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Cost-efficient, automated, and sustainable composite profile manufacture: A review of the state of the art, innovations, and future of pultrusion technologies

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ABSTRACT

Over the last 70 years, pultrusion has matured into an industry-leading process when it comes to providing high throughput and automated composite manufacture at a competitive price point. In this paper, we review recent innovations that have advanced pultrusion to a versatile manufacturing technology and thereby allowed composite materials to penetrate markets in, e.g., the automotive, construction, aerospace, and wind turbine industries. We accompany our review with discussions on how pultrusion has enabled new innovations within additive manufacturing and sustainable composite manufacturing, and finally, we provide an outlook and suggestions for where we see the potential for research and new industrial applications of pultrusion technology.

1. Introduction

Pultrusion is a continuous, fully automatable, and increasingly popular process that is used to manufacture fibre reinforced composite profiles with a constant cross section (see Fig. 1). As the name implies, pultrusion is a compound word of the terms “pull” and “extrusion”, but as opposed to extrusion processes where the profile is produced by pushing material through a die, a pultrusion process advances the profile by pulling it.

There is a broadening adoption of pultruded profiles and products. This can be motivated by the cost and energy efficiency of the process as well as the repeatable, high material quality even for high fibre volume contents. Pultrusion has been experiencing some of the fastest growth across the European fibre-reinforced polymer composite industry [1], and it is expected that the global pultrusion market (\$2 billion) will expand with an annual growth rate of 4.9% [2], far exceeding the

near-future global GDP (gross domestic product) growth of 2.3% (in 2023, [3]). With a reported energy intensity of 3.1 MJ/kg, or approximately one-seventh of autoclave moulding¹ [4,5], pultrusion is an industry-leading process in terms of energy efficiency. Together with processes like filament winding and automated fibre placement technologies, pultrusion is a continuous manufacturing process, but it is the only manufacturing technology that allows for endless and uninterrupted composite manufacture [6]. This enables composite manufacture at a very competitive price point, also since pultrusion has a high material efficiency with low waste and no inherent need for disposable items such as vacuum bags and flow mats. Even with the outbreak of COVID-19, and the subsequent disruption of global supply chains, pultruded polyester/glass fibre profiles may today be acquired in bulk from East Asia for as low as roughly \$3/kg, close to the price point of the raw materials².

While pultrusion was first introduced in early variants in the

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¹ It is important to note that the design freedom of autoclave moulding is unparalleled to pultrusion, which is why this reported energy intensity should be taken as a reference point and not a direct comparison.

² This estimate was based on prices of raw materials and pultruded profiles on the e-commerce giants, okorder.com (CNBM International) and alibaba.com (at the time of writing, the 20th of March 2022).

1950's with the work of W. Brandt Goldsworthy in the USA on polyester resins and Ernst Kühne in Switzerland on epoxy resin [6], a lot has happened since the initial concepts 70 years ago. Pultrusion is today a highly versatile process that is compatible with a wide variety of fibre reinforcements and matrix materials, e.g., conventional thermosets and thermoplastics, as well as renewable matrices, reinforced with glass, carbon, aramid, and natural fibres. Many innovations were required to reach its current maturity and flexibility that have facilitated production of, e.g., lightweight parts for the automotive, construction, aerospace, and wind turbine industries. These are reviewed in this paper together with an accompanying discussion on how pultrusion has enabled technological advancements within additive manufacturing and sustainable composite manufacturing. Compared to other review papers about pultrusion (recapitulating mathematical modelling and simulation [7], materials & applications [8], and thermoplastics [9]), we focus on *recent* innovations, their advantages, and how they have enabled applications and penetration of new markets for composite materials. Finally, we also draw on our consortium's extensive experience in pultrusion gained from completing multiple academic and industrial research projects to provide an outlook and suggestions for where we see potential for new pultrusion-related research and industrial application.

The structure of the review paper is organised as follows. Sec. 2 starts by providing an overview of the pultrusion process and the processing steps. Sec. 3 represents the main body of the paper, and starts with a short review of some of the methodical work that has been completed about pultrusion. Subsequently, we review and discuss recent innovations in pultrusion technology, in particular: new production technology (such as microwave and UV-assisted curing, pultrusion of bent profiles, etc., Sec. 3.1), new layup and material-forming technologies (e.g., pull-braiding, hybrid materials, and multi-step pultrusion, Sec. 3.2), advanced intermediate materials for thermoplastic pultrusion (such as bi-component fibres, Sec. 3.3), how pultrusion is an enabler for additive manufacturing (e.g., pultrusion of thermoplastic strands with fibre reinforcements, use of pultrusion technologies for free-form 3D printing, Sec. 3.4), and how pultrusion can facilitate sustainable composite manufacture (low-waste and energy-efficient composite manufacture, utilisation of renewable materials, etc., Sec. 3.5). Finally, in Sec. 4, we summarise conclusions, outlook, and suggestions for where we see the potential for new research and industrial applications.

2. Process overview and material behaviour

2.1. Process overview

Profiles manufactured using pultrusion processes can be processed using several different process concepts that are compatible with thermoset and thermoplastic polymers, produced from both renewable and fossil resources, reinforced with glass, carbon, aramid, and different types of natural fibres. Table 1 presents common material compositions

and process configurations that reflect the state-of-the-art and versatility of pultrusion processes. While pultrusion processes vary by design, the process generally includes the following substeps [6]:

- **Preparation of layup:** The raw material is drawn from fibre creels or racks. Depending on the required application of the profile, rovings (tows of individual fibres), mats, and fabrics enter the profile layup, but it can also be pre-preg materials, tapes, commingled fibres, or bi-component fibres, as well as other raw materials where the fibres might be pre-combined with the matrix material. Before entering the pultrusion die, the raw material passes a number of guides that organise and shape the layup into the desired cross-sectional shape and ply plan.
- **Impregnation and heating/cooling:** The organised fibre material is now moved towards the pultrusion die. In resin-injection pultrusion (Figs. 2, 3 and 4), injection takes place in the first part of the die. Typically, active heating and cooling are now applied to ensure proper viscosity while controlling the solidification of the profile. In other pultrusion concepts, such as resin-bath pultrusion and open-die pultrusion, the fibre material is pre-impregnated before entering the die, otherwise, the processing steps are identical. A clear advantage of resin-injection pultrusion is the fact that it is a closed-mould process, which limits possible exposure to volatile solvents, etc., from the resin system. Before exiting the die, the outer edges of the profile must be solidified to ensure that the profile remains stable when it is no longer supported by the die walls.
- **Cooling and cutting:** A pulling mechanism advances the process by continuously pulling the solidified profile out of the die to cool down. Finally, the profile is cut into desired lengths.

2.2. Process and material behaviour

Independent of the pultrusion concept, a key ingredient to processing of fibre-reinforced polymer profiles using the pultrusion process is *heat*, and since the rheological behaviour of the matrix material is both time and temperature dependent, tight control of the profile's thermal history is crucial to ensure correct material flow and process quality. Here, we describe the process behaviour of a thermoset resin-injection pultrusion process, and we also discuss how this is different for other materials and pultrusion concepts.

A pultrusion die (Fig. 4) can be designed with or without a tapered section. A taper allows resin saturation to take place at a lower fibre volume fraction, which is a necessity for resin-bath pultrusion where impregnation is started at atmospheric pressure. Collimation, as well as the viscous and mechanical sliding friction, contribute to the pulling force [26,27], but with a tapered die design, the hydrostatic resin pressure during consolidation must be overcome as well (cf., Sec. 6.5.3 in Ref. [10]). For resin-injection pultrusion, the resin system is injected at an elevated pressure allowing for saturation, also in a straight

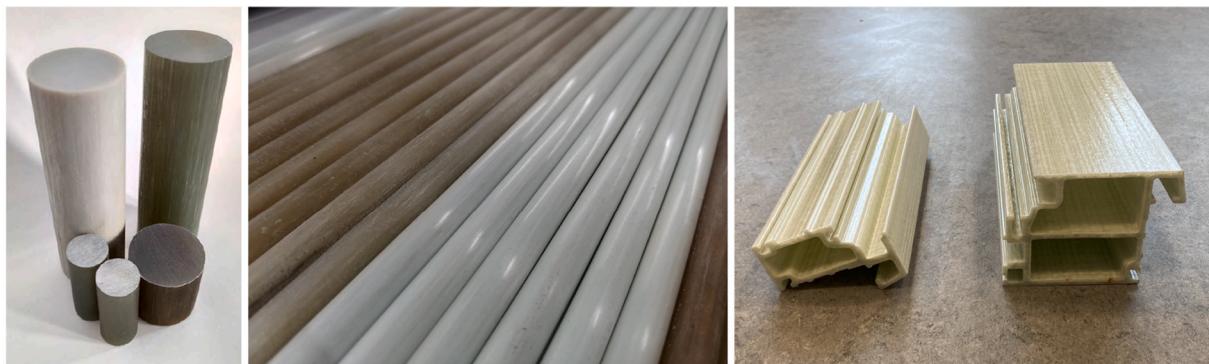


Fig. 1. Fibre-reinforced, thermoplastic rods with different diameters (left and middle, $\varnothing 10 - 40$ mm), fibre-reinforced, thermoset profiles with complex cross sections (right, $w \times h \approx 60 \times 60$ mm 2 , wall thickness 2 mm). All these profiles were manufacturing using the pultrusion process.

pultrusion die. Here, the magnitude of the injection pressure together with the profile-advancing pulling speed is adjusted to achieve sufficient impregnation without resin wastage due to overflow [11,28,29].

Once the fibres are impregnated, the state of the composite profile in a pultrusion process can be derived from the temperature, T , and for a thermosetting polymer, the degree of cure, α , as well. Here, α ranges from 0 to 1, which corresponds to an uncured ($\alpha = 0$) and fully cured matrix material ($\alpha = 1$). A process diagram of a thermosetting resin-injection pultrusion process that was achieved using numerical simulation can be seen in Fig. 5 [11]. This diagram exemplifies how the temperature, degree-of-cure, glass-transition temperature, resin pressure, and viscosity can develop throughout a pultrusion process.

Chemorheology describes the cure and temperature-dependent constitutive behaviour of the matrix material. For a polymer, this behaviour is split into three phases. In the first phase, the viscous phase, the polymer chains are mobile, and the mobility of the polymer chains is characterised by the viscosity of the matrix material. In most thermoset pultrusion processes, impregnation takes place close to or below room temperature, while in some pultrusion concepts, heat is applied to decrease the viscosity and therefore expand the process window suitable for impregnation. For fully polymerised thermoplastics, heat *must* be applied to melt the polymer, otherwise, impregnation cannot take place. Towards gelation, which is the second phase, the viscosity rises as the mobility of the polymer chains decrease. The gelation point is defined as the point where the viscosity rapidly increases, and the matrix starts to support loads without flowing. After gelation, the polymer is in a “rubbery” phase, and as a result of the developed polymer chain structure, straining the polymer introduces a lasting stress state. In loose terms, some may refer to this as the point where the matrix starts to gain “memory” as the response is no longer only viscous but also elastic. For thermosetting polymers, curing is thermally activated (hence the name, “thermoset”) and external heat is now normally needed, however for thermoplastic materials, cooling is needed for the polymer to solidify. In the final phase, further cooling is applied, and the temperature reaches the cure-dependent glass-transitioning temperature (which corresponds to vitrification), $T = T_g$. As Fig. 5 illustrates, resin curing can continue upon die-exit. After cooling down, the process is complete.

3. Innovations and research in pultrusion technology

Over the last 50 years, more than 1400 papers have been published in scientific journals and conference proceedings about pultrusion³, which includes theoretical and experimental studies of various thermoset resin systems [30–40], thermoplastics [20,22,24,41–49], as well as alternative and renewable materials and reinforcements [50–54]. For resin-injection pultrusion, in particular, where analyses of the impregnation flow is essential to ensure saturation [15,38–40,55,56], studies have considered geometrical configurations of the injection chamber [57–61] as well as other process parameters such as the profile-advancing pulling speed [62], resin viscosity [63], and fibre permeability [64]. Using thermo-chemical-mechanical analyses, numerous studies have also looked into process-induced stresses and deformation of thermoset pultrusion [17,65–72]. Clearly, extensive efforts have been invested in advancing the fundamental understanding of the governing physics of pultrusion. These tools today equip engineers to perform the analyses and optimisation of pultrusion processes that are needed to find the most suitable process parameters to reduce costs and defects while maintaining production output and efficiency.

The main limitations of the pultrusion processing window are defined by heat transfer, impregnation, consolidation, die-pultrudate interaction, residual stresses and warpage as well as the alignment and distribution of the reinforcing material in the profile. From the production side, the cost of the overall product remains the main driver for increasing line speeds and the development of new resin formulations, while the need for recyclability and ease of operation puts the focus on thermoplastics in general as well as sustainable and renewable fibre options and bio-based polymers such as PLA. From an application perspective, the limitations in profile geometry as well as the choice of fibres, matrix, and lay-up are the main factors. The following sections (outlined in Fig. 6) address innovations that not only improve the traditional process-related limitations of pultrusion but also extend the application range of pultruded profiles from constant-cross-section profiles to fully functionalised, variable cross-section, multi-material components that keep the advantages of cost-efficient high-quality pultrusion while offering significantly higher design freedom in terms of geometry, material choice and functional integration.



Fig. 2. A state-of-the-art, industrial resin-injection pultrusion line. Here, the material flows from left to right (courtesy of Fiberline Composites A/S). A schematic of the line can be seen in Fig. 3 and a cut-view schematic of the pultrusion die can be seen in Fig. 4. This figure was taken from Sandberg [10].

³ This statement is based on a web-of-science search on titles that included “pultrusion” or “pultruded” on the 9th of February 2022.

3.1. Advances in production technology

Reducing energy and time consumption for manufacturing is a common goal for the whole composite material community to increase efficiency and decrease the environmental impacts. Here, energy-efficient, high-quality, and fast manufacturing methods are needed, and in this section, we review advancements in pultrusion production quality to achieve just that.

3.1.1. Alternative curing/heating technologies

In conventional pultrusion processes, a metal die with electrical heating elements is used to cure and consolidate the pultruded profile (Fig. 4). Even though conventional pultrusion processes are considered to be energy efficient, efforts have been made to further advance the efficiency of pultrusion using alternative heating/curing methods [73].

Radiation curing is an alternative for thermal curing, in which the polymerisation of thermosets is induced by ultraviolet (UV) radiation, gamma X-ray, electromagnetic (microwave), or electron beams [74]. UV radiation is less hazardous, has lower energy consumption, and needs lower initial investment cost compared to the others, and therefore this is one of the most promising choices of alternative curing method for the pultrusion process [74]. A critical factor of UV-curing is the opacity of the resin and fibres, which limits the effectiveness of curing composites with carbon reinforcement [75], for example. For pultrusion, studies assessing UV-curing techniques have been limited to glass fibre profiles.

The first trials of adapting UV curing into pultrusion was presented in the 80s using a UV-transparent die. Later on, UV curing was tested as an out-of-die curing method in which UV radiation was applied at the exit of a forming die. Here, any excess resin was removed, and the impregnated uncured profile was compacted/formed into the desired cross-sectional shape in the forming die. Out-of-die curing has been reported to require two orders of magnitude lower pulling force than conventional pultrusion [76], and it also allows post-die manipulations to apply curved regions on the pultruded profile [77] (known as “bent pultrusion”, to be discussed in Sec. 3.2.4 and Fig. 11). A relatively low peak temperature that is typically reached during UV-cured pultrusion makes it possible to use temperature-sensitive reinforcements such as Nylon6 [78]. Another route is microwave-assisted pultrusion. Here, the material properties together with the electromagnetic characteristics define the microwave interaction [79], meaning that selective heating of the profile with limited loss of energy to the die can be achieved by using a partially microwave-transparent die [80] (Fig. 7). Direct heating of the profile using microwave heating can result in 5 times higher pulling speeds and 1.7 times lower energy consumption as shown in a recent theoretical study by Barkanov et al. [81]. In addition to the energy efficiency, microwave heating can ensure more homogeneous curing throughout the cross section which results in lower residual stresses [82].

In addition to these alternative heating/curing methods, new strategies that exploit material-based technologies to increase efficiency are also investigated in the composites community. For example, an emerging energy-efficient composite manufacturing route is frontal polymerisation. Here, the strategy is to exploit highly reactive resin systems that allow for polymerisation within a very narrow process window. The idea is that with the right curing kinetics, once initiated, a very localised and self-sustained reaction will emerge and propagate

throughout the entire material layup without the need for any additional heating [83]. With well-controlled frontal polymerisation, the energy input can be minimised while producing high-performance composite materials [84]. In theory, frontal polymerisation can allow for pultrusion with no or limited external heat source after initiation of polymerisation.

3.1.2. Vacuum, fibre spreading, and fibre treatments to advance product quality

Fibre-matrix bonding, void content, and microstructural uniformity are some of the key aspects that govern the quality and performance of a pultruded profile and composite materials in general. To optimise pultrusion processes, some of the well-known concepts that are widely applied for the general processing of polymer composites now find their way to pultrusion processes. An innovative example of this is vacuum-assisted pultrusion, where an appropriate vacuum pressure can lead to void reduction by extracting entrapped air in the tow during impregnation [85]. Lapointe and Laberge Lebel [86] showed that the vacuum usage reduces the void content and increases the shear strength as a result thereof. Fibre-surface treatment is another commonly used application for fibre-reinforced polymers to enhance impregnation by capillary effects [87] and mechanical performance by improved fibre-matrix bonding [88]. Some of the surface treatment methods that have been applied with positive outcomes for pultrusion are coupling agent treatments [89], liquid nitrogen treatments [90], and corona discharge treatments [91]. Whereas some of these fibre-surface treatment methods should be done before the pultrusion process, corona discharge treatment, for example, can also be performed continuously in the pultrusion line.

Well-dispersed rovings lead to a more homogeneous fibre distribution, and this is another critical aspect to improve the impregnation and mechanical properties of composites. Even in conventional pultrusion, it is common to have a set of static bars (fibre guides, as referred to in Sec. 2.1) on which rovings are organised and spread when passing. However, more advanced techniques can be necessary to enhance fibre spreading. Here, wrap angle, pre-tension of rovings, temperature, and pulling speed are some of the effective parameters related to mechanical fibre spreading [92]. A higher wrap angle or increasing the number of spreader pins result in better fibre distribution and thinner rovings that will lead to an improved impregnation quality. Irfan et al. [92] developed a modified pultrusion process which consists of a fibre spreading unit. This spreading unit provided better lateral spreading and reduced roving thickness, and this resulted in better impregnation and lower void content [93]. Van De Steene et al. [94] applied passive and active spreading methods into a small scale pultrusion die. They designed a spreader-impregnation device for melt impregnation of Polyamide12, and showed that the dispersion quality with both passive and active spreader units is much better than an equivalent unmodified pultrusion process. On the other hand, adding mechanical resistance to the fibre material increases both the probability of broken fibres and the pulling force needed to advance to profile. Therefore, the trade off between these drawbacks and the potential benefits of mechanically-aided dispersion of rovings should be taken into consideration.

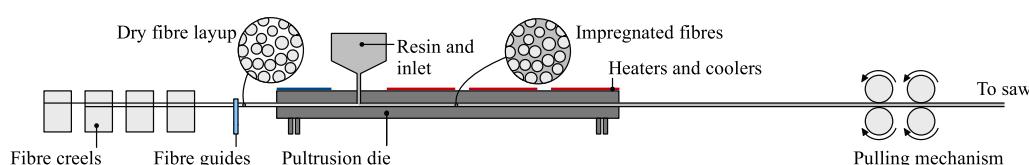


Fig. 3. Schematic of a resin-injection pultrusion line. In the figure, the material flows from left to right.

3.2. New layup and material forming technologies

In addition to the production technologies that we reviewed in Sec. 3.1, significant research efforts have been invested in technologies that can enable pultrusion of certain material layups, configurations, and fibre architectures. In this section, we review some of these latest advancements, and how these have helped the pultrusion industry to penetrate new markets and application areas.

3.2.1. Complex lay-ups, cross-sections, and braidtrusion

The major fibre orientation of pultruded profiles is traditionally along the profile axis to yield the highest packing of fibres and to provide the tensile strength needed to advance the profile by pulling it. To overcome this resulting transverse strength deficiency, continuous strand mats can be used to provide randomly oriented fibres in combination with surface veils for a better surface finish. Recently, more complex non-woven multi-axial stitched fabrics are used to manufacture complex, load-tailored layups even with fully transversal plies as the stitch yarn itself is designed to take the loads in the pulling direction. Different applications include a U-shaped pultruded profile using triaxial +45°/90°/-45° layup mats [95], or combinations of different biaxial and triaxial mats [96] to manufacture road decks. Cost-efficient, high-volume production of an automotive rear bumper profile [97] has been achieved by the combination of different biaxial, triaxial, and UD non-crimp fabrics. Other potential application fields for cost-efficient manufacturing of complex cross-sections include the production of airfoils for unmanned aerial vehicles [98] as well as wind turbines blades [99]. A different approach called patch-pultrusion combines the pultrusion of conventional reinforcing materials with load-tailored fibre patches that are applied to a textile carrier material using the Tailored-Fibre-Placement technology [100]. This enables the local reinforcement of pultruded profiles for highly loaded areas resulting in an overall lighter and more cost-effective profile.

Braidtrusion is another approach to further improve the multiaxial loading capabilities of pultruded profiles. Braidtrusion (also called “braiding pultrusion” or “pullbraiding”) is a compound word of “braiding” and “pultrusion”, and the process works by combining these two processing techniques. In a braidtrusion process, rovings are intertwined by braiding (similar to the processing of textiles) around a mandrel or core, while the profile is advanced by pulling it. Different kinds of material and processing techniques can be utilised in braidtrusion. For example, Michaeli and Jürss [45] demonstrated the use of thermoplastic (PP) commingled with glass fibres. Similar configurations

have also been used to manufacture curved, thermoplastic composite profiles [101], as well as complex cross-sectional shapes such as L-shaped profiles [102]. Braidtrusion has also been conducted with the use of impregnation strategies, for example, resin-bath impregnation [103]. Compared to the use of, e.g., commingled fibres, resin-bath impregnation enables the use of a larger variety of materials, including thermoset resin systems. Recent work has also been completed on more methodical aspects of braidtrusion. For example, Ghaedsharaf et al. [104] investigated the resulting internal geometry and fibre orientation distribution as a result of the material forming that takes place during braidtrusion. Related to braidtrusion, pultrusion-winding is another approach in which a hoop layer is wound around a pultruded core to achieve the helical contact surface that is ideal for composite rebar applications [105,106].

3.2.2. Multi-stage and multi-material pultrusion

Multi-stage pultrusion consists of multiple pultrusion stages, where additional material is added in each stage around an initially pultruded profile as depicted in Fig. 8. For thermoset materials, the proposed benefit of a multi-stage system is the better homogeneity of the curing process in each layer as the curing reaction is initiated from both the previously pultruded core and the heated die walls. This concept has been proposed and investigated by Gorthala and Flynn [107] and modelled by Albayati and Gorthala [108], showing a higher degree of cure for a multi-stage die compared to a single-stage process. These multi-die systems can also be used to continuously increase consolidation pressure for the same profile geometry, without adding additional material in the different stages, resulting in a lower void content and higher shear strength [23]. For thermoplastic pultrusion, multiple stages can also be used to improve heat extraction efficiency, as the ratio between available cooling surface and the pultruded volume increases with the number of stages. This has been successfully demonstrated for solid rods using glass-fibre PET commingled yarns as shown in Fig. 9(a) with a two-step pultrusion process [109] as well as for tubes using braided commingled preforms in a three-stage process [110].

Multi-material pultrusion consists of the combination of different reinforcing fibres, polymer matrices as well as embedded metallic structures or sensors. For thermoset materials, pultrusion of hybrid rods with a carbon fibre core and E-glass fibre shell can increase the tensile strength of overhead electrical conductors by 150%, while reducing the thermal expansion and therefore sag by up to 400% [111]. A different application of a multi-material concept is the combined use of carbon and glass-fibre rovings, multi-axial fabrics, and piezo sensors to optimise

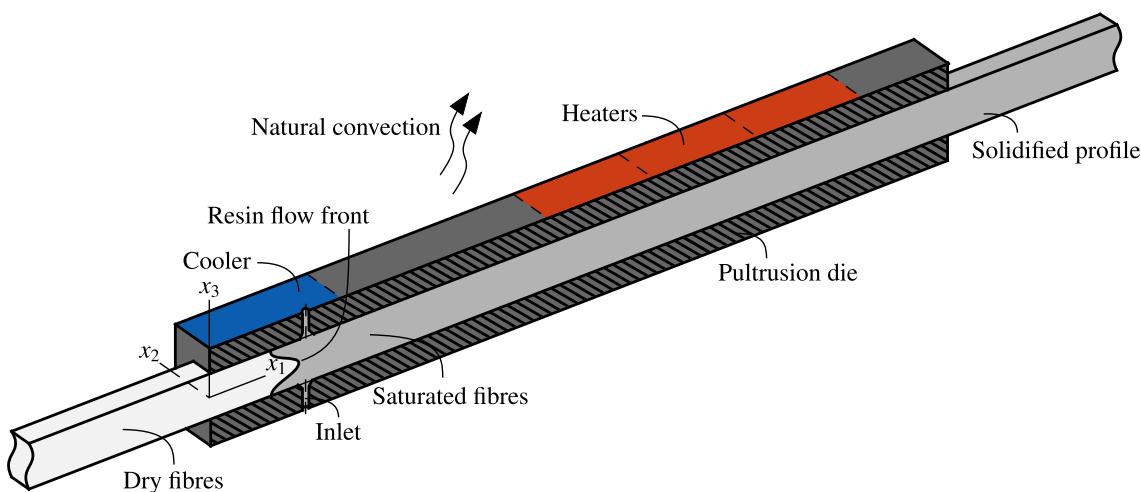


Fig. 4. Cut-view schematic of a pultrusion die that is used for the resin-injection pultrusion concept. Here, the material flows from left to right. The figure indicates placements of heaters, coolers, and resin inlets, and it illustrates how a flow front splits the dry and saturated fibre material. This figure was taken from Sandberg et al. [11].

Table 1

An overview of selected work on pultrusion that reflects the different material compositions and state of the art considering speed and fibre volume fraction for both thermoset and thermoplastic resins, fibre architecture, as well as profile dimensions.

Material technology/fibre architecture	Fibre material	Matrix material	Profile cross section	Fibre volume fraction	Pulling speed mm/min]	Ref.
Unidirectional (UD) Braid	carbon	epoxy	1.75 × 46 mm, 46 × 3.35 mm, T	56–65%	50	[12]
UD	Jute glass	unsat. polyester	Ø12.7 mm	70%	195	[13]
UD (intra-yarn, interlayer hybrid)	Carbon glass	epoxy	Ø12.7 mm	70%	–	[14]
Air text.(UD)	glass	polyurethane	80x80 mm	44%	182	[15]
Cont. strand mat (CSM)	glass	unsat. polyester	750 x 200 mm (hollow)	31–43%	–	[16]
Biaxial fabric (BGF)						
Quadraxial fabric (QGF) UD						
Fabric UD	glass	vinylester	80 × 80 mm, L	59%	200–600	[17]
Tape (TP)	glass carbon	PA12 PEEK	15 × 3mm	50% (PA12) 60% (PEEK)	300–3000	[18]
Parallel (PL)	glass carbon	PP	10x1.5 mm	35–57% (CM)	80–7500 (CM)	[19]
Commigled (CM)		PA6	Ø 8x1,	42–53% (PD)	80–20400 (MT)	
Enveloped (EL)		PES	Ø14 × 1mm tube	27–73% (MT)	80–2240 (PD)	
powder (PD)		PEEK	10 × 50mm U			
Melt (MT)						
Commigled Powder	glass	PP PA12	20 × 2 mm Ø2 mm rods	45%	1000–10000	[20]
Commigled	jute	PLA	23x1.5 tube	43–52%	18	[21]
Melt	glass	PA6	50 × 4 mm 80x4 Omega	45–55%	500–700	[22]
Commigled	flax	PLA	Ø4.76	40%	50–220	[23]
Commigled	glass	PP, PA12, PET PEI, PC	Ø5-Ø40mm	58–65%	0–250	[24]
						[25]

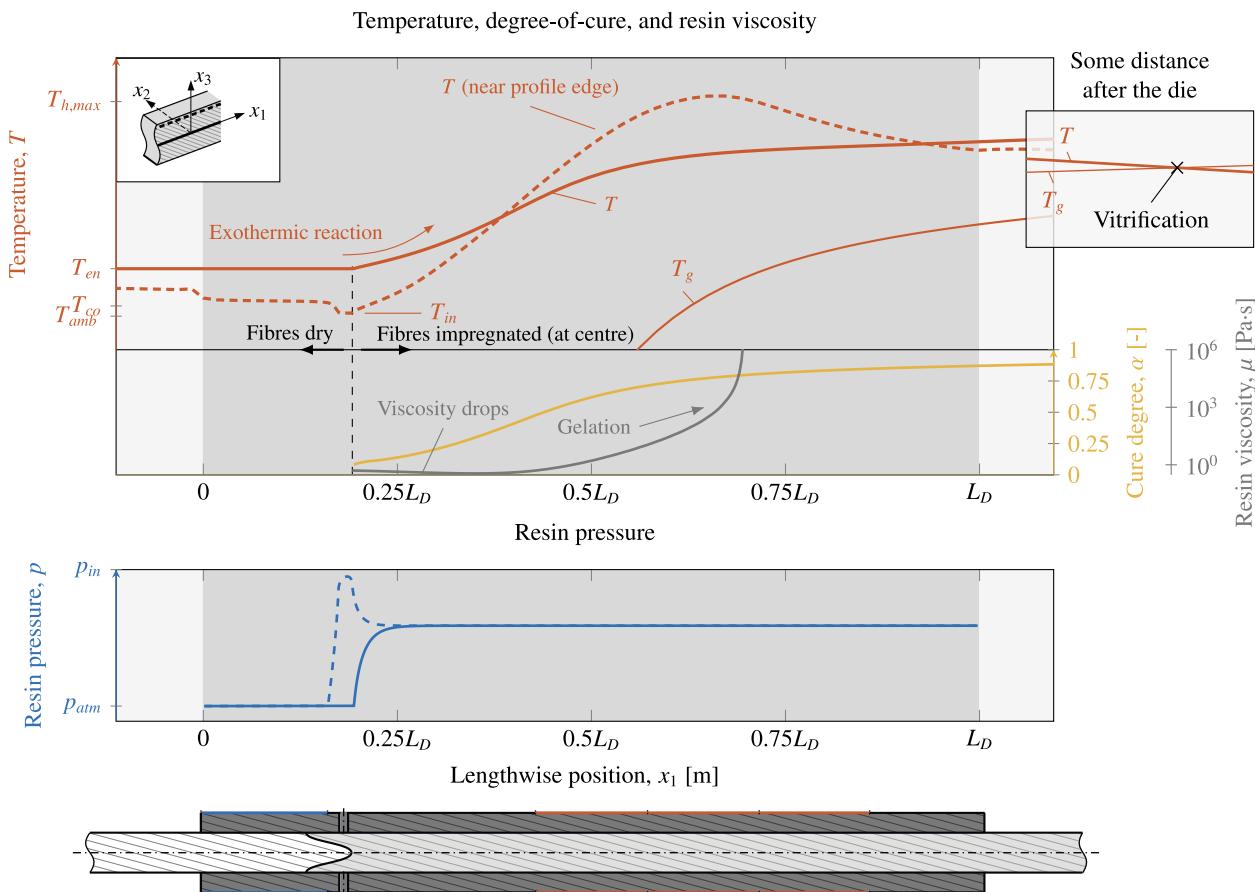


Fig. 5. Typical process diagram for a thermoset resin-injection pultrusion process. In the figure, L_D is the length of the die, and the following subsets apply: “in” = inlet, “atm” = atmospheric, “amb” = ambient, “en” = entrance, “h,max” = max. heater temperature, “co” = cooler. The solid line marks the centre of the profile and the dashed line is near the edge of the profile. This figure was adapted from Sandberg [10].

the mechanical response of a ski-roller while reducing cost and weight [112]. The introduction of a steel sheet into flat and multi-cavity pultruded profiles highlights the benefits of this approach in terms of crash-energy absorption of hybrid metal-composite profiles [113] (an

example of such a hybrid profile can be seen in Fig. 9(b)). For thermoplastic pultrusion, a multi-stage process has been used to successfully combine different fibres, matrix polymers, and metal inserts as shown in Fig. 9(b-d) to manufacture hybrid profiles [109]. These profiles allow

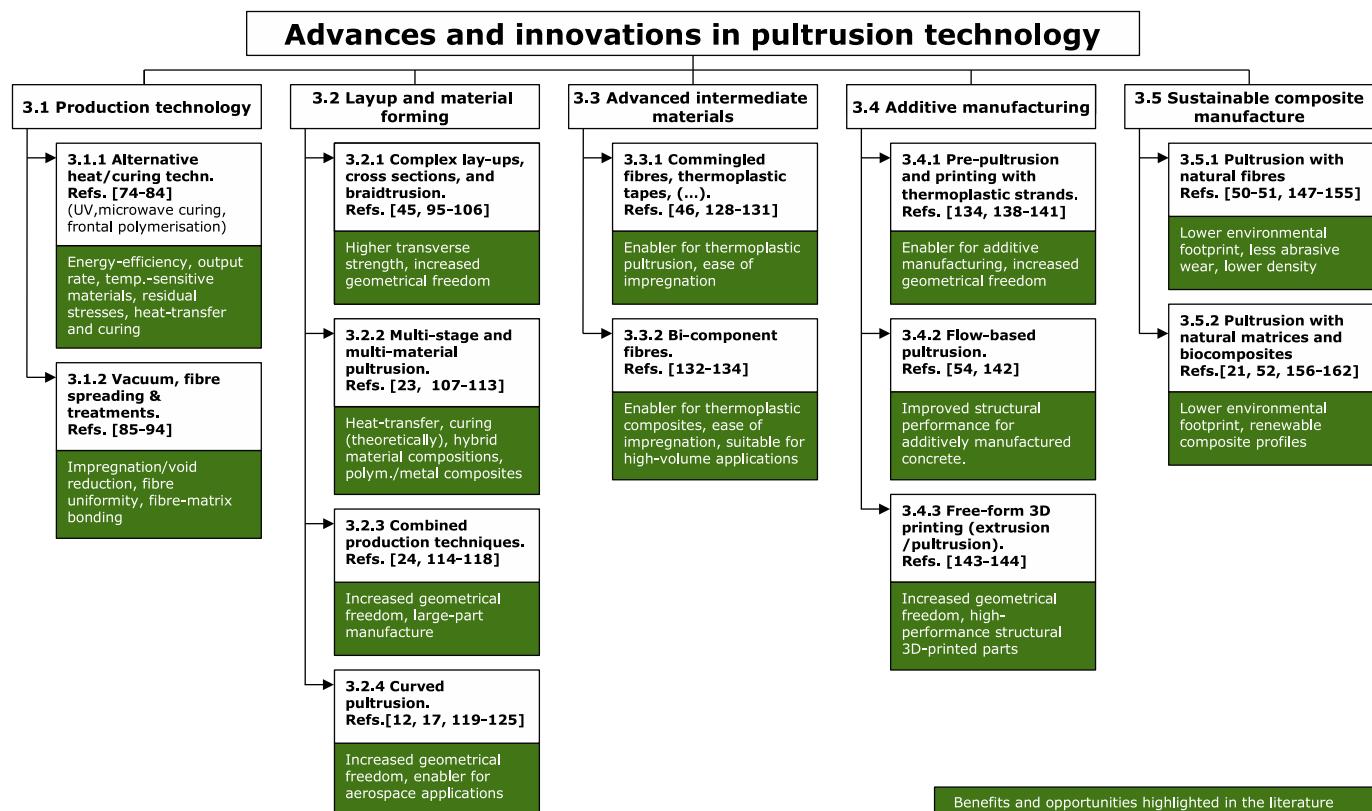


Fig. 6. Outline of the advances and innovations in pultrusion technology that are covered in this paper.

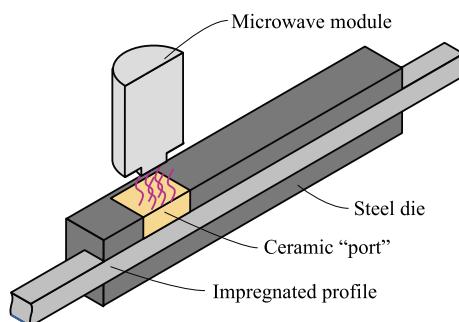


Fig. 7. Illustration of a pultrusion process that utilises microwave technology to enable the curing step of a thermoset resin system. As illustrated, this can be done by integrating a ceramic “port” into the steel die in which microwaves can be transmitted.

engineers to tailor the functionality of the pultruded profile to specific applications, where, e.g., both the electrically insulating properties of glass fibre composites and the electrical conductivity of metals are required. It is important to note that even though multistage thermoset

pultrusion has been envisioned in the patent by Gorthala and Flynn [107], and the theoretical work by Albayati and Gorthala [108], empirical evidence proving the actual feasibility of the concept is currently not available for processes that utilise thermoset materials.

3.2.3. Combined production techniques

By combining pultrusion with traditional manufacturing techniques, one can achieve both the cost efficiency and material quality of pultrudates while enabling the shape flexibility of traditional forming processes. The combination of pull-braiding and blow-moulding, for example, using a hybrid two-step curing thermoset matrix system, allows pultrusion of a fibre-reinforced tube with a B-stage rubbery resin consistency that is then inflated with a bladder and cured in a closed mould (Fig. 10(a–b)). This allows large scale production of high-quality composite parts (500K/year) while reducing the cycle time to 3–4 min and minimising post-processing and material waste [114]. A different strategy consists of using injection moulding to locally deform a pultruded profile, utilising the reformatability and weldability of a thermoplastic matrix. Examples of this include adding out-of-plane components to a pultruded profile such as gears to a pultruded shaft [115] or by partially/completely overmoulding it. By using pultruded polyamide 6 profiles as load-carrying elements that are supported by an

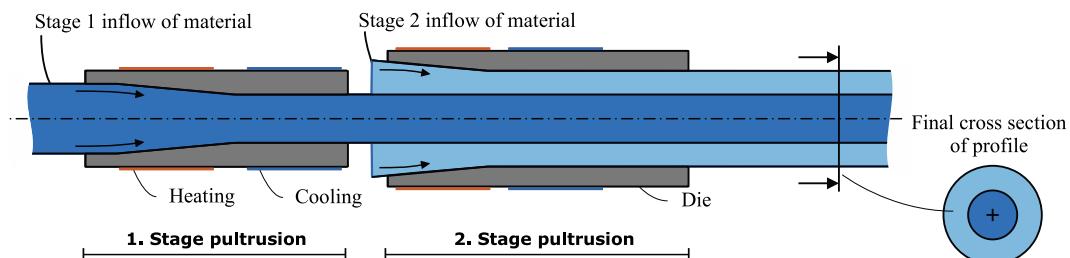


Fig. 8. Multistage pultrusion concept, starting with an initial in the first stage and radial material addition in the second stage.

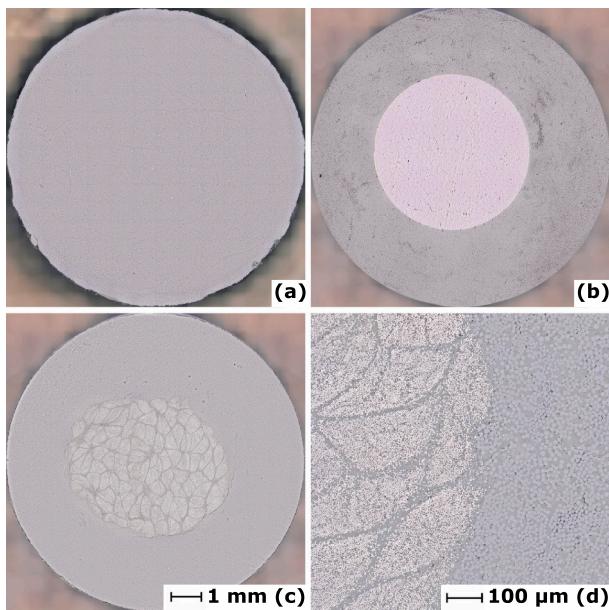


Fig. 9. Micrograph of pultruded rods made from different material combinations in first and second stage: PET/GF (a), aluminium-PET/GF (b), PA12/CF-PET/GF (c, d). This figure was adapted from Volk [109].

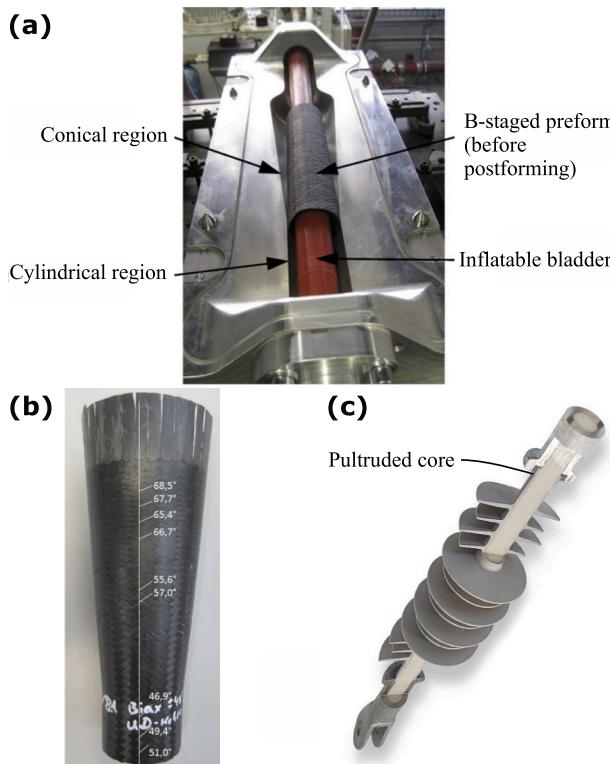


Fig. 10. (a-b) An advanced combined use of two manufacturing processes (pull-braiding and blow-moulding) that enabled new options in terms of shape flexibility (these figures were adapted and reprinted with permission from Bezerra [114]). (c) A high-voltage insulator with cut-out sections. The core of the insulator is a Ø38 mm rod pultruded from commingled thermoplastic and glass fibres (the figure was taken from Volk [109]).

injection-moulded structure with included cable routing and other relevant functionality, a cost-effective solution for an automotive roof section has been successfully brought into mass production in the BMW iX-series of electric cars [116]. A comparison of different approaches to

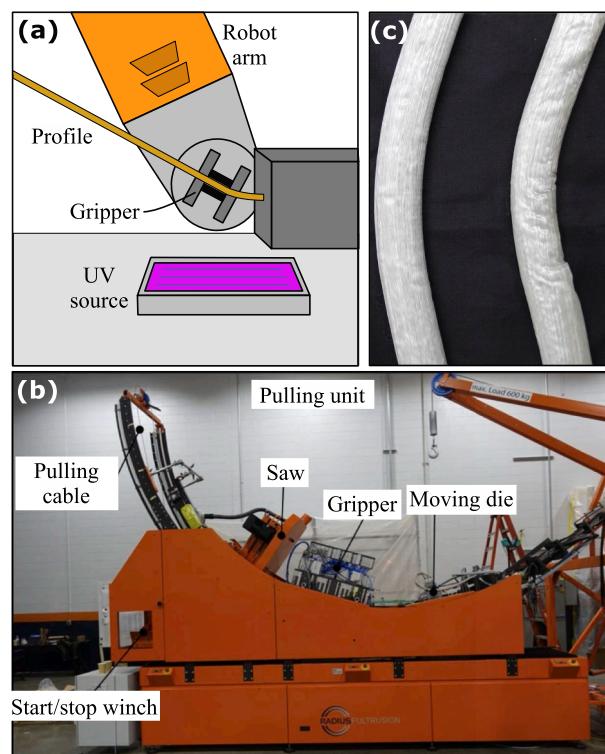


Fig. 11. Three ways to manufacture curved, pultruded profiles: (a) Post-manipulation with a mobile arm and out-of-die UV curing, (b) a mobile die that is moving in a circular motion (this figure was adapted and reprinted with permission of Jansen [120]), and (c) stretch-bending (this figure was adapted and reprinted with permission of Haas and Bose [125]).

combine pultrusion with other post-forming technologies to manufacture thermoplastic composite rebars has been performed by Böhm et al. [117]. Among the studied procedures are contour milling of the surface, form pressing, and helix creation by either reforming the profile or helical winding of an additional tape around the circumference of the rod.

Pultrusion can also be combined with filament winding to increase the diameter of solid pultruded rods to even larger diameters, which can be beneficial for, e.g., high-voltage insulator applications [118] (a cut section of a high-voltage insulator with pultruded a core can be seen in Fig. 10(c)). For cost-efficient pultrusion of smaller series of profiles, the batch-pultrusion approach allows manufacturing profiles of limited length. Conventional continuous pultrusion requires an extensive bobbin creel, yarn guidance system as well as a large material stock, but batch pultrusion circumvents the space requirements and associated equipment and material cost by winding material from a single bobbin into one thick loop that is then fed through the pultrusion die to achieve the required throughput [24].

3.2.4. Curved pultrusion

Pultrusion of curved profiles can be achieved through a post-curing process once the material has exited the die [12,77], for example, by use of out-of-die UV curing [119] (Fig. 11(a)). Alternatively, the profile can be produced through a mobile die that is moving in a circular motion while the profile remains stationary [120] (Fig. 11(b)). This technology enables pultrusion to cover an even wider field of possible applications and has been successfully applied to automotive structures such as bumpers [97,121], sports equipment [112], aerospace components [122], marine [123] and infrastructure applications [124]. An alternative post-forming approach called stretch-bending has been developed for thermoplastic pultrudates which allows for wrinkle-free bending of tubes by selectively pre-tensioning the sections close to the centre of the profile [125] (Fig. 11(c)).

3.3. Advanced intermediate materials for thermoplastic pultrusion

Thermoplastic pultrusion offers numerous advantages in terms of pultrusion speed [20], recyclability, and shelf life while enabling different processing routes based on the weldability and reshaping of thermoplastics [126]. Over the years, the challenge related to the high melt viscosity of thermoplastics impeding efficient impregnation has been addressed through new, innovative approaches. While reactive thermoplastic resins and low viscosity polymers allow a similar impregnation strategy to thermoset pultrusion, combining the polymer matrix and reinforcing fibres into one intermediate material reduces the impregnation length while offering a wide choice of polymer types [127] (Fig. 12). This section reviews different thermoplastic intermediate material types and their processing strategies.

3.3.1. Cmingled fibres, thermoplastic tapes, prepregs, powders, and towpregs

The processing of thermoplastic intermediate materials is achieved by preheating the material in a preheating oven close to the final processing temperature. Then, in the heating die, the polymer melts and impregnates the reinforcing fibres as depicted in Fig. 13(b–c) before being cooled until full consolidation in a cooling die. One of the most popular approaches is commingled yarns, where the reinforcing fibres are intermingled with thermoplastic fibres [128] in individual rovings. A lab-scale setup to achieve these processing steps can be seen in Fig. 13. Other strategies include the use of fully impregnated materials such as thermoplastic tapes or prepregs [129,130] as well as powder impregnated yarns or towpregs (Fig. 12(c)). Here, the neat polymer can be applied inline by use of, e.g., electrostatic powder spray [131] before preheating is applied. A significant challenge related to this technique is to apply a sufficient amount of polymer without losing it in the subsequent processing steps. This can be addressed by introducing a polymer sheath around the reinforcing fibres and the polymer powder [46]. However, the limited drapeability due to the shear resistance of the sheath has prevented the wide adoption of this approach.

3.3.2. Bi-component fibres

To further increase the production output while maintaining a high material quality, bi-component fibres is a novel material technology where each reinforcing fibre is individually clad with thermoplastic polymer (a thermoplastic “sheath”, see Fig. 12(b)). The application of this “coating” occurs directly within filament melt spinning, which makes the manufacturing of such fibres highly efficient, and thus suitable for high-volume applications. The advantage of this new concept lies within the very fast consolidation and low void content of the manufactured material as only extremely short flow lengths (smaller than the diameter of a single fibre) are needed for complete impregnation [132,133]. In terms of pultrusion, initial trials showed that bi-component fibres can readily be processed in setups designed for commingled-fibre pultrusion. In a test case of pultrusion of Ø5 mm thermoplastic rods, bi-component fibres showed a significant effect of lowering the void content [134]. This demonstrates that there is a clear

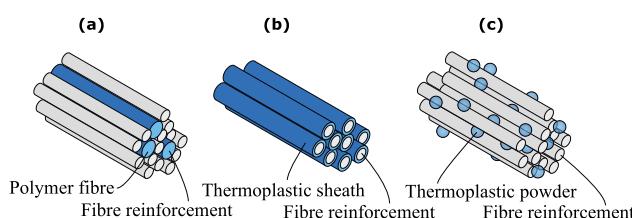


Fig. 12. Overview of some of the strategies that have been used to integrate thermoplastics as an intermediate material in pultrusion processes, (a) commingled fibres, (b) bi-component fibres, and (c) power-impregnated fibres.

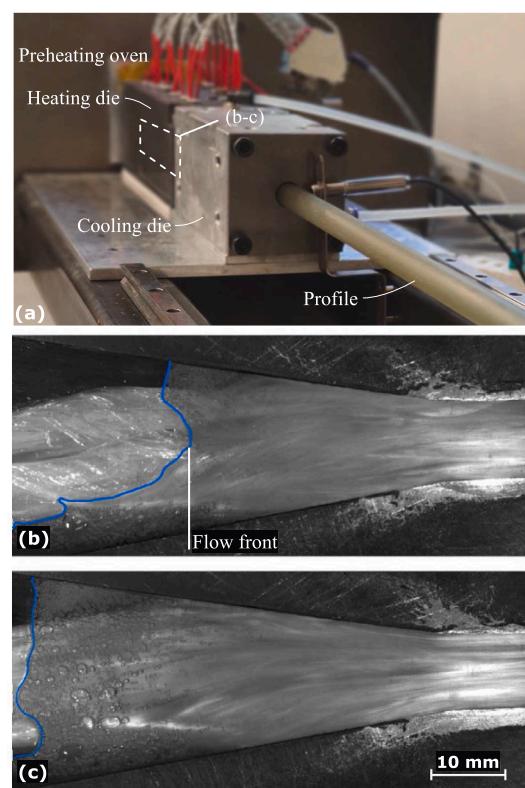


Fig. 13. (a) A lab-scale setup for pultrusion of thermoplastic, commingled fibres. Pictures (b–c) depict the impregnation flow that takes place when the commingled thermoplastic fibres melt. These pictures were adapted from Volk [109] and captured using a transparent die.

potential for bi-component fibres to become a material concept for high-speed pultrusion of low void content profiles.

3.4. Pultrusion as an enabler for additive manufacturing

Three-dimensional (3D) printing or additive manufacturing has gained extraordinary attention in the last 30 years and opened up many opportunities for end-user products [135]. Fibre reinforcement is a promising way to improve 3D printed polymers and other materials by enhancing their mechanical properties [136,137], and here pultrusion has proven as a useful process to enable the implementation in some additive manufacturing technologies.

3.4.1. Pre-pultrusion and printing with thermoplastic strands

The manufacture of fibre-reinforced filaments for 3D printing can be realised by means of melt impregnation of the dry fibres [94] or using intermediate materials such as commingled yarns and tapes [138–140]. The latter has advantages such as better fibre matrix adhesion and a potential of higher fibre volume fraction. In principle, fibre-reinforced filaments need to have a uniform fibre distribution, low void content and low fibre volume fraction to achieve a good printing quality. The effects of variation in the roving/yarn can to some extend be compensated for with smaller strand diameters. Another route is multiple die applications [141] or bi-component fibres [134], but with larger nozzle diameters and high reinforcement content, intra- and inter-strand porosities together with insufficient drapeability at corners can arise.

3.4.2. Flow-based pultrusion

Flow-based pultrusion is a layer-by-layer, extrusion-based additive manufacturing concept (Fig. 14). While it differs from conventional pultrusion, it does lend from the technology as the name implies. Flow-

based pultrusion can be used for 3D printing with continuous fibre reinforcements filaments [54,142], and it is a rather unique additive manufacturing and pultrusion concept since it works without any motorisation to advance the reinforcement fibres. It does, however, require certain rheological properties of the printing material, which can be the reason why the technology has so far only been applied to fresh concrete, a non-Newtonian (viscoplastic) material. For concrete, the rheology develops during extrusion, making the viscosity low enough to permit sufficient fibre impregnation. At the same time, the concrete will be viscous enough in the printing head to pull the fibres and advance the filaments. In Ref. [54], Demont et al. reported successful printing with reinforcement ratios as high as 10%, but even adding very few fibres ($\approx 0.2\%$) was enough to drastically increase the consistency of the printed material and avoid avalanche and slug effects. Nevertheless, it is important to underline that these materials do not have the same fibre composition nor structural properties compared to fibre-reinforced polymer composites.

3.4.3. Free-form 3D printing (extrusion/pultrusion)

In contrast to additive manufacturing techniques that allow for layer-by-layer deposition of material, continuous lattice fabrication (CLF) enables the orientation of continuous reinforcing fibres in all spatial directions. This can be achieved through a two-stage process, where a commingled, stretch-broken filament is first impregnated and consolidated in a pultrusion stage and then fed into an extrusion unit that allows for printing on a polymer substrate as well as altering the shape of the pultrudate in other spatial directions [143] (Fig. 15(a–b)). This technique does not depend on support structures or moulds and therefore enables the creation of free-form structures with a load-tailored design to fully exploit the potential of composite materials. A different approach called pultrusion-winding [144] enables the manufacturing of core-less structures by intermittent ultraviolet pultrusion. It consists of a pultrusion head that can freely move in all spatial directions as well as a winding fixture. For straight sections, the profile is fully cured in the pultrusion head while around winding points the curing can be interrupted so that the yarns remains pliable (Fig. 15(c)).

3.5. Pultrusion as an enabler for sustainable composite manufacture

Our climate calls for urgent action to reduce the risks of future natural disasters [145]. This concerns the associated risks related to the continued accumulation of CO₂ in our atmosphere, but waste and resource management demand immediate changes as well. As discussed, pultrusion is an enabler in the processing of thermoplastic materials (reviewed in Sec. 3.3), which is a step towards new and better recycling routes compared to the use of thermosetting polymers. This is because thermoplastics can be remelted, making it easier for recyclers to recover the polymer and fibres from the composite part after its end-of-life use [146]. Pultrusion is also a material-efficient processing technique as it has no inherent need for disposable items (vacuum bags, flow mats, etc.), making it a step towards zero-waste processing. Nevertheless, composite processing, use, and

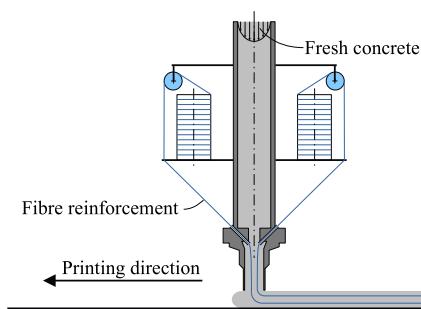


Fig. 14. The concept of flow-based pultrusion for additive manufacture of fibre-reinforced concrete by Demont et al. [54] and Ducoulombier et al. [142].

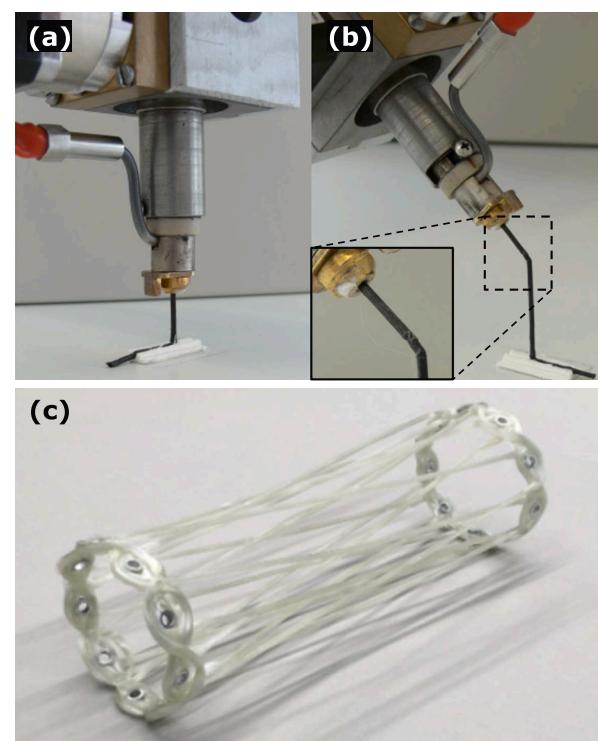


Fig. 15. Additive manufacturing using continuous lattice fabrication (CLF). (a) shows printing of a free-standing structure, and (b) demonstrate the ability to change angle on-the-fly during printing (these figures were adapted and reprinted with permission from Eichenhofer et al. [143]). The printed strand was approximately $\varnothing 1.5$ mm, given by the diameter of the nozzle. (c) demonstrates an example of the design freedom that can be achieved with pultrusion-freeform winding (this figure was reprinted with permission from Mindermann et al. [144]). The printed strand was approximately $\varnothing 3$ mm, given by the diameter of the nozzle).

recycling do leave an environmental footprint since the processing of synthetic fibres and petroleum-based polymers yield a product that has consumed non-renewable materials and is difficult to recycle. But overall, pultrusion technology can allow the industry to move towards more sustainable composite manufacture, also related to utilising renewable materials, which we will review in this section.

3.5.1. Pultrusion with natural fibres

The utilisation of natural fibres in fibre-reinforced polymer composites has several advantages. Natural fibres are biodegradable, have a low density, cause less abrasive wear on equipment, and are safe to handle (e.g., low risks of skin irritation/irritation of airways). Life-cycle analysis studies have confirmed that composites reinforced with natural fibres are environmentally superior to composites with glass fibre reinforcement [147]. With a current annual growth rate of 10.5%, the global market for biocomposites is expected to grow to \$52.72B by 2027 [148], far exceeding the current growth of the fibre-reinforced polymer composite market as a whole (4.5%, [149]).

The most common natural fibres are seed, leaf, and stem fibres [150]. Natural fibres are normally not stable above temperatures exceeding 200 °C, but thermosets, thermoplastics, and natural matrix materials (reviewed in the next subsection) that allow for processing within this temperature range can be used as matrix material [151]. Some thermoplastic polymer matrices, such as PP and PET, are hydrophobic and offer low compatibility with natural fibres, so appropriate surface treatment is needed to obtain a sufficient fibre/matrix interface strength [152]. Natural fibres are staple fibres (of discrete length), but quasi-unidirectional fibres can be processed into a continuous tape by utilising special joining techniques (demonstrated for, e.g., flax and

bamboo [153–155]). Flax yarns commingled with PET/PP fibres has been processed using pultrusion [50,51]. The reported studies concluded that pultrusion technology offered good processability for these flax/thermoplastic composites, but sufficient pre-heating is crucial to ensure a sufficient process window when melting the thermoplastics.

3.5.2. Pultrusion with natural matrices and biocomposites

One way to introduce renewable materials to a composite, is to introduce them as a filler. While fillers will generally decrease the mechanical properties of the final part, they can be relevant for non-structural components such as secondary or tertiary structures, panels, packaging items, etc. [156–158].

Short natural fibres, wood flour, corn starch, natural rubber, and soy [158], for example, can be used as fillers in thermoset and thermoplastic polymers derived from fossil resources such as polyethylene, polypropylene, polyester, and epoxies [159,160]. However, surface treatment is frequently highlighted as a requirement to obtain sufficient interface strength [152,158]. Compared to a conventional, homogeneous polymer, adding a natural filler increases the viscosity [161,162]. Pultrusion is a proven technology to process viscous matrix systems, and the use of conventional oil-based polymers with natural fillers is something that has been explored for pultrusion. For example, Zhu et al. [53] analysed the effects of using 30 wt% of soy resin as matrix content (i.e., a resin extracted from the soybean) to a pultruded glass fibre composite. Here, it was found that the profile maintained its main mechanical properties, and the profile-advancing pulling force was reduced due to the natural lubrication from the soybean.

In a truly “green composite”, the matrix and fibre reinforcement are both made from renewable materials and the final composite is biodegradable. According to La Mantia and Morreale [158] “biodegradable polymers can be classified according to their origin, i.e., into agro-polymers (e.g. starch), microbial-derived (e.g., PHA) and chemically synthesized from agro-based monomers (e.g., PLA), or conventional monomers (e.g. synthetic polyesters)” [158,163]. For pultrusion with PLA, manufacturability has been demonstrated with flax fibres (Linganiso et al. [52]) and jute spun yarns (Memon and Nakai [21]) as reinforcements. In both cases, the matrix material (i.e., PLA) entered as a commingled fibre, which allowed for sufficient impregnation.

The above-mentioned literature about processing natural fibres and matrices in the pultrusion process is not by any means comprehensive. This may appear surprising taking into account the extensive research efforts on the topic for the composite industry as a whole [164]. Also, taking into account that the first successful report of pultrusion with natural fibres is today more than twenty years old [50], and out of the more than twenty natural fibres types that are suitable for processing into a composite [164], the literature so far only looks into the use of flax and jute fibres. With the current and new potential offered by surface modifications to improve the fibre/matrix interface [152], this is an area where pultrusion offers potential as thermoplastic pultrusion has demonstrated that secondary materials can be added continuously inline the process [46].

4. Conclusions and outlook

In this paper, we have reviewed some of the recent advancements in the pultrusion industry, in particular, innovations in production and material forming technologies, new material technologies for thermoplastic pultrusion, and how pultrusion is an enabler for additive manufacturing as well as renewable composite manufacture. With these innovations, we have argued for a promising outlook and potential for further penetration of new markets of pultruded products but also composite materials more broadly. Based on our assessments and discussions throughout the paper, and with our consortium’s experience from completing multiple academic and industrial projects about pultrusion, we identify the following areas where we see potential for further research and industrial application of pultrusion technologies:

4.1. New curing, material, and processing strategies

As reviewed, frontal polymerisation is a curing strategy that is currently finding its way into composite processing. Frontal polymerisation has not yet been utilised in pultrusion processes, but it has the potential of enabling pultrusion with no or limited external heat source. This would reduce costs related to energy released to the ambient surroundings, but also further limit the CAPEX of pultrusion equipment as heater units would become redundant. Since frontal polymerisation techniques utilise rapid-curing resin systems, this curing strategy may also help to increase the production output. To realise frontal polymerisation in pultrusion, new polymerisation kinetics models and resin curing behaviours are needed to be implemented, predicted, and analysed since tight control of the resin cure is required to exploit this strategy. Also, since fast-curing resin systems will introduce steep temperature and cure gradients, the potential impact on process-induced stress and deformation from choosing this cure strategy needs to be fully understood.

Pultruded profiles are traditionally composed of one material composition, but recent studies have investigated the use of different fibres, matrix systems, as well as metals to further improve the cost-efficiency, versatility, and mechanical performance of pultruded profiles. While the initial proof-of-concept results have been promising, a holistic scientific approach is needed for the analyses and syntheses of the different multi-material concepts to understand the challenges and outlook of this technology.

4.2. Advanced lay-up and combined processing technologies

Combining pultrusion with other manufacturing techniques offers the potential of achieving the cost-efficiency and quality of pultrusion, together with the design freedom of other conventional processes. As reviewed in the paper, the geometrical freedom of braiding, blow-moulding, injection moulding, hydro-forming, or tailored fibre placement technology can be exploited together with pultrusion processes. Although mostly proof-of-concept cases make up the literature, the first commercial applications are emerging and are expected to have a major impact on the composite industry. In addition to this, curved pultrusion is a processing concept that has expanded the application area of pultruded profiles from straight geometries to curved geometries. After several academic studies, this technology is now successfully being utilised to manufacture critical structural components for high-performance automobiles, e.g., the BMW iX-series of electric cars. Additional development of curved pultrusion towards variable radii pultrusion including helical geometries will remove further barriers and enable additional application areas of pultrusion.

4.3. Thermoplastic pultrusion

Thermoplastic pultrusion offers numerous advantages in terms of pultrusion speed, recyclability, and shelf life while enabling new manufacturing routes through weldability and reshaping. Until recently, the industrial applications of pultruded thermoplastic materials were limited to tapes, but now large cross-sectional pultruded profiles that were pressed and overmoulded have found application as structural components in mass-produced automobiles.

Another reason for the increased adoption of thermoplastics is their reaction and solvent-free processing that limits chemical exposure to the environment and the technical personnel compared to thermoset plastics. This, together with new and novel material options such as bi-component fibres, sets a strong outlook for thermoplastic pultrusion. On the other hand, for the pultrusion of thick thermoplastic profiles, heat extraction after die exit becomes a major challenge. Here, multi-stage pultrusion, which has already been utilised for both homogeneous and multi-material pultrudates, has potential. However, there have been very limited studies about the governing multi-physical

phenomena, in particular, the development of residual stresses between the different material stages.

4.4. Pultrusion of renewable materials and sustainable composite manufacture

We discussed in the paper that with pultrusion, the industry can take steps towards more sustainable composite manufacture. Nevertheless, fibre-matrix compatibility is a serious issue for the mechanical properties and performance of natural composites in general, but this challenge can be addressed with the use of different compatibilising agents (for example, surface treatments of the reinforcement fibres). The literature about pultrusion has shown that secondary materials and surface agents can readily be applied inline a pultrusion line due to the continuous nature of the process. This technique has been demonstrated possible for thermoplastic pultrusion and expanding this concept to pultrusion of natural material has the potential of lowering the barriers for further adaption of sustainable composite processing.

Credit author statement

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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