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Sustainability in the manufacturing of composite materials: a literature review and directions for future research

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Abstract:

Composite materials showed great potential in replacing metal components in several applications allowing adequate component strength with reduced weight. From a sustainability point of view, a significant number of studies demonstrated that a component made of composite materials is one of the best responses to the recent global legislation for the reduction of energy consumption and CO₂ emissions. The sustainability of the production of composite components should be assessed by structured approaches that consider the whole life cycle of the component from the raw material production, the manufacturing process and post processes to the end-of-life (EoL). The purpose of this paper is to present the state-of-the-art life cycle inventory (LCI) data available in the literature for a composite product. Works evaluating the embodied energy of the most common fibres and polymeric matrixes are collected. Each manufacturing technique is reviewed regarding energy efficiency by considering the specific energy consumption (SEC). Among the potentialities which characterise the EoL of a composite product, a focus is given to the recycling techniques. Future research challenges are proposed and discussed. The outcomes revealed a considerable dispersion in embodied energies and SEC values for both the reviewed materials and technologies. The SEC is the only descriptor for process efficiency. However, there is a lack of investigation into the relationship between the process parameters, processed materials, component size and energy consumption. In particular, for additive manufacturing processes, no data were found. In addition, the literature on using natural fibres as a sustainable alternative and recycling methods and their impact is extremely limited.

Keywords:

Additive manufacturing, Embodied energy, Specific energy consumption, Sustainable manufacturing

1. Introduction

The attention to the efficient use of energy and resources is growing among the scientific community and political authorities of the world [1–3]. The governments embraced this view, and manufacturing

companies and cities signed the COP26 declaration to accelerate the transition to 100% zero-emission cars and vans by 2040 to promote the Sustainable Development Goals [4,5]. Virtuous practices, such as those proposed in the circular economy (CE) system, may reduce the energy demand and CO₂ emissions by up to 90% with respect to the production of the same good using the traditional model [6]. Because of that, the research community is also paying attention to developing energy-saving production routes and implementing the life cycle assessment (LCA) logic on a broad spectrum [7,8]. Such methodologies can be applied in the linear economy system (e.g., produce-use-dispose) [9,10] or in a more complex context which includes the application of CE strategies such as repairing [11,12]. In this context, innovative and high-performance (e.g., with low weight and high resistance) materials can significantly reduce energy demands and the relative CO₂ emissions during the component life [13]. Among these materials, composites made by a polymeric matrix and reinforced by fibres such as carbon (CFRPs) or glass fibres (GFRPs) showed the potential to easily replace metal components in a large number of applications [14]. For example, the lightening of systems, thanks to composite materials, leads to global annual primary energy savings for passenger cars up to 1100 PJ and for aircraft up to 300 PJ [14]. Because of that, this market is growing in different sectors. Fig. 1 shows the composite actual and forecasted 2030 global market by application in terms of dollars and the corresponding cast shadow in tons [15]. The U.S. Department of Energy estimated a carbon fibre market in 2010 of about: 9.9 million lbs for wind energy, 6.6 for aerospace, 2.2 for automotive and 1.7 for pressure vessels [16]. For glass fibres, the estimations were about 485.9 million lbs for automotive, 62.7 for wind turbines, 94.0 for aerospace and pressure vessels [17]. The application of CFRPs and GFRPs to automobiles is also encouraged by the regulations for the CO₂ emission reduction per unit distance [18]. In Europe, the EURO 6 fixed the CO₂ emission limit to 95 g/km. More in general, only for North America and Europe, the forecast of CFRPs was expected to increase by 40% (approaching 60000 tonnes) and 36.8% (overcoming 80000 tonnes) respectively from 2012 to 2020, with an estimated U.S. market value of \$12 billion in 2020 [19]. However, only in Europe in 2018, a market of around 1.14 million tonnes was achieved [20]. Considering the current data, the global composite market is estimated to reach \$40.2 billion by 2024, with a compound annual growth rate (CAGR) of 3.3% and an estimated market of the end products of \$114.7 billion by 2024 [21].

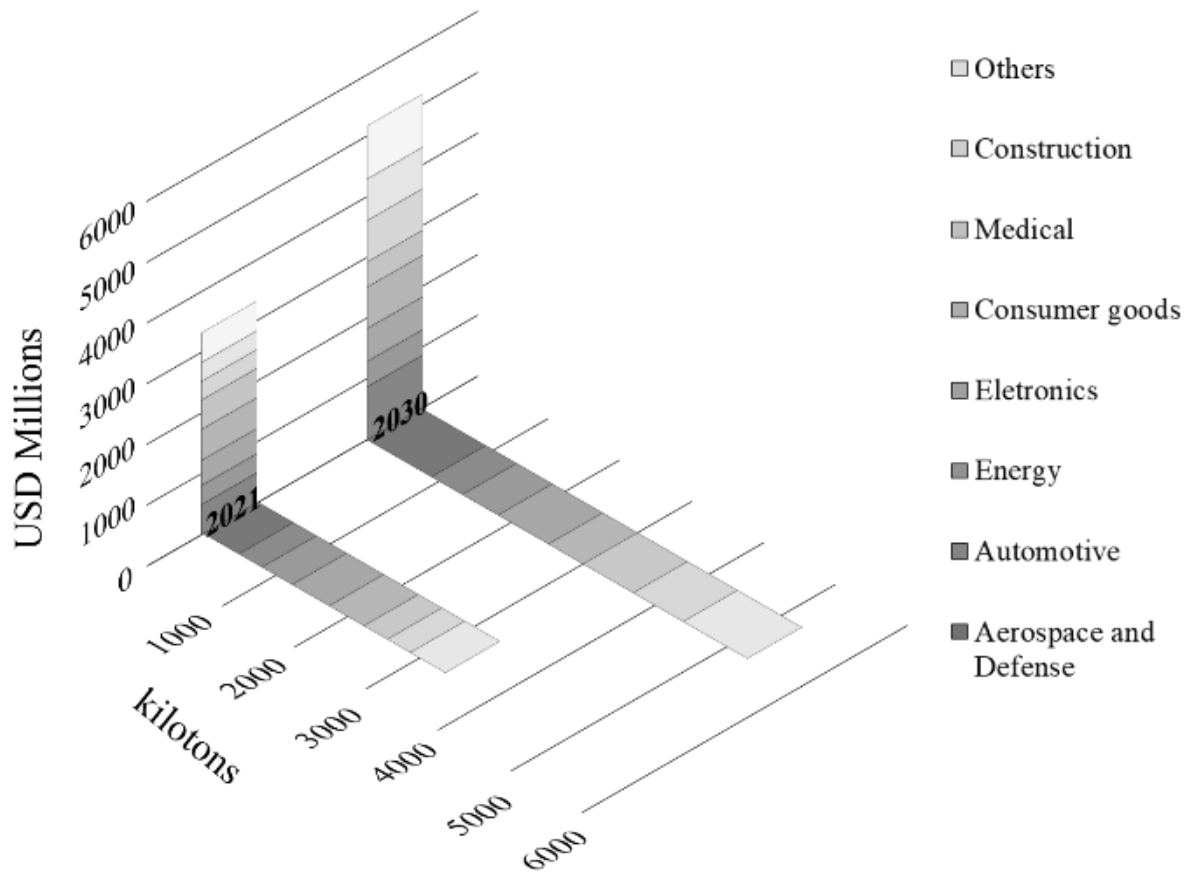


Fig. 1. Global composite market by application, historical data 2021 and forecasting 2030.

The evaluation of the sustainability of a composite through a complete LCA analysis becomes crucial [22]. Fig. 2 gives a schematic overview of this concept. The proper evaluation of each contribution plays a crucial role in energy-saving and total CO₂ emissions. However, the fight against adopting such methodologies faces a major obstacle: the data are dispersed, lacking and far from being measured accurately and comparable.

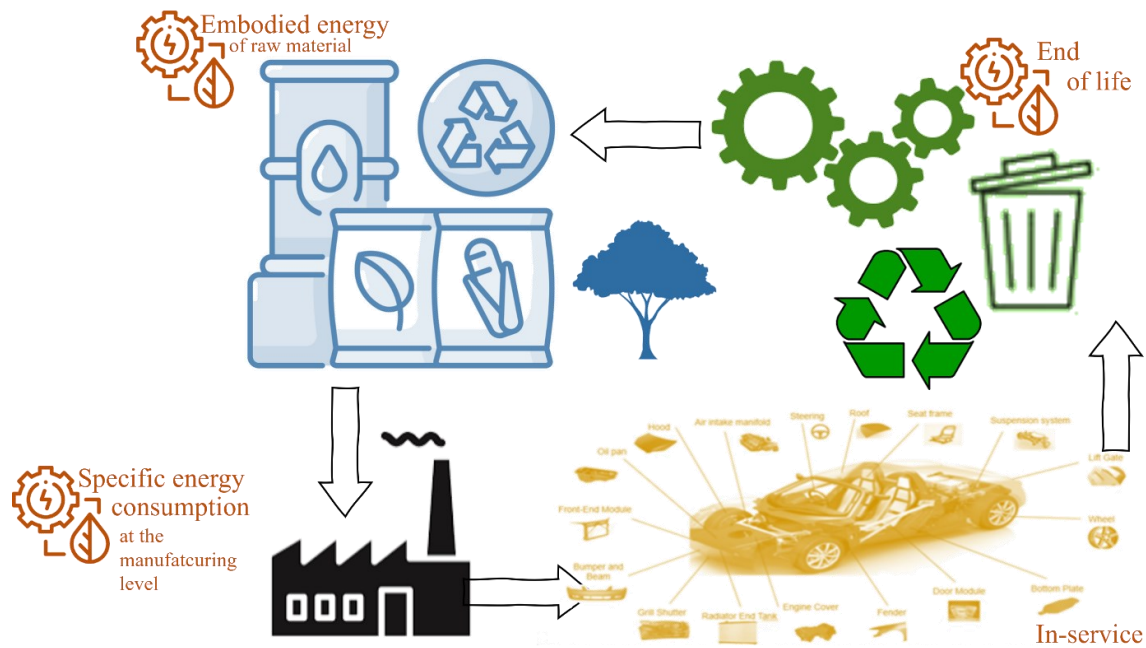


Fig. 2. LCA of composite product: from raw material production, manufacturing, and post-process to end-of-life (EoL) assessment. Each phase is described by an index contributing to the total product sustainability assessment.

This paper presents the state-of-the-art life cycle inventory (LCI) data available in the literature for such products. Therefore, a well-ordered data collection supports future research to develop proper LCAs on composite components. Chapter 2 provides a classification and a brief description of the most common manufacturing processes. Considering the raw material production, the embodied energy of the most common fibres and polymeric matrixes is reviewed in Chapter 3. Chapter 4 focuses on assessing the energy efficiency of the manufacturing techniques using specific energy consumption (SEC). Chapter 5 is centred on the recycling techniques characterising the EoL of a composite product. This chapter also briefly introduces other actions that characterise a circular economy (CE) framework (e.g., repairing, remanufacturing) for a composite product. Chapter 6 presents the current and future research challenges.

2. Manufacturing processes

The manufacturing of composite materials involves complex technologies and different manufacturing routes. According to that, different classifications are possible. Fig. 3 shows a graphical summary of the most common technologies currently used to produce composite materials grouped according to the polymeric matrix. Fig. 3 also classifies the manufacturing processes into (i) conventional, (ii) automated and (iii) additive manufacturing (AM) technologies. The conventional and the automated processes indicate manufacturing processes that utilise one or more tools or moulds to achieve the final component shape [23]. The AM processes collect all manufacturing processes in

which the part is built layer by layer directly into the manufacturing system without using any additional tool, mould, or post-processing operations [23].

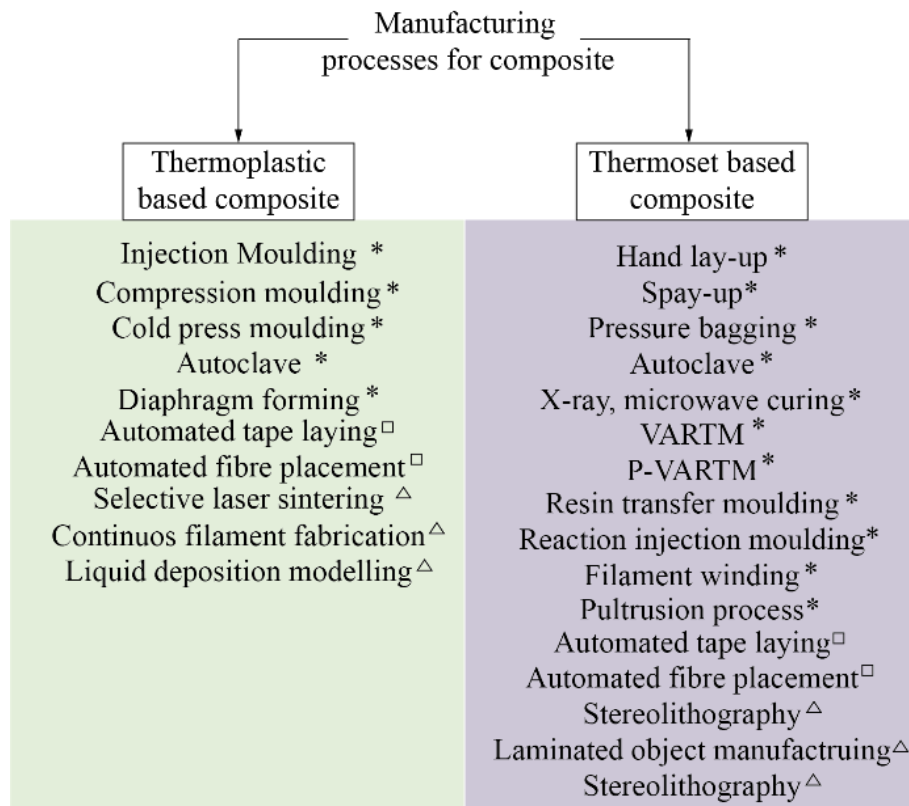


Fig. 3. Manufacturing processes for composite material production. *conventional manufacturing processes, □ automated processes, △ additive manufacturing processes.

The selection of the process depends on the component size and application. Because of that, only a restricted number of processes detect the most significant part of the global volume distribution (Fig. 4) [24]. In particular, lay-up processes led the composite market, followed by injection moulding, compression moulding, resin transfer moulding, and filament winding. The remaining processes occupy only around 5% of the global market because they can produce parts with limited dimensions and complexity. On the other hand, in some instances, such as AM [25] and nanotechnology [26] processes, the application is still limited because the technology readiness level is today too low to be considered for large industrial applications.

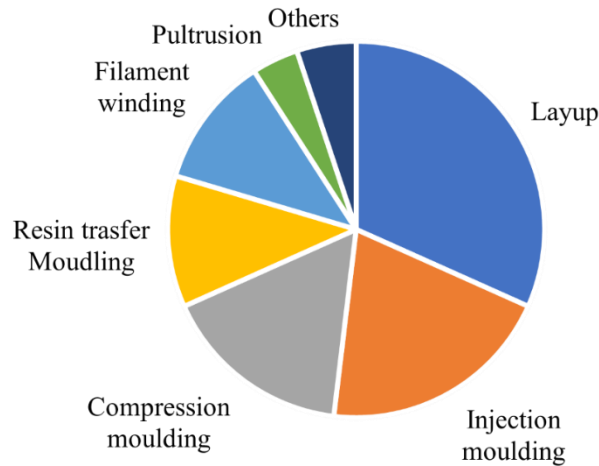


Fig. 4. Global market distribution by manufacturing processes.

To discuss the embodied energy of raw material (see Chapter 3), it is also convenient to group the technologies according to the impregnation step. The techniques in which the fibres are impregnated directly with resins during the process are called fibre on-situ impregnation processes. These techniques can also include manual operations to wet the fibres with the resin. On the other hand, other approaches use previously prepared materials already impregnated with the resin. For these techniques, the standard composites are prepregs and ready-to-mould compounds such as sheet moulding compounds (SMCs), bulk moulding compounds (BMCs) and glass mat thermoplastics (GMTs). Prepregs are plies easy to peel, and the high viscosity assures easy handling at room temperature, even though they are made of uncured resin (typically epoxy) [27]. Typical cure temperatures for prepregs range from 60 °C to about 180 °C; the curing times range between 1 and 6 hours, and curing pressures vary between 2 bar to 7 bar. These materials can be reinforced (typically with a CF fraction up to 50%) along one direction (unidirectional prepregs) or in multiple orientations, e.g., cross-ply (plies oriented at 0 and 90 degrees) or quasi-isotropic (a ply stacking sequence 0 degrees, 45 degrees, 90 degrees, -45 degrees) [28].

SMCs are fibre-reinforced thermosetting semi-finished sheets with a thickness between 1 mm and 3 mm. SMCs include short fibres or fibre bundles impregnated between two layers of a resin paste. SMCs are usually made by a mixture of polyester or epoxy resins and inert fillers, fibre reinforcement, catalysts, pigments and stabilisers, release agents and thickeners. After mixing the paste components with the fibres, a dry bed, called mat, is moulded. The mat is then sandwiched between two paste layers and two carrier films, compacted, and impregnated, and the paste wets the fibre bed [29]. The SMC is a putty-like material for which the handling is facilitated.

BMC is a mixture of a thermoset resin, such as polyester, vinyl ester resin or epoxy resins, and a blend of inert fillers, glass fibre reinforcement, catalysts, stabilisers, and pigments. The components

are mixed at room temperature and stored at low temperatures to slow down the curing before moulding. With respect to SMC, BMCs are highly filled and reinforced with short fibres. The volume fraction can achieve up to 30% for glass fibres, with fibre lengths between 6 mm and 12 mm [30]. BMC is used mainly in compression, injection, and transfer moulding.

GMTs are thermoplastic resin sheets or blanks reinforced with glass mat or unidirectional continuous fibres. These blanks are heated under pressure to a semi-molten state and shaped through a steel tool. The matrix is generally polypropylene (in more than 95 % of GMTs) and, more rarely, thermoplastic polyester. High-performance GMTs are being developed with matrices such as polyethersulfone, polyetherimide, polyamide-imide, PPS, and PEEK. While thermoplastic resins were overlooked in the past because of their poor ability to wet the fibres, their recycling potentialities are attracting attention, and today their use is rapidly increasing [30].

2.1. Conventional manufacturing processes

In Fig. 3, the conventional manufacturing processes are marked with a star.

Hand lay-up is a moulding process where the fibre reinforcements are laid by hand into the mould and then wet with a resin (Fig. 5a). Before distributing the fibres, a gel is spread on the mould surface [31]. The resin is a thermosetting liquid resin such as polyester or epoxy. The resin on the fibre reinforcements is spread using a brush or a roller, layer by layer, up to the desired thickness. Synthetic fibres such as glass, carbon and Kevlar are used as fabrics. The reinforcement weight fraction of the final laminate is over 30-50% [31].

Spray-up is similar to the hand lay-up moulding technique, but a mixture of chopped fibres and resin is sprayed directly onto the surface of the mould (Fig. 5b). Similar materials to those used in hand lay-up are used for both resin and fibre reinforcement. As with hand lay-up, a roller can be used to consolidate the mixture. The reinforcement percentages by weight of the final laminate can range between 15 and 40%. The typical fibre length is around 10 mm to 40 mm. The production rate of hand lay-up is between 0.12 components to 0.5 components per hour (component mass of 1 kg) [32]. The spray-up process has a higher production rate, up to 3 components per hour (component mass of 1 kg) [32].

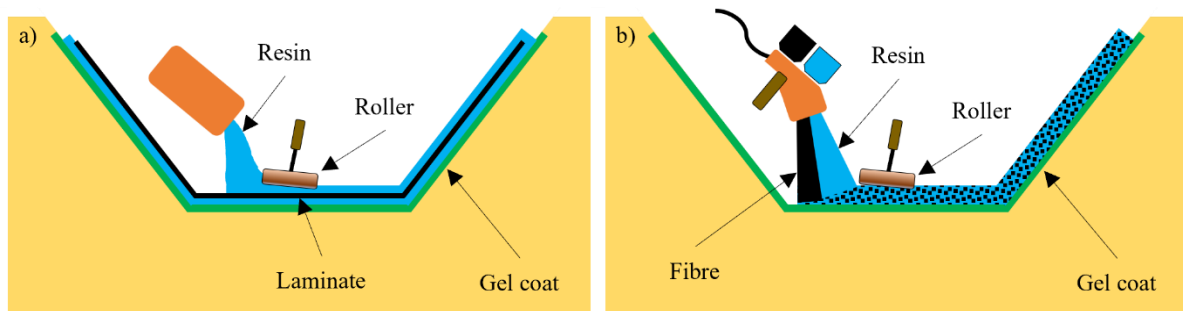


Fig. 5. a) Hand lay-up and b) spray-up.

In the vacuum/pressure bagging process, the reinforcement and the resin are applied to the mould surface by hand or spray lay-up techniques (Fig. 6 a). The laminate is then plugged into a deformable bag with a connector from which a pump applies the vacuum. Furthermore, the bag can be packed inside a case where additional pressure is applied (Fig. 6 b). These techniques are useful to speed up curing and reduce porosities. In vacuum bagging, the pressure differential values between inside and outside the deformable (clamping pressure) range between 0.4 and 0.8 bar. In pressure bagging, it is possible to reach up to 2-3.5 bar. The production rate of this process ranges between 0.05 components and 0.1 components per hour (component mass of 1 kg) [32].

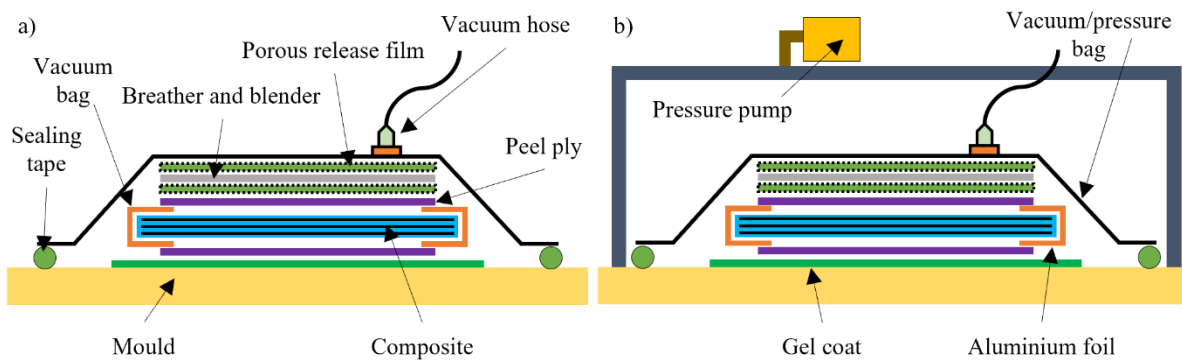


Fig. 6. Vacuum/pressure bagging process: a) standard, b) with a case.

In the autoclave moulding process, the laminate and the mould inside a vacuum bag are placed inside an autoclave and subjected to an external pressure between 3.5 bar and 7 bar (Fig. 7). The autoclave is often heated to accelerate the cure and increase the productivity [33]. Therefore, high-temperature matrix resins such as epoxies can be processed. The produced composite is also denser and void-free. Because of that, more than 98% of aeronautical composites are fabricated using autoclave curing technology [34]. Owing to the long cure cycles, the production rate is low and between 0.05 components to 0.1 components per hour for a mass of 1 kg [32]. The part size is constrained by the autoclave dimensions. To overcome this issue, researchers are currently working on alternative curing techniques such as high-energy X-ray curing [35], microwaves [34] or cyclic heating and cooling direct heating [36].

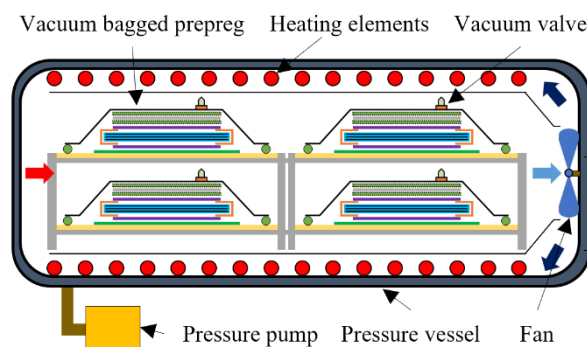


Fig. 7. Autoclave moulding process.

The compression moulding process uses preforms, SMCs, and BMCs (Fig. 8a). The mats are placed directly into a heated mould and cured inside the press. The application of heat and pressure vary according to the charge type [32] and thicknesses [29]. For SMCs, the curing cycles range from less than 1 min to about 5 min depending on the used thermoset polymer and the characteristic thickness of the part. The mould is closed with a pressure between 3 MPa and 7 MPa with a typical moulding temperature between 130 °C and 160 °C. For BMCs, the pressures used vary between 0.5 MPa and 15 MPa depending on mould dimensions and the material being processed. The mould is heated between 140°C and 160 °C [32]. The SMC is fed cold and is heated only in the press to process [29]. The compression moulding process can also be applied to GMTs, known as thermoforming or stamping [37,38]. In this case, the GMTs are heated before entering the moulds, and the mould is not heated during the process (Fig. 8b). The production rate (a component of mass of 1 kg) ranges from 1 to 4 components per hour [32]. The preform matched die moulding process is a variation of compression moulding in which two metal dies with a male-female are used [37].

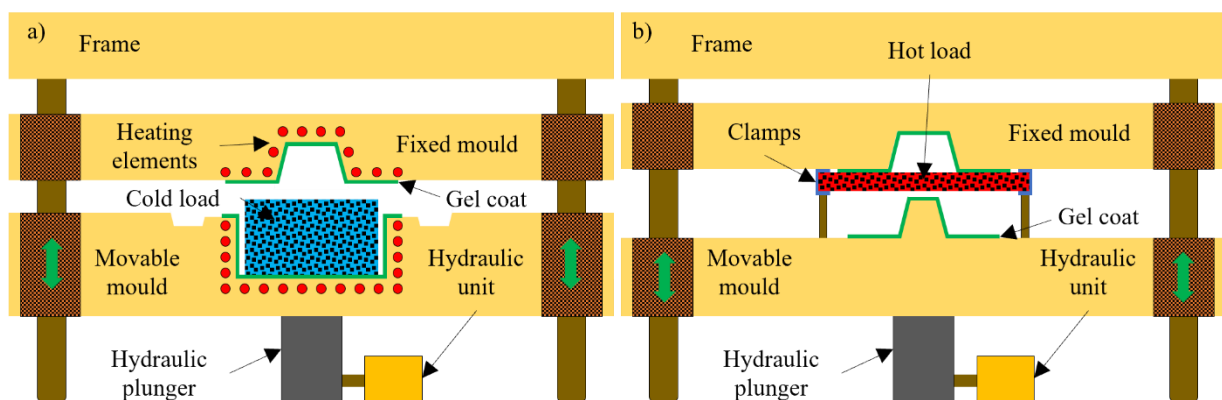


Fig. 8. Compression moulding process for a) SMCs or BMCs and b) GMTs.

Thermal press curing (TPC) is another type of compression moulding in which a composite laminate is clamped between a temperature-controlled mould (curing mould) and a matched mould (compression mould) coated with a rubber mask. Compared with the autoclave process, the components produced by the TPC process show similar material properties but are less expensive, require significantly less energy to be produced (1 kWh per part against 77 kWh per part), and have a faster cycle time (4.3 h against 9 h) [39]. Generally, the production rate for a compression moulding process can reach up to 60 components per hour [32].

The cold press moulding process combines hand lay-up and compression moulding (Fig. 9). The reinforcement is cut into mats and placed in the mould. Then the cold-curing resin system is poured, and the press is closed. The pressure is used only to impregnate the reinforcement with catalysed resin, which cures at room temperature [38]. The exothermic heat generated during gelation, together with the pressure, causes the resin curing. A lightweight hydraulic press is used as the pressures do not exceed 5 bar [32]. The production rate of this process is much lower than that of compression

moulding of SMCs or BMCs: only from 0.5 (component mass of 1 kg) to 3 components per hour can be formed [32].

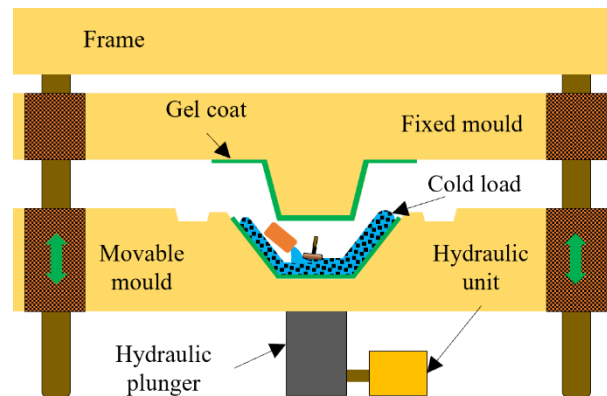


Fig. 9. Cold press moulding.

In the diaphragm forming (DF) process, thermoplastic prepregs are fixed between two diaphragms (Fig. 10a). The system is then clamped between two moulds. A heating system heats up the cold prepregs and the lower mould. The forming process occurs by applying pressure using a gas (typically argon) flow perpendicular to the diaphragms. In this way, the pressure pushes the system against the lower heated tool. The temperature of the thermoplastic matrix is close to the melting point. The cooling phase is carried out under pressure until the structural stability of the component is achieved.

Instead of a heating system located inside the moulds, a variant of the process, called cold diaphragm forming (CDF), uses direct heat sources (e.g., IR lamps) to heat the cold prepregs (Fig. 10b). This process modification leads to higher energy efficiency and lowers the cycle time [40,41]. These processes mainly work with thermoplastic composite materials, but epoxy-based prepregs were also used with curing temperatures ranging between 55° C and 110 °C [42]. In general, thermoplastics and thermosets resins are processes with a hydrostatic pressure of up to 10 bar [42–44]. In the case of thermosets also vacuum is used [45]. The processing time is usually longer (around 2 hours) than other forming techniques [42] but lower than autoclave curing [44]. This process can also be carried out with a single diaphragm [45] (SDF). However, SDF is not suitable for realising complex components.

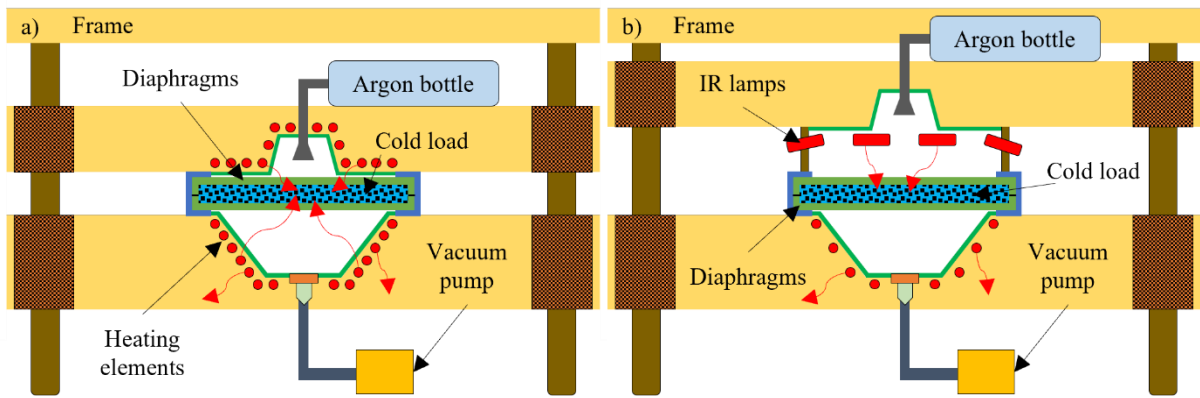


Fig. 10. a) Standard diaphragm forming process, b) Cold diaphragm forming.

The vacuum-assisted resin transfer moulding (VARTM) process (Fig. 11a) is also known as Seemann composite resin infusion moulding process (SCRIMP), resin infusion under flexible tooling (RIFT), vacuum-assisted resin injection (VARI), vacuum resin transfer moulding (VRTM), resin injection recirculation moulding (RIRM) and vacuum injection moulding process (VIMP) [32]. It is low-cost tooling to manufacture composite using only half mould. Dry continuous strand mats and woven reinforcement are laid in the bottom mould, and the whole lot is vacuum bagged. The resin is released and sucked into the bag by the vacuum, flowing through and impregnating the fabric, and then cured by the heaters into the mould. The thickness and the fibre volume fraction of the final piece cannot be fully controlled due to the use of a vacuum bag instead of a rigid mould and the significant variations of the preform volume when vacuum and pressure are applied [46]. On the other hand, because the resin flow and cure are separated, the quality of the produced components is high [46]. The curing can be performed using microwave heating sheets or thermal ovens [47–49]. The compaction pressure on the vacuum bag is limited to 1 bar. Common matrix resins are polyester, vinyl ester, epoxy, and phenolics. The production rate of this process is between 0.2 components to 4 components per hour (component mass of 1 kg) [32]. The pressurised VARTM (P-VARTM) process is a variant of VARTM performed at higher pressures (Fig. 11b). The application of an external pressure facilitates the process and improves the part quality. In addition, with this method, the fibre volume fraction can be higher; therefore, the manufactured part shows better mechanical performance [50].

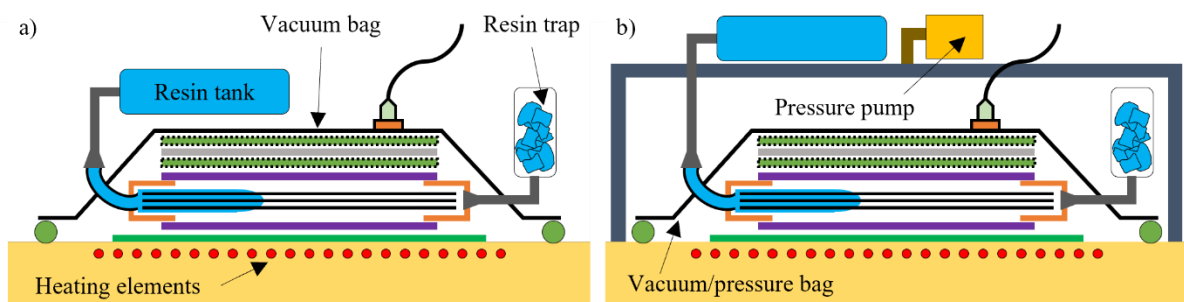


Fig. 11. a) Vacuum-assisted resin transfer moulding (VARTM), b) Pressurised VARTM.

Resin transfer moulding (RTM) is another resin injection technique. The process occurs without assisted vacuum environment and uses two rigid moulds. The pressure is up to 2 MPa. The production rate of RTM and VARTM is up to 8 components per hour (component mass of 1 kg) [32]. The high-pressure RTM (HP-RTM) process is a high-pressure variant of RTM that facilitates short cycle times of complex parts: the production rate is about eight times higher than conventional RTM [32]. This process can also manufacture composites in which the reinforcements are previously placed inside the moulds (Fig. 12) or directly injected with the resin.

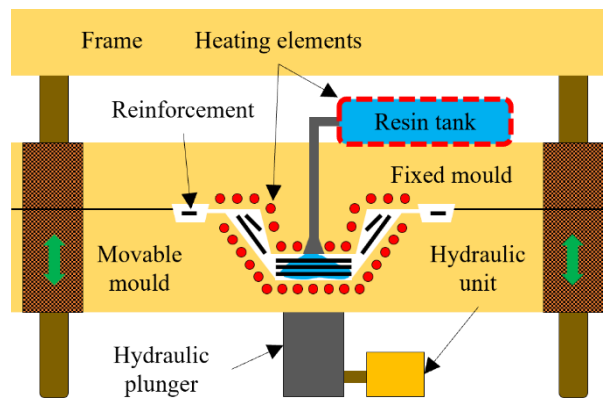


Fig. 12. Resin transfer moulding.

Reaction injection moulding (RIM), also known as liquid reaction moulding (LRM) or high-pressure impingement mixing (HPIM), is a two-step process that uses two reactive resins stored in two separate tanks (typically one for polyol and one for isocyanate). The two resins are mixed at high velocity, heat and pressure. The mixture is then injected into a closed mould. Common RIM parts include automotive components and appliance housings. Structural reaction injection moulding (SRIM) and reinforced reaction injection moulding (RRIM) are the most used variants of the RIM process. RRIM utilises short-milled or chopped fibres to produce more isotropic products (Fig. 13), while SRIM is based on fibre mats, meshes, and preforms, that are placed into the mould before the resin injection. Both processes use low pressure because of the chemical reactions. However, SRIM requires high temperatures (between 150°C and 175 °C) and low-viscosity materials to ensure that all fibre mats or meshes are adequately covered. For this reason, thermoset polyurethane became the most widely used resin. As for RTM, the curing principle of SRIM relies on chemical reactions rather than merely thermal influences. SRIM works with lower pressures and temperatures than the RTM process. Also, SRIM has a short cycle time of between 3 and 5 minutes. However, parts produced using SRIM typically require post-mould paint operations because of the poor surface quality.

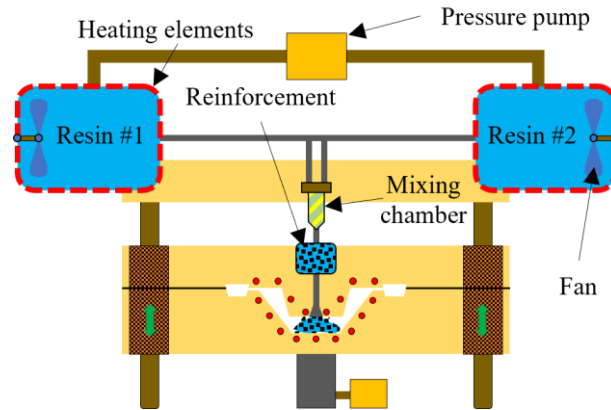


Fig. 13. Reinforced reaction injection moulding.

The filament winding process consists of winding fibres of glass, aramid, or carbon impregnated with resin on a rotating mandrel in predetermined patterns. Because of that, only axisymmetric parts can be produced. Depending on the application, the winding pattern could be helical, hoop or polar. According to the treatment of the fibres, the filament winding processes can be used in wet or dry methods. In the wet method (Fig. 14), the fibres are soaked with low-viscosity resin either by passing through a trough or a metered application system. In the dry method, the reinforcement is in the pre-impregnated form. Typical thermoset resins include polyesters, vinyl esters, epoxies, and phenolics. Thermoplastic resins with high viscosity and elevated temperatures make in-line (wet) impregnation impractical. Some preheating is usually necessary to improve the laydown on the mandrel [37]. The production rate of this process ranges between 0.12 (component mass 1 kg) to 10 components per hour [32].

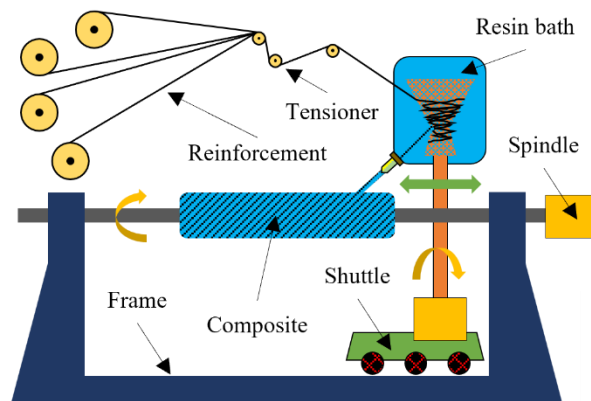


Fig. 14. Filament winding processes in the wet method.

The pultrusion process is continuous and produces a part with a constant cross-section (Fig. 15). The fibres impregnated in the resin are called fabric. The fabric is pulled through a series of forming dies. The profile is determined by the die cross-section, which could be round, rectangular, square, or various other shapes. The final die is heated to cure the resin, producing a rigid composite section. The speed of travel through the die is determined by the resin viscosity, thickness and curing [32].

Many resins could be used, including polyester, polyurethane, vinylester and epoxy. The production rate of this process ranges between 0.002 m/s to 0.03 m/s [32].

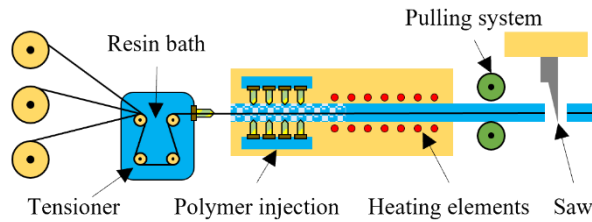


Fig. 15. Pultrusion process.

2.2. Automated manufacturing processes

In Fig. 3, the automated processes are marked with a superscript square. Automated tape laying (ATL) and automated fibre placement (AFP) are similar processes (Fig. 16) and can deliver the composite material directly onto the surface of the component to be reinforced.

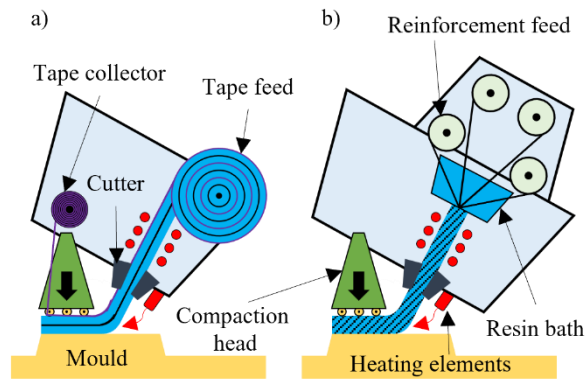


Fig. 16. a) Automated tape laying and b) automated fibre placement.

ATL process (Fig. 16a) is based on delivering heated prepreg tapes onto a surface while the ply backing is automatically removed. The typical widths of the prepreg tape are 75 mm, 150 mm, or 300 mm. The tape courses are placed adjacently with a gap between 0.5mm and 1 mm. The linear layup speed can achieve 1.00 m/s, and most used speeds are not below 0.85 mm/s, with acceleration around 0.5 m/s^2 . The typical pressure applied to the surface depends on the resin and is 0.1 MPa for thermoset tapes and up to 3.5 MPa for thermoplastics[51].

AFP (Fig. 16b) uses a heated band of narrow prepreg slices, collimated on the head and then delivered together. AFP is usually employed for complex shapes, such as fuselage sections and wing skins with valleys and pad-ups. The complexity of the component influences the material width and tow count, which affect productivity. The average contact pressure applied in AFP equals around 0.6 MPa [52]. The AFP head can be driven by each tow individually. The tows can be clamped, cut, and restarted during the process. Therefore, each tow could be deposited at a different speed, allowing layup over complex geometries. Typically, the width of the tows is 3.2 mm, 6.4 mm, and 12.7 mm. Even if these values are lower than ATL, AFP delivers several tows at once in termed bands, up to 32 tows in

parallel, with a linear speed of up to 1 m/s. High accelerations are also achievable in the linear axes with typical values around 2 m/s^2 . For example, the productivity for layup of a fuselage section by AFP is 8.6 kg/h, which is about half of the ATL one [51].

Both processes can produce large components with planar or large curvature radius surfaces [52,53]. However, small curvature radiuses cause the so-called wrinkling phenomenon [53]. The processes also include heating systems such as hot rollers, gas torches, and laser and infrared irradiation techniques are available [51,54,55]. Other heating methodologies were recently developed: ultrasonic consolidation [56] and ohmic heating [57].

2.3. Additive manufacturing processes

Additive manufacturing (AM) is a production approach in which an object is produced layer by layer directly from a 3D CAD model without using any additional tool [58]. The main peculiarities of this technology are the great flexibility [59] and design freedom that can be achieved [60]. Considering the scenario of composite manufacturing, the AM approach is innovative because no moulds are needed for shaping the component, and, in some instances, the fibre can be deposited selectively, guaranteeing tailored local mechanical properties [61]. In Fig. 3, the AM processes are marked with a superscript triangle.

The laser beam powder bed fusion process for polymer (PBF-LB/P) [62], better known as selective laser sintering (SLS) process, is a powder bed process in which a laser with low power is used to sinter thermoplastic powders (Fig. 17). At each layer, the build plate is lowered by a quantity corresponding to the layer thickness. Then, a mix of polymeric powder and short fibres is spread on the build plate by a recoater. The formed powder bed is preheated by external electrical resistances and sintered by a laser according to the 2D geometry. The steps are then repeated up to the part is completed. To avoid thermal deformation, the build tank is also heated up to a temperature close to the crystallisation point of the thermoplastic matrix. Once the process ends, the part is cold down to room temperature. After the cooling, the part is removed from the machine and cleaned from loose powder. Among the AM processes, SLS is the easiest way to produce a composite material because it is enough to mix polyamide (PA) powder with reinforcement fibre [63]. Unlike other AM processes, thanks to the preheating phase and the use of a thermoplastic matrix, no support structures are required to build the parts. Therefore, complex geometries and nesting operations are easily allowed. The size of the reinforcement fibres is mainly particulates because the use of longer fibres could affect the smoothness of the powder bed and compromise the final component density and strength [63]. The maximum volume ratio for CF mixed with PA12 is 50% (38 μm of average fibre length) [64,65]. However, better interfacial adhesion of fibre can be achieved by pre-modifying CF through oxidation [64]. Ternary composites of epoxy powder and CF (33% volume fraction with 55.12 μm of average

fibre length) coated with a layer of PA12 595 nm thin can also improve the composite mixing [64,66]. PA12 mixed with carbon nanotube (CNT) (3 wt%) results in components with higher mechanical properties with respect to the CF short fibres [64,67].

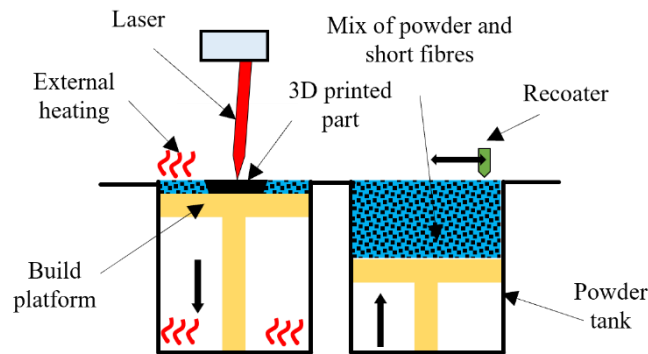


Fig. 17. Selective laser sintering.

Three-dimensional printing (3DP) is also a PB process in which a polymeric binder is selectively jetted onto a pre-deposited powder layer (Fig. 18). Because of that, 3DP belongs to the binder jetting processes [62]. 3DP gives a unique opportunity to locally control the material composition by jetting different materials from different nozzles. These jetting materials can be used either in a molten form or in a slurry form. Composites are made by changing a component of the powder mixture [63].

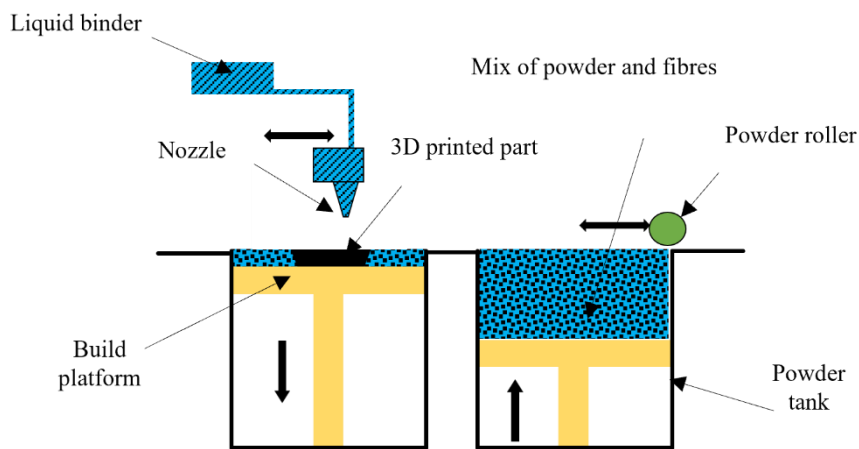


Fig. 18. Three-dimensional printing.

The laminated object manufacturing (LOM) process for composite belongs to the sheet lamination processes [62]. It consists of overlapping sheets reinforced with particulates or fibres [23,64] (Fig. 19). A pressure and temperature cycle (binder burn-out) is applied to avoid delamination. Therefore, this process is similar to the traditional ones in which preforms are used. Layers of different materials can also be used to vary the composite properties [63].

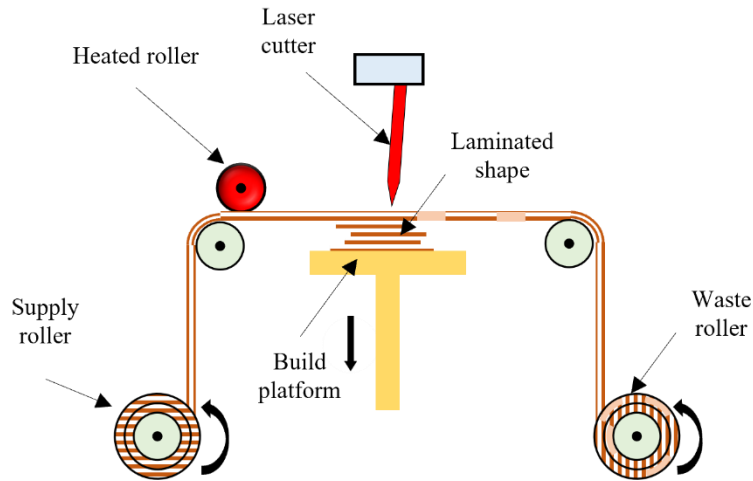


Fig. 19. Laminated object manufacturing.

Stereolithography (SLA) is a vat photopolymerisation process [62] which uses a resin activated by a laser source (Fig. 20). At the end of the process, the photopolymerisation is completed in a UV light oven. The photopolymer is mixed with the reinforcing fibres to realise a composite material. Typical reinforcements are carbon, glass, e-glass and aramid [63]. The shape can be particles, short fibres, continuous fibres and fibre mats [63]. However, in most cases, short fibres are used with 20 vol%. Higher volume fraction led to poorer fibre dispersion and mechanical properties [23]. The use of particles modifies the viscosity properties of the resin, and due to their low weight, they can float in the resins and cause non-uniform dispersion and the formation of bubbles [63]. Another aspect is that the use of fibres can also change the absorptivity of the resin to the laser [63]. For instance, the resin close to the CF could remain uncured; therefore, a long curing time could be required [63]. However, this problem is not present with the GF [64]. The SLA process can also be suitable for nano-reinforcements such as CNT (concentration in the resin of 0.10% w/v) [68].

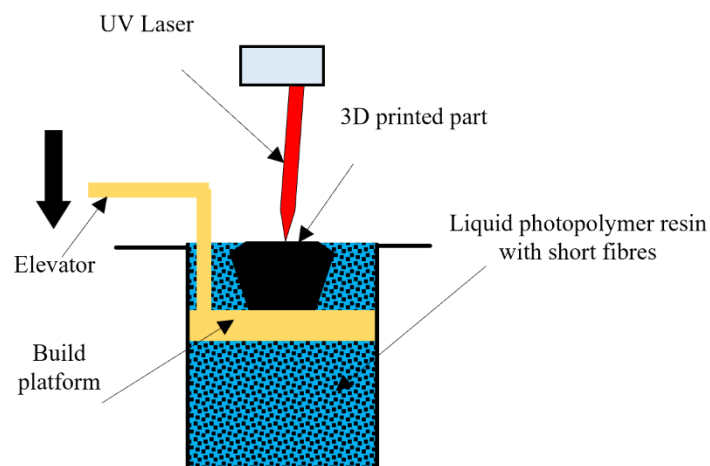


Fig. 20. Stereolithography.

The fused deposition modelling (FDM) process is a material extrusion AM process [62] in which a thermoplastic wire is extruded and deposited by a heated nozzle. The nozzle is heated up to the

material softening temperature, and the material is then deposited. When the layer is completed, the build plate is lowered to allow the deposition of the next layer. The system includes a material feed mechanism, an extrusion head, a gantry structure, and a build platform (the build chamber and platform can be heated if needed). These techniques can be easily expanded to multi-materials printing using multiple nozzles [23]. In the case of composite production, different configurations of FDM can be found in the literature. If the reinforcement is provided through solid filament or paste form, the process is known as continuous fibre fabrication (CFF) [69], while if a fluid-like feedstock is used, the process is called liquid deposition modelling (LDM).

The CFF process (Fig. 21) is a highly flexible technology that can process different fibre sizes (nano, micro and millimetre) or continuous reinforcement [23,64]. The matrix is a thermoplastic material. The fibre is pre-impregnated with a compatible resin [68,70]. The reinforced filament is made by the fibres (continuous carbon fibre, Kevlar, and fibreglass) coated and englobed in a thermoplastic matrix of PA12. To extrude the filament, the nozzle is heated up to the softening temperature of the matrix. An additional nozzle deposits the matrix. Therefore, it is also possible to deposit the fibre selectively [61]. The matrix can also be reinforced with short fibres of carbon. This material is commercially known as Onyx [69].

For LDM processes, the materials are selectively deposited from a syringe controlled by a CNC unit. The reinforcements are in the form of discontinuous fibres dispersed in the resin feedstock. The resin material is liquid at room temperature and, therefore, easy to process [23]. In the literature, different discontinuous reinforcements were investigated as carbon fibres, glass fibres, CNT [23] and silicon carbide (SiC) whiskers [71]. The process using SiC whiskers is called direct ink writing (DIW).

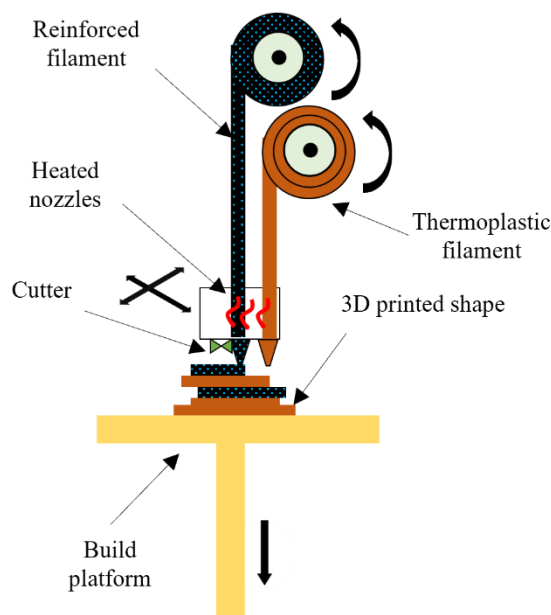


Fig. 21. Continuous fibre fabrication.

3. The embodied energy of composite raw materials

The embodied energy of raw material is the energy the raw material holds for its elementary substances and gains from the manufacturing processes needed to isolate it from nature [72]. According to this definition, embodied energy can also consider the benefits of recycling strategies [73]. Most literature data refer to the electrical energy demand measured at the facility power supply [49,74]. However, all the energy losses from the electric power station to the manufacturing centre must be considered. Electric power can be generated by different power sources (e.g., non-renewable such as coal, natural gas and oil or renewable ones) with different efficiency of the energy production chain. Therefore, to make the reviewed data comparable, all the electrical energy demands were converted into MJ oil equivalent, as proposed by Ashby [72]. According to the European average energy mix [72], the conversion value from electric MJ to MJ oil equivalent equals 0.38. In the following, all the data expressed in MJ should be intended as MJ oil equivalent, unless differently specified. The U.S. Office of Energy Efficiency & Renewable Energy [16,17] gives three values according to the Practical Minimum, State-of-the-Art and Current Typical production. State-of-the-art and current typical data are computed considering energy savings arising from recycling/recovery strategies and improvements on the technological steps. Therefore, the extreme values are reported in this paper.

Fig. 22 shows the global volume distribution of fibres in 2020. Glass and carbon are the most commonly used, while natural fibres still hold a limited volume [75]. Table 1 lists the embodied energies of the most common fibres used as reinforcement for composites.

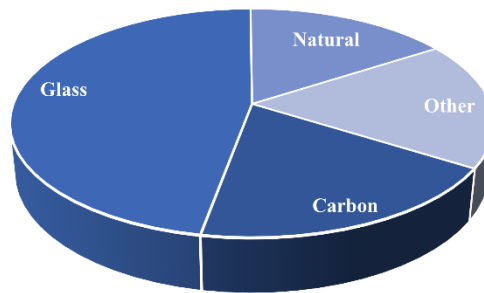


Fig. 22. Global composite market volume by the type of fibre in 2020.

Fibres	Embodied energy (MJ/kg)	Reference(s)
CF	521-1563	[76]
	1468	[74]
	183-286	[22,76,77]
	272-300	[32]
	1000	[49,78]
	704	[49,79]

	855	[49,80]
	286-478	[81]
	190-870	[16]
Recycled CF	27-94	[76]
GF	13-32	[22,76,77]
	49-54	[32]
	7-16	[17]
China reed fibre	3.6	[22,82]
Flax fibre	6.5	[22]

Table 1. The embodied energy of common fibres.

The embodied energy values show a considerable variation range among the fibres. As can be observed, the highest values can be detected for CF. However, the values drop when considering recycling, thanks to the lower energy required for CF production [76]. Also, technology development decreased embodied energy from 478 MJ/kg in 1999 to 286 MJ/kg in 2004 [81]. Relative high values were reported for CF in Ref. [78–80] and reviewed by Witik et al. [49]. Contrasting lower values were found in Ref. [22,76,77]. The reason for such a difference is unclear because the reported analyses lack detailed information. This considerable data dispersion may represent different manufacturing settings and components size. Also, the GF showed a huge dispersion of data [22]. In this case, the studies showed differences in manufacturing technology, production volume, and plant size. In fact, the lowest values (13 MJ/kg) were measured when considering large manufacturers, such as OwenCorning. Considering plants with similar sizes, the main driver that influenced the measured value was the efficiency of the specific facility (e.g., related to the machine tool). As an example, this effect was observed when quantifying the energy consumption of the Vetrotex plants in Germany (32 MJ/kg) compared to other similar plants (25 MJ/kg).

Owing to their nature and the possibility of easy recycling [82], natural fibre, such as China reed and flax fibre, embody the minim value of energy.

Overall, the total embodied energy in the raw material can be explained by considering the specific contribution of the manufacturing step (Table 2). Regarding the use of CF, the major contribution is in processing/assembling stage (247-436 MJ/kg) [81]. When considering the precursor, an organic polymer that, in most cases, is polyacrylonitrile, the production step can be subdivided into more detailed steps (in Table 2: polymerisation of the carbon fibre precursor, spinning, carbonisation and finishing). In this way, it appears clear that the major energy demand is required (142-427 MJ/kg) during the carbonisation of CF because of the need for high temperatures (up to 3000 °C). Similar conclusions can be achieved when considering the GF. The melting phase absorbs the highest energy [17], while the other phases are comparable in energy demand.

It is interesting to notice that the embodied energy of glass fibre (7-16 MJ/kg) is always at least one order of magnitude lower than CF (190-870 MJ/kg). This can be explained by the lower energy

required to perform the sub-processes in the case of GF [16,17]. In the case of natural fibre, the higher contribution to embodied energy is cultivation [82]. However, these values are significantly lower than the typical sub-process for the other fibres.

Fibre manufacturing sub-processes	Specific Energy Consumption (MJ/kg)	Reference(s)
CF		[81]
CF raw material production	39-42	
CF processing and assembly	247-436	
CF		[16]
Polymerisation	0-156	
Spinning	14-212	
Oxidation/Carbonisation	142-427	
Finishing	35-75	
GF		[17]
Batching	1.3-2.5	
Melting	3.4-9.1	
Fiberization	1.4-2.7	
Finishing	0.9-1.9	
China reed fibre		[82]
Cultivation	2.5	
Transport to plant	0.4	
Fibre extraction	0.1	
Fibre grinding	0.4	
Fibre transport	0.2	

Table 2. Contributions to the embodied energy of common fibres.

When considering composite materials prepared for direct shaping, the embodied energy depends greatly on their form (see Table 3). The lowest values were found for SMCs and BMCs, typically GF composite, because of the relatively simple manufacturing process. The minimum value refers to only the manufacturing steps necessary to produce the CF prepregs (40 MJ/kg) [81], but the embodied energy of the raw material was not considered. Analysing the manufacturing sub-processes, significant energy is required to control the environment when processing the material (20.8 MJ/kg) and when it is stored (11.5 MJ/kg) [81].

Mats require much lower energy than prepregs. In particular, the natural flax fibre mats (9.5 MJ/kg) with one order of magnitude lower than the glass fibre mat (54.7 MJ/kg) [82]. Also, in this case, the total values are influenced to a great extent by the high temperature in the melting phase of the GF and the actual mat production forming (Table 5) [82]. Contrarily to Table 4, data in Table 5 also considers the embodied energy of the raw material.

Preforms	Embodied energy (MJ/kg)	Reference(s)
Prepreg	95-124	[16,17]
Prepreg	40	[81]
SMC/BMC	8-11	[16,17]
SMC	3.5	[22,76,77,81]
GF mat	54.7	[82]
Flax fibre mat	9.5	[82]

Table 3. The embodied energy of common preforms.

Prepreg manufacturing sub-processes	Specific Energy Consumption (MJ/kg)	Reference(s)
Resin blending	0.1	[81]
Resin coating	1.4	
Resin impregnation	2.1	
Prepreg winding	0.2	
Atmosphere control	20.8	
Raw material storage	11.5	
Prepreg storage	3.4	
Release coated paper production	0.5	

Table 4. Contributions to the embodied energy of prepregs.

Mat manufacturing sub-processes	Specific Energy Consumption (MJ/kg)	Reference(s)
Glass fibre mat		[82]
Raw materials	1.7	
Mixture	1.0	
Transport	1.6	
Melting	21.5	
Spinning	5.9	
Mat production	23.0	
Flax fibre mat		[82]
Seed production	~0.0	
Fertilisers	1.0	
Transport	0.9	
Cultivation	2.0	
Fibre separation	2.7	
Mat production	2.9	

Table 5. Contributions to the embodied energy of common mats.

Besides the pure fibres, the resins should also be considered in assessing the energy demands (Tables 6 and 7) [16,17,22,32,77,81]. In some instances, no significant differences can be detected between the thermoset (Table 6) and thermoplastic (Table 6) resins. However, it should be considered that thermoplastic resins are recyclable, and thermosets usually need higher temperatures during manufacturing. In addition, it is worth noticing that the average embodied energy found for a fixed resin rises by increasing its service temperature (Tables 6 and 7), influencing the temperature required to process the material [32].

Thermoset resins	Typical service temp. (°C) [32]	Embodied energy (MJ/kg)	Reference(s)
Epoxy	130	76-80	[22,77,81]
		47-93	[16,17]
		126-139	[32]
Phenol	150	33	[81]
		73-81	[32]
Polyester	120	63-78	[22,77,81]
		68-75	[32]
Polyurethane	n.a.	32-64	[16,17]
Polyurethane (flexible foarm)	75	67	[81]

Polyurethane (rigid foarm)	155	88-97	[32]
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Table 6. The embodied energy of common thermoset resins.

Thermoplastic resins	Typical service temp. (°C) [32]	Embodied energy (MJ/kg)	Reference(s)
Polycarbonate	110	80-115	[22]
		102-113	[32]
Polyether ether ketone	250	286-315	[32]
Polyethylene (low-density)	90	65-92	[22]
		76-84	[32]
		20	[81]
Polyethylene (high-density)	120	11-34	[16,17]
		75-82	[32]
Polylactic acid	50	49-67	[32]
Polypropylene	90	24	[81]
		72-112	[22]
		11-27	[16,17]
Polystyrene	80	66-80	[32]
		16-42	[16,17]
		82-90	[32]
Polyurethane	70	71-118	[22]
		78-86	[32]
Polyvinyl chloride	55	53-80	[22]
		13-35	[16,17]
		49-72	[32]

Table 7. The embodied energy of common thermoplastic resins.

4. Specific energy consumption at the manufacturing level

The specific energy consumption (SEC) value is widely used in literature to quantify the energy intensity per unit mass required by a system to manufacture a product [83]. This value is also helpful during an LCA analysis when quantifying the energy demand of the product manufacturing. It can be used as a process selector tool to help the designer choose the most sustainable production route [84,85]. For this reason, the research community is dedicating attention to the correct assessment. Among the several challenges, the correlation between the SEC factor and the typical parameters of the process is the most critical. Fig. 23 shows a graphical summary of the analyses of the manufacturing technologies in terms of energy efficiency. In Ref. [86–88], subtractive processes were characterised by correlating SEC values to the material removal rate (MRR). Similarly, the energy efficiency of non-conventional technologies, such as electrical discharge machining and friction stir extrusion, was evaluated by correlating the SEC to the MRR [89] or the extrusion rate [90]. An empirical methodology to define the SEC parameter is reported for injection moulding [91,92], where this parameter was correlated using a hyperbolic law to the throughput provided by the machine. Those results have been confirmed in a more recent study carried out by Kara et al. [93]. Considering AM, a hyperbolic correlation between SEC and the average deposition rate (DR_a) has been found for

the electron beam powder bed fusion process for metals (PBF-EB/M) [94]. Similar findings have been reported in Ref. [95] for fused deposition modelling (FDM). Overall, those results correspond to processes dominated by a constant power demand [83]. To the best of the authors' knowledge, no studies have been carried out for composite manufacturing techniques to establish a clear relationship between energy consumption, process parameters and component features. However, some measure of the SEC value has been performed in the literature. The data collected is reported in the next paragraph. For comparison purposes, the literature data were converted into primary energy using a conversion coefficient equal to 0.38 [72]. In the following, all the data expressed in MJ should be intended as MJ oil equivalent unless differently specified.

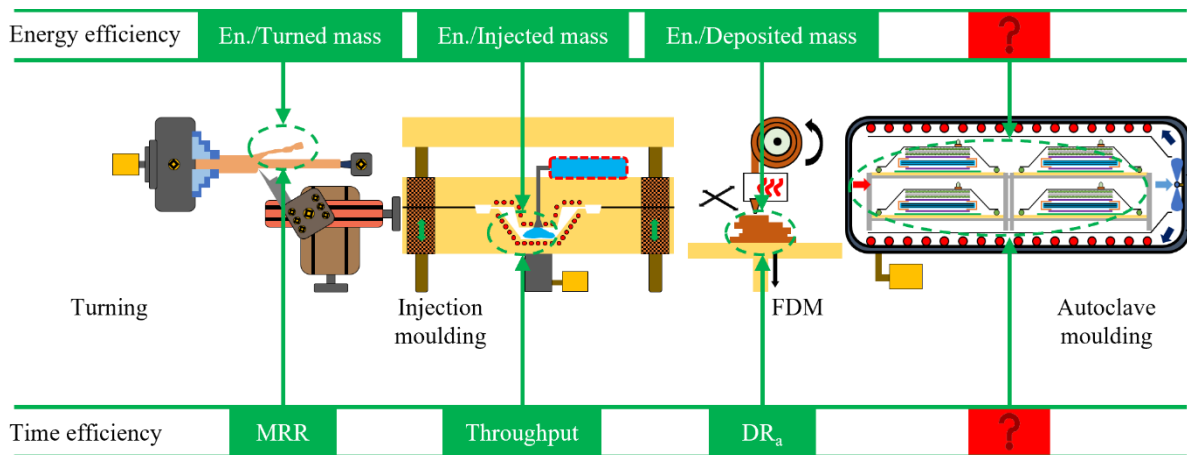


Fig. 23. Graphical representation of the connection between the time and energy efficiency (SEC) for different manufacturing processes.

Table 8 provides a complete overview of the SEC values collected from the literature about the manufacturing processes for composite material.

For the autoclave process, the SEC values ranged from about 20 MJ/kg to 600 MJ/kg. The lowest values refer to large mass production, while the highest values are related to low saturation of the production volume of the pressurised oven or, in general, low production volumes. Because of that, the highest values were registered for the production of aeronautic components (higher than 600 MJ/kg) [81]. In addition, if thermoset resin is considered, the SEC parameter of techniques is at least three times higher production with thermoplastic resins [74]. Even in this case, the processing temperature was the main process parameter influencing the total energy demand. The other processes led to significant energy savings and a more sustainable route. Considering the production of the same and single component, the energy demand for the X-ray cure was around an eighth lower than the corresponding autoclave production [35]. Comparable values were obtained if the number of parts produced in the autoclave was increased [35]. Analysing the specific contribution to the autoclave process, as an example, for thermoset matrix and CF as reinforcement [49] (Table 9), it can be observed that the major contribution (75 % of the total SEC) is due to the curing step. In fact, it has

been demonstrated [34,36] that energy efficiency could be significantly enhanced using a new microwave curing technology. The energy consumption was reduced from 3% of the autoclave curing (1800 kWh) to 50% of the conventional microwave curing technique (52 kWh) [36]. Moreover, a reduction in the curing cycle time was achieved as well. The autoclave and conventional microwave techniques process took over 10 hours and 8 hours, respectively, while the new microwave method curing time was below 5 hours. Another possibility to reduce electrical energy consumption is using nanostructured out-of-oven conductive [96]. In the case of aerospace applications, the electrical energy consumption was reduced from 3.80 kWh for the traditional curing to 0.03 kWh with such a new approach [96].

For hand lay-up and spray-up techniques, the U.S. Office of Energy Efficiency & Renewable Energy [16,17] provided an average range between 8 MJ/kg and 13 MJ/kg. However, in the literature, the higher values were associated with hand lay-up (19 MJ/kg) [81]. This can be explained considering that in the case of hand lay-up, the fibres are applied with a gun, speeding up the manufacturing time. On the contrary, the high values reported in Ref. [49] are not representative because only one single component was considered. As for the autoclave, the major contribution to the energy absorption in the hand lay-up is due to the curing step (Table 10), especially if performed using a microwave oven [49]. However, with respect to the autoclave, in the case of hand lay-up, the contribution to the total energy of the impregnation step becomes significant.

For the compression process (hot press) for SMCs and BMCs, the SEC values (3-18 MJ/kg) indicate an interesting alternative to the autoclave processes, especially in the case of high production volume [16,17,32]. Thermoforming (GMTs, hot compound and cold press) requires higher energy (30-79 MJ/kg) [16,17]. This may be related to the higher compactness of hot press systems and the resultant lower energy losses. The SEC values for the cold press process (12-36 MJ/kg) are higher than those for compression moulding (hot press) [16,17,22,77,81]. The difference could be attributed to the curing method that occurs under pressure for a higher time during the cold process. The energy demand for the preform matched die process (about 10 MJ/kg) is close to that of the compression moulding (hot press). On the other hand, huge energy demand is reported in Ref. [43,74] for CDF (up to 85 MJ/kg). These values may be explained by the selected thermoplastic resin, which showed a high melting point and a long curing cycle time. The injection processes showed lower energy demands and higher production rates than autoclave processes. The VARI process [16,17,22,32,77,81] requires a lower amount of energy if compared to the RTM technology (i.e., 3-11 MJ/kg vs 12-13 MJ/kg) [16,22,32,74,76,77,81]. The difference can be attributed to the lower pressure required by the VARI process, even if higher heat dispersion occurs because of the single mould. However, Witik et al. [49] found a value up to 82-91 MJ/kg for a component made of a

thermoset matrix and CF using the VARI process. Similarly, for RTM, Katsiropoulos et al. [74] found higher values, up to 27 MJ/kg and 81 MJ/kg, for a thermoset and thermoplastic matrix, respectively. Table 11 shows the contribution of each step to the overall energy for the thermoset matrix and CF production using microwave and thermal oven curing according to the VARI process [49]. Even in this case, the main contribution is due to the curing step, around 84% of the total.

The filament winding and the pultrusion technologies reported the lowest energy demands [16,17,22,32,76,77,81]. Comparing all studies, the filament winding technology showed slightly lower SEC values than the pultrusion technique (3-8 MJ/kg versus 3-10 MJ/kg). A considerable lack of data regarding energy consumption was found in the literature. This can be explained by considering the wide variability of fibre placement systems and heating solutions for the AFP and ATL processes. The feasibility of ATL assisted by laser heating was investigated in Ref. [97]. The tape used was a unidirectional (UD) carbon-reinforced PPS added on carbon woven fabric reinforced PPS laminates. The placement velocity varied between 75, 100 and 125 mm/s, while the laser power was fixed to 1300, 1500 or 1700 W. The specific electric energy demand was computed considering the only contribution of the laser input energy to the process, with values ranging between 7.4-14.3 MJ/kg. Brecher et al. [98] evaluated the laser-assisted ATL process for UD carbon fibre tape on substrates made of polyamide six reinforced with chopped glass fibre rovings. The overall energy demand was evaluated for a 250 mm tape length and for the following couple of parameters: (i) 200 mm/s and 1400 W, (ii) 600 mm/s and 2400 W and (iii) 800 mm/s and 2800 W. The results obtained were 37.8 Wh, 18.5 Wh and 14.2 Wh, respectively. The electric specific energy values for unit length were 544 J/mm, 266 J/mm and 204 J/mm, respectively. These results highlighted qualitatively how the choice of process parameters could be driven by energy efficiency evaluations that aim to reduce the cycle time and energy impact [55,98].

Manufacturing process	Specific Energy Consumption (MJ/kg)	Reference(s)
Autoclave (thermoset)	47	[74]
Autoclave (thermoplastic)	141	[74]
Autoclave (thermoset, CF)	386	[49]
Autoclave	21-23	[22,32,76,77]
Autoclave (aerospace component)	>600	[81]
Hand lay-up/Spray-up	8-13	[16,17]
Spray-up	15	[22,77,81]
Hand lay-up	19	[81]
Hand lay-up (thermal oven cur., thermoset, CF)	220	[49]
Hand lay-up (microwave cur., thermoset, CF)	265	[49]
CDF (thermoplastic)	85	[43,74]
Thermoforming (hot compound)	30-79	[16,17]
Preform matched die	10-11	[22,32,77,81]
Cold press	12	[22,77,81]
	28-36	[16,17]

Compression moulding (hot press)	3-4	[32]
	14-18	[16,17]
RTM (thermoset)	27	[74]
RTM (thermoplastic)	81	[74]
RTM	12-13	[22,32,76,77,81]
VARI	10-11	[22,32,77,81]
	3-5	[16,17]
VARI (thermal oven cur., thermoset, CF)	82	[49]
VARI (microwave cur., thermoset, CF)	91	[49]
Filament winding	3	[22,32,76,77,81]
	6-8	[16,17]
Pultrusion	3	[22,32,76,77,81]
	3-10	[16,17]
Laser assisted automated tape laying	0.5-1.4 (MJ/m)	[98]

Table 8. SEC of typical composite manufacturing processes.

Autoclave sub-processes (thermoset, CF)	Specific Energy Consumption (MJ/kg)	Reference(s)
Impregnation	80.3	[49]
Cutting	1.4	
Vacuum generation	10.2	
Autoclave curing	294.3	

Table 9. SEC of autoclave sub-processes.

Hand lay-up sub-processes (thermoset, CF)	Specific Energy Consumption (MJ/kg)	Reference(s)
Impregnation	83.2	[49]
Cutting	1.5	
De-bulking	39.2	
Vacuum generation	15.9	
Thermal oven curing	85.0	
Microwave curing	121.7	

Table 10. SEC of hand lay-up sub-processes.

VARI sub-processes (thermoset, CF)	Specific Energy Consumption (MJ/kg)	Reference(s)
Weaving	3.7	[49]
Cutting	1.1	
Vacuum generation	9.9	
Microwave curing	70.5	
Thermal oven curing	77.1	

Table 11. SEC of VARI sub-processes.

5. End-of-life considerations

As the use of composite is growing in several manufacturing sectors, the problem of handling the relative waste is also becoming essential. According to Naqvi et al. [19], CF waste for the main high-value industry sectors will rise from 10 thousand tonnes in 2010 to over 180 thousand tonnes in 2060 [99]. However, the composite nature of these materials makes them difficult to be recycled. Oliveux

et al. [76] and Zhang et al. [100] grouped the recycling techniques for composite components into (i) mechanical, (ii) thermal and (iii) chemical processes.

Mechanical techniques are based on grinding the matrix to the side of the composite after an initial crushing or shredding. Mechanical recycling consists of machining or other techniques such as high voltage fragmentation. These processes are often applied to glass fibre (SMC and BMC), CF, and Kevlar aramid-reinforced composites [76]. Short fibres are generally obtainable using mechanical approaches.

Different techniques are available for thermal processes, such as pyrolysis, fluidised bed pyrolysis, and microwave-assisted pyrolysis. Typically, materials such as fibres, fillers and inserts are recovered. The same result does not apply to monomers that could be reused to produce resins. The typical process temperatures range between 450°C and 700°C according to the resins. The higher temperature values are used with thermoplastics and epoxides.

Among the chemical approaches, solvolysis uses solvents to degrade the resin. A wide range of solvents, temperatures, pressures, and catalysts make this technique usable with different composites. Furthermore, solvolysis can isolate monomers [76]. Both thermal and solvolysis techniques are more suitable for CF that are not subjected to strong mechanical degradation [101] with respect than for GF [102]. In particular, the reduction of tensile strength of CF recycled is around 20% [101], while it is about 60% for glass fibre [102].

Table 12 lists the SEC of different composite recycling processes. The electrical SEC is generally evaluated by varying the processing conditions, such as the milling rate [77,103]. In the case of CFRP, the obtained SEC values varied between 0.25 MJ/kg and 2 MJ/kg, where the highest value is associated with the lowest milling rate (10kg/h) [77]. Similarly, for unsaturated polyester reinforced with glass fibre chopped strand mats, the electric SEC value varied between 0.35 MJ/kg and 5.5 MJ/kg, where the highest value was obtained for a processing rate equal to 30 kg/h [103]. In general, the relationship between the electric SEC factor and the processing rate is inverse, as for other manufacturing processes, and the lowest values are due to the much higher processing rates. To prove this evidence, Mativenga et al. [103] investigated a processing rate ranging from 10 kg/h to 150 kg/h for the GFRP. The results showed an electric SEC range of 1.9 MJ/kg and 0.15 MJ/kg, slightly lower than that found for the CFRP in the same processing range. This difference was explained because of the better machinability of GFRPs with respect to CFRPs. The SEC values are reduced drastically when considering mechanical recycling because the energy needed is much lower than the embodied energy of virgin CF and GF (see Table 12 in comparison with Table 12). Thermal and chemical techniques demand much more energy than mechanical recycling because of the higher complexity of the recycling route [103].

Recycling techniques	Specific Energy Consumption (MJ/kg)	Reference(s)
Mechanical recycling - CFRP	0.7-5.3	[77]
Mechanical recycling - GFRP	0.4-5.0	[103]
High voltage fragmentation	4	[103]
Pyrolysis	3-30	[103,104]
	63	[74,76]
Microwave	10	[105]
Chemical	61-93	[103]

Table 12. SEC of different composite recycling processes.

Another aspect that should be considered is the simultaneous assessment of the economic and environmental impacts of different strategies for the end-of-life handling of reinforced composites[104,105]. Landfilling, incineration, and co-incineration with energy recovery are less virtuous strategies with respect to recycling via grinding, pyrolysis, microwave, and supercritical water. In fact, recycling techniques are commonly expensive options, especially for the investment cost [105], but they guarantee lower global warming potential (GWP) impacts. Joustra et al. [106] listed the aspects of the design of composite products that can facilitate their implementation in a circular economy scenario. Modularity, simplification, and standardisation are considered the key aspects. However, robust methodologies, analyses and data are still missing in the literature. Sectors such as renewable energy could benefit to a great extent from such strategies because most of their components are realised by composites [107,108].

6. LCA application, conclusion, and outlooks

The methodology proposed by Suzuki et al. [81] for LCA is applied here to provide an example of using the reviewed data. The method considers the weight of the polymer matrix fraction and that of reinforcement on the embodied energy of a composite component (E_{b_C}) and is summarised in Equations (1) to (4) [81]. The embodied energy of a composite is computed as a weighted average ($w_{f_{PM}}$, $w_{f_{RF}}$) of the embodied energy of the polymer matrix ($E_{b_{PM}}$) and reinforcement ($E_{b_{RF}}$). The contribution at the manufacturing level using the SEC parameter (SEC_C) is also included. The terms E_{b_C} and SEC_C are both multiplied by the composite mass, which is the sum of the polymer matrix mass (m_{PM}) and reinforcement mass (m_{RF}). The overall energy of the composite component (E_C) is given by the sum of the terms E_C^{rw} and E_C^m , which represents the energy embedded in the composite raw materials and energy needed to manufacture a composite component, respectively.

$$E_{b_C} = w_{f_{PM}} \cdot E_{b_{PM}} + w_{f_{RF}} \cdot E_{b_{RF}} \quad (1)$$

$$w_{f_{PM}} = \frac{m_{PM}}{m_{RF} + m_{PM}} \quad (2)$$

$$w_{f_{RF}} = \frac{m_{RF}}{m_{RF} + m_{PM}} \quad (3)$$

$$E_C = E^{rw}_C + E^m_C = E_{b_C} \cdot (m_{RF} + m_{PM}) + SEC_C \cdot (m_{RF} + m_{PM}) \quad (4)$$

Where:

E_{b_C} : embodied energy of the composite (MJ/kg);

wf_{PM} : weight fraction of polymer matrix (-);

$E_{b_{PM}}$: embodied energy of the polymer matrix (MJ/kg);

wf_{RF} : weight fraction of reinforcement (-);

$E_{b_{RF}}$: embodied energy of the reinforcement (MJ/kg);

m_{PM} : mass of the polymer matrix (kg);

m_{RF} : mass of the reinforcement (kg);

E_C : overall energy of the composite component (MJ);

E^{rw}_C : energy embedded in the composite raw materials (MJ);

E^m_C : energy needed to manufacture a composite component (MJ);

SEC_C : specific energy consumption of the manufacturing process (MJ/kg);

The selected composite component has a volume of 0.01 m³, and the LCA was performed using two reinforcement fill percentages, 5% and 40%. Two polymer matrixes were selected: PEEK with an embodied energy of 300 MJ/kg [32] and epoxy with an embodied energy of 70 MJ/kg [16,17]. Two reinforcements were evaluated: CF with an embodied energy of 530 MJ/kg [16] and GF with an embodied energy of 12 MJ/kg [17]. Therefore, the analysis has been performed comparing four composites: 1A) PEEK-CF, 2A) Epoxy-CF, 1B) PEEK-GF, and 2B) Epoxy-GF. Autoclave, hand lay-up and thermoforming were considered as manufacturing processes, with an SEC parameter respectively equal to 600 MJ/kg [81], 11 MJ/kg [16,17] and 55 MJ/kg [16,17]. This analysis used the average values of the range reported above. Fig. 24 shows the results distinguishing the raw material production and the manufacturing level contributions. Considering the raw material production, the results show that increasing the reinforcement fill percentage reduces the energy demand only when GF is considered (1B and 2B cases). The opposite trend is visible when the CF reinforcement is present (1A and 2A cases). This result is explained by the higher embodied energy of CF fibre. The importance of the matrix polymer increases for low fill percentages and reveals a higher impact of the PEEK material (1A and 1B cases) with respect to the epoxy resin (2A and 2B cases). At the manufacturing level, the autoclave process contributes the highest respect to the other manufacturing technologies. Moreover, contrary to the other techniques, it holds a higher share of the total energy with respect to the raw material production contribution. This LCA example does not include the contributions of post-processing and EoL scenarios.

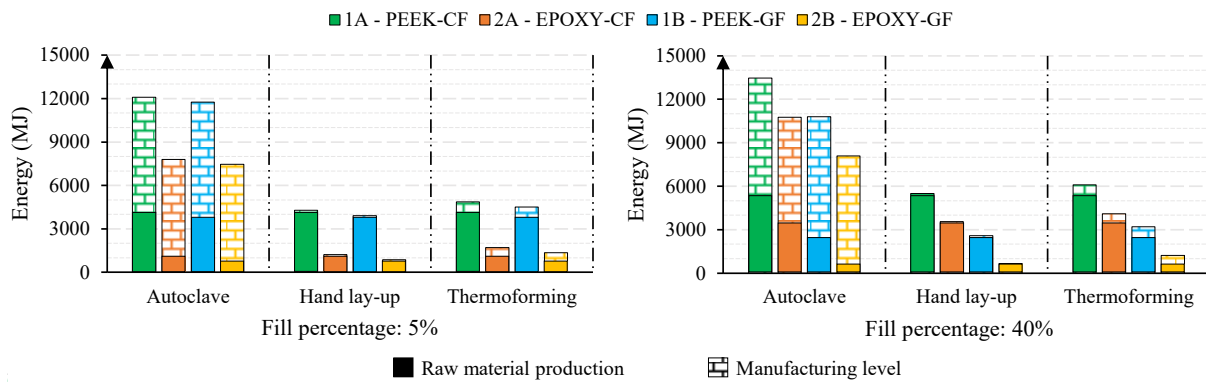


Fig. 24. Example of energy LCA applied to a composite component (volume 0.01 m³).

To conclude, this paper presented the state-of-the-art life cycle inventory (LCI) data available in the literature for composite products and their practical use for performing LCA for a conscious choice of product materials. The embodied energy of the most common fibres and polymeric matrixes and the SEC parameter of the leading manufacturing techniques were considered. All in all, the presented review highlighted that there is still considerable work and research to be accomplished before LCA studies are carried out with reliable data. The collection of these data is an uncompromising challenge that must be addressed to consider the suitability of composite to replace traditional materials not only for their mechanical performance but also from energetic metrics such as the cumulative energy demand (CED) of the product. This review highlighted several critical points which should represent the direction for future research:

- The available data present huge dispersions in embodied energies and SEC values for both the reviewed materials and technologies. Often the comparison among the data is impractical because a well-structured approach for the measures acquisition and basic and detailed information are missing in the published works
- Considering the manufacturing level, the available studies use SEC only as a process descriptor without considering the specific application. Therefore, there is a need to identify a clear relationship between the process parameters, processed materials, component size and energy consumption
- Among the manufacturing processes, no data are available for AM, even if such techniques reached the technological capability to be successfully applied to produce composites
- Few studies analysed the End-of-life methods for composites material and their impact. However, the availability of this data is crucial for the application of a broader logic, such as those of the circular economy
- Over the past decade, the concept of utilising green materials has become more mainstream. However, the literature on the use of natural fibres is still extremely limited [109]. Nowadays,

focusing on these fibres appears to be a priority for several reasons. As an example, compared to glass fibre-reinforced components which hold today the largest market share (Fig. 22), natural fibres are more sustainable and recyclable at their end-of-life, lighter and safer in handling, nonabrasive to the machine parts during processing and show higher specific tensile properties, higher thermal and acoustical insulation [82,110].

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