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Advances in polymers based Multi-Material Additive-Manufacturing Techniques: State-of-art review on properties and applications

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ABSTRACT

Recently, the demand and studies related with Multi-Material Additive Manufacturing (MMAM) is continuously increasing. To uncover the essential knowledge hidden in the current mess of research works, this study, based on a simple review methodology, helps to identify and discuss the current knowledge about: limitations of software and hardware, the interface bonding strength of dissimilar materials, polymer reinforcement with continuous fiber, polymer-based multi-materials, and future and challenges. The review method starts with a list of topics to check: MMAM of polymers, the bonding strength between materials, knowledge gaps, and new development lines in polymer applications. Then, it continues with the search procedure on electronic databases and the inclusion and exclusion criteria definition. Finally, to help the discussion and assessment, the information is collected in tables.

1. Introduction

The addition of materials, usually layer by layer, to produce goods from 3D models defines additive manufacturing (AM), known in non-technical contexts as 3D printing. AM shows a reduced manufacturing lead time to obtain complex products with few or no post-processing steps, little scrap, and a direct connection with 3D product models. Call to be a driving technology of Industry 4.0 [1] and a tool for the repatriation of industrial activities [2]; nowadays, AM is generally used in rapid prototyping and is growing in the production of final products [3]. Sometimes a product's performance depends on the combination of different materials. For most AM processes, which produce one-material parts, obtaining a multi-material product involves an additional step: assembly of individual components. Nevertheless, to maintain AM's advantages in manufacturing multi-functional products, the whole process must be carried out in one step. Multi-Material Additive Manufacturing (MMAM) technologies can print components by joining more than one material in one operation. MMAM solutions have a wide range of potential applications, such as fabric [4], large format equipment [5], drones [6], automotive [7], electronics [8], or droplet deposition [9]. Traditionally MMAM approaches are based on elaborate laboratory set-ups, and further efforts are needed to substantiate their use in actual industrial applications. Scientifics have always seen the reduced adhesion noted at the joining area of materials with different chemical and physical properties as a critical point [10]. The improvement and extension of MMAM to final products foster the research related to this topic. A straightforward search in the Web of Science [11] reveals an increment in the number of jobs about MMAM technologies (Fig. 1). Regarding review works, the first studies appeared in 2013, and currently, their figures (cites and publications) grow slowly. Studies [12–14] classify the existing AM technologies and the key issues that are helping in the manufacture of multi-material components. Other reviews discuss how to print functionally graded materials (FGM), which gradually experience a variation in composition and structure over the volume [15,16]. While other systematic reviews [17], such as Zheng et al. work [18], give an overview of the MMAM of polymers technologies and applications, our study focuses on how the process parameters influence the mechanical properties of MMAM printed products. Besides classifying technologies, the study identifies substantiated statements, investigates issues, discusses practical applications, and analyzes the bonding's weakness problem between dissimilar materials, key for polymer materials. Moreover, a call for an advance in the

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standardization of design rules and tests for multi-material additive manufacturing is done. Paper's Section 2 is devoted to our review procedure; the results are portrayed in Section 3 and discussed in Section 4. Finally, the conclusions are drawn in Section 5. Table 1 contains the meaning of every acronym included in the paper.

2. Review methodology adopted

The review methodology follows several recommendations of the Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) [19]. MMAM is a recent research topic, so this does not apply time limits to the searching process. On the other hand, only papers in English have been selected. Fig. 2 shows the main steps of our review procedure. The literature included in this review has been obtained from online databases. In the first stage, the Elsevier, Springer, Wiley, and Emerald databases have been consulted, with different combinations of the following keywords used for the searches: MULTI-MATERIAL AM, MULTI-MATERIAL 3D PRINTING, INTERFACIAL BONDING, MULTI-NOZZLE 3D PRINTING, MECHANICAL PROPERTIES, FUNCTIONALLY GRADED MATERIALS, FEA, and CONTINUOUS FIBER REINFORCED PLASTIC. To avoid the bias effect, the same criteria of quality, inclusion, and exclusion were used. The search process records studies of research papers, proceedings of renowned conferences, and book chapters. Afterward, all records by the number of citations were filtered and the most cited ones were selected. An in-depth read of the abstract, the conclusions, and the materials and method sections provided the most meaningful information. Later, the results section was analyzed to check numerical values. It is considered out of the present paper's scope those studies that approach 3D printing of composite materials based on a short fiber-reinforced polymer matrix [20], usually printed in FFF (fused filament fabrication). This review takes into account studies that:

- Show or analyze MMAM technologies and approaches for polymers.
- Study the properties achieved.
- The influence of the parameters in multi-material parts made of polymers.

The above classification fits with the topics defined in the introduction section: parameter influence, mechanical properties, MMAM technologies, and applications.

3. Technological developments in additive manufacturing

In a first search, an amount of 33 papers were extracted. However, we rejected 4 records because they analyze the printing of fiber-reinforced polymer filaments and other homogenous combinations of materials within the same filament. Thus, just 29 papers remained for further review. In a second search, we got a total of 55 new valid papers. Altogether, 84 papers have been included in the review, which collects 6762 citations, representing an average of 80.5 citations/papers. The majority of these studies have been analyzed qualitatively (Tables 2–4). Simultaneously, those that measure process parameters' influence are included in a quantitative synthesis (Tables 5–7). In Fig. 3, a flow diagram represents the flow of information through the search phases. The following subsections show the collected main results for each topic considered in Section 2: MMAM technologies, parameter influence, and printed part mechanical properties.

Table 1
List of acronyms.

Acronym	Meaning	Acronym	Meaning
ABS	Acrylonitrile butadiene styrene	LCC	Local control composition
AM	Additive manufacturing	LM	Layered manufacturing
AJ	Aerosol jetting	MRI	Magnetic resonance imaging
ASA	Acrylic styrene-acrylonitrile	PA12	Polyamide 12
BJ	Binder jetting	PA6	Polyamide 6
CAD	Computer-aided design	PC	Polycarbonate
CCF	Continous carbon fiber	PCL	Polycaprolactone
CF	Carbon fiber	PET	Polyethylene terephthalate
CFR	Continuous fiber reinforced	PLA	Polylactic acid
DIW	Direct ink writing	SCRT	Short carbon reinforced termoplastic
EES	Electrical energy storage	SFF	Solid freeform fabrication
FEA	Finite element analysis	SLA	Stereolithography
FEM	Finite element method	SLTAT	Single-layer temperature adjusting transition
FGM	Functionally graded materials	SLS	Selective laser sintering
FDM	Fused deposition modeling	TPMS	Triply periodic minimal surfaces
FFF	Fused Filament Fabrication	TPU	Thermoplastic polyurethane
GF	Glass fiber	UC	Ultrasonic consolidation
IJ	Inkjet	UV	Ultraviolet
KF	Kevlar fiber	XFEM	Extended finite element method

3.1. MMAM technologies

The first studies of MMAM appeared in the late '90 s, after almost two decades of advances in conventional additive manufacturing. At that time, hardware and software systems were not prepared to print more than one material within the same process step. This lack of

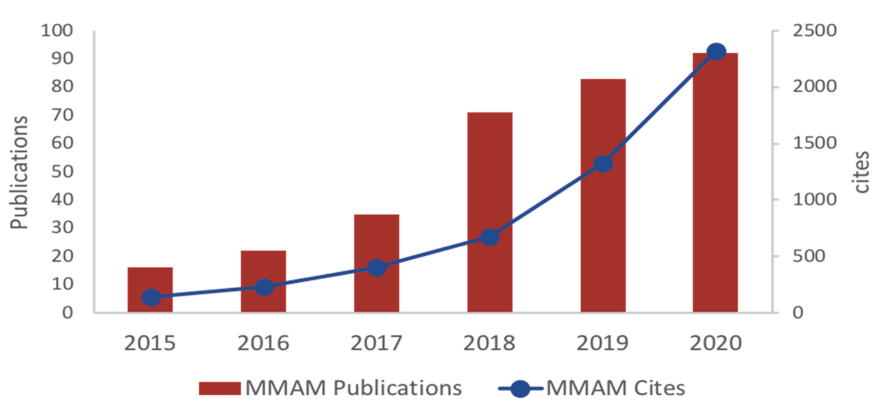

Fig. 1. The number of publications on MMAM and the annual cites, from 2015 to 2020.
Data source: Web of Science, Search TITLE: ("Multi-material" Or "Multimaterial" Or "Multi-Material additive manufacturing" Or "functionally graded") AND TOPIC: ("Additive manufacturing").

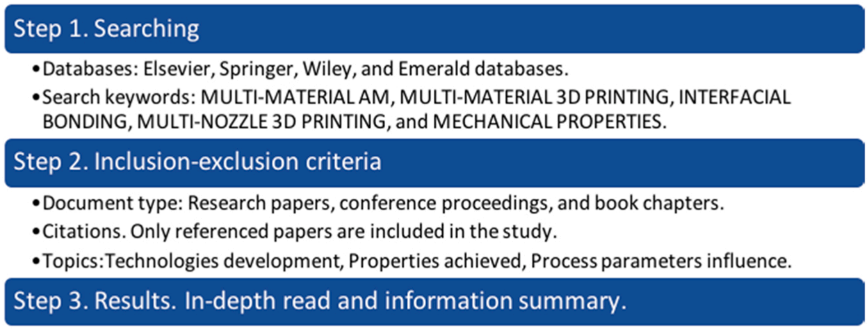

Fig. 2. Main steps of the proposed review methodology.

Table 2
Synthesis of research on the adaptation of software systems to MMAM.

Kumar et al. and Rajagopalan et al. [21,22]	Wu et al. [23]	Bhashyam et al. [24]	Cho et al. [25]	Qiu and Langrana [26]
Obtaining optimal distributions	Integrated system for design of geometry and composition	CAD system for heterogeneous components modeling	Developing a 3D representation system for editing geometry and composition	Optimization of nozzleś paths

Table 3
Synthesis of research on new concepts of manufacturing CFR plastics.

Study	Process	Results
[74]	Assisted printing heating interlayer bonding by plasma-laser	Reduce delamination of CCFR-PEEK
[75]	Compaction process during deposition by RepRap-based FFF 3D printer modified	Reduce voids and increase the strength of parts
[76]	3D printing of continuous CFRTPs using prepreg composite sheets (new approach based on Laminated Object Manufacturing)	High tensile strength parts, (up to 663,8 MPa). Able to produce from small parts ≊ 10 mm up to 5 m.
[77]	Pre-process using ultrasound treatment to increase wettability between fiber and polymer	Improve of tensile strength up to 34% and flexural strength up to 29%
[78]	Modification of commercial 3D printer that incorporates: impregnating, printing, and curing modules	High mechanical performance (tensile strength up to 1432 MPa) and reduction of voids.
[79]	Application of DIW to thermally or Ultraviolet curable resins	Parts obtained with similar mechanical properties that in molded samples.

technology motivated the researchers to work in this field. The first studies addressed the adaptation of software to represent and slice multi-material objects. These works (Summarized in Table 2) were carried out within the Solid Freeform Fabrication (SFF) framework. After developing the software systems carried out by these and other authors, the majority of papers focused their efforts on physical setups. There are considerable differences between the systems developed according to the kind of material printed. Heterogeneous objects were successfully modeled by [21,22], combining an iso-parametric interpolation system with non-linear programming algorithms. Wu et al. [23] worked on an integrated system to allow the design and fabrication of components with local composition variation, especially in generating the post-processing slice of the Local Control Composition (LCC) model. Bhashyam et al., [24] developed a 3D-CAD system that uses libraries of material composite function and planning algorithms for the modeling geometry and composition of heterogeneous objects. In the work of Cho et el., [25], the authors developed and implemented control abilities allowing the editing of geometry and composition during the design phase simultaneously. Qiu and Langrana [26] developed a CAD system for Multi-Material Layered manufacturing processes to provide a high-quality tool path during part manufacturing.

3.1.1. *MMAM: polymers printing*

Stereolithography (SLA), Fused filament fabrication (FFF), Direct ink writing (DIW), and PolyJet, alone or, more frequently combined have been tested for multi-material printing of polymers. For many years, researchers and engineers have adapted commercial machines to handle multiple materials, mainly with these four technologies.

SLA was the first technology adapted to MMAM, employing the solution of the multi-vats that it has been employed in, Digital Light Processing (DLP) technology [27] because today is the most effective

Table 4
Synthesis of research on the adaptation of hardware systems to MMAM.

SLA	namdar et al. [28] Carousel system with multiple vats	Arcaute et al. [30] Just one exchangeable vat	Choi et al. [29] More sophisticated carousel system	Han et al. [31] Lesser process times thanks to dynamic fluid control	
FFF	Espalin et al. [34] Mobile platform between two printers	Ali et al. [35] Creation of a printer with 5 extrusion heads	Khondoker et al. [36] Creation of an extruder with two entries and a mixing chamber	Ren et al. [33] Developing a system for the creation of continuous gradients	
Other systems for MMAM of polymers	Sitthi-Amorn et al. [40] PolyJet system with 10 printheads and an integrated 3D scanner	Laumer et al. [41] Developing an SLBM system for polymer powders	Kokkinis et al. [50] DIW system with low magnetic fields to control particle orientation	Li et al. [51] Combination of DIW with microfluidics	Roach et al. [32] A combined system with FFF, PolyJet, and AJ heads with robotic arms
Hybrid systems for polymer-metal parts	Tang et al. [88] FFF and SLM with ultrasonic welding in sequential processes	Matsuzaki et al. [90] Combination of FFF and metal electroforming within the same step	Ambrosi et al. [92] Combination of FFF and an electrolytic system	Chueh et al. [91] Combination of FFF and SLS within the same platform	

Table 5
Synthesis of research on the properties and parameters in multi-material PolyJet. Legend: ↑ Increasing of property, ↓ decreasing of property, – without effect on property, SD strong dependency.

Study	Materials tested	Influence factor	Analyzed properties	Results
Wang et al. [113]	TangoPlus VeroWhite	MMAM vs reinforced elastomer	Stiffness Compress. Strength Toughness	↑ ↑ ↑
Moore and Williams [126]	TangoPlus VeroPlus	MMAM vs pure elastomer	Fatigue life	–
Dalaq et al. [131]	TangoPlus VeroPlus	MMAM vs pure elastomer	Stiffness Compress. Strength Toughness	↑ ↑ ↑
Vu et al. [129]	TangoPlus VeroPlus	Print orientation	Tensile strength Toughness	SD SD
Boopathy et al. [130]	TangoPlus VeroWhite	Adding layers	Toughness	↑
Mirzaali et al. [127]	Agilus30 VeroCyan	Continuous vs discrete gradients	Tensile strength Stiffness Ductility	↑ ↑ ↓
Tee et al. [128]	Agilus30 VeroMagenta	Reinforcing elastomer with thermoset particles	Tensile strength	↑

method to manipulate the 3D printing resins. The first approach required considerable process time [28,29], developing carousel systems with multiple vats of photopolymer, able to rotate and locate over the printing platform to allow the use of different materials (Fig. 4a). Another system, with only one self-aligning mini-vat setup, was proposed by Arcaute and co-workers [30] employing a platform that fixes a cylindrical vat, and micropipettes add the photopolymer. However, with this system, the processing time is not solved either.

Recent developments reduce the processing time using a micro SLA setup based on a high-pressure hermetic vat [31], with dynamic flow control that allows rapid exchange of the photopolymer; the main advance of this system is the absence of cleaner liquid, Fig. 4b. In the case of FFF technology, the multi-material systems suppose an increase in the number of extrusions via employing a multi-nozzle configuration, Fig. 4c. The first step was adapted from two different commercial machines in a two extrusion head FFF system [34]. One platform moves between the two heads, and the setup enables both the printing with two-layer thickness and the deposition of two materials employing ABS and PC. The manufacturing with several filaments at the same time could be carried out using different extrusion nozzles, driven by two motors and mounted on a platform [35]. Consequently, a noticeable reduction in processing time is obtained. Recently, in 2018, Khondoker et al. [36] customized an FFF extrusion head. It had two heat sinks for two different filaments, which led to a mixing melt chamber and one nozzle. The printing of a mixture between two immiscible filaments was possible using static intermixes introduced in the mixing chamber; The manufacturing of plastic parts with continuous materials gradients is highly desired in a wide range of applications. The researchers work in various gradient patterns of color [37], particle concentration gradient [38], and graded material mechanical properties [39]. The first approach employing the extrusion methods develops a new system that integrates function-driven modeling, gray-scale representation, and multi-material mixing [33], Fig. 4d. Later, Pitchaya et al., [40] presented a photopolymer system with an integrated 3D scanner with a machine vision installed to auto-calibrate the system, scanning the component's current state and providing a feedback loop able to do corrections in real-time, thanks to the 3D scanner. Other researchers have developed power bed-based systems for multiple polymers. Laumer et al. [41] developed a simultaneous laser beam melting (SLBM) system using polypropylene and polyamide 12 powders. Firstly, each layer is added, and both materials are preheated almost to their melting temperature using a laser beam. After that, a digital light processing chip performs a selective control of the beam to achieve a flexible melting of both preheated polymers. To get high-quality distributions, small particle size and high bulk density were demonstrated to be necessary [42], the authors applied electrophotographic powder transfer to polymeric particles obtaining a high-quality distribution of different powders within the same layer.

Systems that allow the MMAM of polymers via DIW where a viscoelastic ink solution is deposited on the platform by the extruders can be classified based on the number of printing cartridges [43]:

- Single-paste cartridge: the part is manufactured by multi-material ink using a single cartridge employing one nozzle [44,45];
- Multi-pastes cartridges: alternatively extruding formulations allowing different compositions using several cartridges and nozzles [46, 47];
- Co-extrusion of pastes: concentric nozzles printing filaments based on core and shell with different compositions [48].

The reduction of the effect of high temperature and controlling the anisotropy of composite materials in polymers is one challenge, specifically in biomedical applications [49]. Kokkinis et al. [50], developed a system where the deposited ink particles' orientation could be controlled with low magnetic fields, Fig. 4e. More recently, combined DIW with microfluidics; this multi-material 3D system allowed the manufacture of textured composites with liquid inclusions [51]. FFF, InkJet (IJ), Aerosol Jetting (AJ), and Direct Ink Writing (DIW) were integrated into a single system developed by [32], (Fig. 4 f). The printing heads move on the Z-axis; meanwhile, the platform adjusts the X and Y-axis positions. Additionally, two robotic arms were installed for pick-and-place operations. With these arms, selectivity between electrically insulating regions and conductive, electrically functional parts

Table 6
Other research on the properties and some applications of MMAM.

Study	Process	Object	Materials	Results
Barthold and Rotthaus [101]	None	Developing of FEM system	None	A promising method for strengthening weak interfaces
Bartlett et al. [132]	PolyJet	Manufacturing of a robot case	Elastomer and thermoset	Successful variation of stiffness in 3 magnitude orders
Rocha et al. [46]	DIW	Manufacturing of graphene-based electrodes for EES application	Thermoresponsive inks	High capability of the method tested
Watschke et al. [138]	FFF	Design of standardized test specimens	ABS and PLA	Correct quantification of the properties
Sakhaei et al. [93]	PolyJet	Redesign of the ratchet mechanism to remove some parts	Tango and vero series	Good performance and strength
Falck et al. [140]	FFF	The direct joining of polymer over an Al plate	ABS, PA6, and Al	Greater shear strength and deformation than with adhesive joining
Afshar and Wood [142]	FFF	Creation of weather-resistant coats in a single operation	ABS and ASA	Improving structural integrity against aggressive environments

Table 7
Synthesis of research on the properties and parameters in multi-material FFF. Legend: ↑ Increasing of property, ↓ decreasing of property, – without effect on the properties.

Study	Materials tested	Influence factor	Analyzed properties	Results
Mansouri et al. [134]	TPU, Bayblend	MMAM versus pure bayblend	Compress. strength Stability Elasticity	↑ ↑ ↑
Lopes et al. [135]	PLA, PET, TPU	Interface presence	Tensile strength, Stiffness	↓ ↓
Yin et al. [10]	ABS, TPU	Increasing the printing temperature and speed and bed temperature	Tensile strength	↑
Lin et al. [139]	PLA, PCL	Applying SLTAT method	Tensile strength	↑
Khondoker et al. [36]	PLA, ABS, HIPS	Using intermixers	Tensile strength	↑
Ribeiro et al. [112]	PLA, TPU	Using interlocked interfaces	Tensile strength	↑
Singh et al. [136]	ABS, HIPS	Raster angle, density, layer height, and number of tops and bottom layers	Tensile, flexural and impact strength	↑ with 15/105°, 80%, 0,25 mm, 3 layers
Kumar et al. [137]	PLA, ABS, HIPS	Increasing printing speed and density	Tensile strength	↑
Baca and Ahmad [141]	PLA, ABS, HIPS	Using multiple extruders versus a single extruder	Tensile strength Process time	– ↑

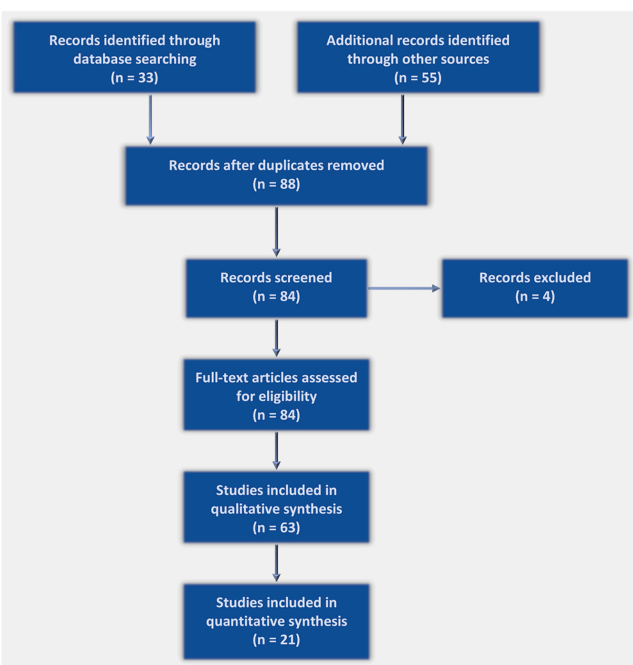

Fig. 3. Flow diagram for the proposed review procedure based on PRISMA statements [19].

such as electrodes, resistors, and sensors could be easily inserted.

3.1.2. MMAM: CFR polymers

The mechanical strength of plastic parts manufactured by additive manufacturing is continuously improved, this field is one of the ten challenges in additive manufacturing [52]. Continuous fiber reinforcement (CFR) materials are a FFF process and one of the key solutions to increase the strength and stiffness of 3D printing components. Fig. 5, shows the evolution of the number of publications in this field during the last years, this is highly important to provide a context for the utility of the technology providing an insight into new concepts and future directions of research. Today, reinforcing these parts with continuous Glass, Kevlar, or Carbon Fiber Filaments [53] improves up to 10x the mechanical performance. Although the use of Short Carbon Reinforcement in Thermoplastics (SCRT) produces parts with a considerable increase in stiffness [20], the strength of the parts is not significantly increased, due to the fracture develops in the interface fiber-matrix originated to high dissimilar stiffness of these two materials [54] generating a pull-out mechanism. On the other hand, CFR can increase the content of fiber up to 95%, being able to obtain parts with a high level of resistance and stiffness, unlike the SCRT that do not usually exceed 40% of carbon fiber content, due to the embrittling of the filament, which cannot be wound without breakage, generating continuous machine stops in the 3D printers.

The efforts of the research community have been directed mainly towards three ways of introducing the continuous fiber into the polymer matrix [55]: 1. Fig. 6a. CF is embedded inside the structure when the deposition of the remaining layers would be completed, which implicates the need for prepreg material [56,57]; 2. Fig. 6b. CF is Embedded into the matrix material inside the extruder body [58–66]; 3. Fig. 6c. CF is deposited after the matrix material nozzle passes [67–72].

Fig. 7-a,b) shows an example of the first concept, a glass fiber layer of ASTM 3039 tensile specimen with the orientation of 90° and 45° respectively over Nylon matrix thermoplastic fabricated with Markforged® two 3D printer. The fiberglass has been deposited on the top of a nylon layer, then a new nylon layer or another fiberglass layer can be applied to form a stack. This printer has two extruders that can print continuous fiber (first extrusion) embedded into a Nylon thermoplastic with chopped short carbon fibers (second extrusion). It is possible to stack several layers of continuous fiber or alternate, with layers of the polymeric matrix, being able to create a sandwich structure with a defined core density. Today, this concept can be applied layer by layer and only inside the part between the matrix polymer. The configuration and density of continuous reinforcement are configured by the software eiger®. The second concept allows the generation of multi-material in a single process, Fig. 7-c) shows CFR embedded in the PLA matrix. This concept allows the generation of components with high resistance in the direction of deposition of the fiber, although the strength of the reinforcement-polymer interface is weak when carbon fiber and thermoplastic are used [73]. The third concept Fig. 7-d) embedding carbon fiber bundles by an automated selective deposition method using an ultrasonic embedding apparatus. In this case, the continuous fiber reinforcement can be placed most efficiently and not only layer by layer. Table 3 shows a list of new concepts which are developed using continuous fiber reinforcement.

3.1.3. MMAM: printing of hybrid parts

The joint of dissimilar materials without adhesives is a group of traditional technologies that have been widely investigated during the last years [80]. The main aim of this technology is to take the advantage of dissimilar materials with different mechanical, thermal, or electrical properties in a single component and with a single manufacturing process in the automotive industry, avoiding the competition of these materials in many automotive engineering applications [81,82]. In additive manufacturing, the investigation of joining metal and polymers is seldom discussed, but in all cases, the strength of metal/polymer interfaces is a challenge, and the majority of the investigations use a sequential manufacturing process, failing in the printing of complex geometries in only one integrated MMAM process, or at least, joining with an acceptable interface strength [83,84]. To improve the interface strength, laser-assisted joining methods were employed in metal/polymer parts [85,86]. Recently numerical and experimental

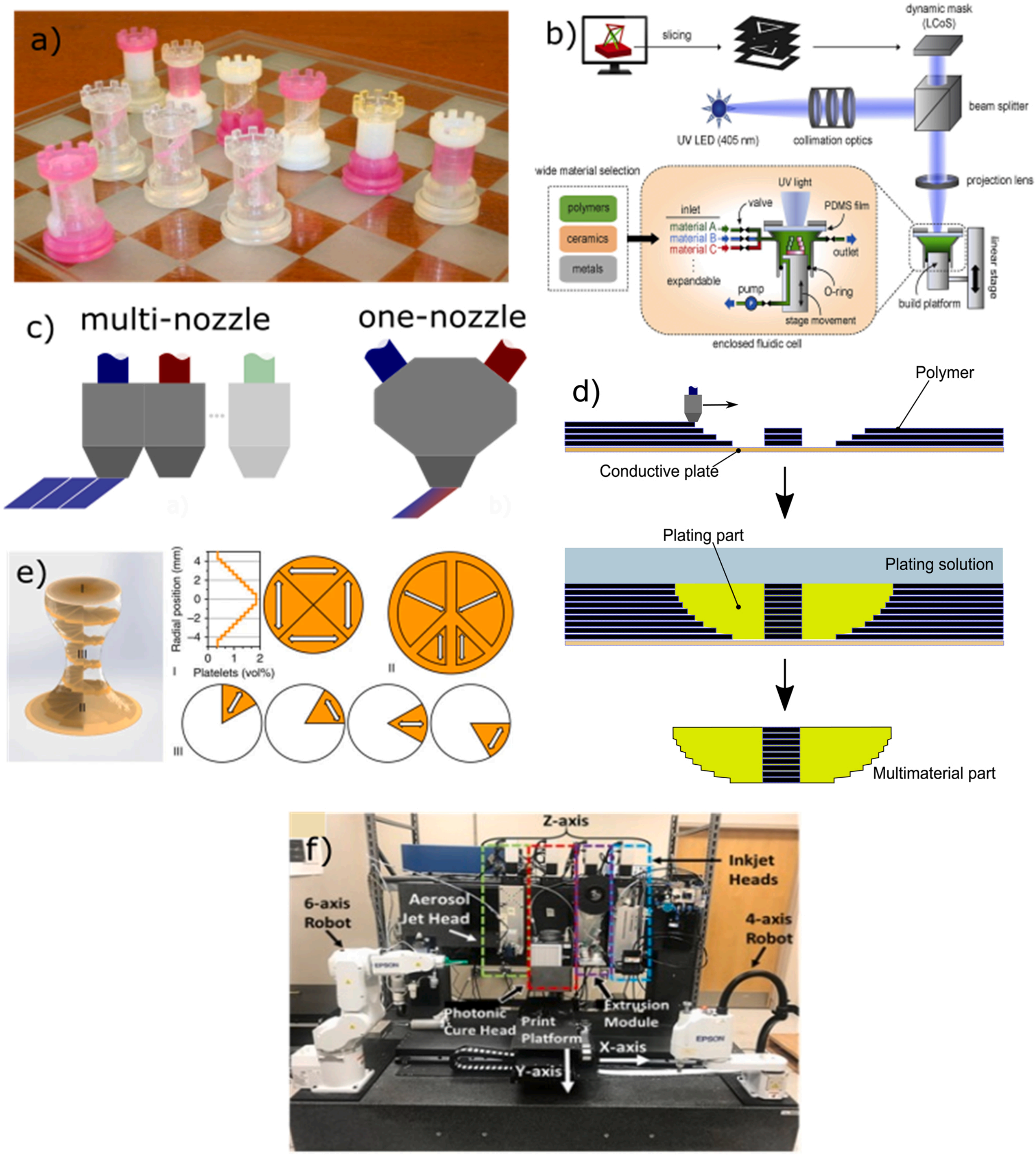

Fig. 4. (a-f) Schematic system. b) Illustration heterogeneouslyintegrate micro-scale.
(a) (Reprinted from**.** (a-f) Schematic process of MMAM and parts. a) Chess pieces printed with multi-material SLA system. b) Illustration of the MM-PμSL system process that allows heterogeneously integrate multiple functional materials in three-dimension at micro-scale. c) Multi-nozzle and one-nozzle FFF approaches; d) Gradient 3D printing and control schemes.; e) Heterogeneous composite sample manufactured by multiple ink writing equipment and scheme of the changes in local texture and platelet concentration. f) An image of the m4 hybrid printer with robot and motion axis. (a) (Reprinted from [29] with permission from Elsevier). (b) (Reprinted from [31] with permission from Elsevier). (d) (Reprinted from [33] with permission from Elsevier). (e) (Reprinted for free with no permission required under http://creativecommons.org/licenses/by/4.0/ article doi: 10.1038/ncomms9643. (f) (Reprinted from [32] with permission from Elsevier).

investigations were developed to joint PLA/Aluminum by laser-assisted metal surface treatment [87] and to jointly print FFF polymeric parts with metallic parts printed in SLM [88]; these two samples of polymer-metal hybrid improved the interface strength but needed a subsequent operation to obtain the final part. Anyway, the roughness surface of the metal part is intimately linked with the increase of the interface plastic-metal strength [89,159], which generates micro-holes, increasing the contact surface of the polymer during the bonding process.

To produce complex functional parts in one same step, Chueh et al. [91] integrated the FFF and SLS processes over the same platform, which deposit metallic (SS316L/Cu10Sn) and polymeric (PLA/PET) layers

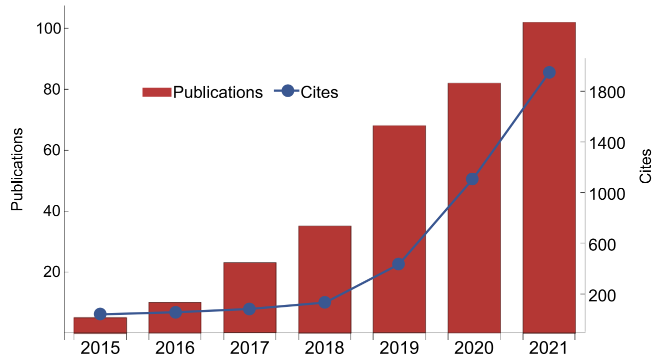

Fig. 5. Number of publications and reviews using “continuous carbon fiber composite additive manufacturing”, from 2015 to 2020. 2021 estimated.
Data source: Web of Science.

enhanced by laser heating. Electroforming based process was developed by Matsuzaki et al., [90] and in this process, the printing polymer and metal are in the same step. The authors apply the FFF process to print the polymeric part (PLA) and a sacrificial mold, then the metal is added by electroforming, with a surface quality affected by the layer height of the polymeric mold. A schematic representation of the process steps can be visualized in Fig. 8a. The procedure followed in manufacturing a Cu/PLA gear is shown in Fig. 8b. An attempt using electrochemical systems was performed by Ambrosi et al. [92]. The use of multi-material PolyJet in practical applications was also investigated by Sakhaei et al. [93]. The authors redefined the classic ratchet mechanism removing the spring, obtaining the compliant mechanism of Fig. 9a, based on a flexible polymer. Finally, Table 4 lists the studies discussed in Section 3.1.1, and 3.1.3, highlighting the main technological innovation.

3.2. Applications, properties and process parameters in MMAM

Robust additive manufacturing with multiple materials requires an excellent bonding strength among the materials to maintain the structural integrity of the part. Several studies analyze the quality of the interface bonding in AM and compare the strength with conventional manufacturing processes, concluding that the lack of compaction and homogeneity in the material due to the absence of pressure generates weaker interfaces compared with parts manufactured by the injection molding process [94,95]. Most authors try to define the influence variables of the final bonding resistance in FFF [96] that is the most widely used technology to generate multi-material-based polymers in AM. In the case of DIW, the limitations imposed by the post-printing steps (drying, debinding, and consolidation) restrict the combination of materials with very different properties, for this reason, a few examples of DIW can be considered truly MMAM parts [97,98].

Using SLA methods, the strength of the interface is improved straightforward, by adding reinforced material such as carbon nanotubes [99] or metal-filled resins [99,100]. Some preliminary works analyze and design systems to achieve the optimal interface configuration. In that sense, Barthold and Rotthaus [101] proposed a numerical study of the bond strength of interfaces, which combines the extended finite element method (XFEM) technique and novel Nitsche type method [102]. The strength between layers is foreseen as the biggest problem when approaching MMAM. It is already a self-evident problem in conventional additive manufacturing, where interfaces of the same material induce an increase of the stiffness [103] and, hence, propitiating fragile fractures and lower mechanical strength than the pure material. Other studies, on the other hand, have investigated how to enhance the bonding strength of the interface planes. It motivated to modify a conventional 3-axis FFF printer to achieve a total of 5-axes [104]. Printing

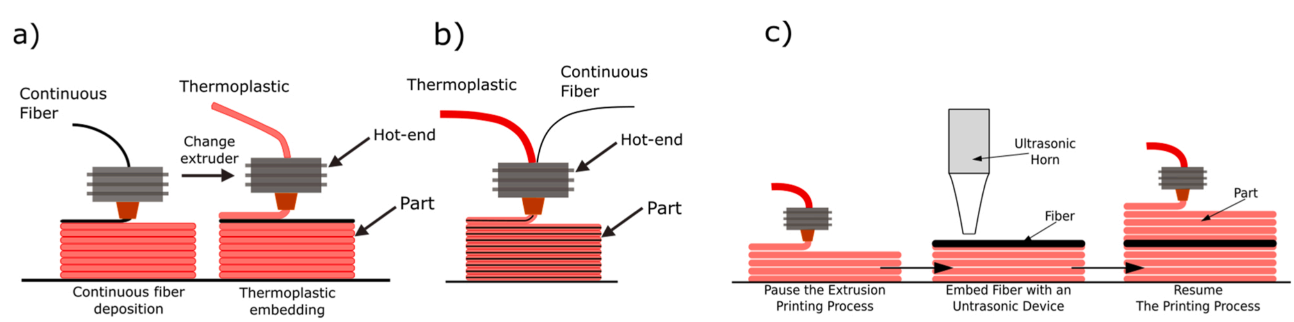

Fig. 6. a) CF is embedded inside the structure when the deposition of the remaining layers would be completed; b) CF is Embedded into the matrix material inside the extruder body; c) CF is deposited after the matrix material nozzle passes.

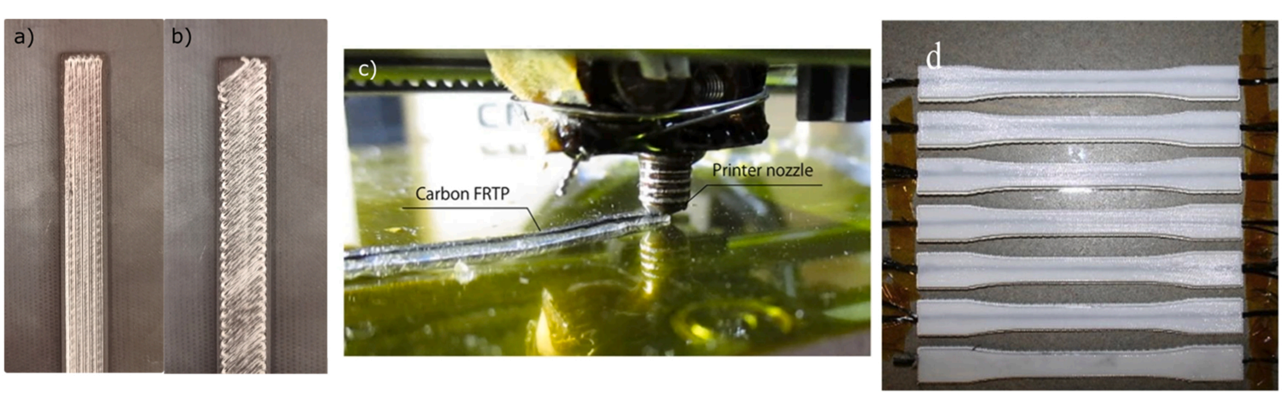

Fig. 7. .
.a) 90º Glass Fiber reinforcement in Nylon matrix thermoplastic; b) 45º Glass Fiber reinforcement in Nylon matrix thermoplastic; c) CFR by in nozzle impregnation; d) Polycarbonate tensile specimens with CFR bundles embedded by Jahangir et. al. [72]. (b) (Reprinted for free with no permission required under http://creativecommons.org/licenses/by/4.0/ article doi.org/10.1038/srep23058). (c) (Reprinted from [72] with permission from Elsevier).

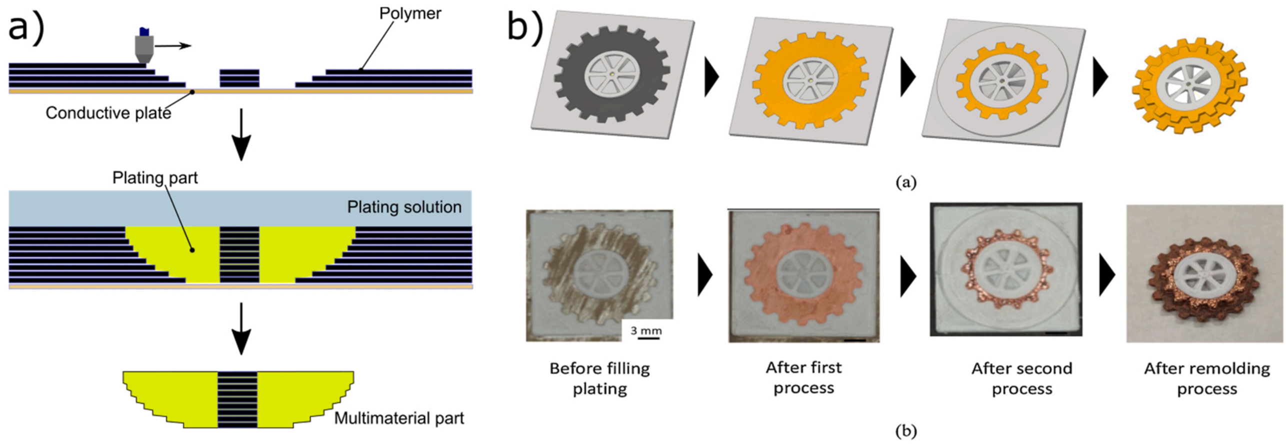

Fig. 8. (a) Schematic and real parts of the molding process developedby Matsuzaki et al. [90]. b) Matsuzaki etal. [90] procedure to manufacture multi-material parts using electroforming and FFF technologies;
(Reprinted from [90] with permission from Elsevier).

parameters have been targeted as a decisive element that influences the final strength [105–109,158]: higher extrusion temperature and bed temperature improve the adhesion strength in FFF, while the increase of printing speed has different effects depending on the material used: beneficial in those studies using PLA and detrimental with ABS. Likewise, lower layer heights and well-studied orientation of the deposition line, not only increased the strength of parts under tensile loads but also produced smaller voids inside the parts that improved the behavior under impact loads [110,111].

Pre-post-processing in additive manufacturing has been widely used in recent years [114–119]. The authors preheat the surfaces to improve adhesion between layers that is its intimate relationship with temperature and time [120], in addition, the ambient temperature has a greater influence on the bonding potential interface than the extrusion temperature of the nozzle. facilitating the diffusion phenomena [121]. The external input of energy has been largely approached as a method for strengthening the bonding between different layers of various materials, thanks to the enhanced interpenetrating diffusion of the material originated by the higher interface temperatures using solid state-laser [117, 122], with infrared-lamp [115,123], or microwave radiation and coating [124]. Energy input raises the temperature of the already deposited layers above its glass transition temperature just before adding the new layer, with a significant enhancement of the interlayer strength. In some cases, the time parameter has proven decisive, especially in technical polymers such as polyphenylene sulfide where not only the application of heat sources at high temperatures is decisive, but also the heat-treatment time [125].

The analysis of fatigue in the interface of multi-material is still widely discussed, several authors proposed that mechanical properties, especially the fatigue life, depend on the kind of gradient that generates the interface and the effect of abrupt surface between dissimilar polymers. Generally, the fatigue life is conditioned not only by the strength of the interface but by the weakest bonding material. In another work [126–128], the authors analyzed the fatigue life between thermoset and elastomeric parts, concluding that interfaces have similar fatigue life on average to the elastomer separately. Greater Young Modulus and tensile strength were reported when gradients are continuous, whereas better elongation and fracture behavior occurred with discrete gradients. The print orientation has a large effect on anisotropy generating different mechanical properties depending on the manufacturing process; for this reason, strength, and energy released during failure were highly dependent on the print orientation. Generally, the fracture is brittle when the plane that contains the interface is printed parallel to the platform, and ductile if it was printed orthogonal with respect to the platform. Vu et al., [129] studied the print orientation effect on sandwich specimens and fracture energy with Jetting Additive Manufacturing, by specimens with an elastomer core and a cover manufactured with a glassy photopolymer. In the work of Boopathy et al., the authors manufactured sandwich samples stacking 2, 3, and 5 layers of two different polymers to assess the absorption capability under dynamic and static conditions [130].

Each 3D printing technology has advantages and disadvantages concerning the bonding process. The bonding of thermoset polymers with elastomers using PolyJet is well documented [93,113,126–132] (Table 5). Because PolyJet is a Stratasys Patent, part of the composition of the materials used is confidential. The use of multi-materials in manufacturing processes can give materials properties not inherent to their nature; for example, Wang et al. [113] made cubic samples of periodic morphologies (Fig. 9b); In comparison with elastomer samples, adding the thermoset in such structures enhanced stiffness, energy dissipation, and ultimate compressive strength. The use of MMAM is not restricted only to improving the mechanical properties of materials, through the addition of other materials that complement their properties, but it can be used to obtain designs that provide rigidity and flexibility in the same part. Bartlett et al. [132] manufactured a robot protective shield in a single printing step. The design was formed by a thermoset shield on the electronic core and a structure of elastomer in the exterior for safe human interaction.

The use of DIW with different polymers is widely used [43] even graded compositions. Special applications are successfully manufactured in multicomponent graphene-based electrodes for electrical energy storage (EES) applications [46], or flexible electrochemical biosensors [133], these two examples expose the versatility of this technology. Mansouri et al. [134] manufactured cubic samples with the triply periodic minimal surface method (TPMS method), similar to the work carried out by [113,131] using PolyJet technology. The authors combined a hard phase (TPU) with a soft phase (commercial designation Bayblend). The results showed that stability at compression tests was higher than samples created only with the soft phase, increasing strain recovery during unloading. Additionally, Table 6 shows a qualitative synthesis of works regarding some applications and other parameters within the category approached.

3.3. Strength of different multi-material parts

3.3.1. Strength of FFF multi-material parts

The multi-material adhesion is not limited to aesthetic purposes but is employed in functionality components with structural responsibilities. In polymers, the lack of chemical affinity undergoes a reduction of the strength that could be solved with a correct design of the interface

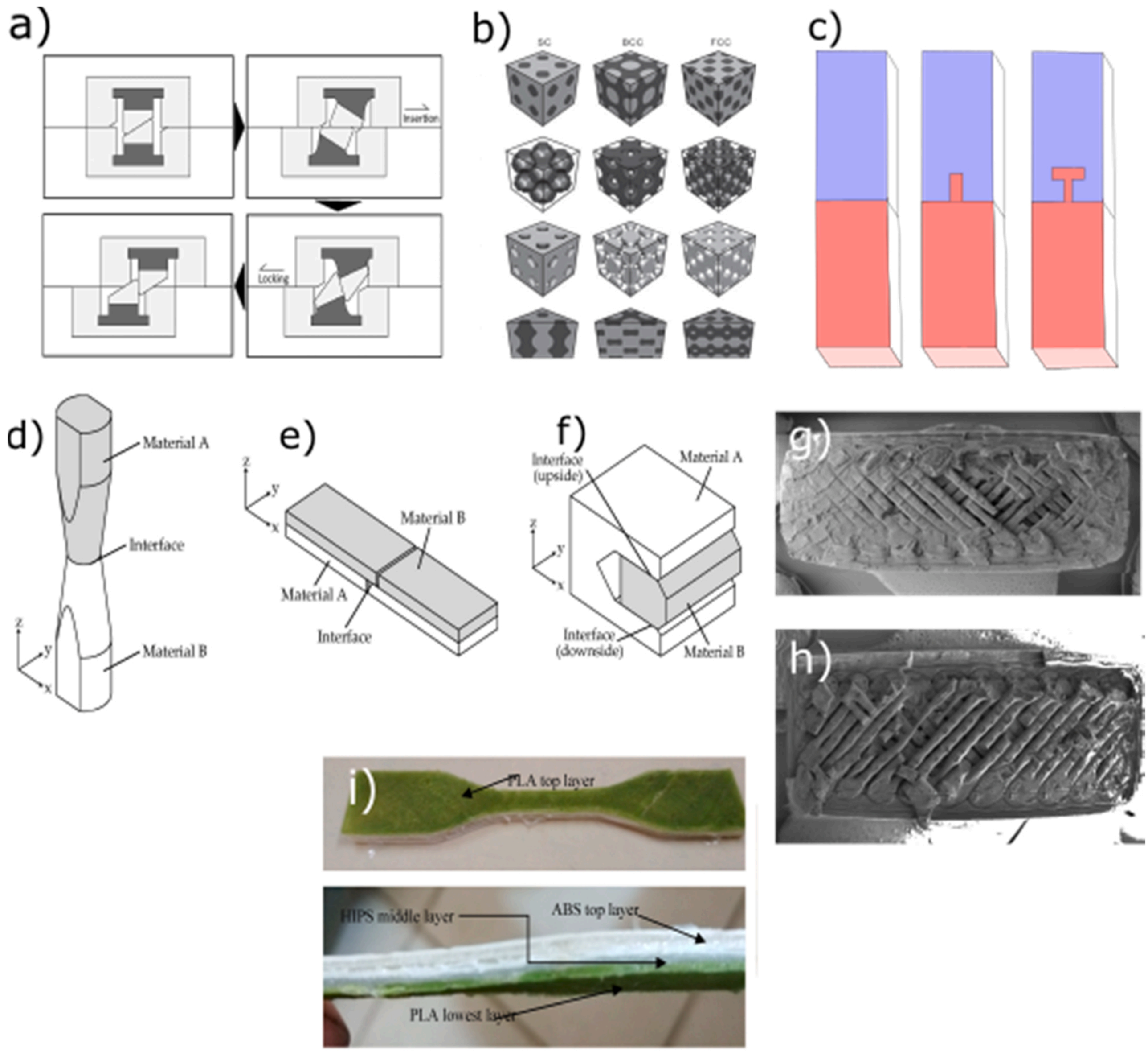

Fig. 9. (a-i) MMAM test specimens and mechanisms.
a) Scheme of the ratchet mechanism proposed by [93], the mechanism causes an interlocking due to elastic deformation of the elastomer (Reprinted from with [93] permission from Elsevier); b) 2x2x2 unit cells of 3D co-continuous composites with different cubic lattice (SC-BCC-FCC) (Reprinted from [113] with permission from Wiley); c) Different interface geometry designs for tensile test specimens employing PLA-PLA and PLA-TPU material pairs by [112]; d) tensile, e) Lap-Shear and f) compression-shear test specimens (4d-f) were reprinted for free with no permission required under http://creativecommons.org/licenses/by/4.0/ article doi: 10.3390/app8081220); g) Fracture surface of PCL-PLA samples generated with a bonding layer transition of 150ºC; h) Fracture surface of PCL-PLA sample generated with a bonding layer transition of 90ºC.; i) PLA-ABS-HIPS Multi-material specimen sample tested according to ASTM 638 type IV(reprinted for free with no permission required under http://creativecommons.org/licenses/by/4.0/ article doi: 10.3390/polym11010062).

between different materials [135]. However, by creating mechanical interlocking at interfaces, as shown in Fig. 9c, the drop in the tensile strength can be diminished, making these interface designs highly recommended in high strength structural applications [112]. The Standard DIN 53504 tensile test specimen is the most commonly used criteria for evaluating strength resistance in multi-material polymers. Other authors [10,136,137], proposed the standard ASTM D638 to evaluate tensile test specimens analyzing the impact of processing parameters on the interfacial bonding strength. Authors such as Watschke et al., [138] developed three different multi-material test specimens by FFF to create standard designs for tensile (Fig. 9d), lap-shear (Fig. 9e), and compression shear (Fig. 9 f) tests. The mentioned studies are based on abrupt interfaces between dissimilar materials, instead of smoothed transitions using gradient zones by static intermixes. Khondoker et al. [36] obtained an increase of 35% of strength using standard ASTM D1708 tensile tests combining PLA, ABS, and HIPS. The studies that use FFF technology exclusively are listed in Table 7. To improve the inter-layer and inter-filament strength in polymers, the authors have focused their efforts on the extrusion temperature due to the increase of molecular diffusión processes and wettability in polymers [96]. For the manufacture of parts with polymers that have large differences in melting point, the effect of shrinkage during solidification must be taken into account, to avoid brittle fracture in the interface bonding and obtain a final strength of the part similar to the softer materials that compose the part. An excellent methodology is trying to find an optimal extrusion temperature for the two materials [139]. Fig. 9g-h, shows a FESEM image of the fracture surface of the PCL-PLA bonding layer with a temperature of 150 °C (Fig. 9 g) and temperature of 90 °C (Fig. 9 h). The increase in temperature enhances the material diffusion between bonding layers. The authors concluded that a temperature of 130 °C for the PCL interface layer obtained the best results. Tensile specimens of three different recycled polymers: ABS, PLA, and HIPS, were printed by Singh et al. [137]. The specimens' final properties were a mixture of the three materials since each material was printed over the previous one, and the three were printed along the entire length of the specimen, Fig. 9i shows a sample tested. Additionally, the authors observed that raising the printing speed and the infill density improve the parts' mechanical properties. In this sense, dos Santos JF Amancio-Filho ST FRGS [140] uses a direct FFF printing of ABS and PA6/PA6 with carbon fibers over an aluminum 2024-T3. The use of multiple or single extruders in FFF has no effect on the final strength of the parts but the use of multiple extruders reduces the processing time [141]. However, with a single extruder, there are no problems related to the nozzle head's calibration and positioning. Afshar and Wood developed a new methodology to create coated parts with a weather-resistant material in a single operation [142]. The authors print acrylic-butadiene-styrene (ABS) as a substrate material and acrylic-styrene-acrylonitrile (ASA) as a coating material. The coating improves the structural integrity of ABS against aggressive environments.

3.3.2. Strength of CFR multi-material parts

The efforts to improve the mechanical performance in reinforced parts with continuous fibers has been focused in pre-impregnated filament technology. Several authors have employed the Markforged® system to evaluate the mechanical properties under different fiber/matrix configurations and load cases, due to the robust manufacturing process of this 3D printer family [143,144], the analysis of continuous carbon fibers is the most used material due to the excellent ratio stiffness/weight compared with kevlar or Fiberglass that shows lower strength and stiffness but are more economical, as seen in Table 8 [53,145].

Different papers has evaluated the properties of carbon, fiberglass, and kevlar reinforcement under tension [53,143,146], compression [147–149], bending [150], impact [151], creep [152], buckling [153], and fatigue [154] during the last years. Fractography analysis is the most used way to evaluate and judge the failure mode, employing optical microscopy or SEM methods [3]. In tension, the dominant failure mode is the fibre pull-out Fig. 10b and Fig. 11a, where the fibers carry out most of the load, and the bonding interface between fiber/matrix undergoes a lack of adhesion. In this load case, the fracture is completely unstable, and the parts do not undergo plasticity. Generally, the fibre pull-out is the most common failure mechanism, to avoid this failure is mandatory to enhance the bond strength between fiber/matrix and the proper wettability of the fibre. Significant differences can be observed in shear tests, where specimens develop plasticity. Fig. 10a and Fig. 11b, and most of the load in the plastic range is carried out by the polymeric material. As a result, the fibers are not clean of matrix polymer, denoting plastic work.

The fiber layer distribution, angle, and the number of layers have a large influence on the strength. A uniform distribution of the fiber layers in the specimen's thickness increases the strength due to equalized distribution of loads that tend to reduce the stress concentrations [155]. In the case of the angle of the fiber disposition in each layer, it is well documented that orientation with the load applied maximized the strength and stiffness of the part [156]. Generally, a compromise in the orientation between tensile (0º) and shear (45º) must be obtained in major industrial applications. The number of layers has a direct influence on the strength and stiffness, increasing with the % of the fiber in the loading direction [157].

Table 8
Material properties of the continuous fiber reinforcement and matrix polymer from Markforged® 2021. Sorted in increase price.

Material	UTS (MPa)	E (GPa)	Tensile strain at break (%)	Density (g/cc)	price ($/cc)
Nylon	36	1.4	150	1.1	0.21
Onyx	51	3.6	25	1.18	0.23
GF	600	21	3.9	1.5	1.53
KF	610	27	2.7	1.2	2
CF	800	60	1.5	1.4	3

4. Discussion on applications, properties of parts, and parameters in MMAM

The major articles reviewed denote that the most significant issue concerning the polymer multi-material printed parts is the weak mechanical properties obtained at the interface bonding. The bonding strength of polymeric parts, specifically for those manufactured by FFF, continues to be under discussion. By using solid and immiscible materials, the intermolecular diffusion is reduced and the interfaces obtained are usually abrupt, causing physical discontinuities and reducing the mechanical properties even when the interface is generated between two parts of the same material. The use of interlocks and temperature adjustments between dissimilar materials increases the resistance of the interface. As a consequence, the correct design of interface geometry that allows mechanical interlocking, so that chemical forces are not the only ones that work, becomes a key factor in multi-material polymers FFF. The interface bond strength is a topic highly discussed by the research community. Nevertheless, although there are combinations of printing parameters that increase it, the final value of the bond strength is lower than the base material; In contrast with the FFF technology, the parts manufactured by polyjet do not present as many bonding problems at the interface. Although it has been proven that the strength of the joint is weighted by the strength of the assembly, a design with gradients is possible, allowing a stronger bond between the materials. In any case, the general agreement about the strength of bond interfaces between dissimilar materials is that it seems to resist compression tests better than tensile tests, regardless of the technology employed. Given that our findings are based on the fact that it is in the tensile stresses that the chemical bonds generated between the layers act to a greater extent, and the phenomenon of delamination appears between adjacent materials.

In the case of DIW, the combination of materials with highly dissimilar properties is limited, for this reason, the formulations with similar composition and rheology are in big demand. This is the major challenge in this technology to obtain truly multi-material parts. Continuous fiber embedment into the plastic matrix of 3d printed model is a recent innovation and is the base of the next generation of new composites, due to its significant contribution to the improvement of mechanical properties such as flexural, shear, and tensile strength. However new experimental applications are developing now, the commercial application is very narrow, due to low interfacial bonding strength, a reduced number of allowable materials (reinforcement and materials), elevated porosity, and highly anisotropic behavior.

5. Conclusions and future challenges

In the present paper, a compilation and review of scientific research about MMAM of polymers have been carried out. Conventional additive manufacturing is a critical technology in Industry 4.0. Further, MMAM adds new advantages to conventional additive manufacturing that are essential in the modern production paradigm. The collected articles have been extracted from different online databases. They have been classified into the development of systems that allow multiple printing, the study of the printing parameters, and the manufactured parts' properties. From all the studies treated, the most helpful information can be summarized in the following observations:

System development.

- The software and hardware limitations of printing with more than one material have been overcome in recent years. There are already

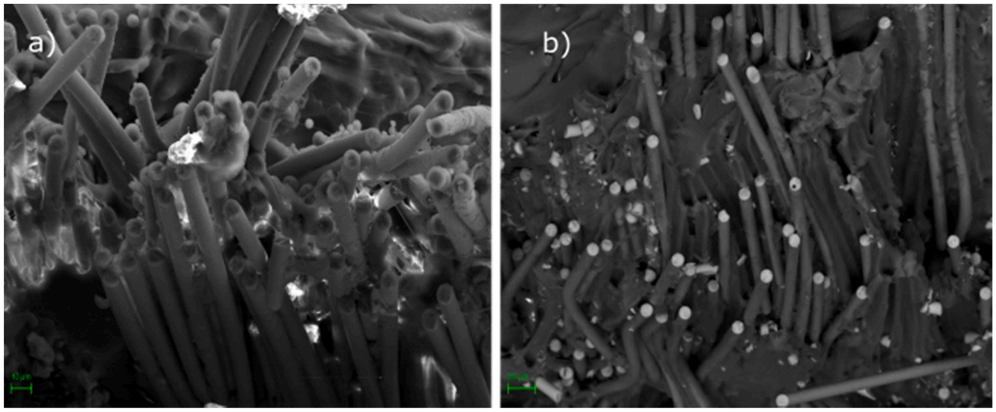

Fig. 10. a) Image of fiberglass breakage in a Shear-test specimen, the fibers adhering to the matrix indicated a ductile fracture; b) Pull-out fracture of fiberglass in a tension specimen.

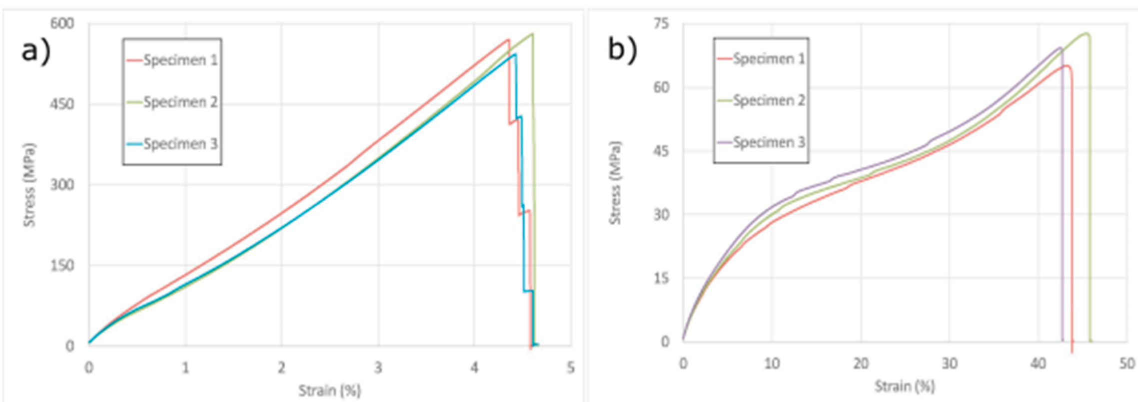

Fig. 11. a) Fiberglass stress-strain curves of the tension test, and b) in-plane-shear test.
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multiple commercial FFF, PolyJet, and DIW systems that use various materials during printing. However, there is a lack of sound commercial stereolithography systems.

- Although the systems and procedures that enable MMAM have already been largely developed, it is necessary to increase the investigation in this field in light of recent events. The 3D printing of hybrid metal-polymer parts is a promising topic, and it is essential to investigate more methods that improve the bond strength of the parts.

Printing parameters and part properties.

- The optimization of one process cannot be generalized to others, so it is necessary to study the bonding forces obtained between each pair of materials and consider the technology and parameters employed for the joint. Likewise, it is possible to continue investigating the combinations and morphologies that obtain the best mechanical performance, and apply more types of tests, since most studies only carry out tensile tests.

- When addressing the properties achieved with parts manufactured by MMAM, the weakness induced at the interface bonding between materials during the manufacture is the most discussed issue. In the case of polymers, specifically with FFF, molecular interdiffusion is usually poor, and the chemical bond weak, so mechanical interlocking characteristics are necessary to strengthen the bonds.
- Parts manufactured with a continuous gradient between dissimilar materials, rather than abrupt interfaces, tend to have better mechanical performance and improved bond strength than parts manufactured with discrete gradients.
- It́ is well accepted that CFR improves the mechanical properties of 3D printed parts. However, meso and microstructural studies evidence poor mechanical performance compared to the traditionally prepared composites. For this reason, cost, time, and scalability are big concerns.

This work has discussed the feasibility of implementing new designs in a wide range of applications by MMAM. Although the weaknesses generated in the interface bonding reduce the resistance of the whole part, in many cases, the resultant resistance is enough to meet the requirements imposed by design. Furthermore, considering the many practical advantages/disadvantages of using MMAM over conventional techniques, we conclude that the substitution of conventional AM by MMAM is usually beneficial.

In our opinion, one of the future challenges for MMAM is standardization. The existence of design guides for multi-material components in additive manufacturing is still scarce. Some papers address this challenge, creating the processes planning and material databases to facilitate the component design. However, the research community still demands the creation of global and standardized design guides. In order to advance in this area, more research is needed to address the relevant issues of printing with certain combinations of materials and parameters.

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CRediT authorship contribution statement

A. García-Collado: Conceptualization, Writing – original draft, Writing – review & editing, Formal analysis. **J.M. Blanco:** Writing – original draft, Software, Resources, Investigation. **Munish Kumar Gupta:** Conceptualization, Data curation, Formal analysis, Validation, Visualization, Writing – original draft, Writing – review & editing. **R. Dorado-Vicente:** Writing – review & editing, Supervision, Software, Resources, Project administration, Methodology, Investigation, Funding acquisition.

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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