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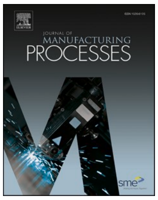

Review

Extrusion-based additive manufacturing technologies: State of the art and future perspectives

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ABSTRACT

Extrusion-based additive manufacturing (AM) has recently become widespread for the layer-by-layer fabrication of three-dimensional prototypes and components even with highly complex shapes. This technology involves extrusion through a nozzle by means of a plunger-, filament- or screw-based mechanism; where necessary, this is preceded by heating of the feedstock material to reduce its viscosity sufficiently to facilitate extrusion. Extrusion-based AM offers greater design freedom, larger building volumes and more cost-efficient production than liquid- and powder-based AM processes. Although this technology was originally developed for polymeric filament materials, it is now increasingly applied to a wide variety of material classes, including metallic, edible and construction materials. This is in part thanks to the recent development of AM-specific feedstock materials (AM materials), in which materials that are not intrinsically suited to extrusion, for example because of high melting points or brittleness, are combined with other, usually polymeric materials that can be more readily extruded. This paper comprehensively and systematically reviews the state of the art in the field of extrusion-based AM, including the techniques applied and the individual challenges and developments in each materials class for which the technology is being developed. The paper includes material- and process-centred suitability analysis of extrusion-based AM, and a comparison of this technology with liquid- and powder-based AM processes. Prospective applications of this technology are also briefly discussed.

1. Introduction

Additive manufacturing (AM) technology, often called 3D printing, is defined by the International Standards Organization/American Society for Testing and Materials Standards (ISO/ASTM 52900:2015) as "the process of joining materials to make parts from 3D model data, usually layer upon layer" [1]. This is in contrast to conventional subtractive manufacturing, whereby material is machined away from a workpiece until the desired shape is obtained [2].

AM technology can be categorised into solid-, liquid- and powder-based processes on the basis of the physical state of the raw material (s) before the AM process commences, i.e. solid (filament, wire or others such as paste (a suspension of granular material), pellets (small and compressed mass of a substance) etc.), liquid or powder. This categorisation has some inconsistency deriving from the use of feedstocks in one physical state in applications developed using a different state. For example, extrusion-based AM processes are regarded as a subcategory of solid-state AM because they were initially developed using polymer filament materials that were heated in the reservoir or heating chamber of the print head to soften or melt them and facilitate extrusion. In most modern extrusion-based AM applications, the feedstock is still filament, wire or other solid materials. However, in some more recently developed extrusion-based AM technologies, the feedstock material is already in a liquid state (e.g. inks) or soft condition (e.g. pastes) before insertion into the AM apparatus. The liquefied material is collected in the reservoir, then pushed out through the nozzle of the print head as a viscous, fluidlike material. The term "fluidlike" refers to the fact that the just-deposited AM material has some characteristics of a fluid for a very short time period, i.e. less than a second, such that it can bond with the previously deposited layer before solidifying.

The first extrusion-based AM process to be developed, "Fused Deposition Modelling (FDM™)", was invented in 1989 by S. Scott Crump [3], the co-founder of Stratasys Ltd. (Rehovot, Israel and Eden Prairie, USA). The FDM™ mechanism uses the extrusion of liquefied filament feedstock through the nozzle of a print head to successively deposit layers and thereby fabricate the desired parts. This invention was patented and commercialised by Stratasys two years later [4]. Until 2012, material options for use in extrusion-based AM were mainly

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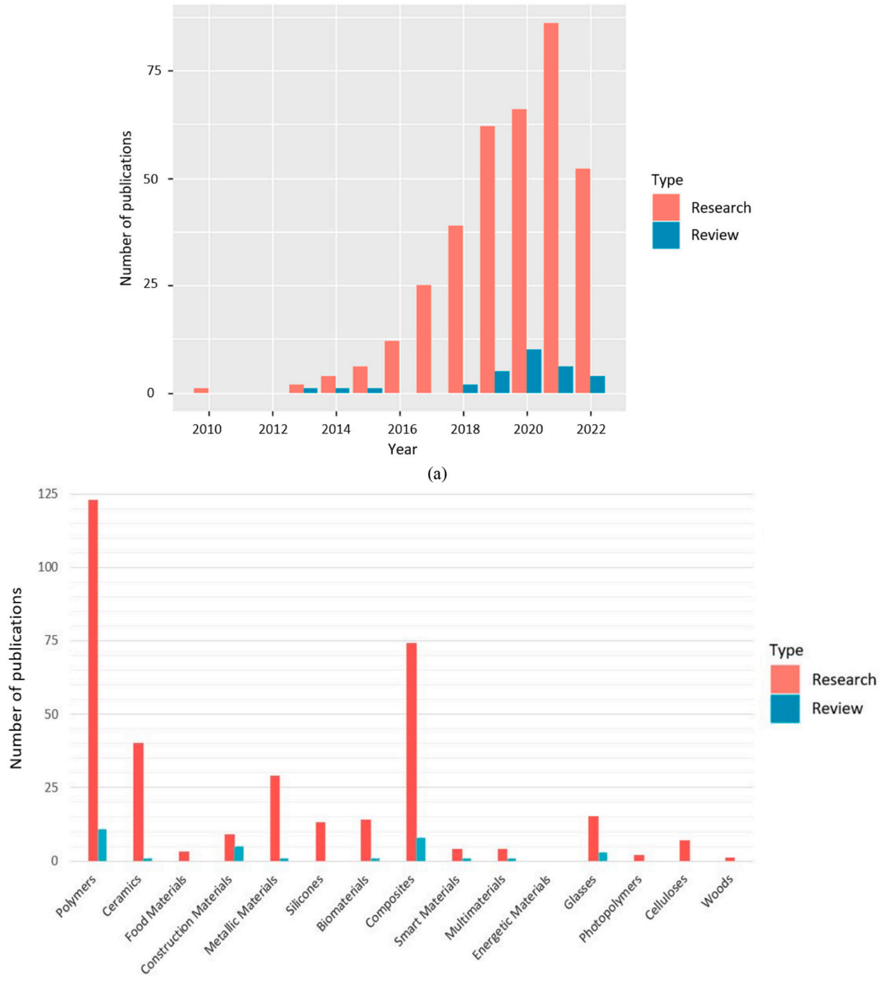
limited to two polymeric materials, namely acrylonitrile butadiene styrene (ABS) and polylactic acid (PLA) [5]; cost, equipment and software limitations precluded the use of other materials in this application.

New developments during the last decade in extrusion-based AM, such as the development of more printing-friendly filament materials, as well as advances in AM equipment enabling higher print head temperatures and faster building, have resulted in a wide expansion in the types of material that can be used as the feedstock for this technology and an improvement in structural integrity of printed parts; this has greatly increased the range of possible applications [2,6]. The main materials now used with this technology are ceramics, food materials, construction materials, metallic materials, elastomers (silicones), polymers, biomaterials, composites, multimaterials, smart materials, energetic materials, glasses, photopolymers, celluloses and woods. Many of these materials, in their conventional form, are not intrinsically printing-friendly; for example, most metals and ceramics have a high melting point and woods would be damaged by exposure to high temperatures. In these cases, AM-specific feedstock materials ("AM materials"), in which the base material is usually mixed with polymers to form composite filaments, are used for extrusion-based AM. This increases the printability of the base material and facilitates or improves the bonding of the successively deposited layers. The technologies of liquefaction/fluidisation and preparation of filaments or feedstock for these materials will be explained in detail in the relevant parts of Section 4.

Fig. 1(a) shows the number of publications on topics relating to extrusion-based AM appearing each year since 2010. The graph was generated by searching the Scopus database for publications whose title, abstract and/or keywords contained either the expression "extrusion-based additive manufacturing" or "extrusion-based AM". The publications have been grouped into "Review" type (i.e. those classified in the Scopus database as "Review", "Conference Review" or "Book chapter") and "Research" type (all other classes in the Scopus database). Since the search was conducted on 18/08/2022, the final pair of bars only represents the publications from just over 60 % of the year 2022. If publications appear at an equal rate throughout the year, a final tally of around 86 research and 6 review publications would be expected for 2022. Particularly for research publications, it is likely that the Covid-19 pandemic and resulting lockdowns will have delayed experimental work and therefore decreased 2020 output below that which would have otherwise been expected, whereas 2021 and 2022 output may represent a catch-up phase. Fig. 1(b) shows the number of publications in the last 5 years, i.e. between 2018 and 2022, on topics relating to all recently available AM materials for extrusion-based AM. This graph was created using the Scopus database search for publications whose title, abstract and/or keywords contained the expression "extrusion-based additive manufacturing of X" or "extrusion-based AM of X", where X represents one of the AM material classes covered in Sections 4.1–4.12. In the same way as in Fig. 1(a), the publications are grouped into "Review" type and "Research" type. It can be seen from Fig. 1(b) that polymers and composites are the most popular material classes for recent publications on



(a)

(b)

Fig. 1. Number of publications on: (a) extrusion-based additive manufacturing per year, and (b) AM materials classes currently available for use in extrusion-based AM (total publications in the last 5 years, i.e. from 2018 to 2022).

Table 1
Selected state-of-the-art papers on extrusion-based AM, and AM materials for use with this technology.

Type of paper	Chief keywords as given in paper	Main research focus or major outcome	Ref.
Comprehensive reviews on extrusion-based AM processes	Fused filament fabrication; metals and ceramics; material extrusion; highly-filled polymers	Various types of material extrusion AM techniques; relevant applications in industry for the fabrication of ceramic and metallic components	[9,13]
	Feedstock extrusion; rapid production; complex-shaped metallic, ceramic parts	Systematic review of all aspects of extrusion-based AM for the fabrication of complex-shaped ceramic and metallic parts	[10,14]
	Fused deposition modelling; polymer-based composites; multifunctional materials systems	Overview of extrusion-based AM processes based on cost efficiency, scalability and material processability	[10,13]
	Extrusion; concrete technology; additive manufacturing; underlying physics	Review of recent relevant knowledge in physics necessary for extrusion-based AM	[14,15]
	3D printing; extrusion-based 3D printing	Novel system of screw-based extrusion AM of thermoplastic materials presented, in which thermoplastic polymer granules were used as the raw material in a plastic processing screw controlled by an open architecture CNC controller	[9,15]
	Extrusion; polymer-matrix composites; polymers; additive manufacturing	Summary of methods to produce polymer and polymer composites for extrusion-based AM; printing conditions of pure polymers, and multi-structure/material filaments	[16]
	Fused filament fabrication; direct ink writing; composite materials; polymer	Introduction of extrusion-based AM techniques; advances in recently developed composite and polymeric materials	[17]
	Polypropylene; 3D-printing; dimensional accuracy; warpage	Recent approaches for the fabrication of polypropylene components by extrusion-based AM; strategies to overcome limitations of dimensional accuracy and printability of polypropylene	[18]
	Construction; building information modelling; additive manufacturing; large scale	Review on systems and AM processes (including extrusion-based AM of construction materials) that are commercially available for use in the construction industry; digital planning techniques for planning 3D-printed building parts	[19]
	Material extrusion; fused particle deposition; additive manufacturing; fused pellet modelling	Comprehensive review on the screw-based extrusion AM process, considering recent technological developments enabling the direct feeding of granulated materials	[20]
	Material extrusion; building construction; energy efficiency; thermal insulation	Review of recent situation in the construction industry; developments in printable AM materials	[21]
Comprehensive reviews on AM materials for extrusion-based AM processes	NA	Summary of state-of-the-art developments and main recent investigations on 3D printing of food, including extrusion-based AM; future perspectives in this field	[22]
	NA	Mini review on the extrusion-based AM, binder jetting and inkjet printing of edible materials, including printability and mechanisms of 3D food printing techniques, as well as some future development directions in this field	[23]
	Customised food; extrusion-based printing; digital gastronomy; personalised nutrition	Comprehensive overview on the 3D printing of food (mainly extrusion-based food printing) focusing on issues such as ingredients, sustainability, legal framework, safety and nutritional values of 3D printed foods	[24]
	Customised food fabrication; 3D food printing; multi-material; platform design	Novel concept designs and workable prototypes to revolutionise the fabrication of customised foods	[25]
	3D food printing; simulation models; material characteristics; quality of 3D printed products	Classification of food materials and their printing characteristics required for extrusion-based AM and other techniques; recently developed technologies for evaluation of 3D printed edible products such as low field nuclear magnetic resonance and novel instrumental analysis techniques	[26]
	Construction industry; additive manufacturing; safety; 3D printing; productivity	Up-to-date review on the recent trends in AM materials and processes, including extrusion-based AM, for use in the construction industry; future research requirements to accelerate the adoption of AM technology in the construction industry	[27]
	NA	Review of recent advances in multi-material AM including extrusion-based AM; recent and potential applications, mainly in the areas of electronics, soft robotics and biomedical engineering	[28]
	Material extrusion; kinematics; machine development; machine frames	Recent trends in extrusion-based AM machines and their potential structure designs, highlighting the strengths and limitations of existing machine designs	[29]
	Fused deposition modelling; sintering; debinding; additive manufacturing	Review of recent state of the achievable quality of parts produced by extrusion-based AM, specifically FDM, considering mechanical properties obtained from the final parts	[30]
	Glass; fused deposition modelling (FDM); direct ink write (DIW); microfluidic; optical devices	Review on the current status, achievements and future perspectives in the development of extrusion-based 3D printing of glass	[31]
	Silicone; material extrusion; soft matter; polysiloxane; additive manufacturing; prosthetics	Review of recently developed AM systems enabling the extrusion-based fabrication of viscous thermosetting silicones, focusing on their roles in biomedical applications; recent developments in the tailoring of silicone, examining their mechanical performance and curing mechanisms	[32]
	Fibre reinforced polymer composite; mechanical properties; 4D printing	Summary of recent developments in the AM (including extrusion-based) of polymer-based fibre-reinforced composites; latest improvements in current commercially available methods	[33]
	Fused deposition modelling; piezoelectric polymers; shape memory composites; shape memory polymers	Wide range of applications (from very simple planar structures to thermo-/hydro-/optical-sensitive lattice components and structures) highlighted, focusing on the properties of components fabricated by FDM using shape memory composites/polymers	[11]
	Smart material; bioprinting; advanced structured materials; shape memory material; 4D printing	Current applications and the classification of responsive and advanced smart materials; models developed to predict their behaviour	[34]
	Additive manufacturing (AM); fused filament fabrication (FFF); polymer nanocomposites	Comprehensive review on high-performance polymer nanocomposites for use in extrusion-based AM	[35]

(*continued on next page*)

Table 1 (*continued*)

Type of paper	Chief keywords as given in paper	Main research focus or major outcome	Ref.
	Bioceramics; metallic biomaterials; biopolymers; additive manufacturing/3D printing; drug delivery	Recent AM techniques (including extrusion-based AM) for fabricating biomaterials; treatment options for these AM materials	[36]
	Bioprinting; polymers; bioinks; 3D printed scaffolds; additive manufacturing; biomaterials; mechanical properties	Summary of recent advances in the extrusion-based AM of polymeric biomaterials, mainly focusing on analysis and improvement of their printability; recommendations and future directions in 3D printing for biomedical; applications of polymeric biomaterials	[37]
	Energetic materials; propellant; additive manufacturing; nanothermite	Recent developments in the characterisation and preparation of energetic AM materials; comparison of AM techniques, including FDM, that can be used for the fabrication of these AM materials, considering binders used and combustion efficiency achieved	[38]
	Reactive materials; direct ink writing; 3D-printing; thermite	Summary of recent applications of reactive AM materials including extrusion-based 3D printed thermite and other reactive AM materials including gun propellants, biocidal agents and thrusters	[39]
	Polypropylene; warpage; 3D-printing; dimensional accuracy	Strategies and methods to make polypropylene easier to print in extrusion-based AM processes; methods of mitigating dimensional issues in polypropylene	[5]
	Multi-material structures; 3D printing; additive manufacturing	Review of wide range of applications of 3D printed metal-metal, metal-ceramic, polymer-based parts, including parts fabricated using extrusion-based AM; discussion on the limitations of the extrusion-based AM of multi-material structures	[40]
Research papers on extrusion-based AM processes	Ionic polymer-metal composites; additive manufacturing; electroactive polymer	Novel FFF technique using electroactive polymer filament material proposed for the fabrication of soft polymer-metal composites	[41]
	Extrusion-based additive manufacturing; silicone elastomer; moisture-cured silicone elastomer	Numerical simulation developed to model and predict the flow rate of silicone during the extrusion-based AM of moisture-cured silicone elastomer; comparison of results of modelling with those obtained from experiment	[42]
	NA	Methodology presented to determine the required process parameters, such as adjacent line spacing, layer thickness, and feed and flow rates, for fabrication of void-free thin-walled moisture-cured silicone structures using extrusion-based AM	[43]
Research papers on AM materials for extrusion-based AM processes	Ceramics; extrusion; functionally graded materials; dynamic mixing	Investigation of feasibility of fabricating functionally graded ceramics produced by ceramic-on-demand extrusion-based AM by controlling the flowrate for ceramic pastes	[44]
	Ceramics; zirconia; material extrusion; mechanical properties	Novel extrusion-based AM process developed to produce 3 mol% Y_2O_3 stabilized zirconia parts with superior mechanical properties e.g. compressive strength and fracture toughness, as well as low porosity	[45]
	Fused filament fabrication; multi-material; polymer; metal; electroforming	Novel AM method combining FFF and electroforming technology proposed for the fabrication of multi-material structures composed of both metal and resin	[46]
	Polyvinyl alcohol; material extrusion; thermal decomposition; three-dimensional printing	Approach for the pellet-fed material extrusion AM of polyvinyl alcohol (PVOH) granules proposed; this does not involve any pre-processing steps but combines extrusion, AM and compounding (i.e. melt blending process for plastics with various additives) enabling multi-material extrusion-based AM	[47]
	Fused deposition modelling; sintering; mechanical properties; microstructure	Advanced extrusion-based AM process called FDMS (fused deposition modelling and sintering) including sintering and catalyst debinding steps proposed for the fabrication of metallic materials	[48]

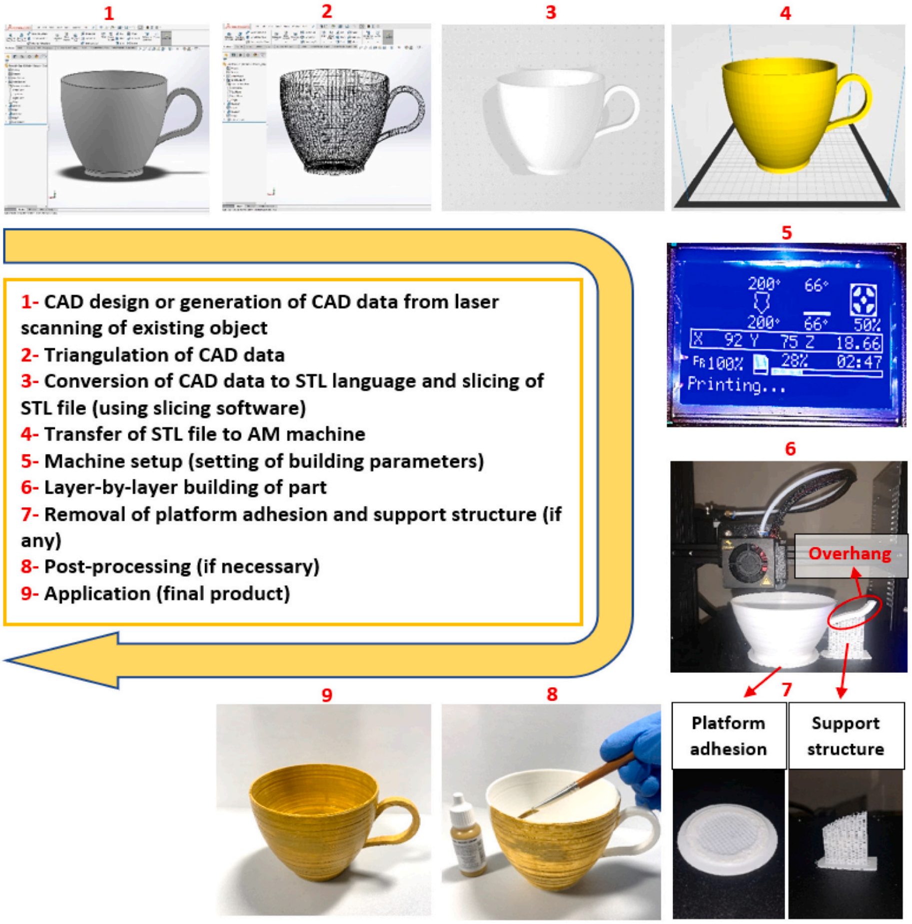
extrusion-based AM.

Despite the recent developments in extrusion-based AM, only a very few reviews [7,8] have so far focused on the developments in material use in AM technology; this is in contrast to powder- and liquid-based AM technologies, which have been reviewed extensively. Reviews focusing on the extrusion-based AM of specific classes of materials, such as metals and ceramics [9,10], polymers and composites [11], and biomaterials and their polymeric composites [12] have been published in the past 3 years; these studies are also discussed in the corresponding subsections of Section 4. There has so far been no comprehensive review covering general aspects of extrusion-based AM, the classes of materials currently used only in extrusion-based AM, and recent developments in these materials. Table 1 summarises selected recent papers focusing on extrusion-based AM process and AM materials used with this technology.

No review has thus far focused on process- and material-centred suitability analysis of extrusion-based AM. Therefore, the primary motivation of the present comprehensive state-of-the-art review is to fill these knowledge gaps, covering all material classes used in conjunction with this technology with particular focus on recent technological developments. Suitability analysis and prospective applications of this technology are covered, and extrusion-based AM is compared with liquid- and powder-based AM technologies. This paper is organised into six sections. In Section 2, the different types of extrusion-based AM process are reviewed; a suitability analysis of these technologies is given in Section 3. Section 4 covers the materials classes currently used in extrusion-based AM. Section 5 reviews new developments in extrusion-based AM, and prospective applications of this technology are considered in Section 6. The main conclusions drawn from the review are given in the final section of the paper.

2. Extrusion-based additive manufacturing

Compared to liquid- and powder-based AM processes, extrusion-based AM is simple to set up, even in domestic environments, and has low equipment and energy costs. As a result, this technology has been adopted by a wide range of users, from hobbyists to large-scale manufacturers in numerous industrial sectors [49]. Fig. 2 shows schematically the steps comprising a generic extrusion-based AM process from CAD design to final product. The first four steps of Fig. 2 are common to most types of AM, while the remainder, i.e. steps 5–8, are specific to extrusion-based AM. In Step 1, a CAD model representing the external geometry of the desired part is generated using: (i) modelling software, (ii) reverse engineering laser scanning techniques such as



1- CAD design or generation of CAD data from laser scanning of existing object
2- Triangulation of CAD data
3- Conversion of CAD data to STL language and slicing of STL file (using slicing software)
4- Transfer of STL file to AM machine
5- Machine setup (setting of building parameters)
6- Layer-by-layer building of part
7- Removal of platform adhesion and support structure (if any)
8- Post-processing (if necessary)
9- Application (final product)

Fig. 2. Generic extrusion-based AM process, from CAD design to final product.

photogrammetry, computed tomography or light-based 3D laser scanning, or (iii) reverse engineering equipment such as the portable Faro Arm or and gantry laser scanning devices, to generate the required 3D CAD model. These CAD models are then converted to the standard triangle language (STL) file format, which is accepted by almost all AM machines, using slicing software (Steps 2 and 3). The STL files are transferred to the AM machine (Step 4), and printing parameters such as layer thickness and printing temperature are set (Step 5); this is followed by the layer-by-layer building of the desired part (Step 6). Once this is complete, platform adhesion and support structures, if present, must be removed from the part (Step 7). In the final step, post-processing may be applied to overcome issues such as anisotropy and undesirably low surface quality. Conventional post-processing methods applied to additively manufactured parts include CNC milling and optical polishing, while non-conventional methods include electroplating, laser surface finishing and ultrasonic abrasion [2,50].

In extrusion-based AM processes, in contrast to liquid- and powder-based AM, a feedstock material is heated until it forms a viscoelastic melt [4,12], which flows in a highly viscous manner as it is forced out through the nozzle of the extruder [2]. The material being extruded can usually be assumed to be a Newtonian fluid, i.e. one whose viscosity is independent of the rate of shear strain, and in which there is a linear relation between shear rate and stress [51]. Most extrusion-based AM processes currently use a single print head, but machines with multiple print heads, enabling multi-material fabrication, are becoming increasingly widespread [50]. Two main approaches to facilitate the bonding of successively printed layers are used in extrusion-based AM processes. The first and more common of these uses temperature to control the state of the material(s) to be extruded; as described above, the feedstock AM material is liquefied and extruded in a softened or molten state to enable flow through the nozzle and deposition onto the building platform or previously printed layer prior to solidification. The alternative approach uses chemical changes to facilitate the solidification and subsequent bonding of successively deposited layers. For example, in the Direct Ink Writing (DIW) process, the extruded AM material (usually ink or paste) is deposited onto a previously deposited layer and curing via chemical bonding begins immediately through interlayer cross-linking; the curing process must be complete by the time the deposited layer is solidified. Chemical curing is achieved using one or a combination of the following techniques: use of a curing agent in the feedstock, reaction of the deposited material with the air, or drying out of the material being deposited [9,24]; in some cases, the curing process is assisted by the application of ultraviolet (UV) light [52] or microwave radiation [53] to increase the curing rate. This chemical-based curing approach is, however, limited to certain classes of soft AM materials, primarily silicones (silicone elastomers and UV- and moisture-curable silicones), and thermoplastic elastomers (TPE). A detailed discussion of how the process is applied to these materials is given in Section 4.6.

2.1. *Classification of extrusion-based additive manufacturing processes*

Extrusion-based AM processes can be classified according to the mechanism [10], i.e. filament, plunger or syringe, or screw, used in extrusion [15]; these three types are shown schematically in Fig. 3. All of these extrusion-based AM processes comprise aspects that differentiate this category of AM from liquid- and powder-based processes, namely [24]:

- Material loading: A continuous feed (filament, wire, pellet or paste) generates a constant input pressure at the nozzle through which the

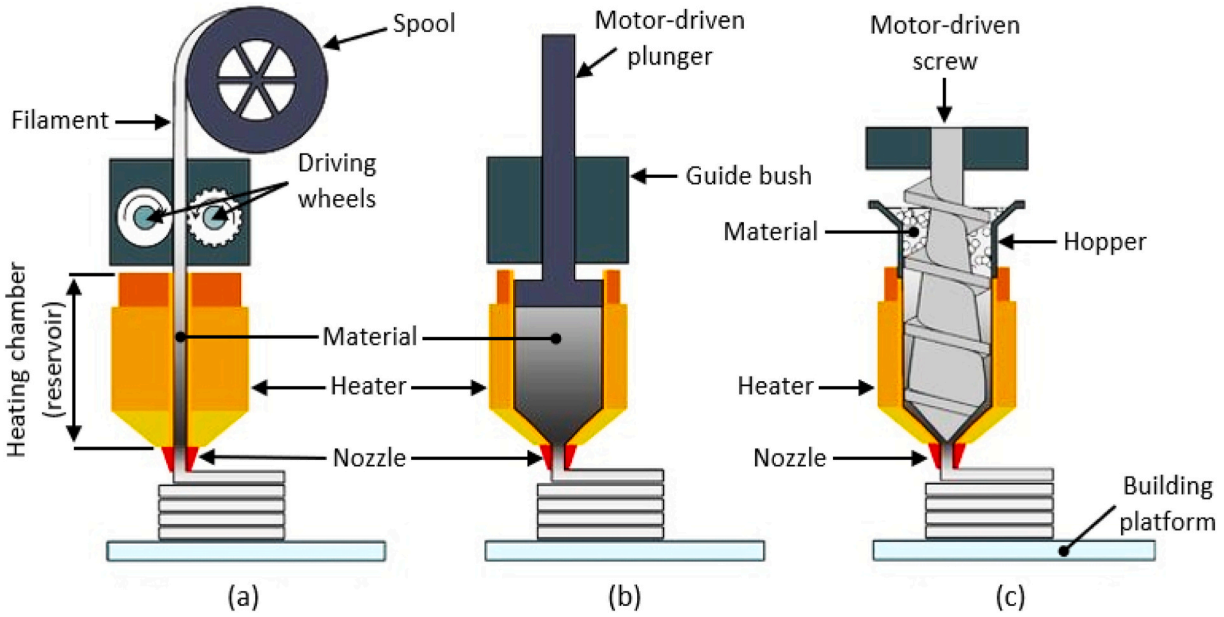


Fig. 3. Extrusion-based AM processes classified according to the extrusion method used: (a) filament-based, (b) plunger- or syringe-based, and (c) screw-based extrusion (adapted from [9]).

molten material is extruded for deposition onto previous material layers on a building platform (Fig. 3(c)). This is in contrast to liquid- and powder-based processes, where parts are built in a resin tank or powder bed that is progressively lowered to allow the loading of new powder or liquid on top of the previously deposited layer.

- Material liquefaction: The molten material is located in a container within the print head; this container is usually called the reservoir in filament-based extrusion mechanisms (Fig. 3(a)) and the heating chamber in plunger-based mechanisms (Fig. 3(b)). Heat is applied by a continuous heat source, most popularly heating coils, located around the reservoir.
- Application of pressure to force the material through the nozzle: In printing-friendly materials, i.e. those able to be melted and deposited at relatively low temperatures and pressures such as certain polymers, sufficient pressure may even be provided by the drive wheels (Fig. 3(a)) facilitating the continuous feed of feedstock material into the reservoir. In other cases, a compressed gas, syringe, plunger or screw is required to achieve the force required for deposition.
- Extrusion of the liquefied material: The size of the extrusion nozzle strongly influences the quality of printed parts; a wider nozzle gives a higher deposition rate and lower fabrication time but a reduction in dimensional precision of the printed product.
- Computer-controlled material deposition according to a pre-programmed print head path codified and stored by the slicing software in an STL file.
- Solidification and bonding of the layers to form a solid dense structure: Ideally, the shape and dimensions of the printed part should exactly match its CAD data (i.e. perfect dimensional accuracy). However, surface tension and gravity can in some cases lead to changes in the dimensions of extruded materials due to cooling and drying effects. Depending on the coefficient of thermal expansion, the material being extruded may shrink and exhibit porosity upon solidification. Shrinkage can be mitigated by reducing the cooling rate or decreasing the temperature difference between the print head and the just-printed layers.
- Involvement of support structures to facilitate the fabrication of complex shapes: AM technologies, including extrusion-based AM, necessitate a means to support unconnected and free-standing features of parts being fabricated. The fabrication of any components having parts with an angle of >45° to the building platform, called overhangs, requires vertically positioned support structures (see Steps 6 and 7 in Fig. 2 for overhang and support structures respectively).

Table 2
Comparison of extrusion-based AM processes.

Extrusion mechanism	Extrusion-based AM process	Largest building volume (X, Y, Z) (mm)			Typical resolution range (μm)
Filament-based	Fused deposition modelling (FDM)	914	610 [54]	914	250–330 [55]
	Composite filament fabrication (CFF)	320	132 [56]	154	200 [57]
	Fused filament fabrication (FFF)	1100	850 [58]	850	100–200 [18]
	Fused granular fabrication (FGF) (pellet printing)	910	910 [59]	1220	1000–2000 [59]
	Fused layer modelling (FLM)	246	163 [60]	155	2.5–**11** [60]
	Fused deposition of ceramics (FDC)	254	254 [61]	254	400 [61]
Plunger- or syringe-based	Robocasting	252	200 [62]	200	100–450 [10]
	Direct ink writing (DIW)	500	300 [63]	100	100–1200 [64]
	Fused filament fabrication (FFF)	250	210 [65]	210	50–350 [65]
Screw-based	Melt extrusion manufacturing (MEM)	255	205 [4]	205	200–500 [4]
	Fused deposition modelling (FDM)	400	400 [66]	300	100 [66]

A comparison of widely used extrusion-based AM processes, in terms of material deposition methods, largest building volumes and typical resolution ranges attainable, is given in Table 2.

2.2. *Process mechanisms in extrusion-based additive manufacturing*

2.2.1. *Filament-based material extrusion*

In filament-based methods, the feedstock material is in the form of a spool of thin filament that is fed into the heating chamber using drive wheels. The FDM process was the first extrusion-based AM process to be developed and is still the most widely used, followed by fused filament fabrication (FFF). The main difference between FDM and FFF is that FDM is conducted in an enclosed, thermally insulated chamber maintained at a constant processing temperature. This gives better mechanical properties in the printed material and enables better layer adhesion (higher bonding strength) but limits the maximum dimensions of parts that can be printed. In the newer FFF technology, by contrast [9], the process is instead carried out in the open air. As a result, materials experience varying temperatures during their thermomechanical cycle, which can result in undesirable residual stresses and worse layer adhesion than is achievable using FDM [67]. To counter this, the building platform is preheated in most cases to reduce the temperature difference between it and the first layer of molten material being deposited. The heating temperature is set before material deposition is started, and heating continues at this temperature until the part is completed. This gives improved adhesion between the first printed layer and the building platform, which is essential to achieve good structural integrity and dimensional accuracy. FFF has become the most widely used simple material extrusion process overall, accessible to hobbyists as well as industry, thanks to the availability of filament materials, the low cost of the process and the lack of limitations on build size [68].

With the exception of a few newly developed FFF technologies, such as syringe-based FFF machines designed for photopolymers [65], most FFF machines can be categorised as ram extruders in which the ram pushes the molten material through the print head. In this case, the filament is forced by means of driving wheels into the reservoir, then the molten material is forced through the nozzle and deposited. The ram provides sufficient pressure for both processes. However, other requirements must be met for deposition to be possible, namely that the motors (Fig. 3(a)) providing the required force to push the rams must generate sufficient torque, and the wheels must have sufficient friction with the filament, to facilitate the transfer of force from the wheels to the filament. This force must then be transferred to the centre of the reservoir along with the melt flow despite the compression and buckling of the filament as it passes between the drive wheels. In addition, the filament must be sufficiently strong to prevent any shearing resulting from pinching at the wheels, but flexible enough to be spooled such that it can be easily fed continuously into the reservoir [69,70]. Only a limited number of materials, mostly thermoplastic-based, fulfil these requirements [9]. The most widely used filament materials for FFF are PLA and ABS, but other commercial thermoplastics are also used with this technology [70–72]; these are discussed in Section 4.1.

2.2.2. Plunger- or syringe-based material extrusion

In this type of extrusion-based AM, instead of the drive wheels used in filament-based material extrusion, a motor-driven plunger or syringe (Fig. 3(b)) is employed to facilitate material flow. This technique is used with printing-friendly feedstocks such as intrinsically fluidlike materials (e.g. pastes) and AM materials with low melting points. The plunger or syringe both forces the feedstock material into the heating chamber of the print head and forces the fluidlike material out through the nozzle for deposition onto the building platform or previously printed layer [9,73]. The process of printing flow begins as soon as the material is passed through the guide bush [30]. It is not necessary in this process for the material to be fully melted; the high pressures permit extrusion at temperatures below its melting point. As a result, this method enables the deposition of AM materials even at low process temperatures if the feedstock used has a relatively low melting point, as does, for example, PLA. Some of the most widely used plunger-based extrusion AM processes given in Table 2 are robocasting and DIW [74], as well as recently developed or modified FFF technologies [65].

2.2.3. Screw-based material extrusion

The stringent requirements for filament materials, discussed in Section 2.2.1, have encouraged manufacturers to use an alternative approach known as screw-based extrusion AM, in which pellets, normally approximately spherical in shape, can instead be used [9]. A screw extruder comprises several zones: in the conveying zone (hopper in Fig. 3(c)), pellets are transferred to a melting zone in which they are softened (without melting) via applied friction and heat. Then, in the metering (pumping) zone, the softened material is forced out and deposited by being subjected to a high pressure using a rotating screw [66,75]. In screw-based extrusion AM, the control of material flow can be a more challenging issue than in plunger- and filament-based methods. As a result, design modifications may be necessary in order to achieve a uniform material flow by ensuring that the pellet material is of a consistent size [76]. Some examples of design modifications in screw-based extrusion AM to enhance the material flow are shown in Fig. 4. Firstly, Kumar et al. [77] modified the design of the screw-based extrusion equipment by using a drill bit combined with the spindle head of a CNC-controlled milling machine (Fig. 4(a)). In another study, Zhou et al. [47] used a simple angular screw but four feeding ports evenly located at different heights to enable uniform material flow from multiple material feeds (Fig. 4(b)). Leng et al. [78] modified the screw-based extrusion AM process by designing a screw of conical shape to improve material homogenisation resulting in a more uniform material flow. Screw-based material deposition has also recently been integrated into other types of extrusion-based AM machines. For example, He et al. [66] integrated screw-based material deposition into an FDM machine for the manufacturing of dense zirconia ceramics (Table 2).

2.3. Control mechanisms in extrusion-based additive manufacturing

Control of extrusion-based AM processes is essential to ensure that printed parts meet design specifications and customer requirements. Control mechanisms in extrusion-based AM process can be associated with feedback-based control systems where parts of the equipment are instrumented to measure quantities such as extrusion velocity, flow rate, thickness of layers and extrusion force. These results are fed back into the system in real time to adjust machine parameters during material deposition. Recent developments in the control mechanisms of extrusion-based AM can be broadly categorised into: (i) control of flow rate of softened or molten material through the nozzle, (ii) control of the force applied to the feedstock material to force it out through the nozzle, (iii) control of the velocity of the motor-driven plunger and screw to provide a constant extrusion flow rate, (iv) model-based control systems, and (v) other recently developed control methods using digital technologies.

Control systems have been widely applied in screw-based material extrusion and freeze-form extrusion fabrication (FEF), particularly in the construction industry. For instance, Valkenaers et al. [15] presented a novel system in which screw-based extrusion AM could be controlled by an open architecture CNC controller. The print head in the proposed system was mounted onto an analogue coordinate measuring machine (CMM) retrofitted with modern control system modules, namely a real-time, field-programmable gate array (FPGA)-based CNC controller, to give more precise control over the print head position. This resulted in better dimensional resolution, i.e. a few μm, in the final printed parts.

Massachusetts Institute of Technology [19,79] developed a digital construction platform (DCP) to control the position and motion of an extrusion nozzle directed by a 6-axis KUKA robotic arm, mounted upon a movable platform with several track rollers. This enabled the printing of a polymeric dome of 14.6 m diameter and 3.7 m height for the construction industry in around half a day. A DCP v.2 platform was subsequently designed to include a 5-axis Altec AT40GW movable hydraulic arm with a 6-axis KUKA robotic arm located at its end point. Both these parts are controlled by micro-macro manipulator robot architecture. The DCP v.2 gave improved system control with better real-time feedback, as well as enhanced system mobility thanks to a digitally controlled tracked base. Recent research and development on the DCP v.2 platform

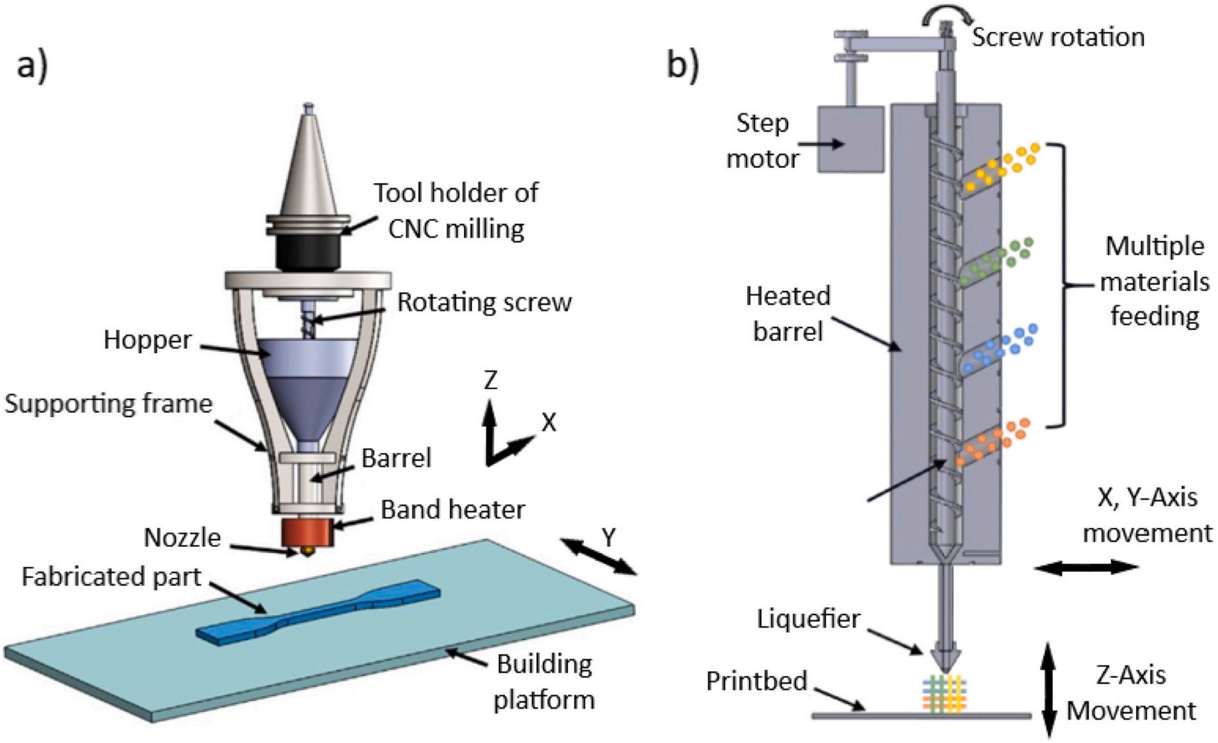


Fig. 4. Some examples of modifications to the screw-based extrusion AM process: (a) drill bit combined with spindle head [77], and (b) angular screw with four feeding ports [47].

has focused on further improvements in the system control [80]. Barjuei et al. [81] have also proposed a control system for the extrusion-based deposition of concrete, which includes a real-time vision-based control framework. This feedback allowed control of layer thickness settings via automatic adjustment of the motion speed of the manipulators during the extrusion process.

Recent approaches proposed to control the extrusion flow rate during extrusion-based material deposition include that of Yuan et al. [82] who presented a novel variable-width extrusion-based material deposition method adapted to a RTPEC for screw-based concrete 3D printing. This work used a Hall Effect Sensor connected to the motor to collect real-time speed data. The constant of proportionality between the pump speed and flow rate was calibrated by experiment. The real-time flow rate control algorithm used in the RTPEC method enabled improved surface quality and horizontal dimensional accuracy, as determined by 3D scanning. Another study [83] has proposed a simplified approach to collect flow rate and gantry speed data during the large scale 3D printing of geopolymeric cementitious-based AM materials (i.e. inorganic polymers able to form solid ceramic-like materials at room temperature) with varying flowability. The proposed extruder enabled control of the compressive stress on the cementitious feedstock. This allowed control of the densification of the printed structure as a function of the flowability of the feedstock. Park et al. [84] designed a nozzle system enabling material deposition at a constant speed using a PID-controlled architectural 3D printer simulator. Similarly, Kazemian et al. [85] developed a strategy in which feedback collected from a vision system was used to control the flow rate during the 3D printing of concrete. However, the main challenge in systems in which the extrusion flow rate is controlled is that in some applications, feedstock material needs to be continuously delivered to the print head. Any adjustment in the time taken to deliver a specific amount of feedstock to the print heads, namely timed dosing, makes the control of feedstock in extrusion-based 3D printing applications very challenging [81]. Due to the effect on printing performance of time delays in the control systems deriving from the nonlinear extrusion dynamics, several studies have proposed feedstock control involving adjustments in the force applied to the feedstock to push it through the system, and velocity of extrusion, i.e. controlling of the force applied to motor-driven plunger and screws to provide a constant extrusion flow rate of feedstocks though the nozzles of print heads.

Strategies focusing on the adjustment of the force applied to the extruders to enable the control of material deposition and extrusion velocity include hybrid extrusion force-velocity control [86], closed loop control [87] and adaptive extrusion force control [86–88]. Deuser et al. [86] have developed an intelligent control methodology for use in the FEF process in which a hybrid extrusion force-velocity controller is used to keep the extrusion force constant during the fabrication. In this study, controllers were integrated with the hybridised control (extrusion force with extrusion velocity) of extrusion-on-demand, enabling fine control of the extrusion force and leading to air bubble release compensation which significantly decreased the severity of gap defects on and under the surfaces of printed parts. The feasibility of this hybrid extrusion force-velocity control system has been demonstrated for both functionally graded and monolithic parts. To control feedstock flow and enable the production of parts with high dimensional accuracy, Greeff and Schilling [87] measured the filament slippage caused by the inadequate gripping of feed gears on the filament feedstock, finding that the implementation of closed-loop control decreased slippage during the FFF of PLA, particularly at lower speed. Zhao et al. [88] developed an adaptive controller to control the extrusion force applied during the FEF of ceramic pastes. They developed a model to describe the extrusion force and used recursive least squares in order to determine the model parameters of extrusion force as a function of the force applied by the driving wheels to the filament feedstocks to push them into the print head. Although these methods improved the dimensional accuracy of printed products by controlling the force applied to the extruders and the extrusion velocity, they were not able to give accurate material deposition throughout the material extrusion. The problem associated with the inaccuracy in material deposition is that these aforementioned developments in the controlling of extruder force indirectly affects the extrusion velocity of extruders, and extrusion force.

As a result of the difficulties involved in material deposition systems with control of extrusion force or velocity, model-based control systems have been proposed to generate a synchronization framework to decrease deposition errors arising from incompatibilities between the extruder velocity and extrusion flow rate, and to maintain the thickness of deposited layers constant. Examples of model-based control schemes include that of Wu et al. [89] who presented two different feedforward control approaches to decrease the number of printing defects (e.g. missing layers, holes and dots on the surfaces, and gaps). These defects resulted from poor synchronization between the extrusion control and motion (positioning) control. The first approach implemented a standard linear extrusion model for FFF applied to the feedforward position control instead of the extrusion control, which resulted in the reduction of some nonlinearities. In the second approach, an empirical nonlinear extrusion model was adopted to design a controller for nonlinear feedforward extrusion. Both approaches improved the dimensional accuracy of printed parts (closer conformity to CAD models) by around 40 % by decreasing extrusion defects. In another study, Zomorodi and Landers [90] suggested a hierarchical control structure integrated with an explicit model predictive controller in both lower (nozzle geometry and paste material) and higher (tracking of extrusion force and ram velocity) levels. The combined control of ram velocity and extrusion force led to more consistent material extrusion. However, the main drawback with these model-based control systems is their limited efficiency; although they can decrease the errors occurring during extrusion-based AM, these control systems are highly dependent on the accuracy and reliability of the models built into the controller structure, i.e. the precision with which parameters such as extrusion force and velocity can be predicted [91].

2.4. Application of digital technologies to extrusion-based additive manufacturing

Digital technologies have been applied to, or combined with, extrusion-based AM to improve the suitability of certain AM materials for extrusion-based fabrication. Examples of both mature and recently developed digital technologies are given in Table 3. These can be broadly generalised into: (i) machine-level, (ii) process-level and (iii) factory-level technologies. Most of the studies currently available in the literature focus on process- and factory-level digital technologies, while few studies focus on the machine level. In detail, digital technologies on the machine level concern operation conditions of the extrusion AM machine, such as process temperature and deposition volume and time. These technologies are mostly applied to control and monitor the build process [92]. Process-level digital technologies normally address the surface conditions and thermal properties during fabrication, and are applied on the micro-scale to control quantities such as the number of layers and the scanning paths [93]. These approaches are mostly used to monitor process performance and diagnose failures in the printed parts. Factory-level digital technologies mainly focus on modelling the whole production system; this typically consists of a process chain involving at least one peripheral (e.g. robotic arms) in addition to an extrusion-based AM machine [94]. These technologies are therefore often used for the control of production tasks involving AM in a production factory, where final consumer products are produced, or a network factory, where components are produced before being sent on for further processing, assembly, etc [95].

Giachini et al. [96] have proposed an approach combining digital processing (fabrication path planning to combine with digital workflow to embed information into design models) and materials engineering to enable the extrusion-based multimaterial AM of tuneable viscoelastic celluloses with multidirectional stiffness gradients. In this work, a combined functionally graded materials (FGMs) approach used mass transport processes together with constructive processes to create continuous gradients. A digital workflow was also created to embed information about these gradients into the design model, in conjunction with fabrication path planning integrated by generating custom G-codes (i.e. codes that control AM machines by instructing them where and how fast to move, and which path to follow) to control the position of the syringe pump and extruder simultaneously. This work demonstrated that using a combination of digital tools and materials engineering,

Table 3
Selected applications of digital technologies to extrusion-based AM processes.

Extrusion-based AM machine	Main focus of research	Digital technology	Focus level	Function of digital technology	Ref.
FDM(Ultimaker 3)	Digital twin development using augmented reality to determine electricity consumption, greenhouse gas emission and cost of extrusion-based AM process	Augmented reality	Machine level	Process monitoring	[95]
FFF (TEVO Tarantula)	Development of an approach involving digital processing and materials engineering to facilitate the extrusion-based AM of tuneable viscoelastic celluloses with continuous multidirectional stiffness gradients	Digital processing and digital workflow	Process level	Position controlling	[96]
FDM (Standard)	Securing the planned production tasks using blockchain technology to improve the security and traceability of AM supply chain	Blockchain, cyber-physical production system	Factory level	Process monitoring	[94]
FDM (Ultimaker original desktop)	Development of an interface using augmented reality to facilitate control and monitoring of extrusion-based AM machine	Augmented reality	Machine level	Process control and monitoring	[92]
FFF (Standard low-cost)	Development of a model for in-situ failure inspection of extrusion-based printed layers using optical sensors and computer vision techniques	Optical sensors, in-process monitoring and computer vision	Process level	Process monitoring and diagnosis	[93]
FDM (JG Z603S)	Runtime mechanism and configuration methodology development for the extrusion-based printer and peripherals of a desktop style AM system	Web-based virtual reality	Factory level	Process monitoring	[97]
FDM (Standard)	modelling of energy consumption in FDM of isovolumetric mechanical parts by means of twelve machine learning algorithms	Machine learning	Machine level	Energy consumption prediction	[99]
FFF (Large custom cartesian printer)	Enabling the real-time monitoring of extrusion-based printing process by means of an advanced interface allowing users to remotely control the printing process	Cyber–physical system and digital twin	Process level	Monitoring and quality assessment	[103]
FDM (Makerbot Replicator 2X)	Development in the algorithmic applications of error detection in extrusion-based AM	Machine vision	Machine level	Error detection	[102]
FDM (Stratasys Dimension 1200es)	Creation of cloud-based intelligent production planning system for use in FDM	Cloud technology	Factory level	Production planning	[101]
DIW (Creality Ender 5)	Development in the optimisation of material deposition and calibration of composite multimaterial extrusion-based AM	Deep learning and computer vision	Process level	Process calibration	[100]
FFF (Standard screw-based)	Control of uniformity of printing strand width during material-based extrusion AM	Machine vision	Process level	Process control	[104]

FGMs with continuous stiffness gradients could be obtained in multiple different ways. Yi et al. [95] proposed an approach in which augmented reality (AR) was adopted at the machine level to develop a novel digital twin for use with extrusion AM printers. In this study, small cylinders were used to roughly approximate the geometry of printed components. This approach, named Volume Approximation by Cumulated Cylinders (VACCY), was used to generate a digital representation of the fabrication process. Four process indicators, i.e. manufacturing cost, greenhouse gas emission, primary energy consumption and electricity consumption, were modelled to assess the environmental and economic performance, and were integrated the proposed AR-based digital twin. The performance of the proposed digital twin was determined using results obtained from tests on the digital twin.

Web technologies have recently been increasingly used in extrusion-based AM, most commonly for the remote control of cyber-physical production systems (CPPS, i.e. subsystems integrated with one another) and autonomous cooperation between extrusion-based printed components across all production stages starting from production using machines and ending with logistic networks. Liu et al. [97] proposed a systematic framework for the runtime of digital twin-based CPPS components incorporating existing techniques such as event-driven distributed control and digital twin modelling, to assist developers to implement their own remote control and building environments for digital twin-based CPPS components.

Prediction of energy consumption in extrusion-based AM processes is important, and many modelling strategies, such as interpretive structural modelling [98], have been developed to tackle this. Machine learning technology has also begun to be used recently in extrusion-based AM. For instance, in [99], twelve different machine learning algorithms were used to predict the energy consumption during FDM. The most effective of these models was determined using four performance criteria: explained variance score, R-squared, root mean squared error and mean absolute error. The model is now available to enable users to predict the settings of printing conditions to minimise energy consumption in any extrusion-based AM process.

Moretti et al. [93] proposed a method for in-process monitoring of component geometries in FDM, using a digital twin for the automatic planning of inspection processes to develop solutions for ultra-high resolution sampling in an extensive range. They demonstrated the potential of using in-process monitoring for computer vision systems using the extrusion-based fabrication of acceptable instances of the previously defined test geometries of components. Blockchain technology can be integrated into extrusion-based AM processes, for example enabling unambiguous tracing of each production process and subsequent product. Mandolla et al. [94] introduced a new concept named “digital twin for AM supply chain” enabling the data generated by the end-to-end extrusion-based AM of metals to be organised and secured; the study also demonstrated the potential role of blockchain technology in the security of companies' supply chains. Eiriksson et al. [92] explored and demonstrated use cases for the application of augmented reality systems in manufacturing environments; it was shown that augmented reality could outperform conventional control mechanisms in these systems.

Wright et al. [100] very recently proposed a novel optimisation framework involving both deep learning and computer vision technologies aiming to optimise the calibration and printing process in the DIW of thermoset composite multimaterials. This framework autonomously and continuously adjusted the printing parameters throughout the material deposition. The novel system optimised the layer thickness and printing speed according to the feedstock being used. This closed-loop system consisted of a computer in communication with the extrusion-based AM system, in-situ imaging camera and high-accuracy convolutional neural networks (CNNs). Wang et al. [101] created an intelligent production planning system for use in FDM; the proposed approach, conducted in a cloud-based AM environment, was found to be beneficial in achieving high part quality. Bauman et al. [102] developed an algorithmic system to detect errors in FDM processes using machine vision technology. The validity and accuracy of the developed error detection algorithms were demonstrated by an experimental showing 60–80 % error detection rate using their video camera data.

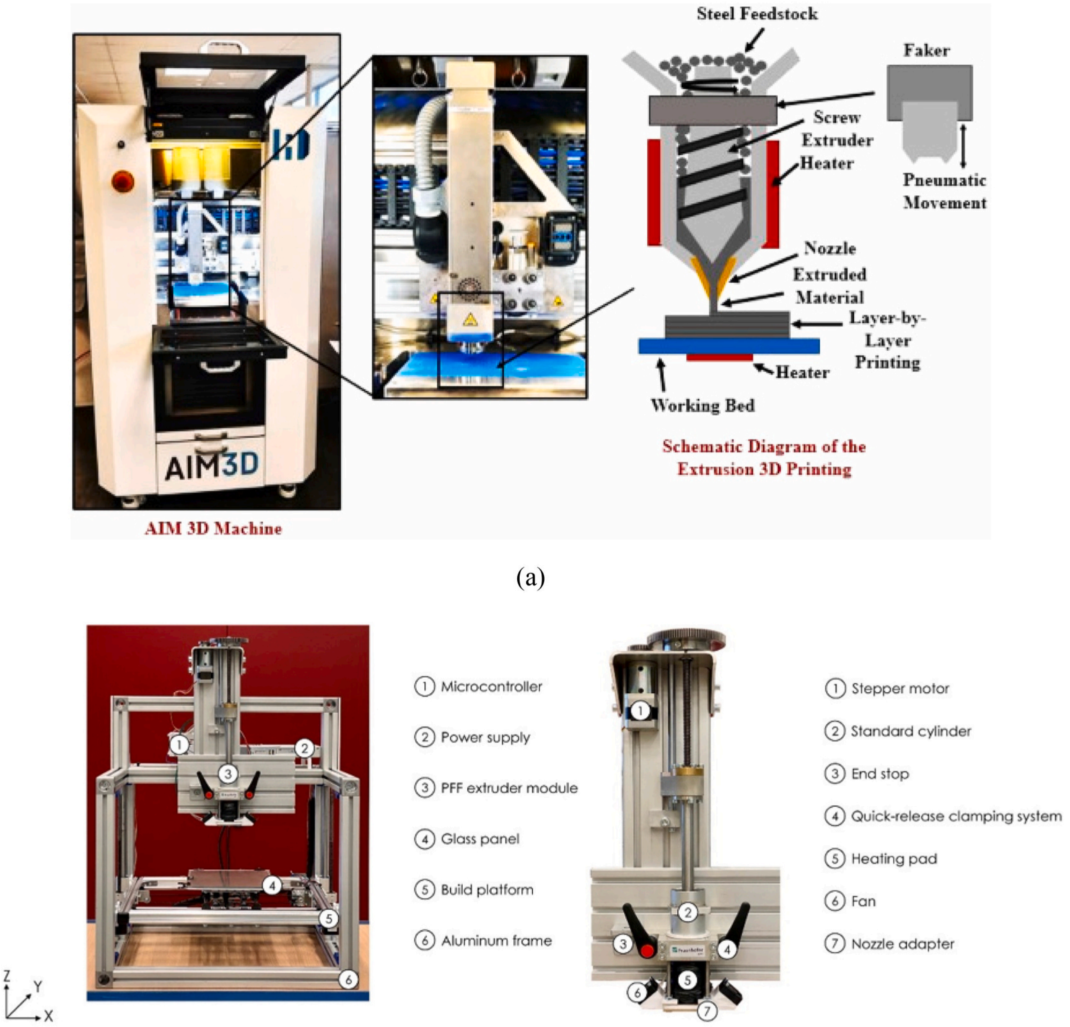
2.5. Commercially available extrusion-based additive manufacturing products

Extrusion-based AM has already been extensively commercially exploited. The FDM process (Section 1) was commercialised under different trademarks as atomic diffusion AM (ADAM), in which filaments made of metal powders are encased within a plastic binder (commercialised by Markforged, Inc., Watertown, USA), and as FFF, a simpler version of FDM (by the members of the RepRap project) [105]. Since the expiry of the original patents relating to extrusion-based AM, 3D printers using polymeric filament feedstock have become widely used and are now even on direct sale to the public for hobby purposes. Numerous commercial extrusion-based AM products are available, many of these having been designed for specific classes of AM materials. For instance, material extrusion AM of metallic materials (commercially known as metal MEX) has recently gained significant interest mainly thanks to its cost-effectiveness and simplicity. This process is similar to conventional metal injection moulding (MIM) [106]. The screw-based version of metal MEX uses granulated feedstock in a similar form to that used in MIM, but provides a feedstock filling system in which the raw material is replenished and continuous feeding is possible throughout the sequential printing of multiple parts, giving a reduction in the total printing time [107]. Well-known commercially available screw-based MEX printing systems for the fabrication of multimaterials, i.e. metals and also ceramics, have been produced by Pollen AM Ltd., using pellet feedstock [108], and AIM3D GmbH, with and ceramic extrusion systems [109]. The AIM3D printer, introduced by Direct3D and also using pellets, comprises a print head integrated with a screw-based printer, as shown in Fig. 5(a). This system allows MIM manufacturers to use their own MIM feedstocks, enabling integration of their debinding and sintering systems [107].

More recently commercialised extrusion-based AM products include a plunger-based extrusion system named bound deposition modelling (BMD) proposed by Desktop Metal Inc. [110]. This employs feedstock with a circular bar geometry which is fed using a cartridge into a continuously heated sleeve before being pushed by the plunger and deposited. The main merit of this system compared to other plunger-based extrusion processes is the improved material handling ability compared to filament feedstock. However, this system also necessitates the additional preparation step of producing the bar-shaped feedstock [107]. Another proposed plunger-based extrusion system is that of Waalkes et al. [111]; this 3D printer, illustrated in Fig. 5(b), was developed for the fabrication of Ti-6Al-4V using MIM feedstock. Using this system, whose cost is similar to that of an open polymer filament-based extrusion system, Ti-6Al-4V parts with very high stability were produced without the necessity of any further preparation of feedstock into filament form. This system is also able to use MIM feedstocks, which increases the range of materials that can be used [107]. In filament-based metal MEX systems, the filaments are composed of metal powders combined with polymer binders; these are transported by a filament transport system into the machine. Commonly used commercially available filaments are 316L metal filament by Anycubic, Filamet® by Virtual foundry and Ultrafuse 316L® by BASF SE [107].

3. Suitability analysis of extrusion-based additive manufacturing processes

The objective of suitability analysis is to determine the best solution to a process-related problem based on specified evaluation criteria, enabling a decision to be made on the most appropriate solution or mitigation for the problem to optimise or improve the process or the product to meet demands and requirements. In the present paper,



(a)

(b)

Fig. 5. Representation of (a) AIM3D printer and schematic diagram of its print head [112], and (b) plunger-based 3D printer and its extruder unit [111].

suitability analysis is adapted to extrusion-based AM to evaluate process-centred suitability (the compatibility of different AM materials with a given process, discussed in Section 2.2.1) and material-centred suitability (the compatibility of different AM processes with a given AM material, Section 2.2.2). The material-centred suitability of the different classes of materials used in extrusion-based AM will be further discussed in the corresponding subsections of Section 4.

A simplified example of material-centred suitability analysis relevant to extrusion-based AM, given by way of illustration, is the optimisation of process parameters, such as layer thickness and printing temperature, to mitigate the problem of porosity in printed parts, thereby enabling the production of sound parts and improving the suitability of the AM material. Results obtained using different values of these parameters are compared according to a set of evaluation criteria, such as pore density and tensile strength, to determine the most appropriate settings of the process parameters to improve the suitability of these aspects of the process. As an example of process-centred suitability of extrusion-based AM processes, DIW uses liquid AM feedstocks, does not involve melting during the process, and can be conducted at low process temperatures, even at room temperature; this is in contrast to FDM and FFF, which involve the melting of AM materials at high process temperatures. As a result, DIW is highly suitable for the fabrication of heat-sensitive photopolymers whose chemical structures can be easily distorted when exposed to high process temperatures.

3.1. *Process-centred suitability*

Certain characteristics of extrusion-based AM cause it to have lower process-centred suitability, and therefore to be less widely used, than liquid- and powder-based AM processes, particularly for applications where high production speeds, structural integrity and dimensional accuracy are required. Characteristic demerits decreasing the process-centred suitability of this technology are its slow deposition rate, vertical anisotropy causing poor structural integrity, and difficulties in preparing filaments. In addition, the spatial resolution of parts produced by extrusion-based AM is dependent on the nozzle diameter, with the typical achievable resolution ranging between 50 and 500 μm (Table 2). As a result, for example, very few of the currently available extrusion-based AM processes can suitably meet the demands for the typical dimensional tolerance of mission-critical aerospace components such as engine parts and propellers, i.e. usually <10 μm [113]. However, an example in which extrusion-based AM has a high process-centred suitability is the low-cost, layer-by-layer production of fluidlike and solid customised foods [25]. The main reasons for this are that extrusion-based AM can: (i) save labour costs and (ii) significantly decrease workload with respect to traditional methods, when automated with robotic systems, as well as (iii) increasing the structural integrity of printed foods thanks to the possibility of cooking during the printing process [25]. For this application, extrusion-based AM is preferred over powder-based AM processes, as the high-power heat source (laser, electron beam or microwave) required for the latter may be unhealthy for consumers exposed to the final products in the long term, and dramatically reduce the nutrient concentration of food products [24]. Extrusion-based AM is easy to apply and automate due to the low cost of equipment. It is often combined with a robotic arm or gantry, most popularly in high-volume food production and 3D printing of construction materials, enabling cost-efficient high-volume production. The automation of extrusion-based AM applications also decreases the production and lead time with respect to conventional processes, especially for the high-volume production of any AM product [27]. Recent increased demand for 3D printed multimaterial components with high complexity and functionality has led to the development of extrusion-based AM equipment with an additional filament or print head [28], thereby making this technology highly suitable for the fabrication of this class of materials. However, extrusion-based AM is not suitable for the fabrication of components having parts with an angle of >45° to the building platform (overhangs). Such components require vertically positioned support structures (see the sixth and seventh steps in Fig. 2 for overhang and support structure). By contrast, support structures are not required for liquid- and powder-based AM processes, because the printed structures in these cases are covered and supported throughout the fabrication process by the surrounding unconsolidated raw material, in the form of liquid resin or powder, within the beds in which fabrication takes place [2]. Table 4 summarises the suitability of current commercially available extrusion-based AM processes. It includes

Table 4
Comparison of suitability of extrusion-based AM processes.

Extrusion-based AM process	Suitability grade (* – *****)	Reasons for suitability grade	Suitable input materials	Suitable sectors	Suitable applications
FDM (and variations)	***** (very high)	FDM can be applied to almost all AM materials that can be melted or softened above RT; mechanical properties of parts are preserved during fabrication due to the use of closed building chamber	Thermoplastics	Design constructions	Housing units and architectural components
			Metallic materials and waxes	Dental	Dental implants
			Titanium	Medical	Osteosynthesis plates for bone fixation
			Gold, waxes, thermoplastics	Jewellery	Complex-shaped earrings, necklaces and rings
			Ceramics, thermoplastics	Electronics, industrial automation	Conductive embedded strain sensors
			ABS technical material, thermoplastics	Hobbies	Hobby applications
			Water, fertilisers (manure, nitrogen, potassium), seeds	Automated farming	Farming and planting applications
			Chocolate, juices, cheese, base dough	Food/Catering	Computer-controlled mass production of food using robotic arms
FFF	**** (high)	High adaptability to 4D printing, easy integration to multimaterial printing, wide material availability	Polymers (thermosetting)	Biomedical	Scaffolds made of thermosetting polymers
			Ceramics	Orthopaedic	Implants for use in orthopaedic applications
			Bioceramics	Biomedical	Scaffold fabrication for bone graft applications
			Metals	Industry	Tribological devices
DIW	**** (high)	DIW can be applied for some AM materials whose melting point is low, and can be operating at low printing temperatures allowing even the printing of heat-sensitive AM materials	Biodegradable and biocompatible AM materials	Healthcare sector	Biomedical implants
			Celluloses	Textile	Fabrication of responsive wearable textiles
				Electronics	Recyclable electronic and photonic cellulose devices e.g. smart labels, disposable smart actuators and sensors
			Nanocelluloses	Biomedical	Cell culturing scaffolds, drug carriers
Robocasting	**** (high)	Many material options, printed materials do not necessitate drying and solidifying	Bioceramics	Biomedical	Porous scaffold fabrication
			Ceramics	Dentistry	Dental crowns
			Metals	Automotive	Stamping dies
				Electronics	Electronic sensor devices
3D concrete printing	** (low)	Suitable only for cementitious materials	Cementitious materials (e.g. cement, concrete)	Construction	Affordable housing construction
				Defence	Military bunkers
Composite filament fabrication	** (low)	Only designed and suitable for composites	Composites	Home appliances	Microwave substrate
				Electronics	Composite circuits and sensors
Melt extrusion manufacturing	*** (moderate)	Limited to polymeric materials and biomaterials, printing temperature cannot be higher than glass transition temperatures of materials being printed	Biopolymers	Biomedical	Tissue engineering for bones and bone replacement
			Biomaterials	Pharmaceutical	Capsules and implants for drug delivery
Ceramic on-demand extrusion AM process (CODE)	*** (moderate)	Designed only for fabrication of ceramics, large and nearly dense parts can be printed with uniform microstructure	Functionally graded ceramics	Oil and gas industries	Gas turbine engines
Fused deposition of ceramics	*** (moderate)	Only suitable for printing a few specific materials	Ceramics	Aerospace	Rocket nozzles
				Space	Missile propulsion nozzles
Bioprinting	*** (moderate)	Limited to biological materials (e.g. living cells and natural human tissues)	Bioinks mixed with cells	Biomedical and health	Printing of artificial organs and skin, drug development

suitability grades and most suitable applications and sectors for each individual material extrusion technique.

3.2. Material-centred suitability

Examples of material-based suitability of extrusion-based AM include photopolymers, i.e. light- and temperature-sensitive polymeric AM materials; changes in light (such as UV) level and temperature may alter the physicochemical and thermomechanical properties of such materials. They can be most suitably printed using DIW, which is a plunger-based process, because this uses viscoelastic inks at or slightly above room temperature such that even very sensitive AM materials can be printed without undergoing any distortion or change in their properties [114]. Extrusion-based AM is also one of the best suited techniques for the fabrication of biodegradable and biocompatible materials; these materials cannot easily be manufactured using powder-based AM processes due to the possible impact of laser power on the crystallinity and molecular integrity of these thermosetting biomaterials [115]. On the other hand, extrusion-based AM has a very low suitability for the production of metallic materials, one of the main reasons being that the kinetics of solidification during extrusion-based AM leads to the formation of microstructures comprising coarse columnar (i.e. elongated) grains. This in turn leads to anisotropy in mechanical properties and the generation of metallurgical defects such as solidification cracks and porosity [116,117]. In addition, above 400–500 °C, cooling fans are not effective, making it very challenging to control the print head temperature and maintain it constant. Certain metallic materials, such as titanium, gold, brass and silver, melt at temperatures of around 1000 °C or more and thus extrusion-based AM processes have a very low suitability for producing printed parts from these materials.

A characteristic feature of extrusion-based AM is that the achievable bonding strength and structural integrity in the printing direction perpendicular to the building platform, i.e. the Z-axis (see Fig. 4 for the axes), is significantly lower than in the other axes. In powder-based AM processes, a high-power heat source is used either to locally melt the material, e.g. in selective laser melting (SLM) and electron beam melting (EBM), or to sinter it, i.e. to heat it below its melting point until the surfaces of the particles adhere to one another. An example of the latter type of process is direct metal laser sintering (DMLS). Such powder-based processes give strong bonding and good structural integrity in the final parts, and are thus the most suitable techniques for the fabrication of metallic products [115]. Extrusion-based AM also has low suitability for the 3D printing of glasses, again mainly due to the very high melting point (1400–1600 °C) of these materials, which is impossible to reach in extrusion print heads, as well as their brittle nature, which leads to very dense cracking during the material deposition and solidification stage of the extrusion process [118]. Table 5 summarises the suitability of AM materials currently available for use in extrusion-based AM processes. It includes their suitability grade, most suitable extrusion-based AM processes and methods to improve their suitability, current and prospective applications, and typical dimensional resolution and printing temperatures. The process- and material-centred suitability of extrusion-based AM processes for specific applications and materials are further detailed in Section 4.

4. Materials in extrusion-based additive manufacturing

As stated earlier, extrusion-based AM was initially limited, particularly for low-cost AM machines, to the polymers ABS and PLA. However, recent years have seen an expansion of this technology to a wide range of materials classes including ceramics, food materials, construction materials, smart materials, energetic materials, metallic materials, silicones, polymers, multimaterials, composites, biomaterials, and other materials such as glasses, photopolymers, celluloses and woods. The application of these material classes in extrusion-based AM will be reviewed in the following sections.

4.1. Polymers

Thermoplastic polymers, particularly PLA and ABS, are the most widely used filament materials in the FDM and FFF processes, and dominate feedstock choices thanks mainly to their melt flow properties and large working temperature ranges. PLA and ABS are the most widely used semi-crystalline and amorphous polymers, respectively, for use as filament. PLA is bioactive and biodegradable, can be made from natural, renewable sources such as tapioca root and sugarcane, with resulting low environmental impact, and has excellent thermal stability and processability. As a result, it is ideal for use in consumer products such as food packaging. However, as it is derived from agricultural feedstocks, it is more costly than petroleum-based polymers [42]. PLA is mainly produced by the polymerisation of cyclic di-ester (lactide) with the aid of metal catalysts or by direct condensation of lactic acid monomer. It can be made into an amorphous solid with a glass transition temperature of 55 °C, or a semi- or highly crystalline solid with a melting temperature around 180 °C [16]. ABS is fabricated from acrylonitrile, styrene and polybutadiene, and is a popular FFF filament material thanks to its mechanical properties (i.e. high toughness and rigidity even at low temperatures) and good printing quality [144]. Reviews of recent material developments in these and other polymers for AM processes are available in the literature [18,35]. In the pyramid in Fig. 6, the polymeric materials used commercially in extrusion-based AM are highlighted in orange, and those that have appeared in the scientific literature but are not yet commercialised are shown in purple. Polymeric materials that are incompatible with extrusion-based AM are highlighted in white. The most widely used materials for extrusion-based AM, apart from PLA, are other thermoplastics, including polypropylene (PP), polycarbonate (PC), high-impact polystyrene (HIPS), aliphatic polyamides (PA, also known as nylon), thermoplastic polyurethane (TPU), ABS, and high-performance plastics, such as polyetherimide (PEI), polyethylene terephthalate (PET) and polyetheretherketone (PEEK) [16]. It can be seen from Fig. 6 that most of the polymeric materials in commercial use in extrusion-based AM are amorphous; the main reason for this is the relatively low coefficient of thermal expansion (CTE) of amorphous polymers, which confers high processability (i.e. good dimensional accuracy between the CAD design and final product).

One of the main issues in the extrusion-based AM of polymers is the challenge of achieving good dimensional accuracy in semicrystalline polymers such as PP where warpage problems occur. During the solidification of the just-deposited polymer, above its glass transition temperature, the free volume (the total volume unoccupied by polymer chains) decreases, resulting in material shrinkage [145]. In amorphous polymers such as ABS, the volumetric decrease is linear with respect to temperature, whereas most semicrystalline polymers such as PP exhibit a dramatic reduction in their free volume as crystallisation occurs in part of the material. The residual stresses resulting from this and other factors, such as the effect of temperature differences in the extrusion AM machine leading to contractile forces in the just-printed strands, cause extensive warpage in the product. Thermoplastic polymers with high degrees of crystallinity are particularly susceptible to volumetric shrinkage and consequent warpage [146]. Studies seeking to mitigate the dimensional accuracy problems caused by warpage include that of Carneiro et al. [147] on PP. Optimisation of printing conditions such as layer thickness, infill, i.e. interior structure of the 3D printed part, and the orientation of printing direction with respect to the system axes were found to help mitigate the warpage. Another method of mitigation is the addition of fibre fillers, e.g. cellulose nanofibrils, or short glass, bamboo, or mineral fibres, to polymer feedstocks. For example, Spoerk et al. [148] successfully incorporated carbon fibre in PP parts fabricated using FFF. This successfully eased the fabrication of customised composite parts with very low anisotropy. Alternative approaches include the introduction of ethylene monomer segments into certain polymers to decrease their degree of crystallinity [149,150], or blending of polymers

Table 5
Comparison of suitability of AM materials for extrusion-based AM processes (RT = room temperature).

AM material	Suitability grade (* – ****)	Reasons for suitability grade	Most suitable extrusion-based AM process(es)	Major limitations	Methods to improve suitability	Current applications	Prospective applications	Typical resolution (µm)	Typical printing temperature (°C)
Polymers	***** (very high)	Cost effective, wide range of material options	FDM, FFF	Nozzle clogging, limitation in the use of minimum nozzle diameter	Developing polymer composites by adding plasticisers, copolymers and rigid fillers	Biomedical applications, scaffolds, implants, bone generation	More highly specialised parts made of polymer composites	200–400 [119]	180–220 °C [120]
Metals	** (low)	Poor bonding, low dimensional accuracy	FDM	Undesirably slow building rate, generation of metallurgical defects	Modification of filament compositions by mixing with polymers	Metal prototypes and products with highly complex geometry, e.g. aerospace components	Industry 4.0 integrated fabrication of large-scale metal components such as whole aircraft wing and fuselage	1000 [121]	210–961 °C [122]
Food (edible) materials	**** (high)	Capability of high-volume production and integration with robotic arms	FDM, FFF	Poor understanding of the effects of food properties on the extrusion-based AM processes and quality of printed foods	Integration of robotic arms for high-volume food fabrication	Customised high-volume food production	Mass production using novel feeding mechanisms, instant food 3D printing during food shortages	1500 [123]	25–85 °C [124]
Construction (cement-based) materials	*** (moderate)	High capability of integration with robotic arms	Robocasting, 3D concrete printing (3DCP)	Maximum component size limited by length of robotic arms, and sizes of gantry systems, anisotropic behaviour of some construction AM materials	Integration of robotic arms and robots enabling high-speed and high-volume material deposition	Low-cost construction	Smart cities covered with 3D printed low-cost houses and tall buildings	100–180 [125]	RT [126]
Ceramics	***** (very high)	Ability to be printed at low temperatures, suitable for several extrusion-based AM processes, challenging microstructure control	FFF, DIW, Ceramic on-demand extrusion-based AM	Problems with ceramic-based filament production, prone to hot cracking	Development of dual paste extruder for ceramics	Engines and exhaust systems, wide application areas in dentistry and biomedicine	Developments in slurry and binder systems, personalised medical products such as hip and knee prostheses	76–2000 [127]	140–185 °C [127]
Photopolymers	** (low)	Very sensitive to temperature and light, only few photopolymers can be printed	DIW	Highly changeable mechanical properties when exposed to high temperature and any light source	Printing at very low temperatures (<RT) using DIW	Dental applications, orthopaedic, nano-composites, optical devices	4D printing of sensitive photopolymers even at high temperatures without damaging their molecular structures	25–150 [128]	25 °C [129]
Glasses	* (very low)	Very brittle nature, highly susceptible to solidification cracks, low dimensional accuracy, very high melting point	DIW, FDM	Challenging temperature control at printing heads at elevated process temperatures	Modification of filament compositions by mixing with polymers	Production of glass products and ornaments	Fabrication of curved lenses and microfluidic glass chips	100 [130]	470–1000 °C [130]
Silicones	**** (high)	Highly viscous nature, high resilience	DIW	Large amount of material required as support structure for larger components being printed	Development of dispensing mechanisms such as mechanical and pressure-actuated (pneumatic) dispensing to print wide range of silicones, from low-viscosity to thick pastes	Stretchable electronics, tissue engineering applications	Personalised organ printing using mixtures that predominantly include silicone	100–400 [131]	65–300 °C [132]
Biomaterials (biocompatible and biodegradable)	**** (high)	Good capability for modifications in chemical composition and structure	FDM, DIW	Structural integrity of extruded biomaterials strongly affected by viscosity of bioink and shear thinning	Modifications in biomaterial inks and filaments for better performance	Bioengineering applications, fabrication of biocompatible scaffolds	Fully personalised organ, tissue and skin printing	5–40 [133]	120–250 °C [134]
Composites	**** (high)	High stiffness and strength, low structural distortion in printed parts	FDM, FFF	Unsatisfactory performance of highly complex composites	Modification of filament compositions by mixing with polymers	Applications in electrical, defence, and aerospace sectors	3D printed fibre composite parts for better performance than	300–800 [135]	180–240 °C [135]

(*continued on next page*)

Table 5 (*continued*)

AM material	Suitability grade (* – ****)	Reasons for suitability grade	Most suitable extrusion-based AM process(es)	Major limitations	Methods to improve suitability	Current applications	Prospective applications	Typical resolution (μm)	Typical printing temperature (°C)
							laminated composite materials for weight reduction		
Smart materials	**** (high)	High manufacturability, self-changing, –sensing, and -healing properties	FDM, DIW	Smart materials produced by AM restricted by small manufacturing scale	Additional rotational degrees of freedom and time to achieve effective smart structures	Production of smart textile products	4D printing integrated fabrication of functional smart bio-medical implants, fabrication of self-repairing systems not necessitating human source	32–64 [136]	20–140 °C [137]
Multimaterials	***** (very high)	Good potential to tailor and customise products	FDM, FFF	Difficulties of using material combinations in a single building environment mainly deriving from differences in melting temperature in compound materials, necessity of post-processing	Developments in multi print heads enabling simultaneous deposition of multiple materials	Production of highly customised low-cost and load-bearing products for use in medical and industrial applications	Highly customised 4D printed components, organs and electrical devices made of multimaterials	200 [138]	120–300 °C [138]
Energetic materials	* (very low)	Tendency to react in a very exothermic and explosive way at printing temperatures	FDM, DIW	Limited range of printing temperatures	Use of inert materials and mixtures to improve suitability	Military and defence applications	Use of mixed inks that are highly flame resistant	388–450 [139]	21–60 °C [39]
Celluloses	**** (high)	Very abundant natural polymer, biodegradable, versatile, able to retain their integrity when swollen with water	FDM, DIW	Minimum nozzle diameter limited by the viscosity of cellulose inks	Modification of filament compositions by mixing with polymers to give composites e.g. polypropylene/cellulose	Applications in electronics and healthcare sectors	4D printed functional nanocellulosic materials	500 [140]	25 °C [141]
Woods	** (low)	Very low bonding capacity of printed layers	FDM, FFF	High filler content increases the viscosity of wood, which results in nozzle clogging	Modification of filament compositions by mixing with polymers	Fabrication of wood-based furniture products	3D and 4D printed wooden buildings and constructions, low-cost customised furniture	178–356 [142]	150–275 °C [143]

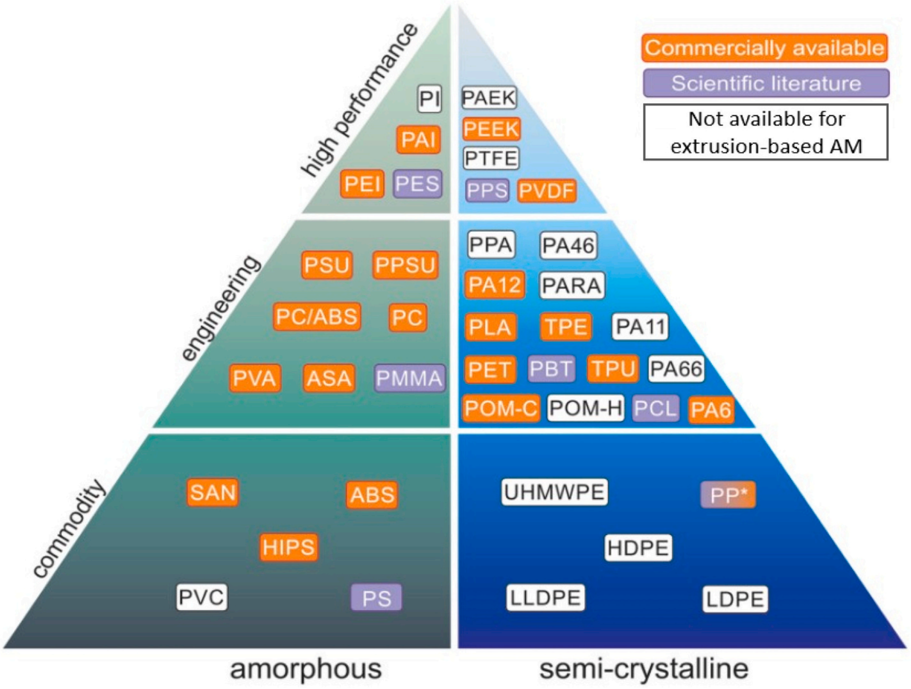


Fig. 6. A selection of common polymeric materials coloured according to the status of their application in extrusion-based AM [5]; reference [153] is recommended for the abbreviations in the figure.

such as PP that have a high tendency to warpage with other thermoplastics such as nylon 6 [150]. A change in slicing strategy [151], in which smaller brick structures are used to decrease the stacking section length during the material deposition, was proposed to mitigate warpage. However, this had a negative impact on the mechanical properties of the product. Deposition of thick layers can also reduce the warpage issue significantly; however, this adjustment degrades both the surface quality and mechanical properties of the product [152].

4.2. Ceramics

Traditional and high-performance (advanced) ceramics have a wide range of applications in the aerospace, automotive, defence and healthcare industries thanks to their excellent corrosion resistance, high hardness, very high melting point, low thermal conductivity and very high thermal stability [154,155]. These properties enable their use in aerospace applications in which they are subjected to extreme operating conditions such as in the nose cones of space shuttles, heat shields and thermal protection systems, engines and exhaust systems, as well as in thermoelectric and piezoelectric devices. Traditional techniques for manufacturing ceramic products include injection moulding and hot pressing. However, significant interest has recently been generated in using ceramics in conjunction with AM technology; this removes the need for expensive tooling and moulds and enables production of components with superior mechanical properties and more complex geometries than is possible using conventional manufacturing processes. Ceramics can be fabricated using extrusion-based AM processes such as FDM [156], FFF [157], FDC [61], robocasting [158] and Ceramic On-Demand Extrusion-based AM (CODE, using paste AM materials, see below) [159]. The FDM and FFF processes used in ceramics are modified versions of the FDM process in which filaments that are mostly composed of thermoplastic and loaded with ceramic powders are melted, extruded and deposited. Alternatively, ceramic components can be fabricated via non-extrusion-based AM processes, including stereolithography (SLA, liquid-based) [160] and SLM (powder-based) [161] both of which use laser melting. However, extrusion-based AM of ceramics usually has the advantage of much lower production costs than for either SLA or SLM [45]. Following the layer-by-layer printing process, further processing steps are often applied to the as-printed (green) parts to achieve final dense products. These include freeze- and bulk drying to eliminate water, and debinding and sintering for consolidation, as detailed in Section 4.5.

The ceramic materials most widely used in solid-, liquid- and powder-based AM processes are zirconia (ZrO_2) and zirconia-based ceramics [162], polymer-derived ceramics (PDC), i.e. those obtained by the pyrolysis of preceramic polymers [163], alumina [164] and bioactive glass [165]. Zirconia and zirconia-based ceramics are widely used in dental applications (implantations) and prostheses due to their non-corrosive and hypoallergenic properties, and as a biomaterial thanks mainly to their biocompatibility and high mechanical strength [166,167]. With the exception of the polymers discussed in Section 4.1, almost all filaments used in extrusion-based AM are composites, consisting of the material to be deposited and polymeric materials added to improve material flow and adhesion of deposited layers. This helps overcome the problems associated with filament production and extrusion of materials with high melting points.

In conventional extrusion-based AM processes for ceramics, the solidification of deposited ceramic layers takes place in air. However, Ghazanfari et al. [159] introduced a novel technique called Ceramic On-Demand Extrusion-based AM (CODE), in which a viscous suspension (paste) of ceramic particles is deposited at controlled flow rates using a circular nozzle into a tank filled with liquid florasense lamp oil for solidification. This study found that the flexural strength (232 MPa) and hardness (15.3 GPa) of yttria-stabilized zirconia (8YSZ) dental material were superior to those produced using conventional ceramic manufacturing techniques such as pressing, tape and slip casting, and injection moulding. Despite the advances in this area, ceramics processed using extrusion-based AM are still prone to hot cracking as a result of the high residual thermal stresses during melting and rapid cooling; this can lead to a reduction in the mechanical integrity of the final ceramic products. These defects arising during the extrusion-based AM of ceramics make the production of ceramics with excellent mechanical properties very challenging. To tackle this, composite ceramic filaments can be produced by the addition of a small amount of graphite.

For instance, Yu et al. [45] proposed a novel extrusion-based AM process for the fabrication of defect-free ceramic parts from yttria-partially-stabilized zirconia with added graphite; these were demonstrated to have superior mechanical properties to those made from the same material fabricated by powder-based AM processes, i.e. binder jetting and SLS. The shrinkage behaviour of the graphite-containing material was also found to be beneficial for achieving good dimensional accuracy.

4.3. Food materials (edible materials)

Four main types of AM process are used for the manufacturing of food materials: extrusion-based AM shown in Fig. 7 (for solid-based and paste (soft) materials), inkjet printing (a form of liquid-based AM suitable for liquid-based materials; note that this is not the same as DIW), binder jetting (powder-based AM method for liquid- and powder-based materials), and selective sintering (selective hot air sintering and melting, and selective laser sintering, both of which are powder-based AM methods for powder-based materials). Selected recent studies covering the 3D printing of food materials are summarised in Table 6; it is apparent from this table that extrusion-based AM is the predominant technology used for this class of materials. The main advantages of extrusion-based AM over the other three types of AM processes are its ease of customisation, minimal food waste thanks to software that specifies the required volume of raw materials for printing, as well as the comparatively low cost of entry-level AM machines [168].

Raw materials for the extrusion-based AM of food can be in the form of paste or solids, both of which have low viscosity. This is in contrast to the extrusion-based AM of polymers in which the feedstock materials are solid and often in the form of filaments. Inkjet printing and binder jetting can only be used with a limited range of raw food materials and require high-cost equipment. Inkjet printing is only suitable for inks (liquid-phase raw materials) with low viscosity, e.g. butter, chocolate, jam and cream; these should also have appropriately high surface tension, ideally below 0.035 N/m [169]. Binder jetting of food materials is mostly limited to food powders with particles of regular shape and uniform size, which are normally necessary for high-quality parts [170]. The high cost of using inkjet printing and binder jetting processes for food materials arises from the capital cost of equipment. In addition, fabrication using these methods requires support structures that are normally neither recyclable nor edible, and must be printed by a secondary print head. These factors result in significant waste; an extra cost is also associated with post-processing to remove these support structures, and they can distort small and thin-walled food products [170]. In the selective sintering of food materials, food powders such as starch

Table 6
Some recent examples of 3D printing of food materials.

Technique	Food materials	Major finding	References
Extrusion-based AM	Hydrocolloids	Prediction of dimensional stability after food printing	[175]
	Egg white + rice flour (blends) and egg yolk	Rice flour addition resulted in enhanced strength	[176]
	Potato flakes	Screw extrusion-based printing of potato flakes displayed complex fluid characteristics, enabling the extrusion of multiphase ink	[177]
	Corn starches, rice and potato	Higher starch concentration led to increase in flow stress	[178]
	Lemon juice gel	Theoretical approach for 3D printing of food materials using Polyflow software	[179]
	Potato starch + xanthan gum and sodium alginate additives	Additives of xanthan gum and sodium alginate ranging between 2 and 6 wt% showed excellent fusibility of potato starch-based composite gel system	[180]
Inkjet printing	Confectionery, decorations on cookies, cupcakes, biscuits, drops on pizza bases, jams, creams	Suitability of inkjet printing for certain food materials with innovative decoration shapes	[181]
Binder jetting	Cellulose + glucomannan (blends) + xanthan gum	Printed foods with complex shapes displayed high porosity, and kept their structural integrity	[182]
	Sugar and starch mixture	Fabrication of customised food with complex structures	[183]
	Sugar and different flavours	Fabrication of sculptural cakes with complex structures	[184]
Selective sintering	Nesquik™ and chocolate powders, sugar	Possibility of food personalisation and selective sintering of wide range of powder materials	[185]

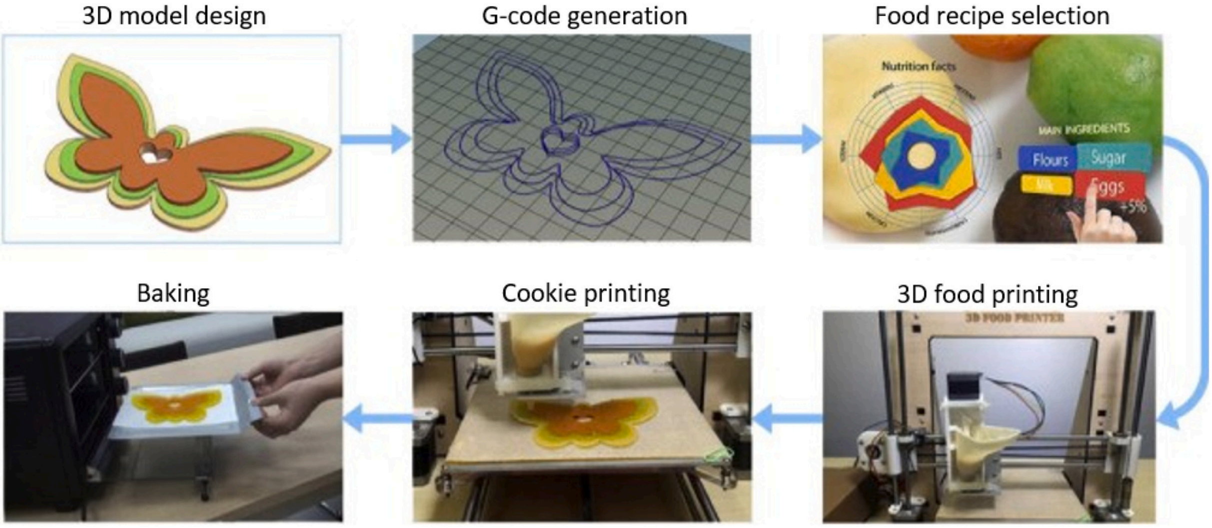


Fig. 7. Schematic illustration of extrusion-based 3D food printing [172].

granules and sugar are locally sintered using a heat source such as hot air or a laser until the production process is complete [171]. However, the high-power sintering sources used in this process can lead to degradation in the nutrient content of the products; for this reason, selective sintering is the least popular process for the AM of edible materials [24].

Extrusion-based AM is the AM technology with the greatest integration of robotics (e.g. robotic arms), especially in the case of food materials, where this enables the fabrication of food products with mass customisation (with regard to the colour, texture, shape and nutrition value of the product) at high production volume. In contrast to liquid- and powder-based AM, extrusion-based AM permits elevated temperatures in the nozzle, enabling cooking while printing, and fast solidification of deposited material, giving excellent structural integrity [25]. Food products that are commonly produced using extrusion-based AM include breakfast cereals, meat analogues, pasta, dry beverage mixes, chewing gums and pet foods [173]. 3D printing of food material can be used to make customised products for specific groups of people according to their nutrition needs, by introducing, modifying or eliminating ingredients [174]. One of the main limitations of extrusion-based fabrication of food materials is the very limited mass flow rate of the food melt, inks or gel. Because an increase in flow rate increases the pressure drop in the nozzles of the print heads, 3D printed food materials conventionally exhibit distortion, such as wrinkling, on the surfaces of deposited gels and melts. An increase in the mass flow rate also has another negative effect called the die-swell (Barus effect) phenomenon; this takes place when the extrudate diameter is greater than the channel diameter during the flow of a viscoelastic food fluid out of the nozzles. As a result of this phenomenon, polymer particles partially recover, in other words "swell back" to their original volume and shape, after deposition thorough the nozzle. To avoid this, the movement in the x, y and z directions and the linear displacement velocity of the nozzles can be adjusted to achieve the desired final shapes and volumes of food materials. The die-swell issue can cause a decrease in the velocity of the ink, necessitating a thermal post-processing curing step to stabilise printed foods. An example is the baking of bread dough following deposition [22].

4.4. *Construction materials*

The construction sector is one of the major contributors to national economies throughout the world, representing, for example, 9 % of the gross domestic product and 18 million jobs in the EU in 2016 [186]. However, the adoption of robotics in this sector, which would facilitate the transition from traditional to more modern construction methods, has been hindered by: (i) high initial investment and maintenance costs, (ii) an unskilled workforce, (iii) lack of government incentives and experts in the field, and (iv) lack of acceptance by labour unions. In addition, poor management of information flow between the different phases of construction have caused this sector to struggle to increase productivity [187].

AM of cementitious construction materials (e.g. self-sensing and self-healing concretes, high- and ultra-high-performance concretes, high-ductility and fibre-reinforced materials, cementitious pavement materials, concrete mixtures, and ultra-high-performance concretes) has many advantages for the construction industry. Firstly, it can reduce the need for construction workers to work in extreme environments, such as military, chemical or nuclear sites or the construction of tall buildings, where they would be exposed to the risks posed by to harmful materials, extreme weather conditions or high altitudes [188]. AM technology instead enables off-site fabrication, i.e. the fabrication of parts which are subsequently conveyed to and assembled at the building site. This eliminates the necessity of crowded on-site labour and enables automation of critical construction tasks, increasing consistency and productivity [168]. Secondly, AM allows custom parts to be produced on demand without long lead times, thereby reducing the number of supply chain steps and decreasing the time taken [189]. Thirdly, it allows the design and low-cost production of complex geometries and structures that may be impossible or costly to produce using conventional construction methods, giving designers and architects greater design freedom without impacting productivity [190].

The pioneering attempt in the extrusion-based AM of cementitious materials was made by Pegna [205] in the late 1990s; a novel layered fabrication technique was used to construct large-scale (>1 m) structures from Portland cement (reactive material) and sand (matrix). Subsequently, a process known as contour crafting AM, which used a gantry system to deposit thick layers of cementitious construction materials for large-scale structures with a high deposition rate, was developed at the University of Southern California [192]. Other types of AM processes in widespread use in the construction sector include concrete printing and 3D concrete printing; a summary of recent studies on extrusion-based AM of construction materials can be found in Table 7. Another advance in a related field was the development, by NASA in 2005, of contour crafting AM for in-situ construction in space environments using regolith rock available on the surface of the Moon [206]. XtreeE, a 3D printing platform, printed a wall element made of ultra-high-performance concrete with a complex shape and dimensions of 1.36 × 1.50 × 0.17 m^3 using a 6-axis robotic arm in about 12 h [58]. Most AM processes are conducted in a thermally controlled environment to achieve high-quality parts (e.g. FDM, conducted in a thermally

Table 7
Recent studies on extrusion-based AM of cementitious construction materials.

Extrusion-based AM technique or project name	Material extrusion system	Cementitious construction material used	Printed construction	Ref.
Contour crafting	Gantry	Concrete	Complex-shaped structures	[191]
	Gantry	Concrete	Smooth-surfaced and complex-shaped objects	[192]
Concrete printing	Gantry	Concrete	Construction components	[193]
	Gantry	Concrete mixture	Curved concrete products	[194]
	Gantry	High-performance fibre-reinforced fine-aggregate concrete	Bench	[195]
3D concrete printing (3DCP)	Gantry	High-performance concrete	Complex-shaped structures	[196]
	Large-scale 5-axis robotic printer	Cementitious composites blended with medium-heat portland cements and high-belite sulfoaluminate	9.4 m-span arch components	[197]
	Small-scale gantry	EMW (electromagnetic wave) absorbing cement	20-layer element with 110 mm height and 28 mm horizontal width	[198]
WinSun	Gantry	Concrete reinforced with special fibreglass	Single-floored office	[199]
TotalKustom	Gantry	Concrete	Affordable houses	[200]
Apis Cor	Robotic	Concrete	Administrative building with two floors (640 m^2, 9.5 m height)	[201]
CyBe	Robotic	Concrete	Wall structures	[202]
XtreeE	6-axis robotic arm	Ultra-high-performance concrete	Wall-element with complex shape	[58]
BetAbram	Robotic	Concrete	Thin wall structures	[203]
WASP	Other	Concrete mixture and concrete	Small house of 12 m^2	[204]

isolated closed printing chamber). In large-scale applications, such as printing of cementitious AM materials, changes in temperature and other environmental factors such as humidity can cause differences in mechanical properties at different stages of printing. To overcome this issue, on-site extrusion-based fabrication AM systems for large-scale cementitious parts have been developed to enable printing at selected temperatures under protective environmental conditions; an example is the work conducted by TotalKustom [200] in a closed printing chamber with the dimensions of 10×20×6 metre.

4.5. Metallic materials

AM of metallic parts was first developed in the early 1990s; the earliest attempts were based on a laser powder bed approach, using metal powder feedstock and a CO_2 laser for the consolidation of the powders. However, the laser power applied was insufficient for full consolidation. Following this, laminated object manufacturing (LOM), in which layers of sheet material are bonded together then cut to shape, was commercialised by Helisys in 1991 [207], simultaneously with the commercialisation of FDM for polymers. Metallic materials, including aluminium and its alloys, brass, titanium and its alloys, stainless and tool steel, silver, gold, bronze, nickel-based alloys, and platinum, are currently used in several extrusion-based AM processes, mainly in filament form. Extrusion-based AM of metallic materials is a low-cost method of fabricating desired metal parts, since the equipment required is easier to operate and cheaper than those needed for liquid- and powder-based AM processes [48]. However, it usually has an undesirably slow building rate and easily results in the generation of metallurgical defects. This is due to the solidification kinetics of extrusion-based AM processes, which lead to the formation of coarse columnar microstructures [10], as discussed in Section 2.2. As a result of the metallurgical defects generally associated with the AM of metallic materials, such as porosity and oxide scaling, some applications can be challenging, and fully dense 3D printed metals cannot easily be achieved [208].

Three extrusion-based AM processes are widely used for the 3D printing of metallic materials, namely FDM (most widely used), FFF and DIW. The FDM process for metallic materials consists of material preparation, printing, and a debinding and sintering post-processing step. The preparation step consists of mixing the metallic powder with a binder to produce a solid filament (shown in the top left image of Fig. 8) or feedstock. After printing, the resultant parts are called “green parts”; the post-processing sequence begins with debinding, i.e. the removal of the binder system. After debinding, these parts are referred to as “brown parts”, and are sintered by compression to reduce the porosity and number of voids, and enhance the strength, after which they are called “final printed parts” (Fig. 8) [30]. The mechanical properties and quality of metallic parts produced by extrusion-based AM depends on the machine type being used and the parameters of the printing process as well as on the properties of the filament or feedstock. Some recent examples of extrusion-based AM of metallic materials and the process parameters applied are given in Table 8. Due to the ductility of metals, buckling of metal filaments is one of the most common issues in the extrusion-based AM of metallic materials. It is particularly challenging to avoid this when the high viscosity of the liquefied metal at low printing temperatures restricts the powder volume fraction that can be used. Therefore, a high powder content is preferable in metallic filaments to enable good quality sintering and debinding [10].

4.6. Silicones

Silicones are siloxane-based polymers which lend themselves readily to use in extrusion-based AM thanks to the high resilience and flexibility of these long-chained, highly viscous materials (viscosity >10,000 mPa.s). The two most common deposition methods for silicones in extrusion-based AM are using mechanical pressure and using pressure-actuated (pneumatic) systems [214]. Applications in which silicones and silicone-based structures have been applied in extrusion-based AM include artificial skin containing deformation receptors [215], tissue engineering [216], stretchable electronics [217] and foams [218]. Duoss et al. [219] conducted the first study in which silicone was printed by extrusion-based AM through a micro-syringe to successfully fabricate porous structures, and demonstrated that strength in shear and compression could be controlled by modification of microstructures and porosity in these structures. Mannoor et al. [220] fabricated a silicone bionic ear to be cultured in vivo enabling the growth of cartilage tissues (Fig. 9). This was the first study in which extrusion-based AM of silicones was used in the new generation of functional prosthetics. Thermally curable silicones have also been used as a raw material in extrusion-based AM processes. For instance, Liravi et al. [32] used a pneumatic nozzle to optimise the printing quality and process parameters of a highly viscous ultraviolet-curable silicone (a subtype of thermally curable silicones), with a viscosity of 3×10^5 mPa.s at 10 s^{-1}. However, Porter et al. [221] showed that curing strands of low-viscosity ultraviolent-curable silicones with 7×10^4 mPa.s viscosity at 10 s^{-1} had low feasibility due to nozzle clogging, which had already been reported elsewhere as the major challenge in the extrusion-based AM of silicones. Many studies have been carried out to tackle this; for example, in the aforementioned study [221], it was found that introducing 0.15–1 wt% carbon black to thermally curable silicones decreased the reflection of ultraviolet light, which was the main origin of the nozzle clogging, and thereby almost completely eliminated this issue. Another critical issue with the extrusion-based AM of silicones is that due to their rubbery nature, these AM materials often require support structures during fabrication. However, the deflection of strands printed onto the support structure, mostly made of poly-vinyl alcohol (PVA), is a very common issue that can be overcome by a using a larger infill density (i.e. >30 % for thermoplastics) [222].

Silicone elastomers are silicone-based materials formed by combining reactive molecules with straight-chained molecules in conjunction with a cross-linking agent to improve mechanical properties such as energy absorption and tear strength. These materials are inert and have a high resistance to ultraviolet radiation, low toxicity and chemical reactivity, and good biocompatibility and electrical isolation

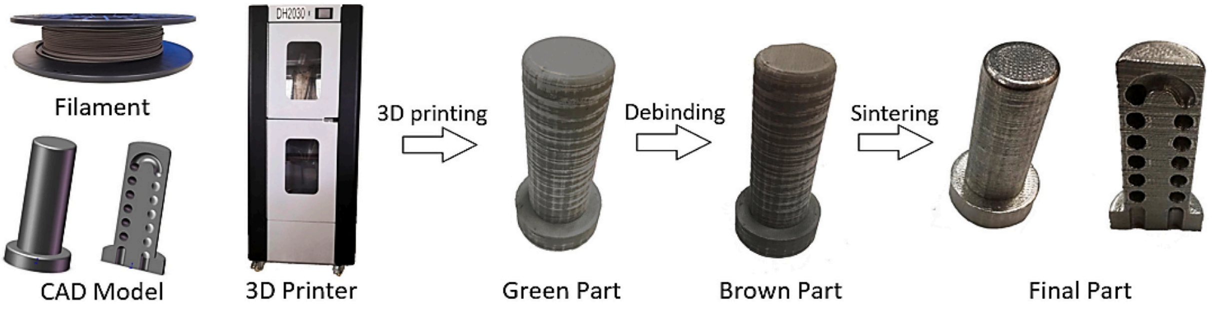


Fig. 8. Steps involved in creating metallic parts through the FDM process [48].

Table 8
Examples of process parameters for some extrusion-based AM processes in metals and alloys.

Extrusion-based AM machines	Metals and alloys	Nozzle temperature (°C)	Nozzle diameter (mm)	Layer thickness (mm)	Ref.
CoLiDo metal 3D printer	Stainless steel (316L)	230	0.4	0.2	[48]
Renkforce RF1000	Titanium (Ti-6Al-4V)	190–210	0.4	0.1	[209]
Prusa i3 MK2	Stainless steel (316L)	250	0.6	0.2	[210]
Duplicator i3 V2	Stainless steel (17-4PH)	235	0.6	0.2	[211]
Pulse 3D printer (MatterHackers)	Titanium (Ti-6Al-4V)	295	0.4	NA	[212]
HYB-AM	Aluminium alloy (AA1050)	NA	1.2	3.25	[213]
HYB-AM	Aluminium alloy (AA6082)	500	1.6	1	[121]

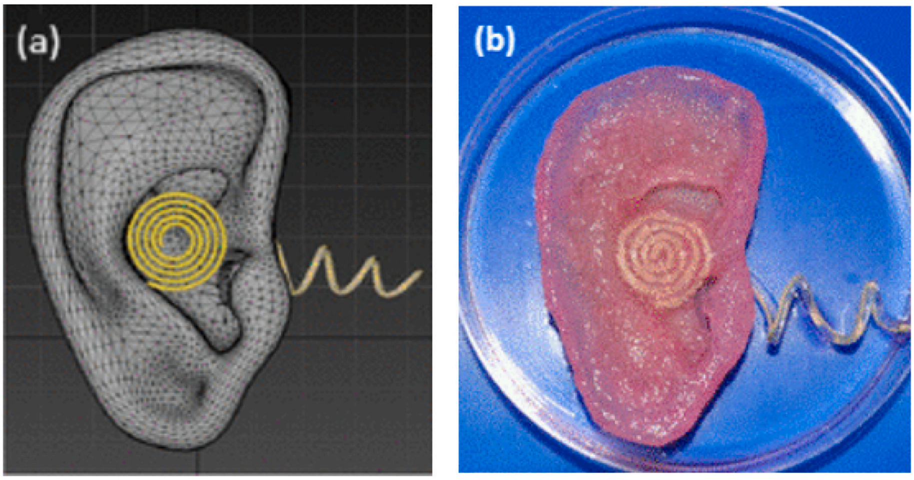


Fig. 9. (a) CAD drawing and (b) printed version of the first silicone bionic ear successfully produced by extrusion-based AM [220].

[223], and as a result are in high demand in the automotive and healthcare sectors. For example, soft and biocompatible silicone elastomers formed using extrusion-based AM are used in plastic surgery and maxillofacial prostheses [32]. The extrusion-based AM of silicone elastomers can be performed in two ways: (a) curing of the material at room temperature (moisture-cured room-temperature vulcanisation) followed by deposition through an extrusion nozzle, or (b) mixing of two types of precursor known as A and B, followed by deposition. Type A comprises vinyl-functional silicones with a platinum catalyst, and type B comprises vinyl-functional silicone with a cure inhibitor and hydrogen-functional crosslinker. The merits of the former method over the latter are the enhanced flexibility and simplicity of soft freeform silicone elastomer [42], which improves the printability by increasing the flow rate of these materials during extrusion-based AM.

4.7. Biomaterials

Biomaterials that can be used in extrusion-based AM include materials of the ceramic, metallic and polymeric classes and their composites which are referred to as bioceramics, biometals, biopolymers and biocomposites, respectively.

In spite of their tendency to hot cracking during extrusion-based AM, ceramics and ceramic-based materials are essential biomaterials, mainly used for biomedical applications such as personally tailored replacement teeth and bones. A ceramic material whose chemical composition is close to calcium phosphate (CaP) forms the main component (around 70 %) of human bones and teeth [36], although the exact fraction ranges from 60 to 99 wt%. Hydroxyapatite ($Ca_{10}(PO_4)_6(OH)_2$) is a form of calcium phosphate (the others being tricalcium phosphate and whitlockite) and is one of the major compounds of human bones. It has been widely used to produce biocompatible and biodegradable scaffolds to support three-dimensional tissue formation. The holding capacity of a scaffold (i.e. the total load that can be supported by the scaffold, including externally applied loads and its own weight), and its cracking resistance, can be increased by increasing the fraction of calcium phosphate [224]. Bioglasses, i.e. silica-based glass-ceramic biomaterials, are a type of bioceramics containing different fractions of CaO, SiO_2, Na_2O and P_2O_5. Their chief merit is their excellent bioactivity (i.e. ability to generate adherent, direct and strong bonds with bone) which makes them an ideal material for bone tissue engineering applications [225]. In a pioneering study, Eqtesadi et al. [226] found that adding carboxymethyl cellulose and poly(methyl vinyl ether) to the water-based 45S5 bioglass (a group of surface reactive glass-ceramic biomaterials) was successful in controlling the rheological properties of inks to meet the rigorous requirements of robocasting extrusion-based AM for scaffold fabrication. A common limitation in the extrusion-based AM of biomaterials is the mismatch of 3D bioprinted tissue structures with the native tissues; this is still challenging in current tissue, skin and organ implantation. This limitation results from the current limited availability of bioinks for use in bioprinting despite the existing classes of hydrogels such as keratin-based [227] and keratin-based [228] hydrogels available for use in tissue/organ bioprinting as part of bioink systems. The main issue with currently available bioinks is the challenge of finding bioinks that have both the desired physical properties for bioprinting and a suitable niche for the cells [229]. Bioinks and AM technologies have been combined to achieve optimal tissue quality and meet the demands of tissue complexity. As an example producing promising results, Kang et al. [230] proposed an integrated tissue-organ printer (ITOP) for the fabrication of human-scale stable tissue constructs at any required shape. Microchannels were incorporated into tissue constructs using clinical imaging data to construct a computer model. This enabled the diffusion limit, i.e. between 100 and 200 μm, for cell survival to be overcome in the engineered tissues and demonstrated the capabilities of the ITOP printer for fabrication of highly complex solid organs and tissues for human applications.

Biocomposites are also widely used in extrusion-based AM. In particular, calcium phosphate bioceramics of different kinds are commonly combined to generate composite biomaterials for the fabrication of highly biocompatible scaffolds [36]. Many recent biomedical devices and metallic scaffolds have been manufactured from titanium, which has excellent biocompatibility and resistance to fatigue, and a high strength-to-weight ratio, but rather poor tribological performance. Titanium biocomposites (naturally biocompatible materials) are therefore an ideal material option for use in load-bearing medical applications such as hip or knee replacements; the poor tribological performance of titanium is compensated by alloying with up to 5 wt% vanadium. Biopolymers have higher manufacturability than bioceramics and biometallic materials due to their lower melting points and greater capacity for modification of their chemical or molecular structures through modification of their crosslinking mechanisms [231]. The term “biopolymers” refers to polymers that have high biocompatibility and biodegradability, whether natural or synthetic. Natural biopolymers (cellulose, chitosan and starch) are in significant demand for use in manufacturing collagen for tissue engineering applications. Extrusion-based AM of collagen biopolymers is the most suitable AM manufacturing technique for 3D collagen architectures; since high-power lasers modify the chemical composition and nature of collagens when applied to localised areas, powder-based AM using laser power cannot be applied to these materials [232]. Gelatines are synthetic (human-made modified) biopolymers obtained from animal by-products such as connective tissues and bones. They have excellent non-immunogenicity (i.e. a low tendency to cause an immune response in human bodies), biodegradability and biocompatibility, making them useful for controlled drug release [232]. Lastly, polymer/ceramic biocomposites are mostly produced using extrusion-based AM processes, predominantly FDM. The preparation of an appropriate composite filament made of polymer/ceramic biocomposites is the chief prerequisite for achieving satisfactory printing results. Such biocomposites can also be processed by FDM to fabricate porous scaffolds for bone tissue engineering applications, achieving a relatively high compressive strength, i.e. 12.7 MPa [233].

4.8. *Composites*

Composite materials are defined as materials combining multiple materials of different chemical and/or physical properties to give a new material with enhanced properties with respect to those of its constituent materials. Mass customisation using extrusion-based AM is commonly and cost-effectively applied to fabricate composite parts for use in sectors such as electronics, biomedical, aerospace and automotive [210]. The main benefits of the extrusion-based AM of composites are the customisation of part geometries and the high dimensional accuracy in comparison to traditional formative manufacturing methods such as moulding [234]. Adding carbon fibres to polymer feedstocks improves the stiffness and strength of the final products, and decreases the distortion (deflection in physical shape) of 3D printed products formed by extrusion-based AM [33,235]. Fibre-reinforced hydrogel composites can also be produced by extrusion-based AM; Bakarich et al. [236] used ultraviolent-curable adhesives and alginate/acrylamide gel as a precursor to reinforce hydrogel (i.e. crosslinked hydrophilic polymer capable of holding a large amount of water) composites, demonstrating an improvement in the capacity for forming complex geometries. Alumina powder can also be used to reinforce polymer matrices produced by FDM to give a more wear-resistant material [237]. Nanocomposites (two-phase composite materials in which a dispersed phase on the nanometre (10^{-9} m) scale is embedded in a matrix of a continuous phase) are widely used in industrial sectors including packaging, electrical and electronics, defence, and aerospace, thanks to attractive properties such as good heat resistance and flame retardancy, light weight and excellent strength [238]. Developments in the fabrication of reliable and cost-effective 3D-printed nanocomposites (Fig. 10) have led to increased production of these AM materials for use in extrusion-based AM. For instance, Weng et al. [239] found that, among different nanoparticles, i.e. SiO_2, montmorillonite and attapulgite, SiO_2 provided the best reinforcement of nanocomposites produced by extrusion-based AM. In another study [240], it was found that a nylon6-Al-Al_2O_3-based nanocomposite filament material had almost the same melt flow index value as standard ABS. This demonstrates the capability of nylon-6-based nanocomposites as alternatives to ABS in filament-based extrusion AM processes such as FDM and FFF. Some of the issues facing extrusion-based AM of composites include the difficulty of maintaining a fully homogenous distribution of the reinforcement material used in the matrix. In addition, defects can be generated due to chemical variations in the composite filament feedstock as a result of secondary materials that are not fully chemically compatible. Another issue is that the choice of process parameters for extrusion-based printing of composite AM materials may lead to variations in mechanical properties and microstructures in the printed parts [241,242]. The introduction of reinforcement fillers to composite filaments also increases the possibility of nozzle clogging, restricting the maximum filler content that can be used in the filament feedstocks. Knowledge of the rheological properties of composite AM materials, and control of filler content in filaments, are essential to overcome these limitations [242].

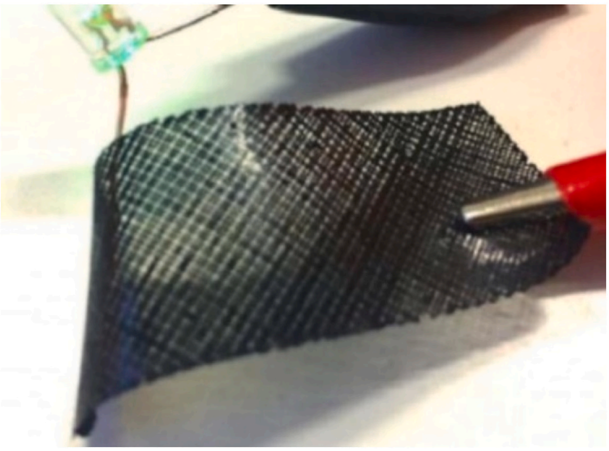


Fig. 10. Multi-walled nanocomposite woven structure produced using extrusion-based AM for use as a conductive element in a simple electrical circuit [243].

4.9. *Smart materials*

Smart materials are materials that are capable of reversibly changing their dimensions or shape (shape-changing), sensing their own deformation and damage (self-sensing), and repairing damage to themselves by changing their properties (self-healing) in response to external stimuli such as light, heat or the presence of solvents [244]. These properties enable them to react in a repeatable and predictable manner when subjected to external effects [245]. The main factor limiting the range of application of smart materials is manufacturability; recently, there has been a growing demand for cost-effective methods of fabricating smart systems and components. The manufacture of smart materials using conventional processes (e.g. solution casting and semiconductor-based fabrication) is laborious and time-consuming and therefore not ideal [246]. The suitability of this class of materials for extrusion-based AM processes has recently increased thanks to the enhanced sophistication of extrusion-based AM equipment and improvements in the availability of different feedstocks. FDM is becoming widespread for the fabrication of smart materials because it offers easy tailoring of molecular chains of polymer- and silicone-based smart materials to improve the chemical and mechanical properties of these AM materials and their composites [247]. The molecular structure and side-chains of polymer-based smart materials are usually tailored by introducing either charged and neutral fillers, or particles having the desired properties, into the material to facilitate the fabrication of extrusion-based AM processed parts with the desired physical and chemical properties [248]. Silicon-based smart materials can also be easily tailored to achieve high performance (i.e. high conductivity and piezoelectric response), and high chemical and thermal stability. However, the extrusion-based AM of these materials is rather costly, slow and non-eco-friendly, resulting in hazardous waste [248,249]. Most of the relatively few studies in the literature on extrusion-based AM of smart materials have focused on a limited number of material types and on materials rather than processes. These studies covered the role of smart materials in 4D printing which includes, in contrast to 3D printing, the fourth dimension of time, and enables printed parts to change their function or form with time in response to stimuli e.g. light, temperature, water and pressure. It usually uses a so-called *input*, i.e. a smart material, with excellent shape-changing capability [250]. For example, Choi et al. [251] have demonstrated the capability of extrusion-based AM for the 4D printing of smart biological shape memory materials, which can be used to target treatments for certain diseases. Smart textile AM materials with a shape memory effect can be fabricated into custom shapes above their glass transition temperature; then revert to their original flat shape on cooling. Leist et al. [252] investigated the suitability of a smart textile material consisting of nylon fabric mixed with PLA for processing via FDM, demonstrating the capacity of functional smart textile products to be self-adaptable to external moisture and heat effects. They showed that the ability of these AM materials to revert to their original shapes allowed textile manufacturers to fabricate custom clothes tailored to the body shapes of individuals. The production by extrusion-based AM of smart AM materials able to respond to diverse external stimuli is restricted to small-scale manufacturing. Models have been developed [253,254] to predict the geometry of smart material filaments considering their responses to the external stimuli during fabrication. The challenges in the 4D printing design of these AM materials are more complex than those in conventional manufacturing processes, since the 4D printing design includes the change strategy design step relating to the functionality demand. As a result, a suitable smart structure that is almost not responsive to stimulus and additively manufacturable was proposed by the studies of Sossou et al. [255,256] for use in applications where dimensional accuracy was essential after printing. Anisotropy is another major problem impacting the mechanical strength of these materials; even the strength of high-strength nickel titanium (NiTi) shape memory alloy produced by FDM is lower than that of conventional steel [257]. Studies with the aim of predicting and mitigating this issue in the FDM of smart AM materials [258,259] used mechanical tests to predict the material properties of smart filaments and the quality of smart products fabricated by extrusion-based AM.

4.10. *Multimaterials*

Extrusion-based AM of multimaterials is a process in which multiple AM materials with different properties (e.g. chemical or physical) are fabricated at the same time to build up a desired product. The constituents of multimaterials preserve their original chemical and physical properties and are locally deposited during fabrication according to the requirements for the final parts. Multimaterials differ from composites in that a composite material is a new material with different properties from those of each of its constituent materials, whereas this is not the case for multimaterials. A specific feature of extrusion-based AM is that multimaterial parts can easily be produced from a majority printing material with little or no additional production cost if the design allows an additional stage such as the use of a secondary filament in either the same or a secondary print head. In this way, different locations on the parts can be printed using filament materials having different chemical and physical properties, colour, or resistance to heat and impact. This additional stage can be conducted according to the requirements for the final product to give locally tailored properties, such as using high-strength materials in the load-bearing locations and low-cost, low strength materials elsewhere [260].

Multimaterial AM including, in some cases, extrusion-based AM, has begun to be introduced in industrial sectors such as electronics, biomedical, mechanical, chemical and aerospace [28]. Extrusion-based AM technologies were originally developed to print only a single material at a time; however, the demand for increasing functionality and complexity of 3D printed multimaterial products (Fig. 11) has led to interest in the development of more sophisticated extrusion-based AM processes. Since the patents relating to FDM expired at the beginning of the 2010s, more affordable FDM machines with multiple print heads have been developed [2], in which the printing speeds, resolutions, filament colours and nozzle temperatures can be adjusted individually facilitating the production of multimaterials [261]. The extrusion-based AM of multimaterials has garnered interest in the aerospace sector for the production of integrated power lines in aerospace structural components and sensors embedded into spacecraft structures [262]. Multimaterial extrusion-based AM technology has also been adopted by companies such as Voxel8, which demonstrated the capability of 3D printers for the filament-based extrusion AM of metal circuitry in a matrix of PLA to fabricate functional components for use in industrial, textile and medical applications, among others [263]. However, extrusion-based AM has limitations for the production of multimaterials, including the necessity of post-processing, unsatisfactory dimensional accuracy and the difficulty of using certain combinations of materials in a single building environment [40]. A particular difficulty is that the difference in melting points between materials classes (e.g. metal/ceramic or metal/polymer) can result in low interfacial bonding strength, as well as different processing energy requirements for the melting and facilitation of material flow in the various components of the multimaterials [46]. Studies have analysed the interfaces between components of printed multimaterials to investigate the poor bonding conditions between them. For example, Guessasma et al. [264] focused on the tensile properties in the interface between two dissimilar polymers (ABS and TPU) on the basis of the porosity generation in the interface, by means of a 3D imaging technique. Lower pore connectivity was observed in a multimaterial fabricated by a droplet-based extrusion AM process than in the same multimaterial produced by filament-based extrusion AM. Further issues that must be considered are material selection and design. Because the mixing of different AM materials with varied and non-uniform properties is very complex, an understanding of the mechanical properties, chemical compositions and other characteristics of the component materials are essential prior to the design of multimaterial fabrication processes [265]. The construction of databases of related AM materials that can be printed using the same processing conditions can help to avoid or mitigate mismatching and poor bonding between printed multimaterial components and components being printed during extrusion-based fabrication [265,266].

4.11. *Energetic materials*

Energetic materials are defined as chemical substances (in solid, liquid or gaseous state), or mixtures of these, that have a large capacity for storing chemical energy, where this can be released rapidly through a reaction caused by an external stimulus such as mechanical or thermal shock waves. Classes of energetic materials include fuels (e.g. diesel fuel and gasoline), pyrotechnic compositions (in military applications, e.g. flares), explosives (in defence), and propellants (in space applications, e.g. rocket fuels and smokeless gunpowder) [267]. The final performance

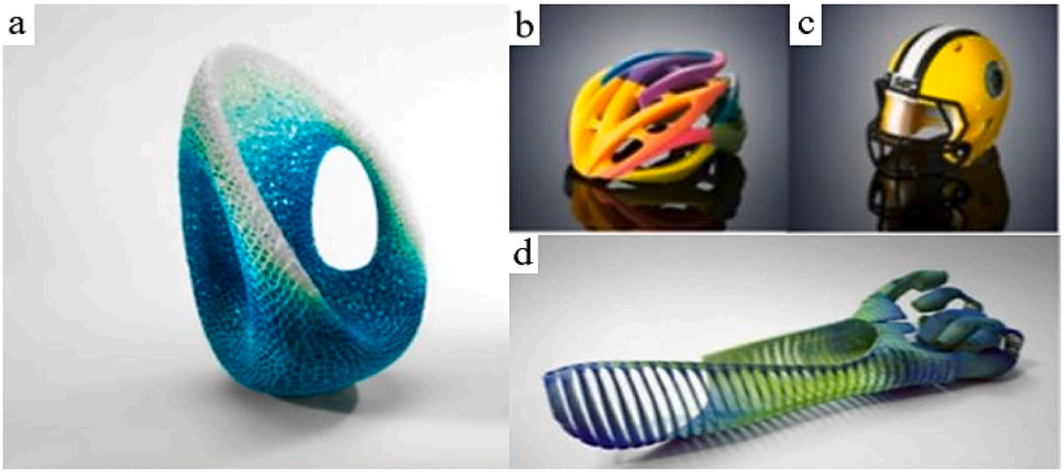


Fig. 11. Examples of extrusion-based AM of multimaterials [40]: (a) Durotaxis Chair by Synthesis Design + Architecture, (b, c) realistic multi-coloured bike and American football helmets by Stratasys, and (d) multimaterial motorcycle glove by Aalto University Digital Design Laboratory.

of these materials is highly dependent on their geometry. AM is one of the manufacturing technologies best adapted to these materials thanks to the design freedom and flexibility that it offers. Conventional methods of fabrication of energetic materials, in contrast, are time-consuming and expensive, limit design options and cause batch variation issues [39]. AM processes allow easy control of the combustion performance of energetic AM materials and tailoring of the functionality of these materials for the desired application [268]. Extrusion-based AM processes that have been applied to print complex-shaped energetic AM materials include DIW, which has been shown to be capable of submillimetre resolution [269]. FDM has also been successfully applied for this class of AM materials. However, because FDM, unlike DIW, requires melting of AM feedstock materials at high temperatures, its application to energetic AM materials is limited due to safety concerns deriving from the tendency of these materials to react in an explosive and highly exothermic way at such temperatures [268]. This highly exothermically reactive nature is the major limitation restricting the range of applications of these materials. Studies on the alleviation of this issue have focused on modifying the filaments used in extrusion-based AM processes. For example, Clark et al. [270] have demonstrated the suitability, in terms of their combustion characteristics, of energetic filament mixtures composed of an ABS binder (including potassium perchlorate ($KClO_4$), Al and molybdenum trioxide (MoO_3)) with nano-thermite energetic AM additives for use in FDM. The presence of ABS in printed energetic thin films was found to reduce the flammability of the mixture by dramatically decreasing the flame speed during combustion. The study showed that ABS was a promising candidate for use as a binder in energetic compositions, thereby also showing the feasibility of ABS energetic thin films for the FDM process at relatively high printing temperatures.

4.12. Other materials (glasses, photopolymers, celluloses, woods)

Extrusion-based AM can also be used to print materials of other classes (glasses, photopolymers, celluloses and woods), but these are much less widely used than the classes discussed in Sections 4.1–4.11 because of challenges arising (i) from the low material-based suitability of these AM materials due to their properties, and (ii) during the extrusion-based fabrication of products made of these other materials, causing low process-centred suitability.

Glasses, unlike polymers and metals, are extremely brittle, and this leads to cracking due to thermal fracture during material deposition and solidification, which decreases the suitability of these materials. Glasses in the liquid phase have a viscosity thousands of times lower than metals, which makes them flow very easily and facilitates deposition [118]. However, the FDM of most standard glasses requires a processing temperature of over 1100 °C, making temperature control at the printing head extremely challenging. In addition, glass products formed by extrusion-based AM normally have relatively low strength and dimensional accuracy, mainly due to the extreme brittleness of glasses, which leads to crack formation. DIW, unlike FDM and FFF, does not require the melting of the raw AM material inside the reservoir or print head; instead, the feedstock AM material, i.e. ink, is normally introduced into the reservoir in either a liquid or a fluidlike state. Thanks to the elimination of melting during the fabrication, glasses processed using DIW experience less crack formation, which makes DIW the most suitable extrusion-based AM process for most glasses. The use of fluidlike material in DIW enables the plunger-based deposition of liquid glass material directly from the reservoir through the nozzle. However, the relatively large nozzle diameter usually required to avoid clogging leads to low dimensional accuracy [31]. Baudet et al. [271] have demonstrated the possibility of using FDM with low process temperatures to print complex-shaped optical components made from chalcogenide glass at its melting point i.e. 350 °C, a much lower temperature than that used in extrusion-based AM of standard glasses. They achieved almost the same thermal and chemical properties in the printed product as would be obtained using non-AM, i.e. conventional, techniques.

Photopolymers, also called light-activated resins, are materials that are very sensitive to light and temperature, i.e. their properties change at elevated processing temperatures and when exposed to any light source (e.g. laser, electron beam or UV). As a result, only certain photopolymers can be printed using the DIW process [114]. Cellulose is an essential component of renewable biopolymers mainly used in the healthcare, textile and electronics sectors. Thanks to developments in processing technology and cellulose chemistry, extrusion-based AM of celluloses is no longer restricted to traditional applications such as paper and forest-originated products. Advanced functional application areas of celluloses processed by extrusion-based AM include the biomedical and electronics sectors, and the fields of responsive wearable textiles and smart health care [272]. Cellulosic materials can be printed using the three main extrusion-based AM processes, i.e. FDM, FFF and DIW. The most widely used forms of cellulose that can currently be used as FDM filament material in industry include: bacterial cellulose, microcrystalline cellulose, lignocelluloses, nanocrystalline cellulose, and other material mixtures including cellulose contents such as ColorFabb recycled wood floor/PLA (15 % cellulose), cellulose fibre/PLA (0–20 % cellulose), micro-nanocellulose/PLA (0–30 % cellulose) and cellulose nanocrystals/polyvinyl alcohol (2–10 % cellulose) [273]. Woods and wood-based materials can also be printed using extrusion-based AM, in the form of filaments made of both wood fill (wood filaments in which PLA-based material is combined with lignocelluloses such as wood dust, cork, or other derivatives, resulting in printed parts with a real wooden look) and a mixture of a polymer with recycled wood-based material recovered from waste from furniture manufacturers [272]. FFF is the process with the greatest potential for further developments in the extrusion-based AM of woods, with new strategies recently generated for using furniture waste to achieve low-cost fabrication of wood-based products [274]. In the large-scale extrusion-based printing of wood-based products such as home furniture, some challenges arise; temperature gradients and resolution control during extended printing times can lead to warpage and low interlayer adhesion in the printed wood products [275]. These issues are more problematic when wood filaments are incorporated with biomass-based filler due to the lack of polymer entanglement and heterogenous dispersion of the filler. In order to tackle these issues, large-scale extrusion-based 3D printers can be made to incorporate tuneable screws, environment control and dual-printing heads so that better control of bonding conditions and temperature gradients can be achieved [276].

5. New developments in extrusion-based additive manufacturing technology and materials

Recent progress in academia and industry in the development and commercialisation of extrusion-based AM processes have expanded the application range of this technology [10], for example in sectors such as electronics, biomedical, aerospace, construction, automotive, food production and healthcare [277]. Extrusion-based AM can now compete with powder- and liquid-based AM processes as regards building rate, cost-effectiveness, spatial resolution and available material options [278].

Most recent research and development work on extrusion-based AM has focused on improving (i) the processing methods to improve the performance of parts, and (ii) the availability and suitability of materials for AM [277]; examples include the multiple print head units applied in some extrusion-based AM processes to enable the fabrication of multi-materials as discussed in Section 4.10. Also, as seen in Sections 4.2 and 4.3, extrusion-based AM has recently been combined with a gantry system or robotic arm for cost-efficient high-volume applications, such as large-scale food production, and 3D printing of multi-floored tall constructions in as short as possible lead times [27]. Recent examples of studies that achieved short lead times include a 12 m^2 house with walls made of a concrete mixture (aggregate and water) and concrete roof that was printed in Italy in 10 days by the WASP project in 2018; it was claimed that the cost of construction was significantly lower than that of conventional construction methods [279]. The WinSun company printed a single-floored office with an area of 242 m^2 in 17 days for the Dubai Future Foundation [280]. These printed parts were then assembled in Dubai in just 2 days. By using a 3D printing technique that was similar to counter crating AM (see Table 7), the total production cost was decreased significantly, labour was halved, and the construction waste was decreased to 60 % of what would be produced using conventional building techniques [281]. Recent developments in extrusion-based AM have enabled the fabrication of dense structural components using extrusion filaments containing alumina [282], zirconia [283], and other ceramic powders. This technology can also be used for the fabrication of complex-shaped, very lightweight structures in the aerospace industry, particularly for high-temperature applications such exhaust and engine systems, which require materials with very high melting points i.e. 2000 °C or higher [284]. Recent developments in the extrusion-based fabrication of multimaterial transducer design have demonstrated the capability of this technology to successfully print multi-layered functionally graded ceramic structures [285]. A very recent development in extrusion-based AM is the production of artificial organs (e.g. heart, skin, eye cornea and kidney) from human tissue. The first attempt to produce a human heart by extrusion-based AM was made in 2017 by ETH Zurich; this printed human heart was artificial and made of flexible materials instead of human tissues. The first 3D-printed whole human heart made of human tissue comprising biological molecules, collagen and vessels was announced in 2019 by scientists at Tel Aviv University [286]. In 2019, the first successfully printed artificial human cornea was announced by researchers at the University of Newcastle. This was customised according to data gathered from the eye of the patient who was to receive the artificial cornea [287]. In another recent application in the biomedical industry, artificial human teeth and dental crowns were printed using Slurry Micro Extrusion (SME), in which around 40 wt % slurry was deposited using a changeable nozzle with 100–800 μm diameter. Numerous other products for the biomedical sector, such as shape memory NiTi catheters, porous implants, artificial heart valves and stents can be printed using extrusion-based AM [288]. The latest developments in extrusion-based AM technology also facilitate the fabrication of energy engineering components. For instance, a 3D alumina impeller was fabricated, showing no visible printing defects, using the extrusion-based deposition of alumina mixture pastes [289].

6. Prospective applications and future perspectives of extrusion-based additive manufacturing

Current research into extrusion-based AM processes is mainly focused on: (i) advances in process competence aiming to achieve improved performance from the printed components, (ii) extension of material availability to increase system integration in extrusion-based AM processes, (iii) numerical analysis to analyse and predict the physical properties of AM materials, and (iv) modelling and simulations of the dependence of material performance on design and printing strategies [277]. As discussed in Section 4, the past decade has been characterised by a dramatic increase in material availability for extrusion-based AM. This is expected to lead to many innovative applications in different sectors.

An unreinforced concrete footbridge manufactured by extrusion-based AM was placed in Venice in 2021 in a collaboration by ETH Architects, Zaha Hadid Architects and Block Research Group [292]. During the deposition step of this compression-only 12-by-16-m arched footbridge structure, the concrete was not deposited in the conventional AM way, i.e. on a horizontal building platform, but orthogonally to the direction of the compression forces. This approach facilitates the pressing of printed concrete layers together under the summation of their own weight and external forces, thus eliminating the necessity of post-tensioning and reinforcement [292]. The successful production of the footbridge represents the proof of a concept which could be applied in other concrete structures.

Some of the main limitations of extrusion-based AM thus far have been low deposition rates and limited building volumes, but the sizes of printed products have been increasing. For example, the largest 3D printed aerospace component to date, which is listed in Guinness World Records, is an aircraft wing of dimensions 5.3 m length × 1.7 m width × 0.5 m height produced by Oak Ridge National Laboratory, USA for Boeing. This component had a mass of 748.5 kg and was printed using a large-area AM machine with a robotic arm in only 30 h at a high deposition rate [293]. The Apis Cor company printed the world's largest 3D printed building to date (Fig. 12) in Dubai in 2019; this was a 640 m^2 administrative building with two floors and a height of 9.5 m. It is expected that around 30 % of all the houses in the city of Dubai will be constructed using 3D printing by 2030 [201]. Also in 2018, the Chicon House company printed the first legally approved 3D printed house in the USA. This was 60 m^2 in floor area; it was claimed that the project demonstrated the potential of AM of construction materials for affordable housing for people with low income [294]. The number of material options for the extrusion-based AM of construction materials has recently increased, and metals have started to be used to replace cementitious construction materials in some applications. The world's first 3D printed (wire + arc additive manufacturing) pedestrian bridge with a steel structure was designed by Joris Laarman Lab and tested by the Alan Turing Institute and Imperial College London. This novel 12-m-

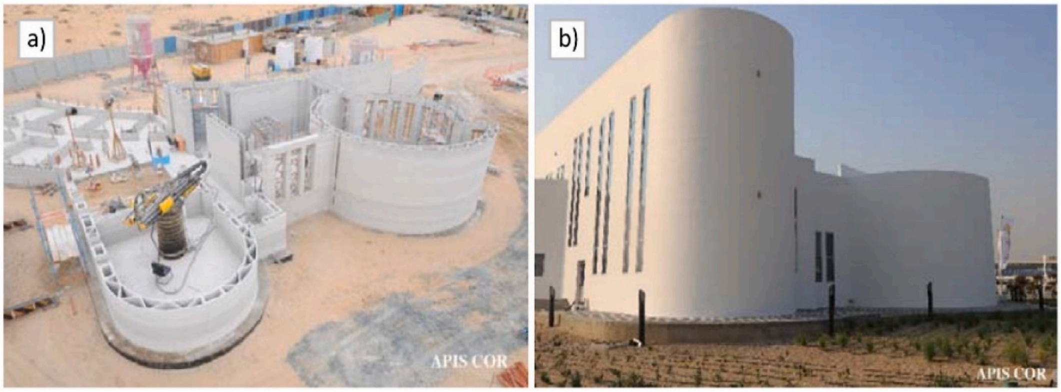


Fig. 12. (a) Printing stage and (b) complete view of the world's largest 3D printed house by Apis Cor in Dubai [21,201].

long bridge structure was then printed by the Dutch company MX3D in 4 years and has been in use since 2021 [295]. Based on these promising early applications, metals are expected to be used in future in extrusion-based AM processes for structures capable of bearing higher loads, such as printed steel bridges for vehicular traffic. A problem in extrusion-based AM processes, particularly for metallic materials, is undesirably low bonding strength in the final parts. In a promising study, Khondoker et al. [296] recently developed a bi-extruder FDM print head with 0.2-mm thick intermixer blades to enable mixing (crushing) and deposition of two different filament materials at the same time through a single nozzle. This approach also enables the layer-by-layer deposition of functionally graded materials, improving the interfacial bonding strength of this multimaterial in the interlocking of interfaces.

Promising applications exist in the biomedical and healthcare sectors. In 2011, the director of the Wake Forest Institute for Regenerative Medicine presented the first successfully bioprinted human kidney, which was produced by extrusion-based AM. Although this kidney did not live for very long, the short printing time of 7 h was a great improvement over previous achievements, and was regarded as very promising for future developments in this area [287]. Although a 3D printed heart has been successfully produced, it is likely to be several years before the transplantation of hearts and other organs produced by extrusion-based AM is possible. The use of patients' own biological molecules and cells to print replacement organs is expected to eventually render organ donation obsolete [286]. Another promising area is the production of human pancreas and skin using extrusion-based AM. According to BBC Research, the total market for bioprinting of skin is anticipated to grow from $24.7 billion in 2018 to $109.9 billion by 2023 [287]. Companies producing biocompatible human skin for transplantation to victims of burns or skin disease have attracted significant investment [293] and clinical testing is underway. These prospective applications of extrusion-based AM to produce biocompatible, personalised human skin and tissues would reduce waiting times for skin transplantation because skin donors would no longer be required [297]. In the future, every hospital is expected to have a bio-printer, and print personalised human skin or other organs according to the transplantation and replacement demands of their patients using their own biological molecules and cells to avoid rejection problems that might occur after the transplantation of donor skin, tissue or organs.

Thanks to the recent developments in digital technologies, the potential and performance of extrusion-based AM is expected to increase, accelerating the commercialisation of this technology in industry. Such developments assisting extrusion-based AM are anticipated to increase the suitability both of the technology and of AM materials, extending the areas of application of this technology and enabling a quality of printed parts that is comparable to that obtainable using conventional manufacturing processes. The optimisation of extrusion-based printing parameters was traditionally carried out using laborious trial and error, but recently developed deep learning frameworks can be applied to any type of extrusion-based AM process to detect defects produced in extrusion-based AM with average accuracies of up to 95.5 % [100].

The contribution of machine learning to extrusion-based AM extends beyond defect detection to the control of process parameters and print head positioning, prediction of cost and energy consumption and process monitoring. This subject has been covered in [290], while many papers such as [291] focused on the analysis of energy consumption and cost on the basis of re-manufacturing and life-cycle emissions in a complex way involving many factors. Machine learning technology has great potential to overcome this complexity, enabling more reliable prediction of energy consumption and cost.

7. Conclusions

This paper has reviewed the state of the art in the extrusion-based AM of all classes of material to which this technology is currently applied, as well as recent developments and innovations in extrusion-based AM technology. It has also covered process- and material-centred suitability, and current and potential future applications of this technology. The following conclusions can be drawn:

[1] The range of materials that can be used in extrusion-based AM has increased significantly during the last decade thanks mainly to: (i) the implementation of multiple print heads and filaments in the process, (ii) developments in AM equipment and software, and (iii) developments in materials technology, such as fibre reinforcement of filament materials to generate composite AM materials, and the use of multimaterial filaments.

[2] Recent developments, particularly in control mechanisms and digital technologies for extrusion-based AM processes, have led to a significant increase in the process suitability of extrusion-based AM and in the suitability of available materials for use in this technology. This is expected to lead to wider use of the technology in more industrial sectors.

[3] In contrast to liquid- and powder-based AM, extrusion-based AM offers a wider range of available modifications to printing equipment and mass customisation of the final product. These capabilities allow manufacturers to meet demands for ever more sophisticated final products. It is expected that in future, the number of available material options for extrusion-based AM will increase further, and multimaterials will be one of the main applications for this technology to meet the increasing demand for high-performance, highly customised products.

[4] The material-centred suitability of extrusion-based AM is more limited than those of liquid- and powder-based AM due to the nature of this technology. Several materials classes, such as metals, cellulose and glasses, cannot easily be printed using

extrusion-based AM, but can be printed more or less satisfactorily using liquid- and powder-based AM processes. However, extrusion-based AM is the most suitable technology for the manufacture of products made from many types of materials including polymers, cementitious construction materials, food materials, photopolymers, smart materials, and biocompatible and biodegradable biomaterials.

[5] Extrusion-based AM technology has prospective applications in the aerospace, construction, food production, medical and electrical sectors; as a result of great interest from the healthcare sector, biomaterials are expected to show faster development than other materials classes. For example, the market for the bioprinting of skin is anticipated to quadruple between 2018 and 2023 and exceed $100bn [287].

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.

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