydrogen peroxide to recover carbon fibers. In all cases, depolymerization took place at ambient pressure, temperature from 50 to 100 °C, and time between 0.5 and 6 h. The recovered CF did not show any signs of degradation, with limited surface oxidation. However, the degraded matrix can hardly be valorized, and no LCA was performed. Recently, Wang et al. [299] used $FeCl_2$, with acetonitrile (ACN) and TBHP to depolymerize epoxy resins; the degraded product was exploited as an adhesive with medium shear strength. Again, no LCA was performed. If the interest in recovering carbon fibers seems obvious, comparing the different approaches and reagents would help in designing industrial strategies.

The selective cleavage of covalent bonds presents a significant challenge. Ester linkages (O-C=O) [301] are more easily cleaved compared to ether groups (C-O-C) and the amine bonds (C-N) found in epoxy-amine networks. Lately, the catalytic cleavage of C-O bonds was reported by Skrydstrup et al. [300]. Using a ruthenium catalyst with isopropanol as a hydrogen donor in toluene, they recovered BPA, with a yield of around 80% in model systems and of about 50% in epoxy resin. The procedure was extended to other (bio)phenolic resins. The fibers and aluminum parts were also recovered from turbine blade samples (see Fig. 35). The remaining resin was considered wasted. This work is the first to provide well-defined compounds from the depolymerization of epoxy. However, the conditions used (24 h at 160 °C) and the

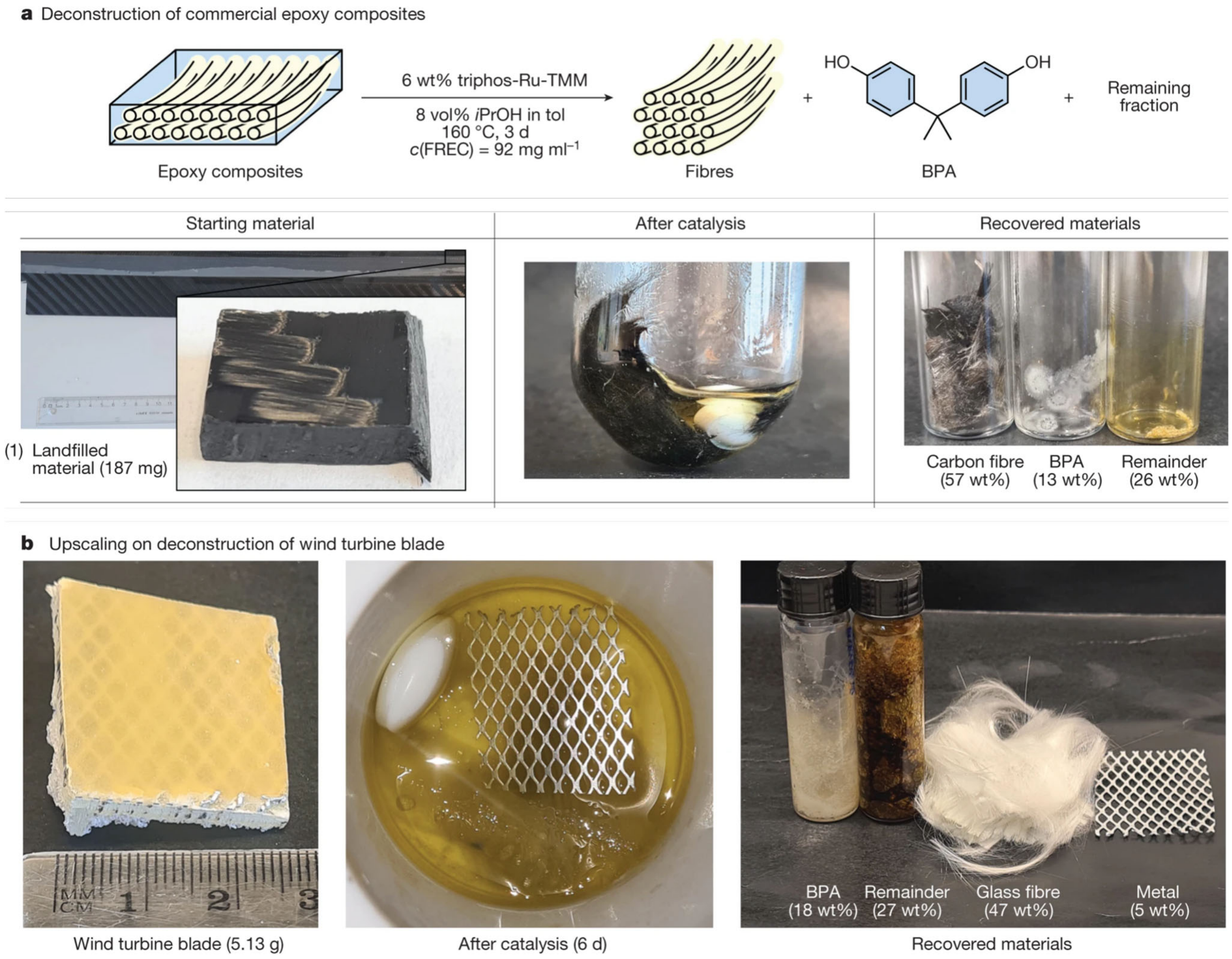


Fig. 35 Recovery of BPA and fibers from commercial epoxy composites using Ru catalysis by Skrydstrup et al. **a** Schematic of the strategy and example of a CFRP previously landfilled subjected to catalysis. **b** Upscaling of deconstruction conditions on wind turbine blade. Reproduced and adapted with permission from [300]

reagents, in particular the availability and cost of the ruthenium catalyst, may limit the approach to the lab scale. The environmental footprint of virgin petro-based BPA and the recovered one should also be considered.

Overall, the depolymerization of epoxy resin and the recovery of synthetic fibers have been shown to be feasible. However, the optimization, as well as economic and environmental relevance, still needs to be addressed. It is important to note that, to date, except for using Recyclamine [302], the recovery of natural fibers has not been achieved. NFs would require much milder conditions as they are less chemically stable, and pyrolysis is not suitable.

6.4 Vinyl ester resins—in between epoxy and UPR

Vinyl ester resins (VER) are thermosetting polymers widely used in composite materials due to their excellent mechanical properties, chemical resistance, and processability [182]. VER monomers are synthesized through the esterification of epoxy resin with unsaturated carboxylic acids (typically methacrylic acid), introducing reactive double bonds at the ends of the molecular structure. Monomers can also be prepared from polyols and phenols by condensation with MA [303]. As the monomers possess high viscosity, a reactive diluent is typically used to lower the viscosity to an acceptable processable range [182]. Like in UPR, styrene is the most common reactive diluent, with the same issues related to its use. The formulation can be one pot and is cured by free radical polymerization, as mentioned in UPRs. The same catalysts (such as peroxides) are typically employed. The most common formulations are based on methacrylated diglycidyl ether of bisphenol A, diluted in styrene.

They are the third most spread matrices in composites and possess intermediate properties between unsaturated polyesters and epoxides. Their structure combines the toughness of epoxies with the rapid curing and easy processing of polyesters [304]. VER exhibits high fracture toughness, superior adhesion to reinforcements, and resistance to moisture ingress, making it suitable for applications in marine, automotive, aerospace, and corrosion-resistant structures [305]. While VER offers enhanced durability and longevity for composite structures—reducing the need for replacements—its environmental impact remains a concern due to its petroleum-based origin and the release of volatile organic compounds (VOCs) during processing. The issues raised by epoxies, such as BPA and ECH use and UPRs due to styrene, are maintained here. Moreover, while the cleavage of the ester bonds, by alcoholysis [306] or hydrolysis [307], for example, is feasible, allowing the recovery of the fibers, it remains an under-investigated field. Moreover, like in UPRs, the free radical polymerization induces the formation of stable C-C bonds, yielding styrene-rich waste to be treated.

Advances in bio-based vinyl esters, reduced-styrene formulations, and improved recycling methods are being explored to enhance the sustainability of VER-based composites [181, 247]. Most efforts focus on biobased starting epoxy and polyols while some biobased routes to MA have been mentioned previously. Additionally, research into greener curing agents and resin recovery technologies is critical for minimizing waste and improving end-of-life recyclability. Yet, more research efforts into bio-based precursors and improved recyclability strategies are essential for enhancing its sustainability profile while maintaining its superior performance characteristics.

6.5 Polyurethanes—rising use and concerns

Polyurethanes (PU) rank as the sixth most produced polymer worldwide. Depending on the starting monomers, PU can also have a wide array of properties, from elastomeric to rigid, but they are mostly used as thermosets for cellular materials (soft and rigid foams).

6.5.1 A large thermosets market but marginal in composites with potential outcomes?

Despite their elevated volume produced and low cost, PUs represent only a negligible share of the composite market. However, PUs exhibit many advantages and potential for use in composites, including low cost, high reactivity, and versatile properties. They can have properties ranging from elastomeric to rigid with T_g up to 120 °C and showcase higher toughness than epoxy, with similar strength and stiffness [180]. They are also more resistant to abrasion and crack propagation than epoxy resins [308].

The limited use of PU in composites can be traced back to the traditional methods of composite processing. Historically, most of the composites were produced handcrafted by hand lay-up [44] either using prepreg or by manual impregnation of dry reinforcement. These manual processes were made in an open environment, with workers easily exposed to volatile compounds. This method is still a common practice used in sports, leisure, and aerospace. Polyurethanes result from a step-growth polymerization between polyols and polyisocyanates [309]. Isocyanates have been known for a long time as potent sensitizers and classified as carcinogenic, mutagenic, and reprotoxic [310]. For instance, methyl isocyanate was involved in the Bhopal disaster in 1984, causing thousands of deaths, injuries, and permanent disabilities with long-lasting effects; the decontamination efforts took 15 years [311]. They are now facing stricter REACH regulations, with a strong push towards their ban [312]. Additionally, typical isocyanates have low vapor pressure, which facilitates the risk of worker exposure. Isocyanates are also sensitive to water, which hydrolyzes the isocyanate into amine, thus requiring dry working conditions (see Fig. 36). Considering such issues, PUs have not been an ideal choice for facile composite manufacturing.

However, the increasing use of injection techniques, like resin transfer molding, has changed the game. The implementation of composites in automotive also pushes towards fast-curing procedures. In that sense, the workers are much

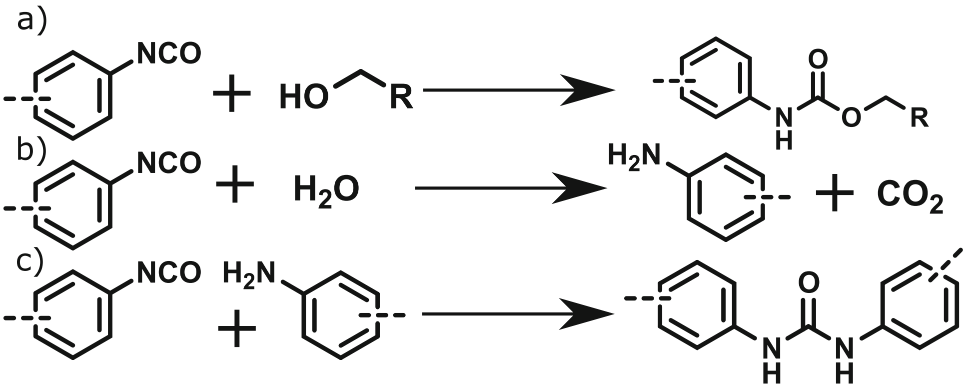


Fig. 36 Polyurethane polymerization and main isocyanates side reaction. **a** Principal urethane formation from isocyanate and alcohol, **b** isocyanate hydrolysis to amine and carbon dioxide, **c** urea formation from isocyanate and amine reaction

less exposed to the resins, and bi-component resins can be mixed automatically during the injection process and cured within minutes [313]. In such processes, PUs showcase advantages over other resins. Polymerization can be achieved within several hours at a temperature between 60 and 100 °C. Using a catalyst (typically a Lewis acid), the time can be reduced to minutes or curing can take place at room temperature [309]. PU suppliers such as Henkel or Huntsmann are now selling low-viscosity formulations suitable for resin transfer molding (RTM, see Sect. 7), and some studies demonstrated promising results, even with natural fibers, particularly considering moisture resistance [314]. Faster production cycles intend to offer potential energy and production savings while generalizing low-cost, high-performance composites, which would help reduce service-life footprint by weight savings.

6.5.2 The problem of the building blocks—sourcing and toxicity

The rise in exploiting PUs in composite structures does not reduce the problems initially mentioned, i.e., the fossil sourcing of the monomers and the toxicity of isocyanates. Catalysts needed for rapid curing, typically tin-based, such as dibutyltin dilaureate (DBTDL), are also highly toxic [309]. Interestingly, stricter regulations concerning PU and the demand for renewable raw materials in typical PU applications have already paved the way for more sustainable alternatives.

For instance, rigid PU thermosets usually require molecular diols and polyols such as ethylene glycol, cyclohexyl dimethanol, and glycerol. Routes towards the bio-sourcing of these compounds already exist, with pilot and industrial scale implementations all over the world [309].

The industrial production of isocyanates is typically performed by phosgenation of amines [315]. The most common isocyanates are MDI, toluene diisocyanate (TDI), isophorone diamine (IPDI), and hexamethylene diamine (HMDI). Access to bio-based amines, particularly bio-aniline, which leads to bio-MDA, has already been discussed, representing a current need and effort for many industries. The second compound for isocyanate production is phosgene. Phosgene is a highly toxic gas that causes acute respiratory distress syndrome. It was used as a chemical weapon during WWI. Phosgene is still the preferred way to produce isocyanates due to its high efficiency and low cost. The phosgenation process, highly reactive, does not necessarily require a catalyst, but the solvents commonly used during the process, like o-dichlorobenzene, are toxic. Intense efforts are being made to develop phosgene-free routes to obtain (bio)isocyanates [309, 315, 316], exploring dimethyl carbonate or urea reactions with amines or thermal decomposition of carbamates [315]. So far, these strategies are still at the laboratory and exploratory levels.

Caillol et al. successfully synthesized a fully bio-derived aromatic isocyanate from bis-guaiacol, through a phosgene-free method [192]. The resulting PU exhibited properties similar to those of analogous petro-based MDI. However, the environmental strategies developed to obtain bio-based and/or phosgene-free isocyanates have not been discussed enough. Even if phosgene has to be avoided regarding its toxicity, the efficiency and the atom economy of its process, which only generates HCl, will make it difficult to replace it. However, the LCA studies using biobased polyols and petro-based isocyanates have highlighted the benefits of bio-sourced PU and the important contribution of isocyanate in the global footprint [317, 318].

Despite the numerous possibilities to avoid the use of phosgene and source monomers from renewable resources, the toxicity of isocyanates will remain a concern. Therefore, new strategies to obtain PU while preventing the use of isocyanates have been explored over the last decade. They may offer decisive advantages in the realm of composites and will be discussed in Section 6.7.

6.5.3 Potential for recycling

PUs are widely used for foams, coatings, and fibers (e.g., Spandex ®). PUs are formed of carbamate moieties that can be cleaved upon appropriate conditions. Therefore, the recycling of PUs is relatively advanced [319], as summarized in Fig. 37.

Most recycling strategies have been developed for PU foams, representing 65% of the market. Mechanical recycling remains the most widespread strategy through crushing and compaction of PU foams resulting in downgraded products [320, 321] but is poorly relevant for composites.

Catalytic chemical depolymerization has been conducted at the laboratory level, but scaling-up is still limited [322]. Depolymerization can be performed by hydrolysis, glycolysis, alcoholysis, and aminolysis. A catalyst, typically a base such as KOH or 1,8-Diazabicyclo[5.4.0]undéc-7-ène (DBU), is required to activate the stable carbamate.

Hydrolysis of PUs releases CO_2 from the carbamate, leading to the recovery of the polyol and the amine. The challenge relies on the depolymerization conditions, often harsh, and the separation of these products. Hydrolysis commonly requires temperatures superior to 150 °C [321] for extended periods (5–24 h), leading to high energy consumption. The separation of amines and polyols usually demands multiple purification steps, such as liquid-liquid extraction, which are both time- and solvent-consuming.

Glycolysis is the most developed PU depolymerization process, widely exploited in industries [319, 323]. Glycol-

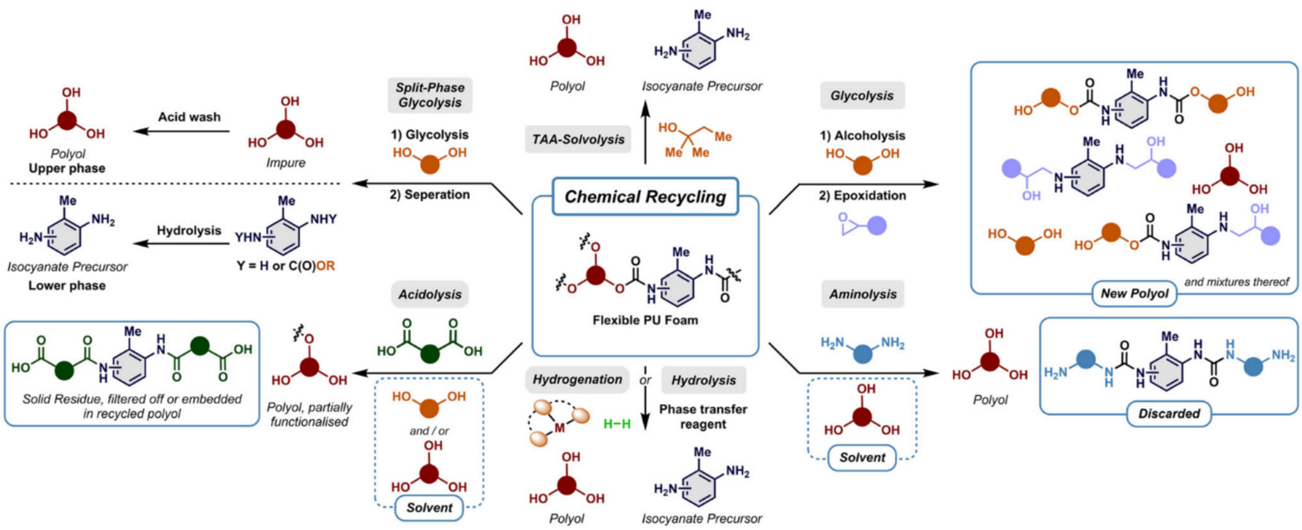


Fig. 37 Chemical recycling methodologies for PU foam. TAA, tert-amyl alcohol. Reproduced with permission from [326]

ysis is traditionally performed by exploiting base-catalyzed transcarbamoylation in the presence of ethylene glycol. This generates short carbamate molecules and the recovery of the polyol. The two compounds are not miscible and lead to an in-situ phase separation [322] into polyol-rich and carbamate-rich phases. This process still requires high temperature and energy that can hamper its benefits, with some LCA analyses demonstrating a negative impact compared to virgin petro-based PUs [324] due to the higher isocyanate content needed to compensate for properties depletion.

Alcoholysis, using methanol, may also offer solutions towards milder depolymerization processes [325]. In general, hydrolysis, alcoholysis, and glycolysis allow high depolymerization yields, but separation and purification showcase low purification yields. Recently, an acidolysis process developed by Bech et al. [326] has allowed the direct recovery of the amine and polyol in high yield (90%).

In some cases, urethanes can be enzymatically degraded [319]. This is only possible under specific conditions and in the presence of specific enzymes such as urethanase [262], and the process should not be expected to occur naturally in the environment within a reasonable time frame. Moreover, PU bulk thermosets may be resistant to enzymatic degradation due to the low contact between the enzyme's active site and the urethane function [327]. Other strategies could also rely on the integration of enzymes during the polymerization to trigger the depolymerization when needed [328]. Up to now, with scientific advances, urethanase and bacterial degradation could offer an alternative pathway to recirculating PU-based composites.

6.6 Covalent adaptable networks—bridging thermosets and thermoplastics, but still a lab curiosity rather than a market reality

Thermoset matrices offer the best properties for structural composites. They can efficiently wet fibers through several processes, particularly infusion-based, and provide improved interfacial adhesion, key parameters towards high performance. In addition, they offer satisfying longevity and environmental resistance, ensuring a long service life and reducing the need for maintenance and replacement. On the other hand, their stability makes the treatment of decommissioned structures problematic.

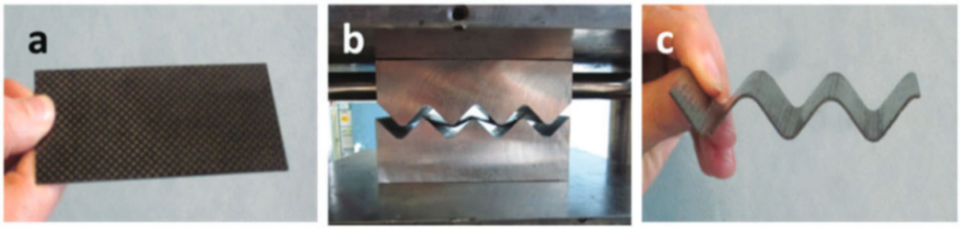


Fig. 38 Thermoformation of cured composite laminate exploiting epoxy CAN with dynamic disulfide. A 2-mm thick carbon fiber-reinforced dynamic epoxy laminate (**a**) was compression-molded in a zig-zag-shaped mold (**b**), rendering a thermoformed wavy 3D part (**c**). Reproduced with permission from [330]. Copyright 2016 Royal Society of Chemistry

Another approach that potentially gathers the advantages of both thermosets and thermoplastics is based on covalent adaptable networks (CANs). They are crosslinked polymer networks, similar to thermosets, but they possess dynamic crosslinking nodes that can be triggered on demand. This allows the network to be reworked, even after gelation[329]. Consequently, CANs display the reprocessability characteristics of thermoplastics (see Fig. 38).

6.6.1 Historical and fundamental background of CANs

The concept of bond exchange in crosslinked polymers dates back to the late 50s [331]. Reversible gels and polymers were reported in the 80s [332] and 90s [333–335], in particular by the Rubinstein group, but they faced limited possibilities and applicability. In 2002, Wudl et al. [336] developed a polymer network that can "re-mend" itself by exploiting the reversible Diels-Alder addition reactions. Such networks are known as self-healing polymers. Another work led by Lehn explored reversible covalent networks such as "dynamers" [337]. Bowman and co-workers introduced the concept of CANs [338, 339]. In parallel, Montarnal et al. exploited transesterification in epoxy through the curing of epoxy with carboxylic acid and a Zn-based catalyst to obtain a permanent crosslinking that can be reprocessed [340]. This specific class of CANs, so-called "vitrimers," possesses a temperature-viscosity relationship following an Arrhenius relationship by analogy to vitreous glass.

From a mechanical viewpoint, CANs differ from thermosets through their ability to relax stress above a specific

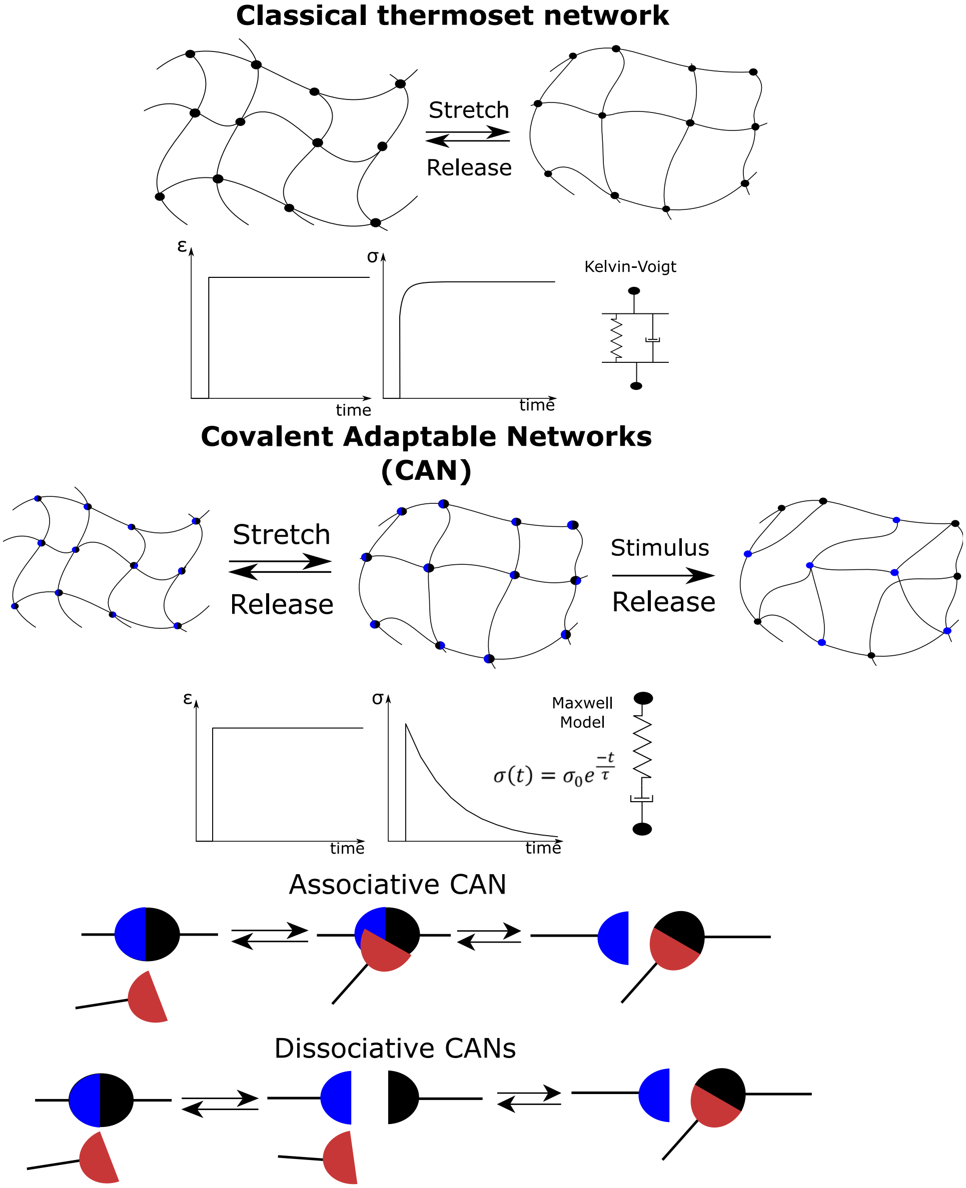


Fig. 39 Schematic representation of thermoset upon applied strain/release and CAN upon strain/release in the presence of an external stimulus to trigger exchange reactions. Figure inspired from [346]

temperature, usually called vitrification temperature or topology freezing temperature (T_v) [341, 342]. If the glass transition is superior to $T v$, the chain mobility is not sufficient to allow relaxation within a realistic timeframe. Typically, CANs, particularly those with a high glass transition, are characterized under small deformations to remain within their linear viscoelastic region [343]. In that case, a Maxwell model can be adequate (see Fig. 39). However, many complex chemical and physical mechanisms can take place at the same time. Therefore, more sophisticated models, such as the generalized Maxwell model (Wiechert model) or stretched exponential models, are sometimes used to better represent the mechanisms that take place. There is still no recognized standard for the characterization of CANs, nor unique methods. The most accepted method remains the stress relaxation experiment, mostly under shear (in a rheometer) or tension (in a DMTA apparatus), to demonstrate the ability of the crosslinked polymer to release internal stress through rearrangement of its internal structure. However, this stress relaxation ability makes CANs more sensitive to creep [344], in contrast to thermosets. Creep must be considered when designing load-bearing structures, which could be detrimental to long-term performances [345].

6.6.2 The diversity of CANs and their relevance in composites

The most common classification of CANs distinguishes dissociative and associative CANs. In dissociative CANs, the dynamic moieties are temporally dissociated to react with the free functional groups [341]. The network connectivity is thus partly reduced. In associative CANs, also called vitrimers, the free reactive functions connect as initial bonds break, maintaining a permanent crosslinking density [341]. It is important to note that if associative and dissociative networks tend to differ in rheological behavior, both can showcase a "vitrimer-like" behavior. The most meaningful point is that upon certain conditions, such as swelling in a good solvent, the equilibrium of dissociative CANs can be switched towards dissociation and thus allow the dissolution of the network [246]. Associative CANs are more stable and cannot be simply depolymerized. Several dynamic moieties can undergo both associative and dissociative exchange reactions, particularly playing on reaction conditions and catalysts [347]. For the sake of simplicity, the distinction between associative and dissociative will not be made in the rest of this state of art. However, it might imply significant

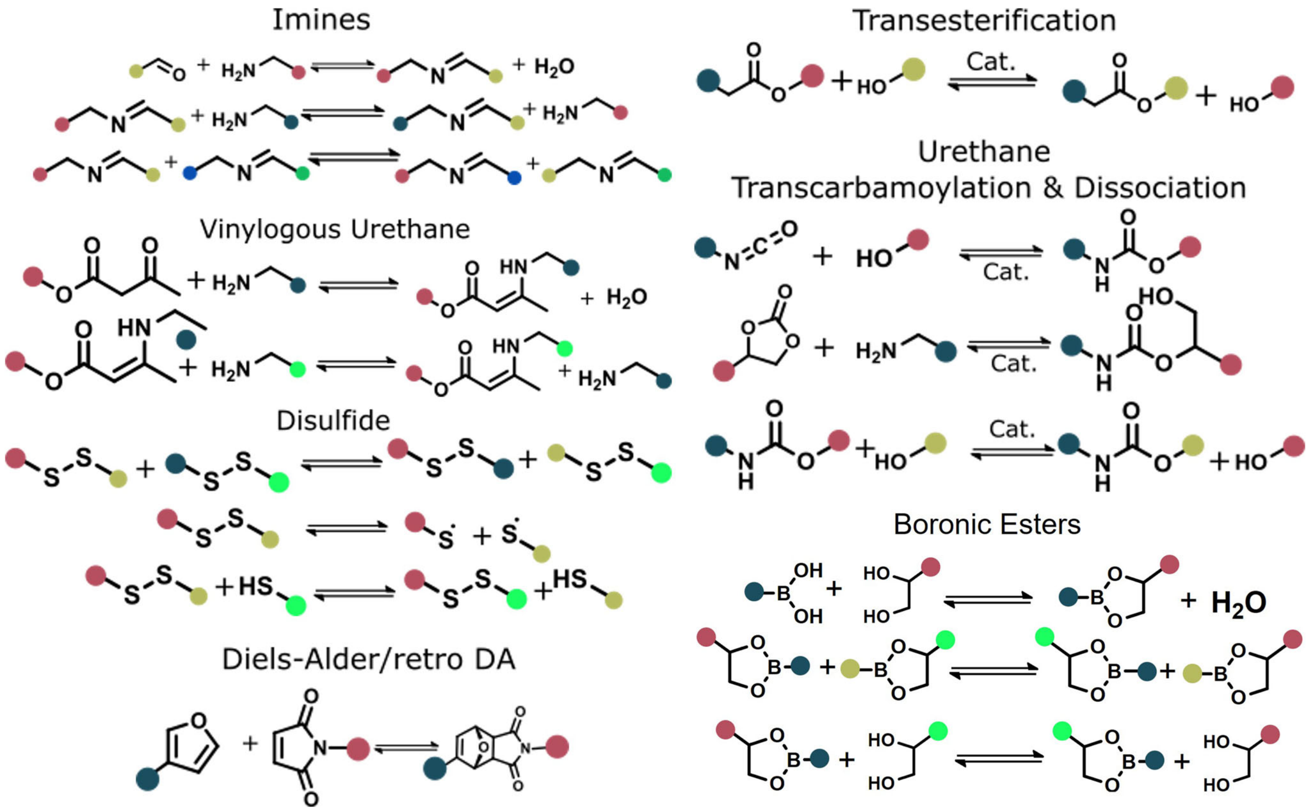


Fig. 40 Most important dynamic chemistries exploited in composites and exchange reactions

changes in chemistry, applications, and limitations. In particular, many dynamic moieties can exhibit both associative and dissociative behavior [347], and the apparent behavior is often close as the kinetics and thermodynamics of dissociation are not sufficient to observe a sol–gel transition [341].

Ushered by high scientific interests, numerous CAN chemistries, strategies, and materials have been unveiled within the last decade (see Fig. 40), with extensive literature reviews [275, 348–350], including bio-based [16, 22, 351] and composites [25]; recently, Du Prez et al. discussed the current status and trends for the implementation of CANs in several market application and highlighted the future needs [23]. Therefore, this section only intends to briefly reposition CANs in the frame of more sustainable structural composites.

CANs remain very scarce and at low TRL for industrial applications, probably due to their limited availability on the market and the associated costs. To date, only Mallinda®, an American start-up, commercializes dynamic EP for composites. LCA has almost not been performed for CANs, with only Vora et al. [352] who assessed the environmental footprint of polydiketoenamines. However, their cost and GWP were yet to meet the results of commodity polymers. By modeling a more efficient and industrial process, they showcase that circular polydiketoenamines can match benchmark polymers. This work highlights the importance of developing LCA-assisted strategies to assess the accurate environmental footprint and cost of CANs, which is severely lacking in the literature.

We propose herein to discuss only two main strategies regarding composites: (a) new dynamic networks with potential in composites and (b) transforming existing thermosets (i.e., UPR, EP, and PU) into CANs.

6.6.3 New dynamic networks for structural composites

The development of new chemistries can leverage significant potential for composites. However, mastering building block synthesis, polymerization kinetics, and properties is complex and usually requires several years of work and increments before achieving a partially optimized form. Moreover, such new chemistries must be scaled before being considered in composites.

This is the case with polyimines, formed from the condensation of (poly)aldehydes and (poly)amines. They were among the first applied to composites [353] thanks to the wide availability of multifunctional monomers. The network is also soluble in an excess of amine, allowing the fibers to be recovered. High mechanical performances were even obtained. Interestingly, imines are highly dynamic and can be processed through transamination in the presence of free amines or through metathesis [354]. The exchange is fast, allowing quick welding processes [353]. However, the implementation of large-scale composites exploiting polyimines is complex, and their sustainability is questionable, although it has not been assessed up to now. The network proposed by Taynton et al. [353] was composed of terephtaldehyde, a toxic aromatic dialdehyde; tris (2-aminoethyl)amine (TREN), a highly toxic polyamine; and several diamines, including ethylene diamine, a volatile and toxic diamine. The condensation of amines and aldehydes occurs spontaneously and generates 1 equivalent of water. To allow the impregnation of the matrix, ethanol was used, which needed to be evaporated, and the formed water also needed to be removed to advance the curing. The formation of imine is reversible; this signifies that in the presence of excess water, they can go back spontaneously to the aldehyde and amine, thus affecting the material integrity. Such questions are fundamental for considering composite applications and hydrolytic stability, and weathering should be investigated if polyimines are to be considered for composites.

Vinylogous urethanes/ureas (VU) were introduced by Du Prez's group as a potential replacement for imine and ester, owing to favorable thermodynamics [355]. VUs are formed through the condensation of (poly)amine and (poly)acetoacetate (AA). Acetoacetate synthesis is fully scalable and can be considered green as no solvent is required, and purification can be performed through simple distillation. AA can be obtained from any polyols, including biobased ones like isosorbide [356], accessing a wide array of sustainable building blocks. Like imine, it forms water that needs elimination for composite manufacturing. However, the thermodynamics strongly favored the formation of the VU adduct, and thus, water does not affect the extent of polymerization. They are much less sensitive to hydrolysis [355]. The exchange reaction requires an excess of amine but no catalyst; full reprocessing can be obtained within minutes at a temperature below 150 °C. The authors demonstrated that they could completely control the dynamicity through the use of an organic catalyst [357]. VU (urea-based) was successfully applied to GFRP by manual impregnation [358], and the welding of cured prepregs was feasible, featuring comparable mechanical properties to epoxy benchmark ($E = 40$ GPa, $\sigma = 800$ MPa). However, two issues might hamper the widespread adoption of VU in composites. The first issue is process-related. The addition reaction between amines and AA is extremely fast and occurs at room temperature; thus, the injection of liquid resin might be challenging to perform in a reasonable time frame before gelation [355], especially impossible for large structures. As aforementioned, the water removal also needs to be implemented in the process. The second issue relates to the highly dynamic nature of VU. An excess of amine is needed for the exchange reactions to take place, but they can occur (at a slow rate) at room temperature. This leads the VU to creep, a common issue for many highly dynamic CANs [344]. To deal with such, Du Prez et al. developed several strategies. Eliminating the excess of amines while inserting secondary amines in the vicinity of the enamine defined so-called "masked primary amines," allowing the fast rearrangement at an elevated temperature while suppressing creep [359]. Another approach has been recently developed that consisted of the control of the topology by intermolecular hydrogen bonding of a hybrid vinylogous urethane/urea network to control the dynamicity [360]. Finally, the design of switchable catalysts [361] also helps obtain low creep at service temperature and high dynamicity at recycling temperatures. Investigating and characterizing such strategies in applied composite materials could bring substantial benefits to structural materials sustainability, but still, an environmental assessment is yet to be performed.

Boronic esters were first introduced as self-healing polymers [362]. Since then, they have been exploited to provide healable and dynamic CANs, mostly elastomeric [363–365]. Boronic esters are typically used as crosslinkers for linear polymers or oligomers by reacting with pendant vicinal diols [366]. The reaction also generates water that requires elimination procedures. Little information is provided concerning the applicability of this chemistry in composite materials or their sustainability. Boronic esters are sensitive to hydrolysis, which can again decrease the stability of the matrix in a humid environment.

Recently, Raman et al. [367] introduced a new strategy to tailor the interface of composites. They inserted dynamic boronic ester functions at the fiber/matrix interface. They created covalent bonding between the fiber and the matrix by functionalizing the CF with a vicinal diol (see Fig. 41) and modifying a commercial styrene-butadiene linear polymer with boronic functions. A highly efficient stress transfer within the material was achieved, while maintaining its toughness. Through this strategy, they increased the strain and stress of the resulting CFRP by 50% compared to a conventional DGEBA-based matrix, with an 84% improvement in toughness. Importantly, as all crosslinking nodes were derived from these boronic esters, the network was degradable in pinacol to retrieve the CFs. However, the requirement of solvents (THF and DMF) to manufacture these composites and the complex synthetic steps needed might make this strategy not ready for large-scale composites. In any case, it could be a promising approach to unify the recyclability, properties, and toughness issues of composite.

Many other dynamic networks exist, in particular those arising from click chemistry, such as thiol-enes, acetals [246, 368], or β-amino esters [369]. However, access to these novel chemistries remains extremely limited due to the complexity of scaling monomer synthesis to the minimum quantities

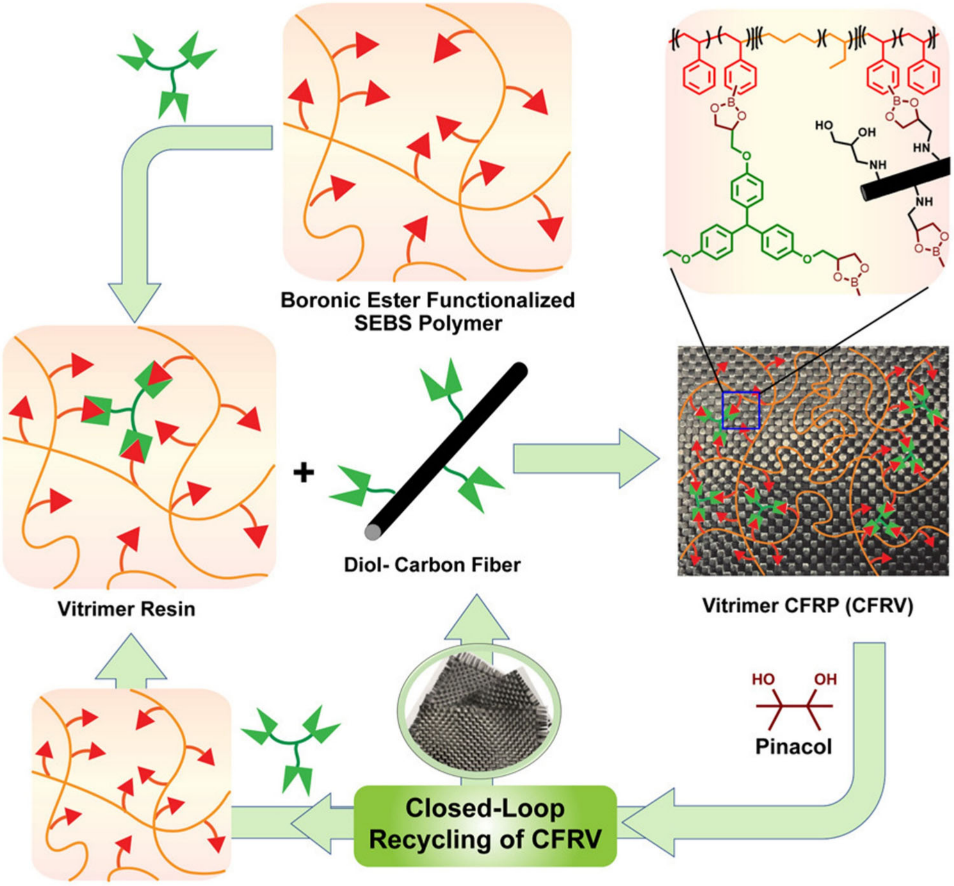


Fig. 41 Design of CAN with implementation of dynamic boronic ester moieties at the fiber/matrix interface to achieve a recyclable, mechanically robust CFRP composite. Reproduced with permission from [367]

required for lab-scale composite manufacturing. Additionally, there is limited data available on the truly sustainable aspects of what is sometimes referred to as "green" chemistry, particularly when using hazardous solvents and catalysts. Besides, the characterization of the materials is often insufficient, with a lack of discussions, which further complicates and slows the selection of potential candidates for composite applications. Therefore, the most advanced dynamic networks rely on modifying well-known thermosets.

6.6.4 Switching existing thermosets to CANs

A crucial strategy consists of inserting dynamic linkages into existing thermosets, ideally with minor changes. This approach facilitates the implementation of CANs into existing processes and known chemistries. The most prevalent dynamic linkages include esters, urethanes, disulfides, and imines.

Unsaturated polyester resin (UPR) CANs have been poorly investigated, with only 5 articles published so far on this topic. Supposedly, the presence of ester groups should facilitate the transformation of UPR thermosets into CANs, by, for instance, introducing free hydroxyl moieties and a catalyst to enable transesterification. However, this might also affect the properties by plasticizing the UPR. Rizzo et al. proposed a straightforward approach by including a titanium-derived catalyst, a cheap, non-toxic metal catalyst, into a commercial UPR formulation [370]. Titanium-catalyst efficiently activates the intramolecular ester metathesis. The resulting vitrimer showcased minimal property modifications compared to the benchmark and good properties retention, with 50% modulus recovery after reprocessing. The strategy was successfully applied to a CFRP [371], allowing the healing of the composite. This could help the direct thermomechanical recycling of these composites, which is of high interest for EoL management of marine and wind turbines.

The carbamate moieties in polyurethane are also dynamic, and they can be involved in transcarbamoylation reactions [372]. However, to adequately perform these exchange reactions, PU CANs require an excess of free hydroxyl groups and a catalyst [372]. Tin-based catalysts are the most efficient but are highly toxic. Interestingly, they also help the polymerization, limiting the multiplication of compounds. The excess of polyol leads to a lower gel content of the polymer decreasing the stability and properties of the networks. Aromatic amines containing dynamic disulfide were successfully implemented into PU-based GFRP, allowing reshaping and welding of the composite [373]. However, the process is solvent-based and appears not suited for large-scale composites. Other dynamic functions such as imines and esters were also implemented [374] but are not relevant for the industrial scale of composites.

Finally, dynamic moieties have been largely implemented into EP resins. Ester dynamic moieties were first introduced by Montarnal et al. [340]. Free hydroxyls and dynamic esters can be directly formed from the reaction of epoxy and carboxylic compounds, creating a CAN in the presence of a catalyst (Zn-derivatives). Using epoxidized diamines and a carboxylic acid curing agent, the catalyst can be avoided, exploiting the internal catalytic effect of the amine [375]. The latter was successfully implemented into CFRP and could be depolymerized in the presence of ethanolamine.

Disulfide metathesis is widely exploited to obtain dynamic EP [330]. They are probably the most advanced for composites [25, 330]. Amine crosslinkers containing disulfides such as aminophenyl disulfide (APDS, aromatic) or cystamine (CSTA, aliphatic, biobased) can be easily incorporated into EP resins. Recently, Schenk et al. [376] demonstrated that APDS can be used to successfully prepare high-performance aero-grade CFRP by resin transfer molding (Fig. 42), with a glass transition of 232 °C. A time-temperature-transformation (TTT) diagram was developed to control the curing progress through infusion, representing the first TTT diagram of CANs.

Based on imines that are constructed from the condensation of aldehydes (or ketones) and amines, several imine-containing epoxies have been explored for composites, primarily exploiting vanillin for biosourcing, and already containing a free alcohol and aldehyde [377–379]. This chemistry is exploited by Mallinda ®, with T_g up to 130 °C obtainable. However, as the formation of imines generates 1 equivalent of water, most processes are conducted by preparing an imine-containing epoxy, which is later polymerized. This strategy leads to high-viscosity monomers that often require solvents [377], representing a drawback in composite manufacturing. Nonetheless, eliminating the formed water by imine condensation is feasible, depending on the composite process, and might be of interest if properties, cost, and environmental benefits compensate for the complexity of the process.

Other dynamic functions have been successfully implemented into epoxy, such as siloxane-containing diamines [380], which allow fast reprocessing of composites, including flax-reinforced ones [157]. The properties were retained after reprocessing. This hardener presents very low viscosity, allowing the efficient infusion of the resin within the reinforcement. T_g up to 85 °C were obtained. One drawback might arise from the need for a catalyst, 1,5,7-triazabicyclo[4.4.0]dec-5-ene (TBD), in high content (10 mol%) needed for the process. TBD is an expensive and corrosive catalyst that is not registered under REACH regulations.

Although dynamic systems encompass many properties, no single CAN can fully meet the diverse and often competing demands for high performance and elevated dynamicity.

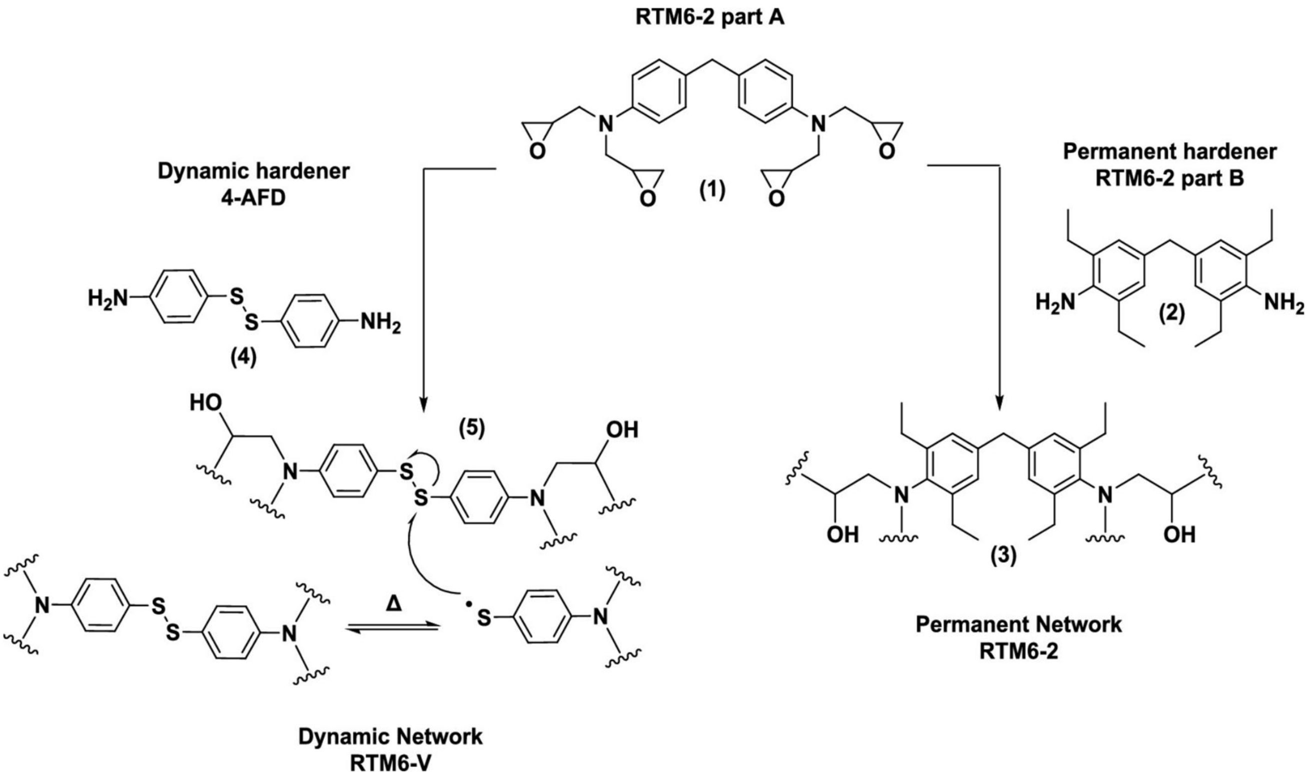


Fig. 42 Aerograde epoxy-disulfide CAN for Resin Transfer Molding of CFRP. Reprinted with permission from [376]. Copyright 2023 American Chemical Society

Various dynamic bonds have been incorporated into these networks to achieve the necessary tunability. Recently, Serra et al. [381] introduced an epoxy vitrimer featuring dual relaxation mechanisms based on imine and disulfide linkages, enabling straightforward access to a biobased epoxy network with balanced intermediate properties. However, the dual vitrimer copolymerization approach necessitates additional complex synthetic processes, posing a significant challenge to industrial scalability.

Significant efforts have been devoted to expanding the understanding and diversity of CANs, including their applications in composites. However, a fully satisfactory solution has yet to be achieved. Key challenges remain in combining competitive properties with availability, sustainability, and recyclability. In this context, investigating other emerging chemistries could prove highly valuable, particularly in the case of non-isocyanate poly(hydroxy)urethane as discussed in the forthcoming section.

6.7 Rising chemistries and alternatives

In parallel to the ongoing efforts to increase the sustainability of composites through the development of biobased equivalents to existing matrices, there are also emerging chemistries that are still at low TRL or marginally commercialized. This section aims to briefly introduce and discuss such emerging strategies in the continuum of the emerging CANs already introduced in a dedicated section.

Phenolic resins, typically phenol-formaldehyde, are commonly used for high-service temperature and chemical resistance [183]. However, the high toxicity and fossil sourcing of both formaldehyde and phenol push towards alternatives. As mentioned in Sect. 6.1.1, numerous lignin-derived phenols can be exploited to substitute petroleum-based phenols [183], such as cresol, resorcinol, or guaiacol. However, the substitution of these naturally occurring phenolic compounds tends to modify the reactivity and the properties. Although potentially biobased, formaldehyde remains a concern. Terephtaldehyde was shown to be a less toxic agent in combination with biobased phenolic compounds to provide sustainable phenolic resins [382].

Alternatively, polybenzoxazines (BXZ) are gaining a growing interest as phenolic resins and are relatively new, with a first commercialization back to 40 years ago [383]. Benzoxazines are pre-polymeric phenolic resins produced from phenolics, amines, and formaldehyde to produce benzoxazine monomers (see Fig. 43a). The stability of the cyclic oxazine ring allows storage at room temperature with long shelf life while the formaldehyde is “trapped” within the

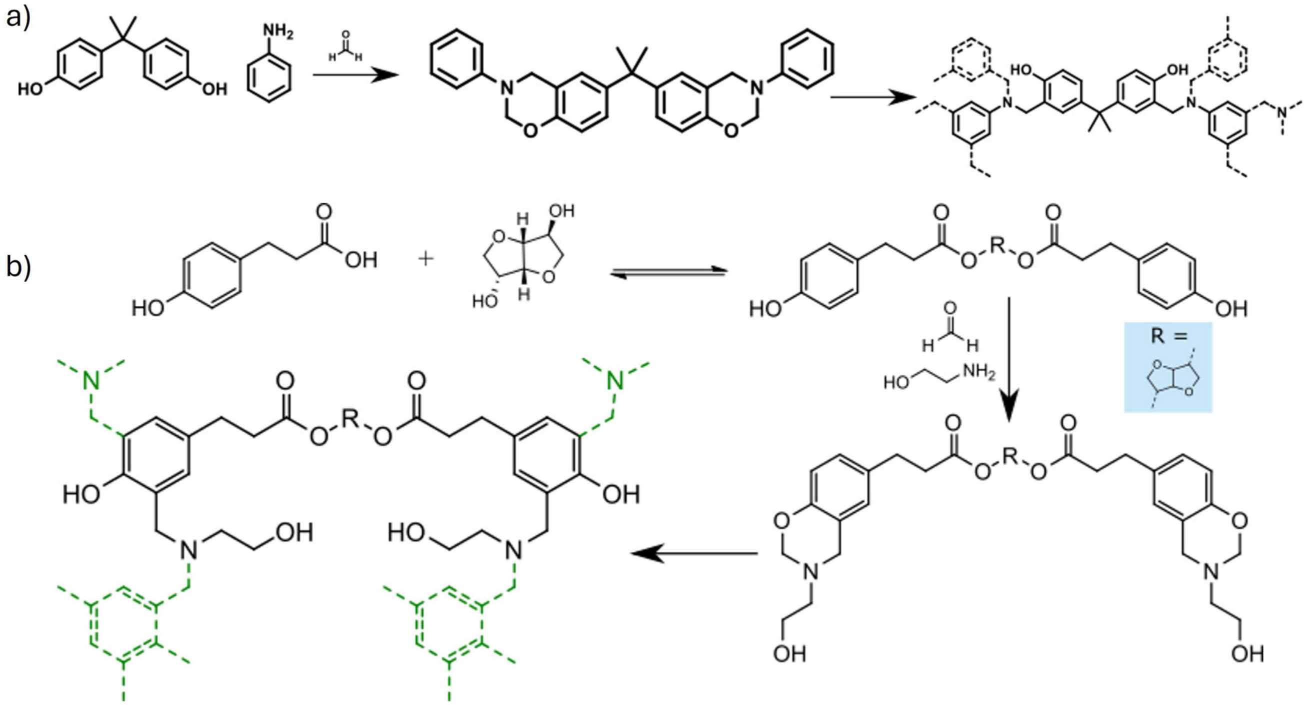


Fig. 43 Benzoxazine resins for thermally resistant matrices. **a** Representative BPA-derived benzoxazine and **b** bio-derived benzoxazine, from isosorbide, phloretic acid, and ethanolamine with dynamic ester moieties for high-performance recyclable matrices compatible with natural fibers [388, 390]

monomer, thus avoiding the handling of hazardous volatile compounds. Typically, benzoxazine monomer is produced by the ring closure of BPA with aniline and paraformaldehyde, usually in solvent conditions. BXZ are polymerized at elevated temperatures to produce high-performance resin with natural flame-resistant behavior. Glass transition superior to 200 °C and char yield superior to 50% are typical [384]. Recently, many bio-derived phenols [384, 385] and amines [204] have been explored to produce sustainable high-performance benzoxazines, including the incorporation of dynamic moieties into CANs [386]. As such, dynamic BXZ has been successfully developed and implemented to produce repairable composites [387]. Recently, our team successfully developed biobased CAN BXZs in combination with natural fiber to produce high-performance biobased composites with flame resistance and strong affinities with flax fibers [388] (see Fig. 43b). The resulting (biobased) matrices combine the interesting performances of conventional BXZ, such as elevated thermal and fire stability, while the dynamicity allows new features and advanced recyclability perspectives. Such materials could be of high interest in applications such as sports, leisure, or transportation industries, among others [389]. However, it is important to note that no environmental assessment has been performed on such emerging matrices, and the use of formaldehyde in the preparation of the monomers might remain an issue.

When discussing the potential of conventional polyurethanes in more sustainable composites, the issue of isocyanates appeared predominant. Their hazardous character, combined with the high toxicity of phosgene required for isocyanate synthesis and their detrimental environmental footprint [317], makes their replacement appealing.

In that aim, a growing interest has surged since 2010 to develop various non-isocyanate polyurethanes (NIPU) [391, 392]. However, only a few appear to be pertinent in the frame of a circular and sustainable economy. Among them, poly(hydroxyurethanes) (PHU), a family of NIPU, has garnered significant attention. These resins can readily derive from renewable precursors and offer modular and high-performance properties, which make PHUs viable and sustainable alternatives to traditional PU [393]. PHUs are produced through a step-growth reaction between (poly)amines and (poly)cyclic carbonates (CC). One of the best promises lies in the synthesis of CC which are easily accessible from the cycloaddition of CO_2 into (bio)epoxy (Fig. 44) [235]. The process has been demonstrated to be easily scalable, with several tens of kg already produced by laboratories [394]. The carbonation of epoxy gave minimal waste with a low E-factor [395].

The (macro)molecular structure of PHUs differs from PUs through the presence of hydroxyl groups pendant along the main chain skeleton. This structural feature imparts a more

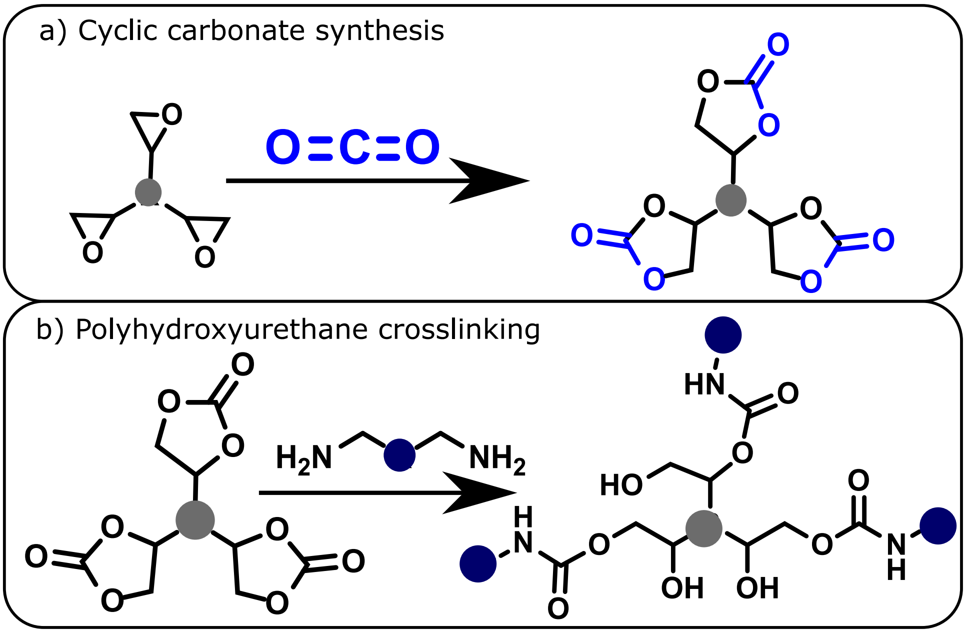


Fig. 44 Polyhydroxyurethanes, emerging sustainable CANs. **a** Obtention of cyclic carbonates from epoxy and CO_2. **b** Aminolysis of cyclic carbonates to polyhydroxyurethane network

hydrophilic character to the resins and enhances the ability of PHU chains to form multiple hydrogen bonds [396]. The presence of these hydroxyl groups is particularly advantageous for composite applications by providing tougher interfacial adhesion and forming intricate interactions with natural fibers [397]. Several authors have reported properties suitable for high load-bearing applications [398–400]. Another exciting aspect of PHUs is their ability to be reprocessed thanks to the transcarbamoylation mechanism already known in PU [372]. If all hydroxyurethane functions can rearrange through transcarbamoylation, the reprocessability of PHUs depends significantly on the network structure and their physical properties [399] and remains poorly efficient. Catalysts were investigated to improve the behavior [401] with DBTDL showing to be efficient to some extent but remaining an unsatisfying solution. Recently, our research team developed synergetic hybridization strategies by assembling epoxy with PHU to obtain a self-catalyzed network, significantly enhancing the dynamicity of the network [402]. By doing so, performance composites were obtained using carbon fibers [402] and natural fibers [403] that could be reshaped but also chemically recycled to separate the fiber from the matrix. The numerous opportunities leveraged by PHUs could make them ideal candidates for more sustainable composites if developed, optimized, and characterized adequately for such applications. More than PUs, the features enabled by PHUs, such as sustainability, dynamicity, and advanced properties, could make them ideal for replacing traditional matrices, such as the aforementioned epoxy and unsaturated polyesters, while being more easily accessible than other CANs.

The shift towards thermoplastic composites, which can be reprocessed more easily than thermoset composites, is another significant step forward. Thermoplastic composites can be reshaped, reformed, and reused, in alignment with circular economy principles. This shift is particularly important for mass production sectors, like automotive, where lightweight and recyclable composites are becoming increasingly accepted [404]. Thermoplastics enable the development of rapid processes, such as injection, 3D printing, or welding, suited for large-scale productions [40]. Yet, the properties in high load-bearing applications over long lifetimes have yet to match thermoset matrices. As discussed in the natural fiber sections, the high temperatures required for the polymer flow limit the option with thermally sensitive fibers and generate high energy consumption [405]. Although thermo-mechanical recycling is intrinsically feasible, it leads to a systematic downgrade of the material properties [406], while dissolution in a solvent for fiber/matrix separation is yet to be performed on large scales. Typical thermoplastics for composites include polyamides, polypropylene, polycarbonates, and polyesters. There are important efforts to produce thermoplastics from sustainable resources such as biobased polyamides [198, 407]. In general, thermoplastic composites are built in several steps as semi-finished products via direct blending in the case of short fibers or via impregnation with long fibers (pre-preg, commingled...). They are then sold as parts to be remelted inside a mold to give their final shape, resulting in energy-intensive steps. Moreover, the viscosity of the melted polymer can limit the efficient wetting of the fibers and thus degrade the part performance [405]. Interestingly, to reduce such issues, new opportunities arise from the in-situ synthesis of thermoplastics. In this case, (liquid) monomers are directly infused into the reinforcement with a catalyst to polymerize and promote efficient wetting of the fibers while maintaining the recycling feature [408]. Polyesters, polyamides, and polymethylmethacrylate (PMMA) were successfully prepared by thermoplastic resin transfer molding with satisfying properties. Currently, PMMA unpolymerized formulations for composite applications are commercially sold by Arkema under the name Elium®and were successfully implemented into large turbine blades [409] and were demonstrated to be economically advantageous due to the lower cost of tooling [410]. More detailed insights into (sustainable) high-performance thermoplastic composites can be found in previous reviews [20, 40, 408, 411].

7 Brief discussion about the importance of manufacturing technology in composites and their sustainability

The manufacturing technology plays a pivotal role in the properties and sustainability of composites. Selecting a specific manufacturing process for composite production involves several key considerations. Developing alternative (sustainable) chemistries should take into consideration such aspects to keep development relevant and ensure that environmental savings due to more sustainable sourcing are not hampered by more impactful processes later on.

On the one hand, the scale, shape, and production rate will dictate which processes are suitable and thereby define their overall costs. For example, autoclaves are adapted for large parts with high mechanical properties but showcase high energy consumption, high costs, and low production rates. For that reason, they are commonly used in aeronautics and aerospace [45]. The thermo-compression process is well-suited for small-scale structures with faster rates and parallel lines, such as sports and leisure [46]. On the other hand, the process will condition the matrix requirements, such as viscosity and curing time, as well as the type and form of reinforcement. Balancing these considerations and optimizing the conditions will contribute to the final quality of the material but also affect the final price and environmental burden [44].

The last two decades have experienced a surge in composite process developments. The historical craft processes, such

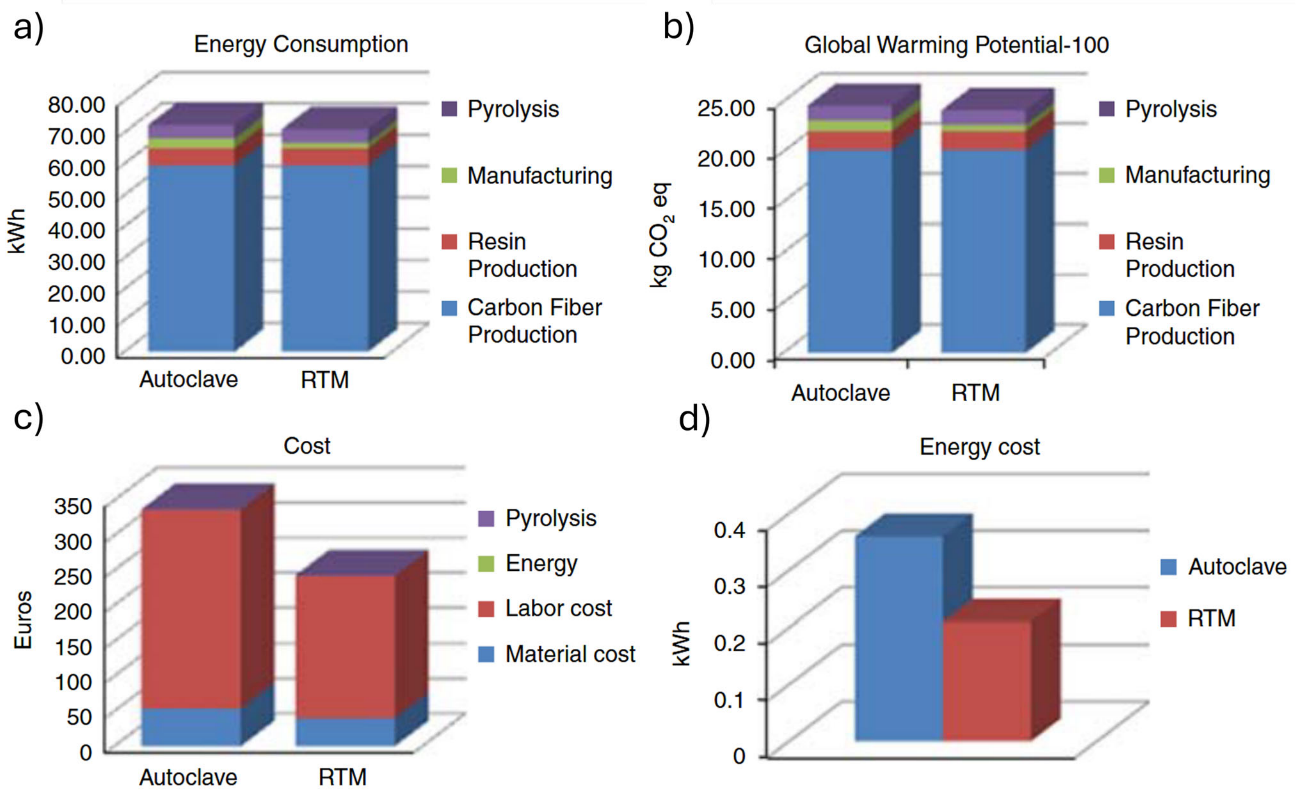


Fig. 45 Comparison of **a** energy consumption, **b** global warming potential and **c** total cost, and **d** energy cost of an aeronautic component manufactured by autoclave or RTM. Reproduced with permission from [419]

as hand lay-up, are increasingly replaced by more automated and controlled processes. The replacement is motivated by the need for a faster production rate, lower labor costs, and limitations of defects and waste. The recent advances in polymer chemistry for both thermosets and thermoplastics, in parallel to automation progress, has enabled the development of automatic fiber placement (AFP) [412, 413], welding [414, 415], pultrusion [416], additive manufacturing [417, 418], and many other processes [26]. We invite the reader to access the aforementioned excellent, specific reviews for more technical information.

Yet, the chosen process will significantly alter the material's environmental footprint due to the energy inputs and waste outputs. Energy, in addition to ecological considerations, remains fundamental in defining production costs. All these aspects push towards the use of out-of-autoclave methods [46], and some thorough LCA analysis on composite processes is now being performed.

As a quick example, RTM demonstrated significant energy and cost savings compared to autoclave [419, 420] (see Fig. 45). Equally, the energy savings, with less generated waste, were shown to enable the decrease of several environmental impacts, in particular, GWP but not only [419, 420], even compared to metallic process [11]. It is important to note that these results might differ from one application to another, as well as the origin of the energy input and process parameters. Some studies also demonstrated better results for autoclave over RTM [421]. Nonetheless, with a near-net-shape process, improvement of resins towards faster curing time and lower temperature would benefit all aspects of the composite value chain.

The process design is at the core of composite sustainability and its relevance in a circular economy to limit energy consumption, waste, and material final properties. Therefore, its central role in sustainability cannot be avoided. For such questions, Lunetto et al. [422] recently published an excellent, exhaustive review on the sustainability in composite manufacturing processes, including specific energy consumption (SEC) of the processes, and we refer the reader to a more comprehensive overview of this important question. We would like to highlight that the authors noted a substantial scattering in environmental data for materials and technologies. Also, they stressed the need to link process parameters, materials, size, and energy consumption more often, while some processes, such as additive manufacturing, natural fiber-related processes, or recycling methods, have limited data availability.

Processing technology has led to significant investments from composite manufacturers that strongly limit the adoption of other technologies. In that sense, new chemistries, including CAN ones, must be adapted to existing procedures to be implemented by industries [25] and supported by a fully integrated LCA.

8 End-of-life scenario, towards nested circular economy—environmental and technological perspectives

There are many pathways toward more sustainable composite materials through the selection of the reinforcement, matrix, processing, and optimized design of the structure. Even in the case of a genuinely green scenario with renewable sourcing, low-energy processes, and extended service life, any composite structure will end up as waste. At this point, end-of-life management becomes crucial in the recirculation and valorization of (some) constituents and imparts non-negligible impacts on the cradle-to-grave or cradle-to-cradle balance sheet.

8.1 General considerations

Many waste management strategies exist (see Fig. 46), though not all are implemented industrially. Currently, most (composite) wastes are landfilled, with some being incinerated, but the inorganic fibers still require landfill [65]. Incineration with energy recovery is sometimes considered but does not necessarily benefit the GWP, particularly in countries with low-carbon electricity (such as France, Sweden, or Paraguay).

A small proportion of composites is reused or recycled. However, landfilling of composite structures is progressively banned worldwide through tax incentives or new regulations, like in Germany in 2009 [47], due to land occupation and long-term pollution [34]. However, landfilling is still, to date, the most economic EoL scenario with an estimated 200–300 €/ton (including transportation, waste management, and gate fees) [424]. Direct repurposing of composite structures also exists, such as converting wind blades into bridges [425], but is marginal. Mechanical recycling is the most advanced and industrially applied recycling strategy, but it only provides low-grade materials because of the reduction of the reinforcement length [426]. This method is generally limited to thermoplastic FRP, which can be melted and reinjected into new applications. Thermoset-based FRPs are reduced to low-quality fillers. Again, the process of grinding tends to make the fillers not even competitive in prices and performance compared with conventional fillers such as carbonates or silica [37]. Nevertheless, mechanical recycling could still be viable through the use of CANs in order to enable higher properties of the recycled products using simple reshaping and welding [25]. The wear of grinding equipment should also be considered for large-capacity plants.

More promising strategies have been envisioned over the last two decades through the development of direct reuse/repurpose value chains, pyrolysis techniques, and chemical recycling. All of them impart different environmental burdens and economic potential, and the same level

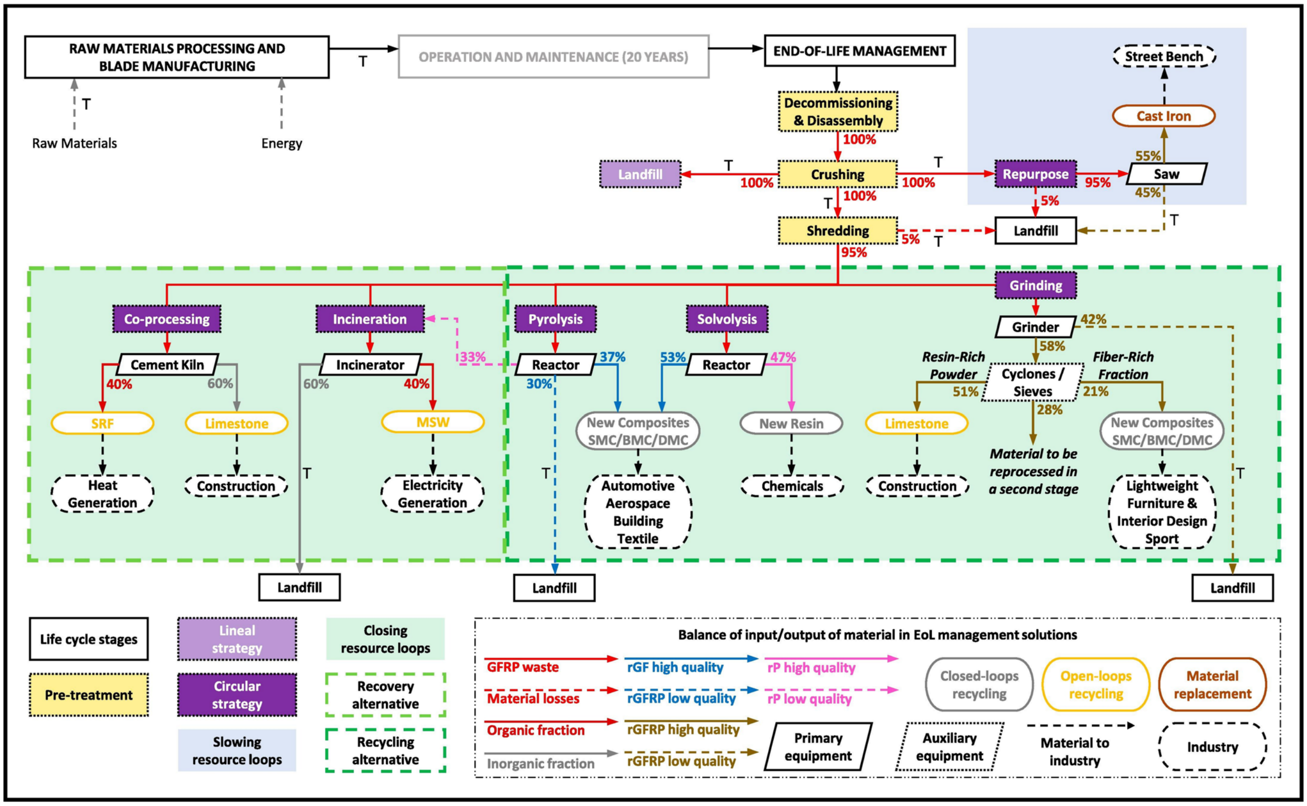


Fig. 46 Life cycle management of composite materials, as applied here to wind turbine blades, and the different EoL strategies applicable. Acronyms: T, transport; SRF, solid recovered fuel; MSW, municipal solid waste; SMC, sheet molding compound; BMC, bulk molding compound; DMC, dough molding compound; GFRP, glass fiber-reinforced polymers; rGF, recycled glass fiber; rGFRP, recycled glass fiber-reinforced polymers; rP, recycled plastic (epoxy resin). Reproduced with permission from [423]

of maturity is not achieved for all. No recycling technique was demonstrated to possess both low cost and low environmental footprint [427]. Moreover, the matrix chemistry, reinforcement type, form, and initial/final application types severely affect and dictate the economic and environmental viability of each EoL scenario.

In composite recycling, retrieving the fibers is the most critical aspect. They commonly represent 40–80% in weight of the laminate and about 60–95% of the economic value [26, 47]. They also convey the majority of the environmental score. Accordingly, fibers drive the environmental and economic interests of recycling (see Fig. 47). Virgin carbon fibers possess a high cost, ranging from 30 to 60€/kg (2018) [428], and a high environmental footprint (100–1500 MJ/kg), which makes them particularly interesting to recover. It was estimated that recovered CF by pyrolysis could range between 5 and 15€/kg to be profitable [427, 428] while grinding costs range between 0.3 and 5 €/kg [427]. As glass fibers typically range between 1–3€/kg for low-grade and 3–30€/kg for high-performance [427], the cost of most recycling methods is challenging to make recovered GF cost-competitive with virgin ones. Cement kiln was considered to be one of the best valorization strategies for GFRP by Hagnell et al. [37]. For both GF and CF, retrieving the fiber helps reduce the footprint compared to virgin materials by 20–90% [114, 427]. However, this value strongly depends on the fibers and process. Supposedly, glass and basalt fibers could be re-melted into virgin continuous fibers, but these have not been considered so far.

Apart from the mechanical recycling of thermoplastic/natural fiber composites [429], and the biodegradation of PLA/NF composites [430–432], the recycling of NFC has been poorly addressed or discussed. In NFCs, the matrix contributes a significant part of the total environmental footprint; therefore, the recovery of the matrix could be critical. Moreover, recycling methods involve harsh conditions such as aggressive chemical reagents, long times, or temperatures higher than 150 °C that cannot be considered due to the lower stability of NFs when compared to CFs or GFs.

Moreover, most studies focus on composite structures and pure composite materials; the importance of material identification, sorting, and the presence of other constituents such as metallic grids and manufacturing waste (uncured pre-preg cuts...) might require specific waste streams [11, 37].

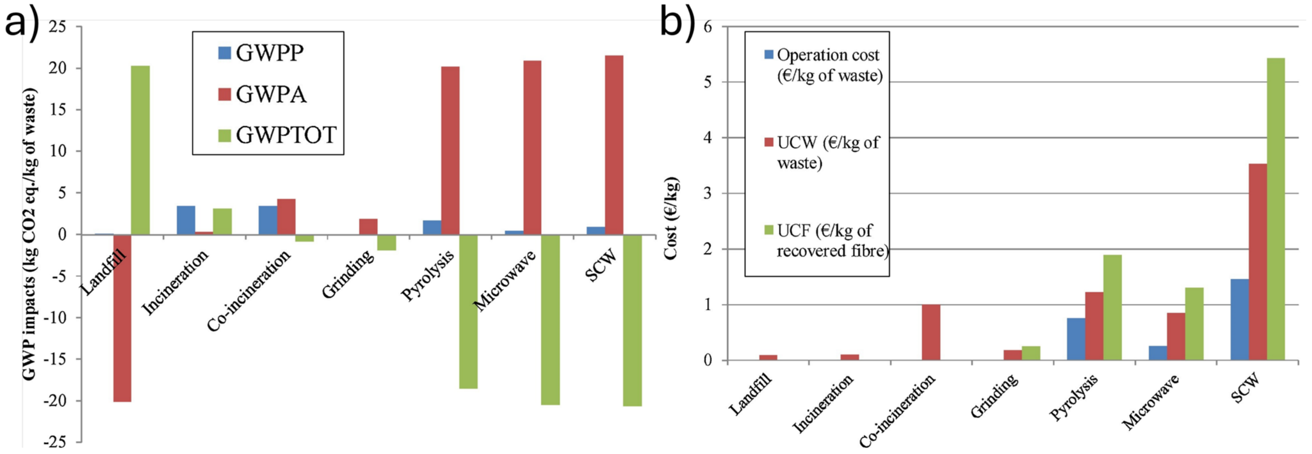


Fig. 47 **a** GWP and **b** cost implications of the EoL management of CFRPs. Acronyms: SCW, supercritical water; GWPP, GWP impact of the process; GWPA, GWP impact of substituted products; GWPTOT, GWP total of the system; UCF, the average unit cost per mass unit of recovered fiber; UCW, the average unit cost per mass unit of waste. Reproduced with permission from [427]

8.2 Pyrolysis—balancing energy inputs and recovery quality

Pyrolysis is one of the most advanced emerging recycling techniques. Before pyrolysis, composites are first collected and shredded (see Fig. 48). The pyrolysis process itself involves heating between 350 and 700 °C in an inert atmosphere; the process degrades the organic matrix into char, gas, and oil (potentially). The inorganic fiber recovered generally needs to be washed and treated for further use [268]. Pyrolysis can be advantageous as it does not necessarily require the removal of other chemical compounds, such as coatings or paint, that could be damageable in another process. It can be adapted to most matrices, although the temperature for some might differ; UPR can be degraded below 400 °C while high-performance resins such as EP or PEEK require higher temperatures [37, 82]. However, pyrolysis requires high energy input and can significantly degrade the fiber properties. Temperatures higher than 400 °C can lead to a 50% reduction of GF mechanical properties [70]. The pyrolysis process is currently cost-effective only for high-value fibers, such as carbon fiber. It is already implemented

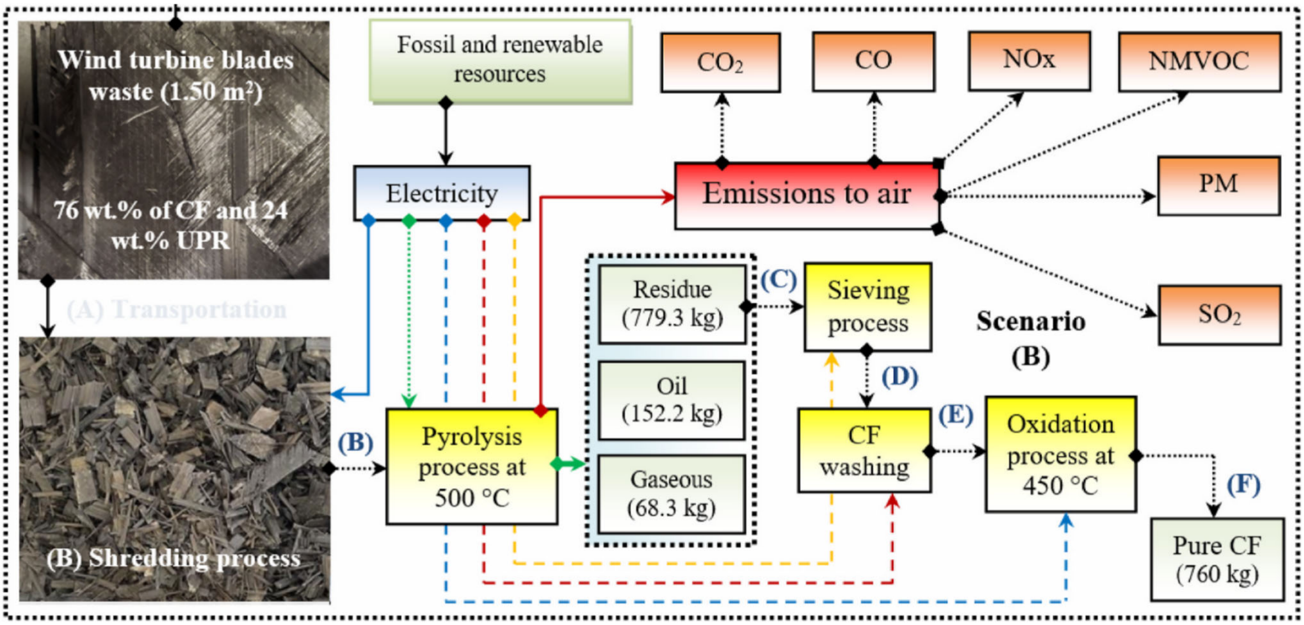


Fig. 48 Schematic representation of conventional pyrolysis of FRP and its outputs. Reprinted from [268] Copyright 2024, with permission from Elsevier

commercially for CFRP (Gen2 Carbon—ELG Group), and some Start-Ups are now implementing new pyrolysis equipment for pilot scale (Composite Recycling, Switzerland), using a mobile pyrolysis unit able to recover glass fibers. The details of the start-up technology, including economic or environmental benefits, are not disclosed. They recently signed a partnership with Veolia Recycling (2024) for pilot-scale pyrolysis recycling with a capacity of up to 2 tons/day.

In all cases, pyrolysis demonstrates more economic potential than incineration for energy recovery as it allows for the retrieving of materials. The oil produced from the degraded polymer might be expected to be valorized in the future [97, 433]. Additionally, using an oxygen-free environment prevents combustion, reducing air pollution.

A new generation of pyrolysis procedures is rapidly being developed [77]. In the latter, a bed of silica sand is fluidized by hot air to rapidly heat the FRP materials, facilitating the recovery of fibers through the erosion of the polymer matrix. Similarly, microwave-assisted pyrolysis is an advanced technique that heats FRP scrap materials by rapidly transferring thermal energy in an inert atmosphere, breaking down the polymer matrix into fuels.

From an environmental aspect, Yousef et al. [268] discussed the environmental and economic benefits of recovering carbon fibers from UPR composites through pyrolysis. They highlighted the potential to reduce several parameters such as climate change and fossil resources scarcity (see Fig. 50). However, the exceptionally high energy consumption (around 2500 kWh/ton of composites) led to detrimental outcomes on other criteria, such as terrestrial acidification and ozone depletion. Using catalytic pyrolysis and artificial neural networks, they successfully obtained aromatic chemicals that could benefit such a strategy [267, 433].

8.3 Chemical recycling

The chemical recycling of thermosets and their composites is appealing, yet it is not as well-developed as other strategies. It generally provides the highest fiber quality [434]. It can be divided into four main strategies: selective catalytic depolymerization, mild oxidative depolymerization, sub-critical solvolysis, and supercritical solvolysis. In all cases, solvents are necessary to dissolve the degraded product and ease the cleaning of the fiber. The solvent cost and environmental score severely affect the relevance of the process but also its efficiency [435]. Ideally, toxic and halogenated solvents must be avoided (e.g., DMF, dichloromethane/chloroform, hexane...), while non-toxic (biobased) solvents (ethyl acetate, ethanol, methanol, water...) with a low boiling point can support the recirculation [436].

The selective catalytic pathways target degradable bonds for high recovery yields. Conditions are typically expected to be below 200 °C; a pressurized step might be necessary if low-boiling-point reagents and solvents are required. However, the depolymerization conditions severely differ from polymers, catalysts, and solvents [296, 300, 437]. Effective selective depolymerization protocols have been developed for thermoplastics such as polyesters, polyurethanes, and polycarbonates, and are still to be implemented for thermosets. They require the use of an organic catalyst (commonly base, acids, organic salts) [263, 438] or a metallic catalyst [300, 439], and a nucleophile (alcohols, glycols, amines). In some cases, the nucleophile can also play the role of the solvent, as in the case of the depolymerization of ester-containing epoxy by methanol [210, 224]. Selective recycling strategies depend on the polymer chemistry and thus require sorting or multiple steps for mixed composite waste.

Unselective oxidative depolymerization has been developed for epoxy-amine-based composites. Interestingly, the fiber can be retrieved within a few hours at ambient pressure and a temperature below 80 °C. As such, it was successfully applied to glass and carbon fibers, but it has never been tried with natural fibers [298, 299]. Both selective and unselective depolymerization strategies were discussed in the specific section corresponding to each type of polymer.

Currently, there is no data on the economic or environmental benefits of these two emerging strategies. This is most probably because they are still at low TRL (2–3) and thus not enough established and generalized. Most of the LCA and DEA investigations for EoL management of composites are performed on sub- or super-critical solvolysis.

Sub-critical solvolysis is generally a hydrolysis strategy where water is below its critical point (200 °C < temperature < 374 °C and pressure < 221 bars). Other solvents (ethanol, methanol) can also be used. Supercritical solvolysis is performed above the critical point, is more efficient, has faster rates, and has higher recovery quality. They are still at the research stage, but their TRL is slightly higher than catalytic depolymerization, with pilot plants being implemented [28]. Difficulties in scaling up such a process could appear, but it remains beneficial for the overall results with optimized conditions [440]. The energy input for the process imparts the most economic and environmental cost. In all case studies, solvolysis was beneficial by comparison with incineration or landfill for carbon fibers [116, 427, 440]. Compared to pyrolysis, the results are somewhat unclear; some authors claimed better results of supercritical solvolysis over pyrolysis [423, 427, 440] (Fig. 49), while others position pyrolysis as more efficient [441]. From an economic viewpoint, pyrolysis remains more competitive at the current development stages [424, 427]. Glass fiber recovery from pyrolysis and solvolysis is not competitive with virgin glass fibers [37, 427]. Pillain et al. [440] took into account the fiber quality in their analysis. Indeed, pyrolysis can significantly decrease

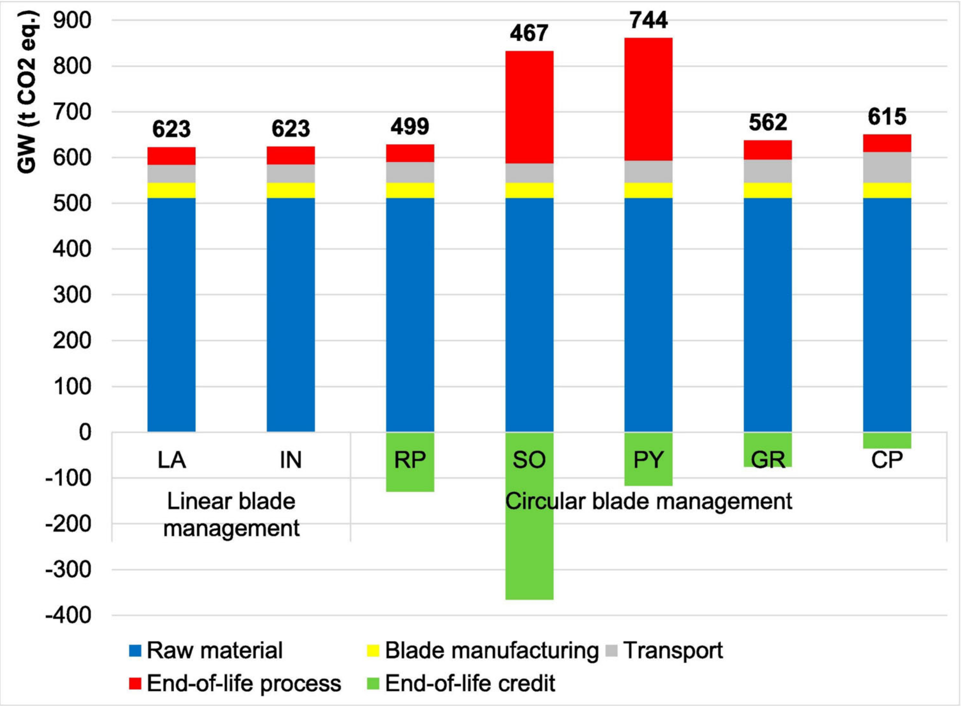


Fig. 49 Influence of the EoL management on the global warming potential of GFRP wind turbine blades. Acronyms: LA, landfill; IN, incineration with electricity recovery; RP, repurposing; SO, solvolysis; PY, pyrolysis; GR, grinding; CP, co-processing in cement kiln. Reproduced with authorization from [423]

CF properties and make them only suited to replace GF. They show that the substitution of GF by pyrolyzed rCF is not relevant when the decrease is superior to 40% (Fig. 50a). On the other hand, supercritical water does not alter the properties and provides significant environmental benefits. If the fiber quality is not differentiated, similar outcomes are obtained (Fig. 50b). The different recycling strategies should, however, be considered complementary to best mitigate environmental footprints and maximize product valorization.

None of these processes has been applied to natural fibers (except for acidolysis of Recyclamine-derived epoxides). Similar to pyrolysis, the conditions for supercritical solvolysis are not suitable for NF. Mild depolymerization conditions could be valuable but should be discussed environmentally (and economically). The environmental footprint of NF is much lower than that of synthetic fibers, and prices are only slightly higher than those of GF. The viability of recovering natural fibers might be questioned, in addition to the existing technological challenges. Upon those considerations, scientific efforts might be more valuable toward the design of fully biobased resins with extended durability and low GWP emissions.

8.4 Ideal valorization of recovered products

The valorization of the matrix-degraded products has been chiefly foreseen as non-viable [442]. The full recirculation of the products into a virgin polymer is not realistic if current developments are considered, albeit certain authors have succeeded at lab-scale [224]. However, the reagents involved and the successive purification steps make the economic and environmental aspects irrelevant. To date, a sustainable 100% recirculation of composites has not been demonstrated nor seems attainable despite significant efforts and breakthroughs. Up to now, unrecoverable organic compounds could be valorized through pyrolysis to recirculate carbon in naphtha and syngas (see Fig. 51), as claimed by Vidal et al. [19].

The fibers hold more promise for recycling. Composite properties are highly dependent on the form of the reinforcement. As reclaimed fibers will be necessarily discontinuous, conventional manufacturing of a randomly oriented mat can provide low-medium grade reinforcement [443]. Woven fabrics can retain the weave depending on the process and can thus be used as such [28]. However, obtaining aligned discontinuous fibers in continuous tape would be profitable for higher performance of the recycled composites. This process allows fiber to be realigned, woven, or transformed into thin isotropic mats. Notably, a sizing agent can be reapplied during this step as it is lost during the recycling [28]. Various techniques utilizing electrical, magnetic, and pneumatic methods have been explored in the past to align discontinuous fibers in a preferred direction, but most of them face limitations to the type of fibers, quality, length, and production rate [444].

To present one promising example widely studied, the University of Bristol developed the HiPerDiF equipment (High-Performance Discontinuous Fiber) [444]. The process explores the fiber suspension in water going through parallel plates with a water jet to obtain highly aligned fibers. About 65% of the fibers were in the ±3° range. The resulting unidi-

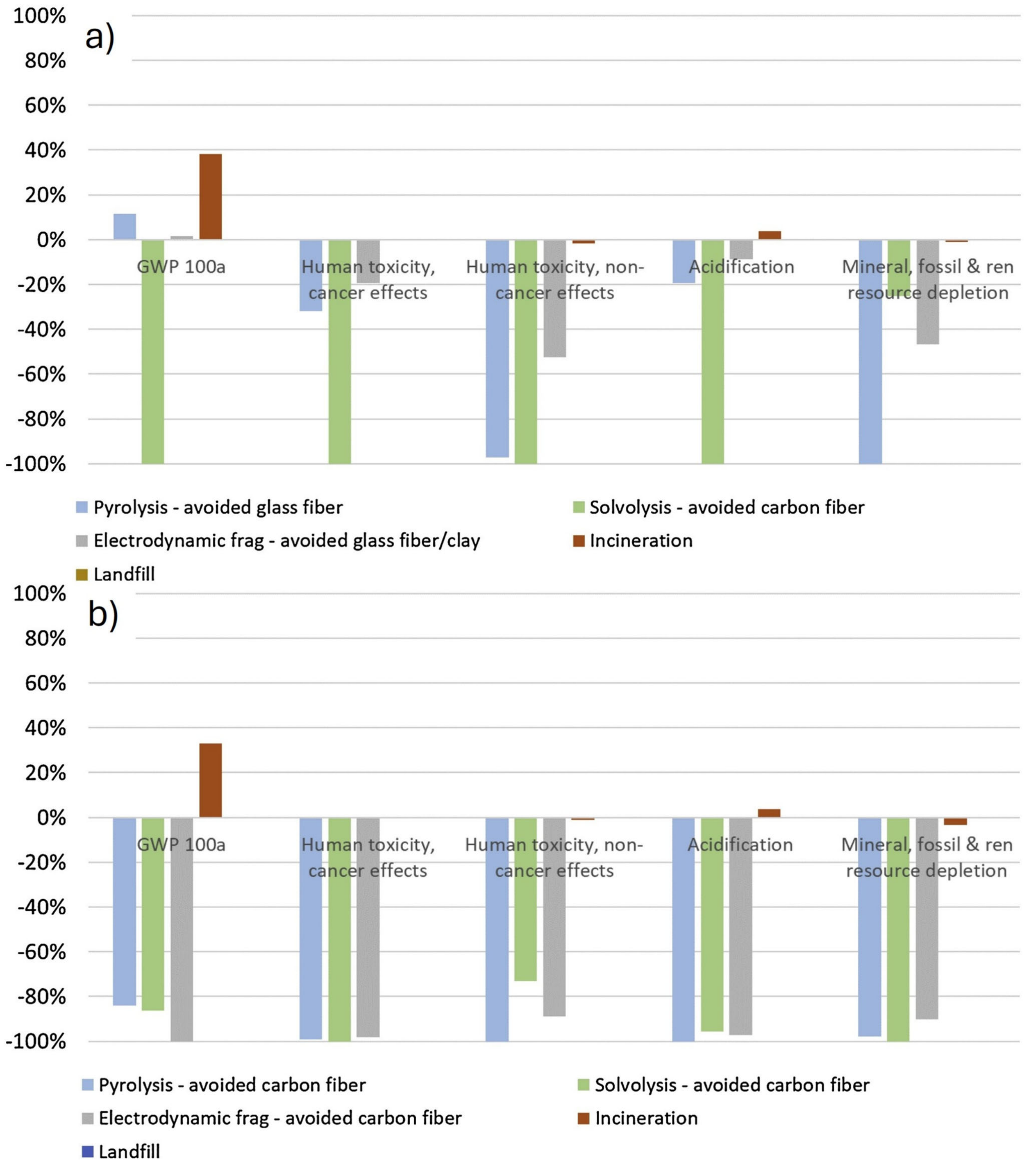


Fig. 50 Environmental impacts comparison between pyrolysis, supercritical hydrolysis, electrodynamic fragmentation, incineration, and landfilling, **a** considering the fiber degradation and thus different substitution product and **b** considering the same quality of carbon fiber (predictions upon optimization). Reprinted from [440] Copyright 2019, with permission from Elsevier

rectional tapes exhibited a tensile modulus of 115 GPa and a tensile strength of 1509 MPa, with a fiber volume fraction of 55%. The technology was successfully applied to a yachting demonstrator [445]. The research team also investigated the hotspot to lower the environmental footprint of the technology, mainly caused by electricity power [446].

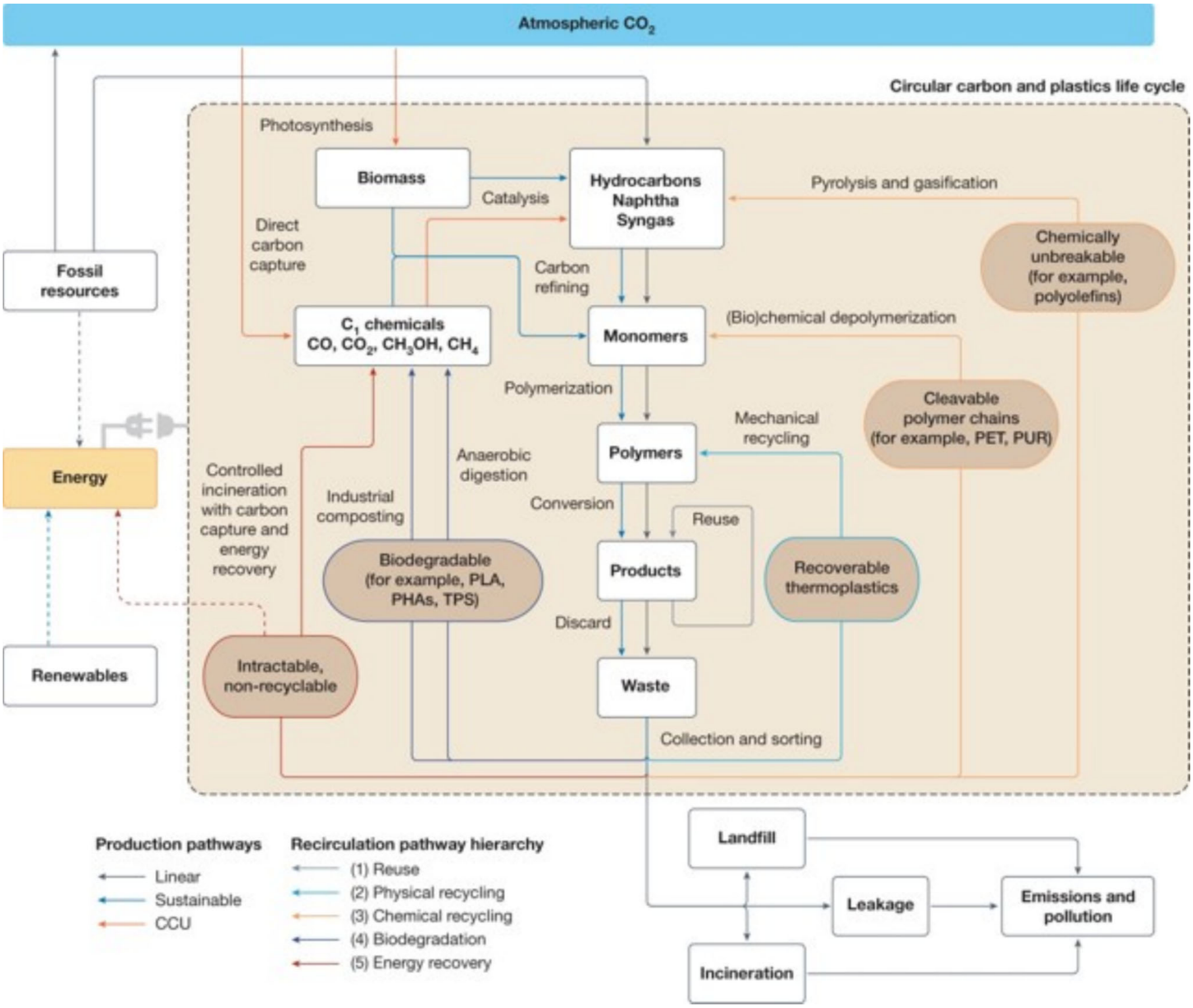


Fig. 51 The global circular carbon and plastics life cycle as proposed by Vidal et al. to replace the current linear flows (represented in black). Acronyms: PET, poly(ethylene terephthalate); PHAs, poly(hydroxy alkanoates); PLA, poly(lactic acid); PUR, poly(urethane); TPS, thermoplastic starch. Reproduced with permission from [19]. Copyright 2024 Springer Nature

Another research team in Bordeaux patented a mechanical equipment to unweave and realign carbon fibers [447]. From woven carbon reinforcement, a continuous tape with 95% of fibers in a ±14° range is obtained. High performances were also obtained [448]. The technology is now under commercial development by the Start-Up Nova Carbon®, in Bordeaux (Southwest France).

9 Summary and conclusions

Despite being known for many decades, the continuous depletion of resources and the exponential increase of the human footprint on the environment have raised awareness of the need to rethink the economic paradigm. The amounts of waste and rapid resource attrition due to our existing linear economy cannot be sustained for much longer than a few decades. The UN global objectives of limiting global warming to +1.5 °C by 2100 impose new strategies. Earth is a closed system, so limiting entropy imposes the use of renewable resources and the recirculation of waste. Efficiency must be improved at all levels through higher energy yields, lower material use, and eco-design.

Fiber-reinforced polymers (FRP) will be critical in achieving ambitious goals. Their exceptional strength-to-weight ratio allows for obtaining much more performant structures. FRPs have become an irreplaceable asset in many applications. Nonetheless, FRPs cannot be considered sustainable materials. Carbon and glass fibers contain high embodied energy and high GWP while causing the attrition of critical resources such as sand and oil. The fossil-based thermoset matrices significantly hamper the recovery of decommissioned composite structures. It is critical to rethink the composite industry from raw material extraction to end-of-life management and valorization. This review reports the current existing solutions to foreseen composites in a sustainable circular economy model.

Many academics have explored different strategies to lower the impact of composites, such as the use of natural fibers and thermoplastics, including the recycling possibilities. Using alternative fibers to glass fibers provides substantial environmental and economic benefits. Flax, hemp, and basalt are promising. Natural fibers are the only ones that demonstrate high properties when considering sustainable criteria, and NFCs are close to fulfilling net-zero carbon goals. Carbon fibers' sustainability seems to rely on the obtention of biobased PAN. Lignin-based CFs could also be beneficial, but are not competitive so far.

The pivotal role of chemistry in the realm of composites has often been occulted or forgotten. Many biobased substitutes for petro-based polymer building blocks have been reported in the literature, but have rarely been developed following the requirements of the composite industry. Only a few have investigated the actual environmental benefits of these compounds, while the chemical steps involved sometimes imply highly toxic or polluting reagents, solvents, and catalysts. An optimized valorization of feedstocks to different products and markets is often required to compete with highly atom-efficient and cost-effective petrochemical processes. Still, bio-based resins, particularly epoxy ones, seem to be promising in reducing environmental impacts and toxicity compared to conventional ones. There is a detrimental gap between the market, dominated by glass fibers (85%) and unsaturated polyesters (83%), and academic efforts, which mostly focus on the recovery of carbon fibers and epoxy chemistry. The direction taken by most academic laboratories in actually working on the biggest problems sometimes seems more driven by trends than rationality. Putting more effort into widely spread chemistries (i.e., UPR) seems critical if researchers want to make a global difference, but would require taking back knowledge into chemistries that have been partly abandoned. Building stronger consortia with industries should be prioritized to address both scientific and technical challenges while keeping solutions pragmatic and applicable.

Some emerging chemistries could benefit more through other complementary sourcing, such as CO_2-derived polymers and Covalent Adaptable Networks. Recyclable by-design thermosets are advancing, but should also be developed and characterized to better suit the composite industry requirements. As most of them are still in their infancy, they have not been investigated for such applications. Especially, there is a lack of comprehensive data available from the literature to demonstrate their applicability in composites within the context of a circular economy. The complex interface between fiber and matrix, crucial to the final composite properties, is often not considered when developing new resins, particularly for natural fibers.

The scaling of building blocks synthesis is often underestimated or unaddressed, while proof of concept will quickly need 1–10 kg synthesis scales. This includes the need to develop resins that are implementable into efficient manufacturing processes such as RTM. Developing low-viscosity resins that can be cured in a short and controlled time at low to moderate temperatures remains the main threshold for their potential implementation. Again, the evaluation of environmental impact and economic worth should be addressed to a larger extent, as only one LCA has been reported for CANs up to now. In addition, the toxicity of building blocks is increasingly being looked at by industries, and their investigation should be systematized; it is extremely rare to observe the assessment of this aspect in studies aiming to replace toxic compounds.

It is also important to note that, despite all promises, these emerging polymers will still require, depending on their current TRL, many years to enter the market. Adding up the lifetime of composite structures, the recycling question will only arise in 30-50 years from now. Therefore, more efforts are needed to address the end-of-life of actual composite structures but also the ones to be built over the next decades at least. The recycling of composites remains complex despite the development of various strategies. The economic and environmental benefits rely on the recovery of the fibers, as the recovery of matrices or functional building blocks is too complex and worthless. Considerable efforts must be conducted in this direction, and new catalytic pathways developed for commodity polymers could benefit the composite industry, but they still need to be validated.

Not all processes lead to the same fiber quality, environmental score, and economic value. The different processes must be developed in a complementary manner to mitigate current drawbacks. Mechanical recycling is the simplest method, but it downgrades the composite to a low-value use. Pyrolysis and solvolysis are the most advanced emerging technologies for recycling as they can provide higher quality fiber, but optimization of conditions is essential to ensure relevant outcomes. The recovery of CFs is, so far, the only valuable one because of its extremely high cost and environmental footprint. The current recycling costs make the recovery of glass fibers not competitive with virgin ones. Improvement in the processes' efficiency, the rise of energy and resource prices, taxes, and regulations could boost the interest in recirculating glass fibers. Innovative recycling methods and catalysts have been recently unveiled. However, their industrial relevance is insufficient, and more efforts are still required. Natural fiber recycling has almost not been studied, apart from biodegradation and mechanical recycling (limited to some thermoplastic matrices). The recycling of NFC with thermoset matrices is still to be established and discussed.

Despite efforts, solutions are still insufficient. To foster the development of more sustainable advanced materials, systematic multidisciplinary investigations and proper quantification metrics are recommended. As pointed out in this review, teams gathering chemists, materials, mechanicals, engineering, and environmental experts seem to be needed. Adding toxicologists and economists would also be beneficial, while industrial advisory boards should help ensure the applicability and transfer.

Also, we would like to emphasize that aiming to develop sustainable materials is neither easy nor straightforward. It is unrealistic to think of one technological solution to tackle all issues. In the same trend, it is unlikely that fully sustainable materials can exist with no impact on all sustainable metrics,

Table 3 Areas of improvement

Area of improvement	Current practice	Suggested practice
Material traceability	Lack of traceability in sourcing of bio-based or recycled feedstocks for matrices and fibers production	Implement cradle-to-gate traceability and certification for renewable inputs (e.g., ISCC+, FSC)
Fiber selection and evaluation	Emphasis on mechanical properties; sustainability criteria are often overlooked for most real-world applications	Combine mechanical performance with sustainability, embodied energy, and EoL options to select fibers. Adopt the use of fibers with lower performance metrics for applications where high-end properties are not critical
Natural fiber integration	Accounting for 5–10% of fiber use, natural fibers are often promoted without solving interfacial adhesion challenges or inconsistencies in their chemical and physical variability	Increase the implementation of natural fibers and improve compatibility via tailored surface modifications or better resin selection and standardize quality for industrial aims
Matrix selection	Non-recyclable, fossil-based resins remain prevalent due to cost, availability, and performance advantages	Promote the adoption of bio-CO2-derived resins and alternative matrices with enhanced recyclability, such as CANs. Also, investigate hybrid matrices to combine properties and explore synergies, improving sustainability and industrial applicability
Recycling pathways	The primary recycling methods currently relevant at the industrial level include pyrolysis, mechanical recycling for short fiber recovery, and cement kiln co-processing mainly for GFRP. Other techniques remain at the pilot stage or are in the early phases of industrial implementation	True closed-loop alternatives should enable the recovery of fibers in their pristine state, along with the regeneration of building blocks for the matrix preparation. Standardized and opened LCA and techno-economic analyses must be considered when evaluating these idealized recycling approaches
Manufacturing	Manufacturing is often optimized for speed, cost, and performance, with little consideration of energy consumption, material waste, or compatibility with sustainable resins and fibers	Develop and assess low-energy, fiber-friendly, and scalable processes (e.g., RTM, compression molding with green matrices). Encourage design-for-manufacturing principles that align with circularity goals, including repairability and remanufacturing
End-of-Life (EoL) Scenarios	The majority of composites are currently landfilled. While the organic matrix may be incinerated for energy recovery, the inorganic components and non-combustible residues are typically landfilled at a relatively low cost, estimated between €200–300 per ton	Ideally, composites should be directly repurposed or incorporate CAN-based matrices to enable direct reuse and reshaping. Lifetime should be extended. Further, advancing recycling approaches that recover both fibers and matrices with minimal economic and environmental impact
LCA and sustainability metrics	Not sufficiently applied. Environmental impacts are too often addressed qualitatively or using outdated references without including actual sourcing. Too much focus on CO2 emissions (GHG) while other parameters are at least as important if not more (biodiversity, toxicity, water use/quality…)	Apply process-based or hybrid LCA approaches that include the use phase, repairability, and end-of-life. Avoid the use of single-score LCA and provide comprehensive analyses. Integrate metrics like GWP and embodied energy across the full value chain

and strategic choices will be required, either by engineers developing structures or by regulatory offices. Academic scientists should focus on investigating pathways and rigorously characterizing them, with a critical mind, objectivity, and transparency. It is possible that some more sustainable materials do not possess the properties needed for an application, or some materials/strategies thought to be sustainable appear worse than the ones currently applied. It is part of the research process, and negative aspects should be reported as much, if not more, than positive outcomes to help the incremental work and address challenges, as a collective effort instead of a publishing race.

This review intends to gather the most recent developments to develop sustainable composites and foster innovation in the field, in particular by trying to gather the polymer chemistry and engineering communities. However, some aspects have been deliberately overlooked, such as the comprehensive aspect of manufacturing (consumables, resources...), post-manufacturing aspects (paint and coatings, assembly...), or service-life (structural health monitoring, repairing, ageing...). Structural composites are highly complex materials with numerous steps of high value and importance. Embracing truly sustainable composites will require considerable effort from various actors with a global perspective.

Finally, we propose some suggestions in Table 3 to help drive composite materials into a more sustainable pathway.

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Author contribution G.S wrote the main manuscript, G.P. and A.G. provided help and writing in the main manuscript in the specific sections related to their expertise, and in figure preparations. H.S., N.A., and J-M.R. supervised the work, suggested corrections, and provided guidance in the conceptualization. All authors reviewed the manuscript.

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Data availability No datasets were generated or analysed during the current study.

Declarations

Conflict of interest Jean-Marie Raquez, a co-author of this article, is an Associate Editor for the journal and was not involved with the peer review process in any way.

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