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# Multi-material additive manufacturing: A systematic review of design, properties, applications, challenges, and 3D printing of materials and cellular metamaterials

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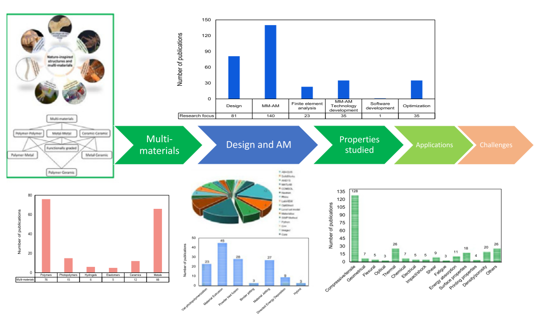
## HIGHLIGHTS

- Recent developments in multi-material additive manufacturing (MMAM) were comprehensively reviewed.
- The applications of MMAM were discussed regarding various materials and AM methods.
- The aspect of MMAM design, modeling, post-processing, and analysis was summarized.
- Limitations, challenges, future trends of MMAM were highlighted.

## GRAPHICAL ABSTRACT

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## ABSTRACT

Extensive research on nature-inspired cellular metamaterials has globally inspired innovations using single material and limited multifunctionality. Additive manufacturing (AM) of intricate geometries using multi-materials provides additional functionality, environmental adaptation, and improved mechanical properties. Recently, several studies have been conducted on multi-material additive manufacturing

*Abbreviations:* ABS, Acrylonitrile Butadiene Styrene; ASA, Acrylonitrile Styrene Acrylate; AJ, Aerosol jetting; AF, Aramid fibre; AM, additive manufacturing; BJ, Binder jetting; $B_4C$, Boron carbide; CAD, Computer-aided design; CaP, Calcium phosphate; CFD, Computational fluid dynamics; CPE, Chlorinated polyethylene; DED, Direct energy deposition; DLD, Direct laser deposition; DMD, Direct metal deposition; DEM, Discrete element method; DMLS, Direct metal laser sintering; DfAM, Design for AM; DIW, Direct ink writing; DLP, Digital light processing; EAP, Electroactive polymer; EPBF, Electron beam powder bed fusion; FEM, Finite element method; FFA, Flexible fluidic actuation; FDM, Fused deposition modelling; FFF, Fused filament fabrication; FGM, functionally graded materials; GelMA, Gelatin methacryloyl; HA, Hydroxyapatite; HIPS, High Impact Polystyrene; IJ, Inkjet; ISS, International Space Station; IN718, Inconel 718; LENS, Laser Engineered Net Shaping; LOM, Laminated object manufacturing; LPBF, Laser powder bed fusion; LMD, Laser metal deposition; LBF, Laser based fusion; MD, Molecular dynamics; ME, Material Extrusion; MJ, Material jetting; MJF, Multi Jet Fusion; MM, multi-material; MMAM, multi-material additive manufacturing; MMFGAM, Multi-material functionally graded additive manufacturing; OLED, Organic light emitting diode; PBF, Powder bed fusion; PneuNet, Pneumatic network; PLA, Polylactic acid; PETG, Polyethylene terephthalate glycol; PJ, PolyJet; PEEK, Polyether ether ketone; PET, Polyethylene terephthalate; PC, Polycarbonate; PCL, Polycaprolactone; PVA, Polyvinyl alcohol; PEGDA, Poly(ethylene glycol) diacrylate; SLA, Stereolithography; SiC, Silicon carbide; SCF, Short carbon fiber; SMMs, Shape-memory materials; TPU, Thermoplastic polyurethane; TCP, Tricalcium phosphate; TPE, Thermoplastic Elastomer; SLS, Selective laser sintering; VW, Vero White Plus; WoS, Web of Science.

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(MMAM) technologies, including multi-materials, methodologies, design, and optimization. However, in the past six years, very few or no systematic and complete reviews have been conducted in this research domain. This review intends to comprehensively summarize MMAM systems and the working principles of its fundamental processes. Herein, the Multi-material combinations and their design, modeling, and analysis strategies have been reviewed systematically. In particular, the focus is on applications and opportunities for using MMAM for several industries and postprocessing MMAM fabricated parts. Furthermore, this review identified the limitations and challenges of existing software packages, MMAM processes, materials, and joining mechanisms, especially at the multi-material interfaces. Finally, we discuss the possible strategies to overcome the aforementioned technological challenges and state the future directions, which will provide insights to researchers and engineers designing and manufacturing complex nature-inspired objects.



## 1. Introduction and background overview

Nature demonstrates incredibly optimized design and fabrication by selecting application-specific combinations of compositional and functional gradation of both materials and structures. Several examples of nature-inspired structures and multi-materials (MMs) for inspiration of designing cellular metamaterials and multi-material additive manufacturing (MMAM) are collectively illustrated in Fig. 1. As captioned in the subsets of Fig. 1, these exemplary cases demonstrate excellent performance by combining the advantages of multiple materials with functionally graded structures. However, the objects designed by humans exhibit simple structures and are manufactured using single materials. Thus, they are not completely optimized for specific applications because of the unique challenges and limitations in design and conventional manufacturing methods [1]. As additive manufacturing (AM) and Design for AM (DfAM) provides unlimited freedom of design and layer-by-layer or particle-by-particle deposition of material, they are deemed as the most potential candidate for addressing the aforementioned challenges [2-4]. Moreover, the recent advancements in AM techniques have enabled the fabrication of MMs and compositions with functionally graded structures in a single component, thereby providing a design and manufacturing platform for imitating nature-inspired MMs and structures.

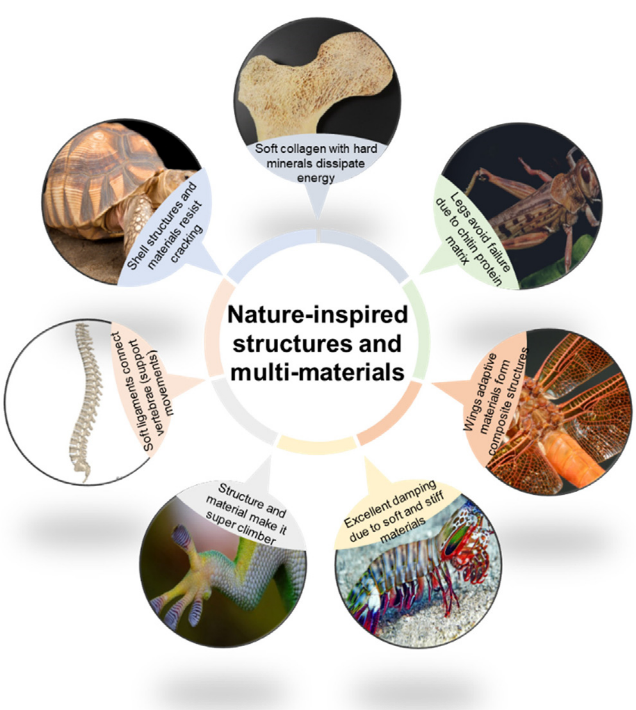


**Fig. 1.** Nature-inspired structures and multi-materials (with composition and structural variations) inspire the design of cellular metamaterials and multi-material additive manufacturing.

In a recent study published in Nature Physics, Yasuga et al. [5] developed a two-step platform—FLUID3EAMS—that can create MM structures for scaffolds and several other applications. They validated the process by fabricating various structures, e.g., extremely soft tissue-like architecture. Inspired by nature, Mirzaali et al. [6] designed bioinspired functionally graded materials (FGM) to investigate the deformation and fracture behaviors of continuous and hard–soft composites, which offer numerous applications in high-tech industries such as aerospace and soft robotics. In several other studies, the nature-inspired multi-material fabrication concept has been adopted to design and additively manufacture objects for numerous applications, such as energy harvesting [7], soft sensors [8], lightweight automotive parts [9], and denture 3D printing [10]. In the last six years, over 200 Scopus and SCI-indexed research articles have been published in the research domain of MMAM.

Based on the significance and progress of MMAM research, several reviews have been reported on specific aspects. However, a complete and systematic review encompassing the advancements in this research domain in the past six years is yet to be prepared. Wang et al. reviewed the interface characteristics, manufacturing challenges, and applications of MM structures fabricated using the laser powder-bed fusion (LPBF) process [11]. Goh et al. reviewed MMAM and the 3D printing process of electronics by exploring the fabrication techniques, design considerations, and challenges of multilayered electronics [12]. In the context of recent advancements in technologies and applications, Collado et al. [13] and Zheng et al. [14] reported a systematic review of MMAM of polymer materials. Wei and Li (2021) reviewed the MM powder-deposition mechanism, molten pool behavior, and process parameters of LPBF [15]. Zhang et al. [16] and Loh et al. [17] reported progress in design, modeling, and AM technologies capable of fabricating FGM. Blanco et al. reviewed the applications of MMAM in the automotive and aeronautical industry [18]. Ravanbakhsh et al. comprehensively assessed the emerging technologies for MM bioprinting [19], following which Viola et al. [20] reported the progress on multimaterial biofabricated tissue structures in the same year. More recently, Mitchell et al. published a review on 4D printing [21]. Ha and Lu reviewed various bioinspired materials and structures for energy absorption applications [22]. Earlier, the AM of MM structures was reviewed by Bandyopadhyay and Heer [23].

To the best of our knowledge, the progress of MMAM in the recent years has not yet been comprehensively reviewed. Consequently, several aspects of MMAM have not been adequately discussed, such as MM design and analysis, recent technological advancements, design, and MM-related developments in finite element analysis software, post-processing, and strengthening methods for MMAM. Thus, the potential applications, technical issues, limitations of MMAM should be comprehensively discussed to ensure the sustainability of this technology.

This article aims to provide a comprehensive and systematic review of all the aspects of the research area of MMAM. Following the introduction and background overview provided in this section, the methodology and procedure of data acquirement and analysis are described in Section 2. The types of multi-materials, including metal–metal, metal–ceramics, polymer–polymer, and other combinations, are reviewed in Section 3. The multi-material modeling, numerical simulations of objects, and structures using MMs are elaborated on in Section 4. The AM processes capable of manufacturing multi-materials, research gaps, and limitations of these processes are discussed in Section 5. In Section 6, the post-processing of MM-manufactured objects are discussed. Thereafter, the MM applications and potential opportunities in several industries are illustrated in Section 7. The limitations and critical challenges of adopting MMAM are discussed in Section 8. Finally, the conclusion of this review and overall future trends in the MMAM research domain are summarized in Section 9.

## 2. Methodology

The literature search methodology adopted in this review article follows several literature search criteria, including literature inclusion and exclusion criteria, as listed in Table 1. An initial search of the keywords listed in Table 1 displayed 27,230 research publications, but not all studies were related to MMAM research. Thus, an inclusion–exclusion criterion was defined in which all publications were filtered to increase the relevancy and reduce the number of searched articles. Furthermore, in-depth reading and filtering were conducted on article titles, abstracts, keywords, conclusions, materials, and methods sections. Consequently, 205 articles relevant to MMAM were screened, which primarily focused on multi-material technology development, mechanical property investigation, types of material, design, and structure, and printing parameters.

The number of articles on MMAM published each year from 2017 to April 2022 is charted in Fig. 2, which indicates an increasing trend throughout the search period and implies the growing significance of MMAM research and development.

**Table 1**
Summary of literature search and inclusion–exclusion criteria included in present study.

| Literature searching criteria | |
|---|---|
| Language | English |
| Publication period | 2017 to 2022 |
| Date of search | May 2022 |
| Bibliography sources | Web of Science (WoS): Elsevier, Emerald, Wiley, Springer, Sage, Nature, JAMA, Taylor and Francis, Science, MDPI, ASME databases. |
| Search keywords | Dual materials, multimaterial, multi-material, multiple material additive manufacturing, multi-material 3D printing, multi-material additive manufacturing |
| Inclusion and exclusion criteria | |
| Publication type | Original research articles |
| Scope | Properties studied, printing parameters, material types, design and structure types, technology development. |

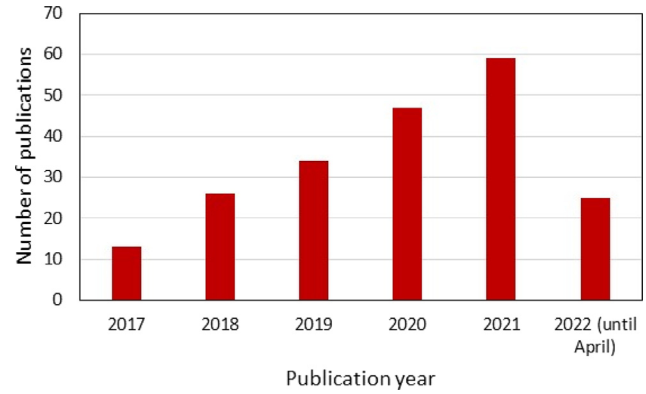


**Fig. 2.** Number of articles published annually on multi-material additive manufacturing from 2017 to April 2022.

## 3. Multi-materials for additive manufacturing

Nature offers excellent functionalities and performance by exploiting the combinations of excellent structures and compositions of materials [1]. Inspired by nature, such excellent personalized performance in engineered designs can be realized through emerging AM technologies, especially MMAM techniques that can enhance the performance of 3D printed components by increasing its functionality and complexity with the addition of multiple materials or varying compositions of the same material within a single component. Thus, MMAM offers several opportunities for designing complex, functional, highly personalized, and high-value products with improved properties, as depicted in Fig. 3. Simple and general combinations of the materials fabricated using a suitable AM process based on material types and the possible improvements in properties are presented in Fig. 3. This integration of materials at various scales can tailor the properties of the components, including electrical, thermal, mechanical, optical, and multifunctional properties [24,25].

As summarized in Fig. 4, almost all types of materials have been utilized in MMAM. The materials combinations being studied either pertain to the same category (e.g., polymer–polymer) or two distinct categories (e.g., polymer–metal). According to the analyzed literature, the two most widely investigated categories of materials include polymers and metals, studied in 76 and 66 publications, respectively. Among polymers, the most commonly investigated MM include PLA [26-39], ABS [32,39-46], PEEK [30,47,48], and PET [36] to improve strength, stability, functionality, etc. For metals, the majority of research on MMs has been conducted on alloys of titanium, copper, aluminum, and steels for improving the functionality of designed components as well as enhancing the characteristics such as hardness, conductivity, and magnetic properties, wear resistance, strength, and thermal performance. As elastomers, e.g., TPU [32,37,38,45] and hydrogels, are the least studied multi-materials. TPU is a difficult-to-print elastomer due to incomplete melting, material feeding issues challenges due to buckling, higher viscosity and melt strength, and incomplete integration between layers and rasters [49]. Moreover, precise AM of hydrogels is also difficult to achieve due to their large gelling temperature ranges, and low viscosity [50]. Therefore, most of the aspects of the combinations of these materials require further investigation.

The selection of a suitable MMAM method for manufacturing multifunctional components requires a comprehensive understanding of the advantages and limitations of each AM process, as described in Section 5. The fundamental concerns for MMAM include the successful delivery of multiple materials and the strong

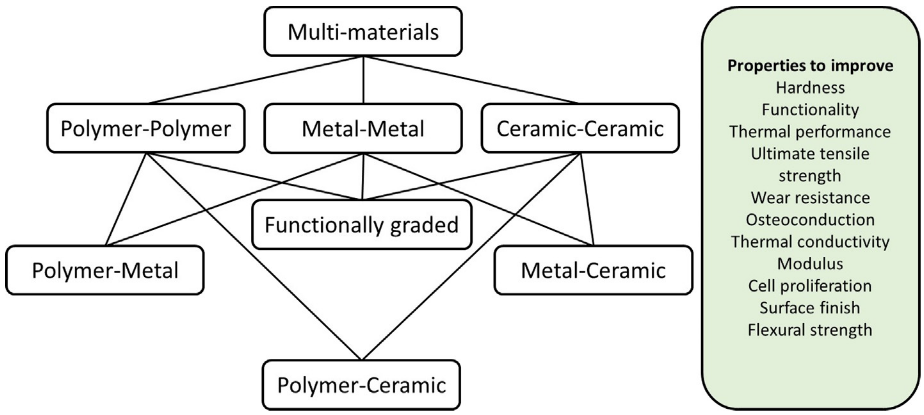


**Fig. 3.** General classification of Multi-materials and their respective improvements in properties.

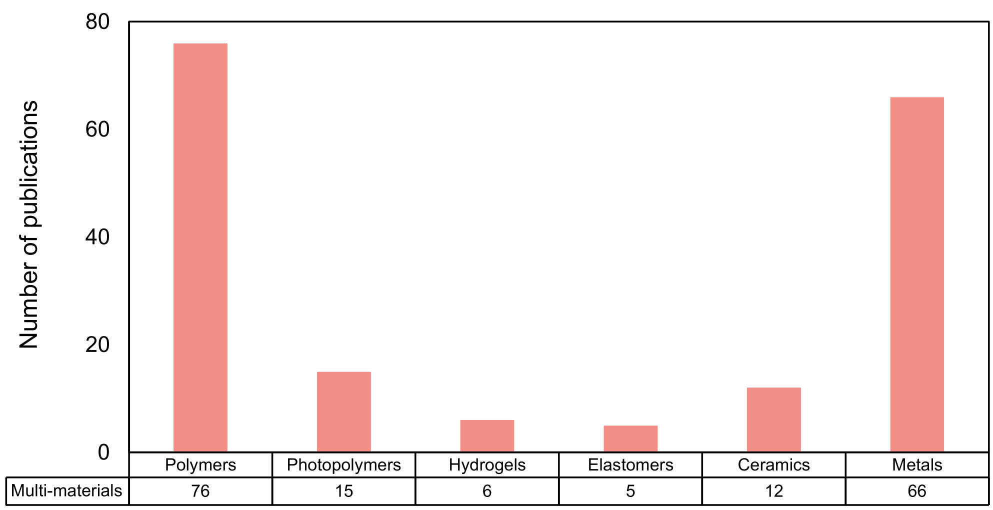


**Fig. 4.** Materials combinations utilized in additive manufacturing of multi-materials literature.

bonding between materials with various types and compositions. The possible variations in material types and compositions studied with the opted MMAM process are outlined in Table 2 and detailed in the following subsections.

### 3.1. Metal–metal

Although numerous AM methods exist for printing metals, most of them are limited to the fabrication of single-metal composition at a given time owing to machine-specific restrictions such as a single powder-feeding unit. Thus, a new powder-feeding design with multiple materials feeding sources can resolve this limitation. Otherwise, premixed powders can be used for printing multifunctional components (Fig. 5a) [119]. Other material- and process-related limitations include dimensional accuracies and post-process requirements. Overall, the major challenge is the bonding of dissimilar materials. Therefore, the compatibility of the MMs is an essential consideration for MMAM.

The PBF process was applied to fabricate 316 L/CuSn10 MM and enhance its structural and functional properties for applications in automobile, rail, and aviation industries [120]. The traditional fabrication of steel–copper MM components involve the following steps: welding, hot-rolling, and compound casting. However, MMAM can produce 316 L/CuSn10 components with improved mechanical properties in a single step under the optimized process condition that prevents the occurrence of cracks at the interface and allows lattice design, which is impossible with conventional manufacturing methods (Fig. 5b). To overcome the multiple powder feeding and weak bonding limitations during the LPBF process, Demir et al. proposed an in-house developed *in-situ* powder mixing unit to fabricate pure Fe, Fe/Al–12Si (volume ratio 55:45), and Al–12Si multi-material products using LPBF, which exhibited adequately strong bonding because of mixed Fe and Al–12Si composition acting as the interface and improved hardness [78]. Although Demir et al. obtained promising results, the processability of Fe/Al–12Si multi-material displayed a lack of dimensional accuracy (Fig. 5c). Wei et al. studied two metallurgically compatible $Ti_5Al_{2.5}$-Sn and $Ti_6Al_4V$ processing using LPBF [75]. A defect-free interface with an elemental diffusion zone exhibited good-quality bonding. Nonetheless, Scaramuccia et al. reported severe cracking between $Ti_6Al_4V$ and IN718, indicating metallurgical incompatibility that results in brittle intermetallic phase formation [87].

Among other MMAM methods, the directed-energy deposition (DED) process has garnered considerable research attention owing to its capability for manufacturing large-scale components with a high building rate and no requirement for powder recycling (Fig. 6a). Moreover, multiple or mixed powder-feeding systems are possible (Fig. 6a&b). Therefore, SS316L/IN718 (Inconel 718) multi-material components have been fabricated by DED to combine the high strength of IN718 and adequately high ductility of SS316L in a single product [79]. The results revealed the brittleness of the obtained IN718, which conveniently facilitated crack

**Table 2**
MMAM materials and their compositions with applied AM processes and properties studied in analyzed literature.

| Material composition | AM process | Properties studied | Refs. | Material composition | AM process | Properties studied | Refs. |
|---|---|---|---|---|---|---|---|
| Polymer–polymer | | | | | | | |
| -ABS<br>-TPU | FDM | -Interfacial bonding | [41] | -PLA<br>-ABS | FDM | -Tensile | [51] |
| -Nylon<br>-Carbon fiber | FFF | -Thermal<br>-Mechanical | [30] | -PLA<br>-Nylon | FFF | -Fracture toughness | [52,53] |
| -PC/ABS<br>-PE | FFF | -Fracture toughness | [54] | -PLA<br>-TPU<br>-PET | FFF | -Impact resistance | [36] |
| -ABS – HIPS<br>-ABS – PLA<br>-PLA – HIPS<br>-ABS – HIPS – PLA | FDM | -Tensile | [32] | -PCL<br>-PLA | FDM | -Printability<br>-Porous scaffolds | [55] |
| -Difunctional urethane acrylate oligomer | DLP | -Modulus<br>-Ultimate strength<br>-Elongation | [56] | -Nylon<br>-PVA | ME | -Thermal expansion<br>-Strength/ stiffness<br>-Thermal stress | [57] |
| -Silicones | SLA | -Shore hardness | [58] | -Aramid fibre (AF)<br>–PA12 | MJF | -Ultimate tensile strength<br>-Young's modulus | [59] |
| -TPE<br>-PA12 | SLS<br>MJF | -Poisson's ratio<br>-Thermal expansion | [60] | -PEEK<br>-CFR-PEEK | FFF | -compression | [47] |
| -PLA<br>-ABS<br>-HIPS | FDM | -Printability | [61] | -ABS<br>-TPU | FDM | -Orthosis design<br>-Printability | [62] |
| -PET<br>-polycarbonate (PC) | FDM | -Poisson's ratio<br>-Thermal deformation | [63] | -TPU - PLA<br>-PLA - CPE<br>-CPE - TPU | FDM | -Tensile | [26] |
| -Monomers<br>- poly(ethylene glycol) diacrylate (PEGDA)<br>-initiators | SLA | -Young modulus<br>-Hardness | [64] | -Albumen /alginate/gelatin<br>-Hydrogel | ME | -Compressive and tensile strength | [65] |
| -ABS<br>-TPU | FDM | -Compressive and energy absorption | [42] | -Polymers<br>-Drying adhesives<br>-Thixotropic fluids | FDM | -Digital anisotropy | [66] |
| -alginate<br>-methylcellulose<br>-Sodium alginate/ Methylcellulose | PJ | -Printability<br>-Rheological properties<br>-Viscosity | [67] | -ABS<br>-Short carbon fiber (SCF) | FFF | Strength and stiffness | [45] |
| -UV curable<br>-silicone inks | ME | -Printability<br>-flow parameters to optimize the mixing process | [68] | -CF nylon<br>-Carbon PEEK-PLA | FFF | -Elastic stiffness<br>-Poisson's ratio<br>-Coefficient of thermal expansion | [30] |
| -PET film<br>-embedded piezoelectric elements | SLA | -Heterogeneous properties | [69] | -ABS<br>-carbon fiber | FDM | -Tensile Strength<br>-Modulus<br>-Porosity | [70] |
| -Resins of different mechanical property | SLA | -Mechanical properties<br>-Material waste<br>-Fabrication time | [10] | -PCL<br>-TCP | FDM | -Osteoconduction<br>-Bioactivity | [71] |
| -biopolymers<br>-hydrogels<br>-chitosan<br>-cellulose<br>-pectin | ME | -Shear thinning ratio<br>-Viscosity<br>-Ultimate tensile stress<br>-Elongation at break | [72] | -ABS<br>-polycarbonate (PC) | FDM | -Tensile | [44] |

(*continued on next page*)

**Table 2** (*continued*)

| Material composition | AM process | Properties studied | Refs. | Material composition | AM process | Properties studied | Refs. |
|---|---|---|---|---|---|---|---|
| -Agilus30 (FLX935)<br>-Vero Magnet (RGD852)) | MJ | -Compressive and tensile<br>-Geometric<br>-Dimensional accuracy analysis | [73] | -silicone rubber<br>-silicone/ethanol composite | Hybrid | -Density<br>-Tensile and tear strength<br>-Elongation<br>-Hardness<br>-Tensile stress and strain<br>-Stiffness | [74] |
| -Acrylonitrile Styrene Acrylate (ASA)<br>-PETG | FFF | -Ultimate stress<br>-Elastic<br>-Modulus<br>-Bonding strength<br>-Hardness<br>-Stiffness | [29] | | | | |
| <u>Metal–metal</u> | | | | | | | |
| -Ti5Al2.5Sn<br>-Ti6Al4V | LPBF | -Strength<br>-Interface bonding | [75] | -Al-12Si<br>-Al-3.5Cu-1.5 Mg-1Si | LBF | -Interface bonding<br>-Flexural<br>-Tensile<br>-Nanohardness | [76] |
| -Zinc<br>-Aluminum<br>-Copper | Fusion bonding | -Printability | [77] | -Fe<br>-Al-12Si | LPBF | -Crack propagation<br>-Microhardness | [78] |
| -SS316L<br>-Ti6Al4V | DED | -Ductility | [79] | -AlSi10Mg<br>-C18400 | LBF | -Tensile strength<br>-Flexural strength | [80] |
| -SS316<br>-SS430 | DED | -Magnetic property | [81] | −316L<br>-C18400 | LBF | -Metallurgical bonding<br>-Tensile strength<br>-Micro harness | [82] |
| -aluminum<br>-silicon | PBF | -Chemical etching | [83] | −316L<br>-CuSn10 | LPBF | -Printability<br>-Hardness | [84,85] |
| -Al<br>-Steel | – | -Young's modulus<br>-Physical density<br>-Poisson's ratio | [86] | -Ti6Al4 V<br>-IN718 | LPBF | -Defect formation<br>-Micro harness | [87] |
| -Al<br>-Steel | – | -Stress | [88] | -steel<br>-nickel<br>-copper | SLM | -Roughness<br>-Hardness | [89] |
| layered 50Cr6Ni2<br>-Stellite X-40 composite | DLD | -Wear resistance<br>-Corrosion resistance | [90] | | | | |
| <u>Metal–polymer</u> | | | | | | | |
| -SS316L<br>-Cu10Sn<br>-PLA | -LPBF<br>-ME | -bone implant interface | [91] | -ABS<br>-Fe<br>-Cu | FDM | -Thermal conductivity<br>-Tensile strength | [92] |
| -Cu-reinforced<br>-PLA | FDM | -Mechanical property | [93] | -Soft elastomer resin<br>-Conductive ink<br>-ABS<br>-Photocurable acrylate-based polymer | Hybrid | | [43] |
| -carbon-fibre thermoplastic composite<br>-aluminum adherent | Hybrid | -Shear deformation | [94] | -Bismuth telluride<br>-TE inks<br>-Chalcogenide metallic binders | Extrusion | Thermoelectric property | [95] |
| -Maraging steel<br>-PLA | ME | -Magnetic property | [96] | -Resin<br>-Metal | FFF | -Surface shape | [97] |
| −316L<br>-CuSn10<br>-PLA) | LBF | -Printability | [91] | -Metal<br>-Rubber<br>-Plastic | DIW | -Young's modulus<br>-Density<br>-Poisson's ratio<br>-Porosity | [98] |

**Table 2** (*continued*)

| Material composition | AM process | Properties studied | Refs. | Material composition | AM process | Properties studied | Refs. |
|---|---|---|---|---|---|---|---|
| −316L<br>-CuSn10<br>-PA11 | LBF | -Tensile<br>-Shear strength | [99] | | | | |
| <u>Ceramic–ceramic</u> | | | | | | | |
| -silica sand<br>-zircon sand | SLS | -Surface roughness<br>-Compactness | [100] | -Boron carbide ($B_4C$)<br>-Silicon carbide (SiC) | DIW | -Yield stress<br>-Viscosity<br>-Hardness<br>-Shear | [101] |
| <u>Metal–ceramic</u> | | | | | | | |
| -Al2O3<br>-Cu–O | LOM | -Mechanical property<br>-Electrical property | [102] | −1.2367 tool steel<br>-$ZrO_2$ + $Al_2O_3$ | SLM | -tensile strength<br>-adhesive strength | [103] |
| -Ti6Al4V<br>-TiB2 | LPBF | -Hardness<br>-Wear | [104] | -SiC<br>−316L | LBF | -surface roughness | [105] |
| -Cp-Ti<br>-CaP | LENS | -Wear | [106] | −16NCD13<br>-TiC | LC | -Hardness<br>-Defects analysis | [107] |
| -Fe<br>−30CaSiO3 | ME | -Osteosynthesis | [108] | -CpTi<br>-Zr | LENS | -Wear<br>-Cell Attachment<br>-Hardness | [109] |
| -zirconia–alumina<br>-steel | SLM | -Tensile strength<br>-Adhesive strength | [110] | -IN718<br>-Cu | LENS | -Thermal conductivity<br>-Diffusivity | [111] |
| -TiB2<br>-Ti6Al4V | SLM | -Nano hardness structural stabilities | [112] | -ceramics<br>($Bi_2Mo_2O_9$)<br>-Ag | selective laser burnout | -Compression -Density<br>-Porosity<br>-Viscoelastic properties | [113] |
| <u>Polymer–ceramic</u> | | | | | | | |
| -PEEK<br>-β-tricalcium phosphate (β-TCP)<br>-PEEK/β-TCP/PLLA | SLS | -Compressive thermal properties bioactivity,<br>-biodegradability<br>-cytocompatibility. | [48] | | | | |
| <u>Photopolymer–photopolymer</u> | | | | | | | |
| -TangoPlus (FLX980)<br>-rigid VeroClear (RGD810) | MJ | -Young's modulus<br>-Tensile strength and elongation at break. | [114] | -Vero Plus<br>-Agilus | PJ | -Flexibility and shock absorption | [115] |
| -acrylic-based photopolymers -Vero White Plus (VW) | MJ | -Young's modulus<br>-Fracture strength<br>-Ultimate tensile strength -Bond strength -Stiffness | [116] | | | | |
| <u>Polymer–wood</u> | | | | **<u>Polymer–glass</u>** | | | |
| -Wood<br>-polymer | Hybrid | -Rigidity<br>-Opacity<br>-Tensile strength<br>-Flexural strength<br>-Elongation<br>-Elastic modulus | [117] | -Polyethylene<br>-Glass fibre<br>-Carbon fibre or Kevlar<br>-Low-density polymer | MJ | -Impact-Transmitted force and penetration | [118] |

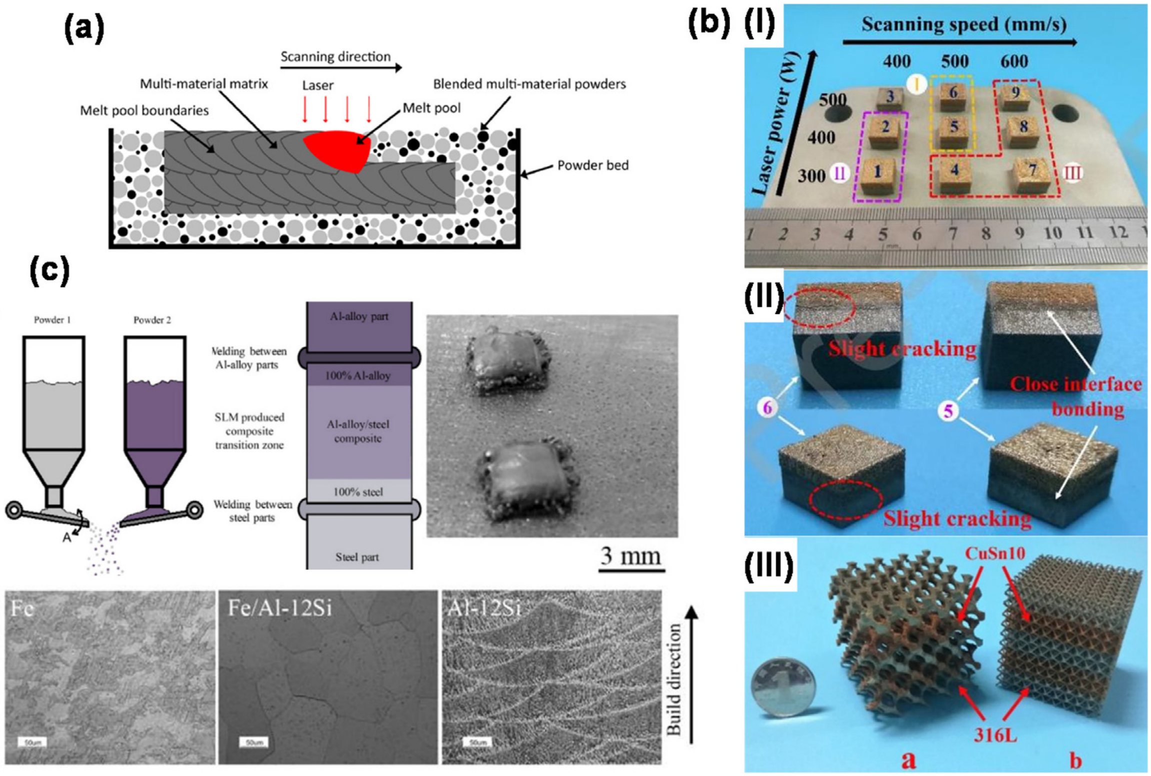


**Fig. 5.** (a) Illustration of LPBF processing premixed dissimilar powders [119], (b) process-parameter optimization of 316L/CuSn10 multi-material processed by LPBF prevents interface cracking under optimum process condition and successfully materializes multi-material lattice fabrication [120], (c) Fe/Al–12Si multi-material processing by LPBF exhibiting dimensional inaccuracy but sufficiently strong bonding [78].

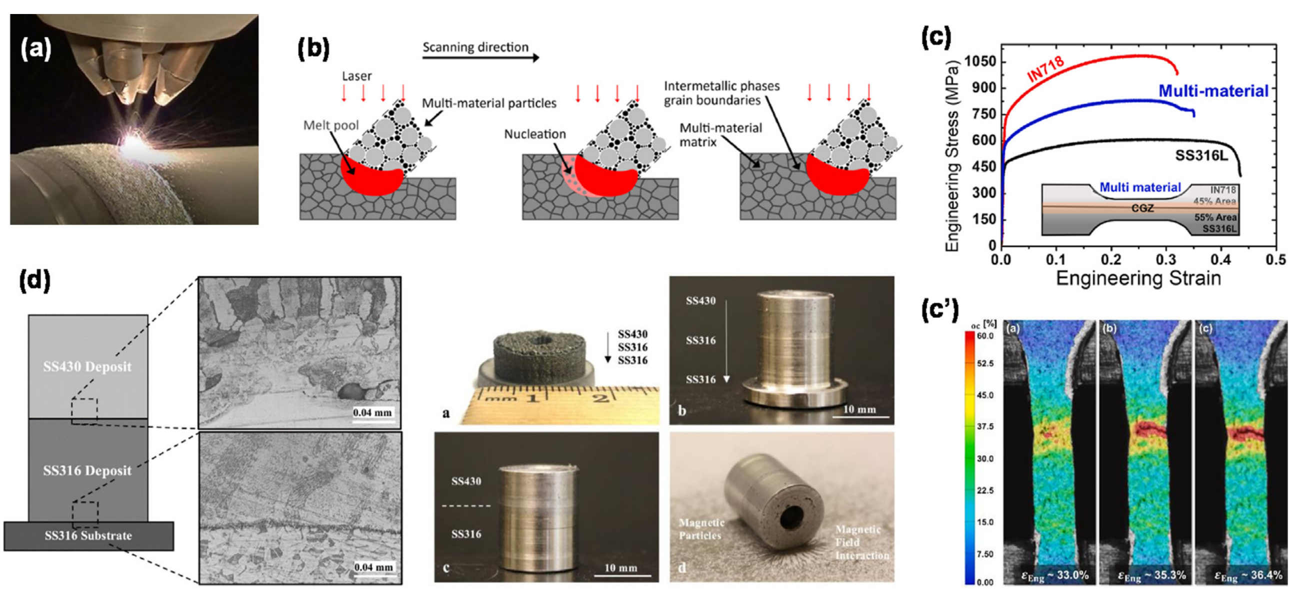


**Fig. 6.** (a, b) Schematics of multi-material DED processing [119], (c, c') tensile performance of DED processed SS316L/IN817 multi-material with corresponding stress distribution [79], (d) SS316/SS430 multi-material with selective magnetic functionality fabricated by LENS with adequate interface bonding [81].

growth. However, the other half of the product fabricated by SS316L manifested ductile properties and supported plastic strain during tensile deformation (Fig. 6c), representing the multifunctionality of the SS316L/IN718 MM component. LENS method with multiple powder-feeding system was applied to fabricate magnetic–nonmagnetic bimetallic structure with compositional grading to enhance the bonding between dissimilar metals (Fig. 6d), realize a single structure composed of stainless steel 316 (SS316) and SS430 with selective magnetic functionality [81].

### 3.2. Metal–ceramic

Currently, the MMAM with metals and ceramics are still in the developmental stage. Forming a strong bond between metals and ceramics is extremely challenging because of the high melting point mismatch and thermal expansion, which results in thermal stresses. At sufficiently high temperatures, the ceramic melt can attain flowability that causes metals to evaporate, creating difficulties in controlling the desired multi-material composition. Moreover, the thermal stress and significant difference in thermal expansion coefficients promote delamination or cracks between metals and ceramics, jeopardizing the mechanical strength and limits the structural application of the metal–ceramic MMAM components.

Laminated object manufacturing (LOM) is a sheet lamination method that can successfully fabricate metal–ceramic structures *via* its low-temperature processing (Fig. 7a) [119] and prevention of thermal stress-related limitations. The composite structure of $Al_2O_3$/Cu–O was fabricated using LOM with applied heat treatment. The results displayed dense material with appropriate mechanical and electrical properties, while ensuring reasonable bonding quality (Fig. 7b) and the fabrication of geometrically complex components such as gears (Fig. 7c) [102].

Recent studies have investigated a mixture of commercially pure Ti with 1.6 wt% Bromide (B) powder processed using LENS [121] and a mixture of $Ti_6Al_4V$ powders with 3 wt% $TiB_2$ powders processed using PBF [104]. Both MMAM processes improved the hardness and wear properties of titanium. Furthermore, LENS fabrications of $Ti_6Al_4V$ with 5 wt% hydroxyapatite (HA) [122] and Cp–Ti with 10% calcium phosphate (CaP) [106] were studied for biomedical applications. The results exhibited enhanced wear properties with the formation of the corresponding $CaTiO_3$ and $Ca(PO_4)_3$ phases as a tribological protection layer on the biomaterial surface. However, controlling the reaction phases, e.g., $Ti_5P_3$, $CaTiO_3$, and $Ca(PO_4)_3$, is crucial for the mechanical performance of biomedical components because these fragile phases induce vulnerability to crack due to the thermal differences in metal and ceramic. Furthermore, the material extrusion MMAM method was applied to fabricate lattice bone implants composed to Fe-30$CaSiO_3$ (in wt%) [108]. Fe-30$CaSiO_3$ biomaterial significantly improved the *in-vivo* osteosynthesis compared to pure-Fe lattice structure and exhibited promising results for bone cancer therapy (Fig. 8b).

### 3.3. Metal–polymer

AM processing of polymers has been extensively studied because of their low temperature and more manageable processing condition than metals, which are applied in medical, automotive, and aerospace industries [123]. Among various polymers, PLA has been extensively researched and applied by AM technologies as a filament material. The low working temperature of the FDM technique enables the use of materials with low melting temperatures, e.g., PLA [124]. Considering the advantages of PLA with the mechanical strength of metals, metal-polymer MMAM has been implemented for bio-inspired soft robot fabrication [125]. However, the insufficiently low process temperature fails to form a strong bonding between metals and polymers. Therefore, metal–polymer MMAM has primarily focused on using metal–polymer hybrid filament to enhance the properties of polymer materials. Herein, metals are considered fillers to improve the functionality of polymers. Nonetheless, the dearth of research on mechanical properties related to these polymer filaments incorporating various filler metals is concerning. Upon increasing the metal ratio in metal–polymer MMAM, shrinkage and pore formation emerge as the main limitations.

Kottasamy et al. studied Cu-reinforced PLA using the FDM method [93]. With 25 wt% Cu addition, Cu–PLA composites exhibited the most suitable mechanical properties and implied a possible alteration of the properties with the adjustment of the metal filler content. Diaz-Garcia used maraging steel as a filler for PLA to induce magnetic functionality within the multi-material fabricated *via* the extrusion AM method [96]. Upon increasing the metal ratio to 85 wt% with 15 wt% of the polymer [126], the products printed with the bronze–PLA filament exhibited approximately 20% shrinkage and significant porosity after sintering (Fig. 9a). Although the literature on metal–polymer MMAM is limited, a combination of the fused filament fabrication, extrusion-based AM method, and electroforming process has successfully demonstrated a metal–plastic multi-material structure [128].

As discussed earlier, the realization of metal–polymer bonding with a single AM process is challenging. Therefore, utilizing two distinct AM processes to manufacture metal–polymer structures is proposed (Fig. 9c&d) [91]. Several MMAM components composed of SS316L, Cu10Sn, and polymer (PLA, PET) were successfully fabricated through integrated extrusion and PBF methods, whereas poor metal–polymer bonding and surface quality posed a significant problem. However, the metal–polymer interface exhibited reliable strength with an interlocking design, indicating adequate infiltration of the polymer into both macroscopic design and microscopic mechanical structures.

The IN625 alloy fabricated using the binder jetting process with an intentional reaction during the debonding of polymer resulted in the formation of a strengthening Cr3C2 phase (Fig. 9b) at the polymer binder–metal powder interface [127]. This confirmed the significance of the reaction between the polymer and metal

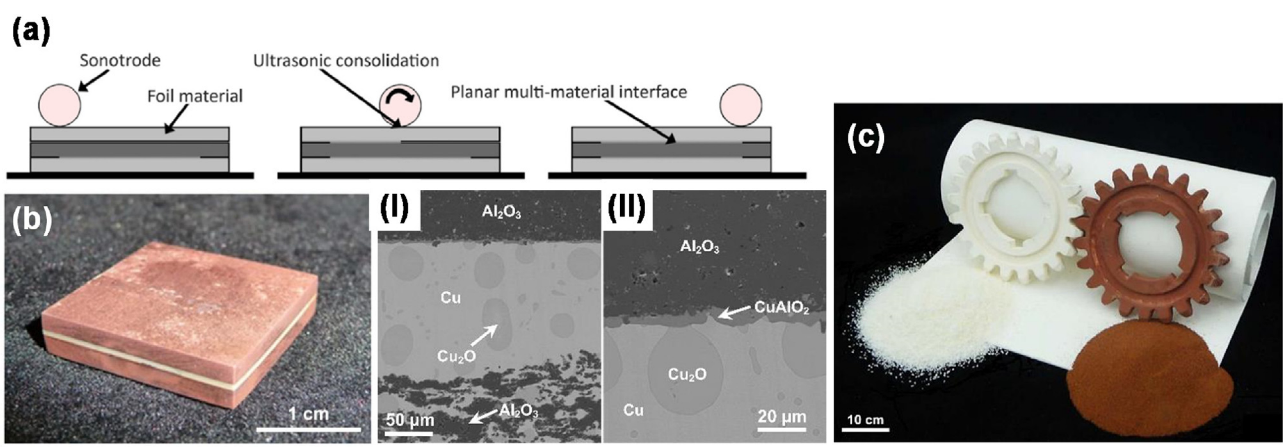


**Fig. 7.** (a) Illustration of multi-material sheet lamination process [119], (b) successful fabrication of $Al_2O_3$/Cu–O sandwich structure by sheet lamination process with interface observations, and (c) demonstrative gear shape fabrication [102].

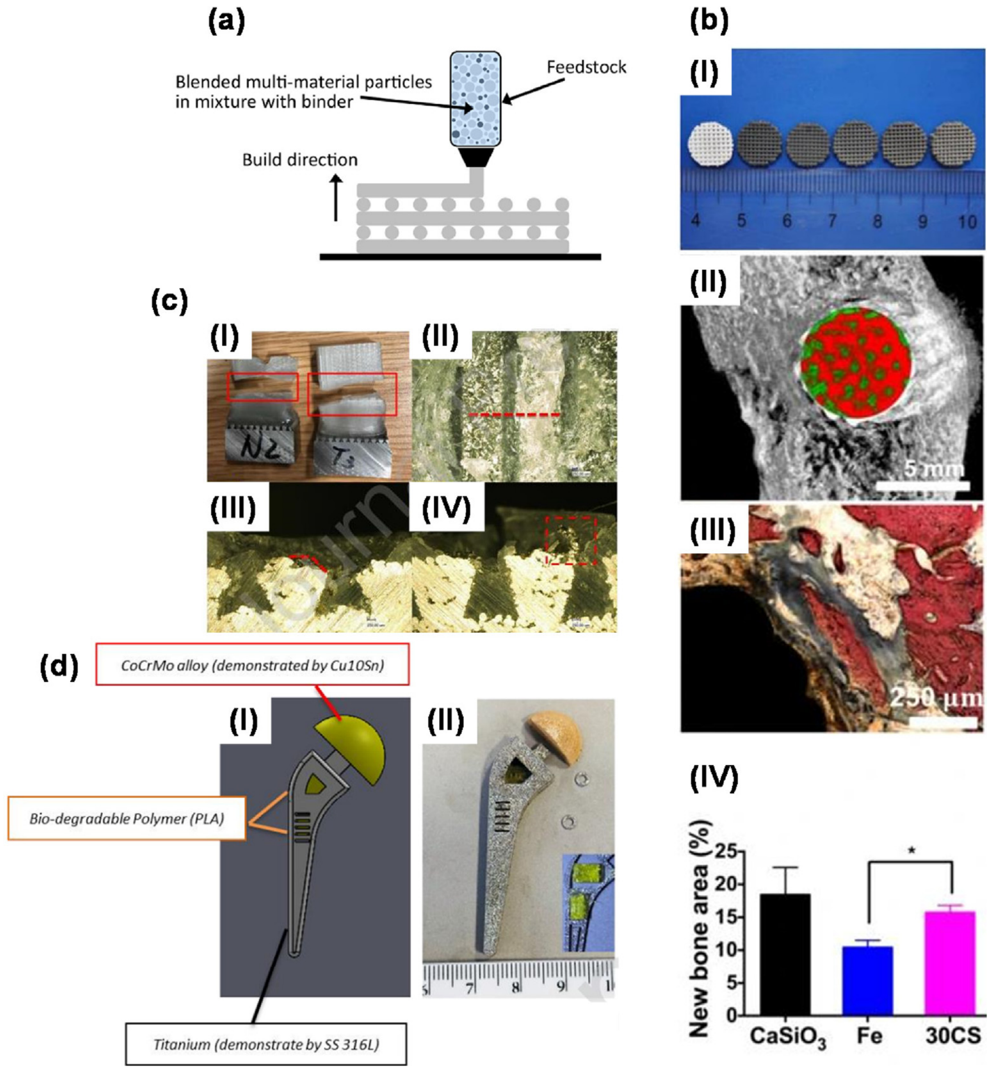


**Fig. 8.** (a) Illustration of multi-material processing of premixed powders by material extrusion [119], (b) 3D-printed scaffolds with gradient composition (left to right: $CaSiO_3$ 100%, 40%, 30%, 20%, 10%, and 100% Fe) and promising bone regeneration results of MMAM processed 30% CaSiO3/70% Fe scaffold [108], (c) MMAM processing metal–metal–polymer with mechanical interlock to enhance metal–polymer bonding and (d) its successful application to bone implant [91].

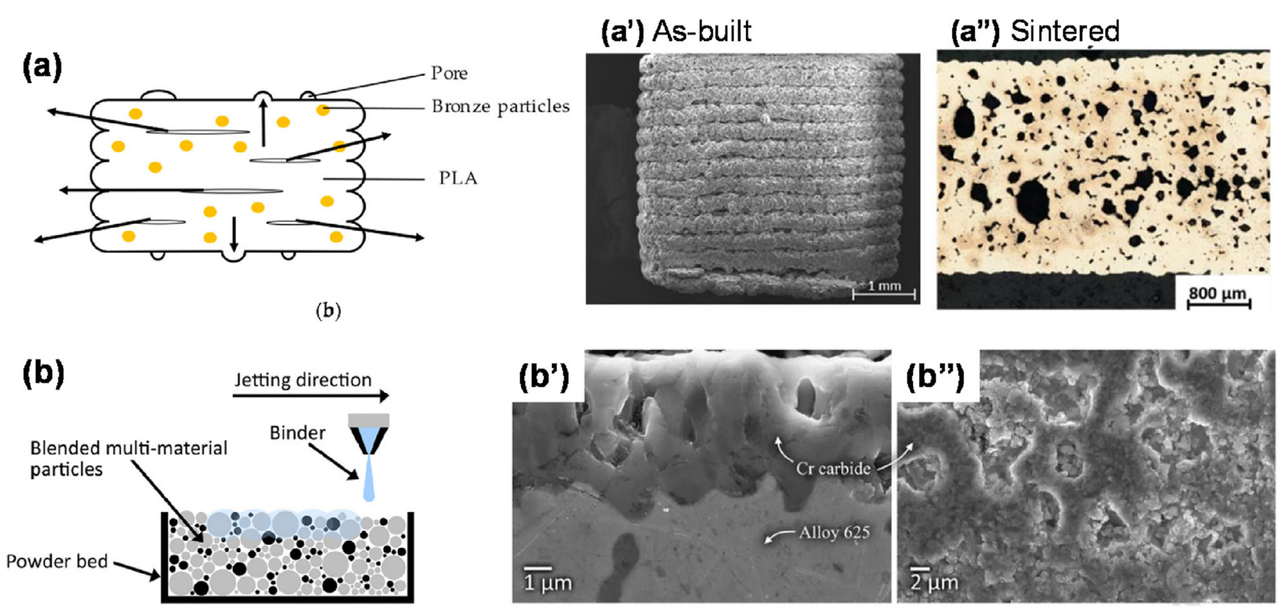


**Fig. 9.** (a, b) Illustration of metal-reinforced polymer extrusion and premixed multi-material binder jetting, respectively, with interface and porosity observations [126,127].

that can alter the properties of the final components. To achieve the goal of producing higher-quality metal components, future research should focus on optimizing the metal–polymer ratio, diminishing the differences in the melting points of metal–polymer MMs, and optimizing the sintering conditions to minimize the porosity of the printed components [129].

### 3.4. Polymer–polymer

In the context of the material extrusion AM, several materials have been used to fabricate multi-material yet monolithic components. The MMAM process can yield functionally gradient structures with multicolor regions/shades and spatial variations of mechanical, thermal, electrical, and optical properties. Depending on the application and requirement of the manufacturing process, various combinations of the polymers (refer to the related section of Table 2) can be observed in the analyzed literature. Thermoplastic polymer chains offer considerable challenges in mobilization *via* chemical reaction, which can facilely yield the counterpart of thermoset plastics. The combination of the thermoset and thermoplastic polymer is seldom used in the MMAM system. The dominant use of thermoplastic polymers in hot-melt extrusion-based system promoted the growth of the MMAM system across diverse research areas, including the study of the affinity of dissimilar materials and their mechanical properties in fabricated yield of a monolithic structure.

The interface formed between the discrete materials at their geometrical boundary is regarded as the critical element in multi-material 3D printed components, which varies with the properties and printing conditions of the materials [130]. To date, no significant research work has been reported regarding multi-material fused-filament fabrication-based AM, focused on the relationship between printing conditions and dissimilar materials interface quality in comparison to those of standard (single material) extrusion 3D printing [36]. The scarcity of information concerning multi-material extrusion-based 3D printing persists to be an unexplored area, specifically regarding the limitations of equipment, component design, and printing materials.

The major process limitations related to the equipment are stated as follows: (a) reduction of the available printing area and calibration of the extrusion heads; (b) precise alignment of the two (or more) extrusion heads during the deposition process. For component design, multi-material printing enables the development of various geometrical interfaces in the component. However, discrete material properties at the interface produce inferior bonding strength compared to the bulk material strength. Moreover, the filament's residence interval inside the extruder forms an additional aspect of the design-related challenge. In general, the residence interval impacts the thermophysical and rheological properties of the extruded material and does not provide sufficient opportunities for optimization through the flow rate in a dual extrusion system. Thus, the plastic tends to drool more often in the traditional extrusion system [131].

The materials-related challenges are associated with the chemical structures and formulation of the thermoplastics used in the extrusion process. The discrete polymer materials exhibit distinct chemical structures, i.e., thermoplastic materials often comprise a mature polymer chain, unlike the thermosets that execute the crosslinking of the open monomer and dictate the curing process. Dissimilar plastic materials (refer to Fig. 10) can create problems associated with the mismatch between the thermal expansion coefficients. Therefore, dissimilar shrinkage upon cooling eventually results in distortion and low dimensional stability of the components. Furthermore, the mechanical integrity expected at the interfaces between various printed materials reduces in case of chemical incompatibility or low chemical affinity [36].

### 3.5. Functionally-graded multi-materials

FGMs offer remarkable applications in which the service conditions and material requirements vary spatially throughout the component and a material gradient is required [133]. As thermal gradients are desired in components in aerospace and aviation applications, engineers and designers leverage the interaction between the material properties. For instance, the temperature in the center portion of a turbine blade is relatively low, and thus,

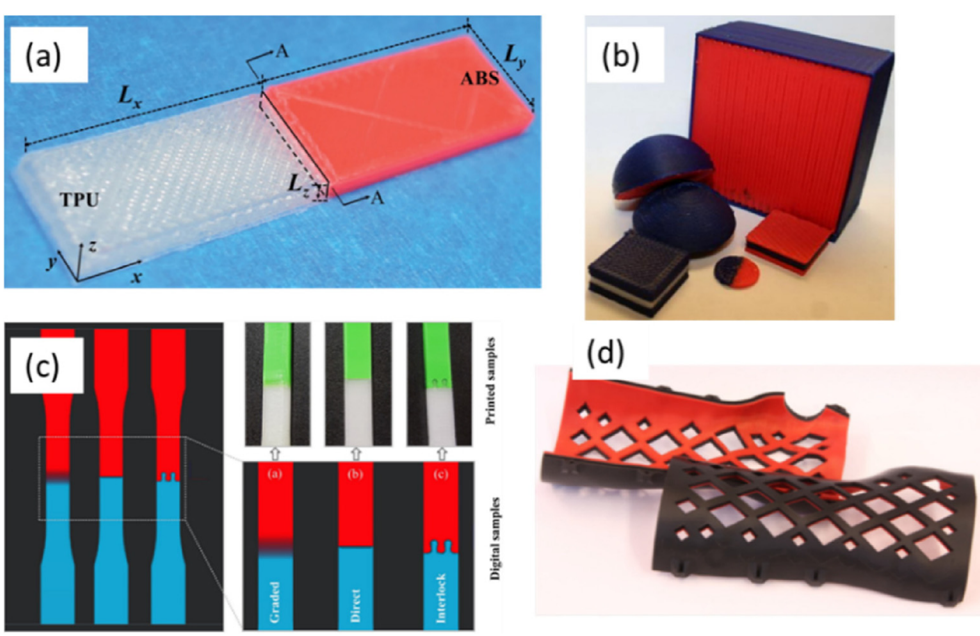


**Fig. 10.** Representative polymer–polymer MMAM example, (a) ABS and TPU specimen fabricated using FFF [41] (b) ABS and PC parts fabricated using modified FFF system [130], (c) functionally graded multi-material components fabricated in FFF system using ABS and PC [132], and (d) FFF of wrist orthosis using ABS and TPU [62].

the center portion is required to exhibit adequate tensile strength and fatigue resistance, which is designed to obtain specific physical or chemical performances [134]. Thus, the functional grading of two or more materials is required [133]. FGMs can be classified as follows: (a) FGMs *via* construction, such as addition of layers to manufacture a component, and (b) FGMs *via* mass transport or diffusion to create a material gradient [133].

Current AM methods can adapt to the constructive method of preparing FGMs with infinite combinations of ceramics, polymers, metals, and even composites designed with complex geometries. The fundamental goal of using AM in FGM production is to produce performance-based freeform components driven by their gradient in material properties [135]. In the DED process, also known as laser metal deposition (LMD), the powder is fed into the melt pool under a moving laser, and it can be used for fabricating FGMs by varying the powder composition between layers. This method can be conveniently managed using conventional or robotic DED machines equipped with two or more powder feeders [133], and thus, it has been utilized for preparing graded structures of several alloys such as titanium alloys, nickel alloys, steels, and metal matrix composites. In particular, DED offers several benefits compared with traditional manufacturing techniques, such as casting or powder metallurgy. The components are constructed layer-by-layer, reproducing interior features and channels design. Simultaneously, it does not require special tooling such as that involved in casting and powder metallurgy. Therefore, it can efficiently fabricate complex geometries and coatings [136]. As FGMs are classical MM structures involving compositional gradient, DED can rapidly produce MM components when coupled with multinozzles. During DED, multinozzles can satisfy this requirement by adjusting the powder types fed and their relative percentages. Compared with other AM processes, such as PBF, this is a beneficial advantage of this method [136]. Recently, scholars have attempted to produce FGMs using PBF–AM techniques [137].

Functionally graded MMAM addresses the MM aspect through the dynamically generated gradients approach or complex morphology. The geometry and material arrangements control the functions and properties of the final component, which aims to improve the interfacial bonding between dissimilar or incompatible materials, as depicted in Fig. 11b. A compositional transition from a dispersed to an interconnected second-phase structure—layered and graded with discrete compositional parameters or smooth concentration gradients—can be obtained to eliminate the severe properties of two dissimilar materials. Thus, this method can avoid the common failures of conventional multimaterial AM, e.g., delamination and cracks induced by surface tension owing to the discrete variations in material properties (Fig. 11a) [17].

After a three-dimensional fusion between two materials using a dynamic gradient, the printed component exhibits the optimal properties of both materials. It can be transitional in weight yet retain its toughness, wear resistance, impact resistance, or physical, chemical, biochemical, or mechanical properties. Heterogeneous mixtures of materials no longer require a compromise of their intrinsic properties to achieve the desirable properties of the component [17]. MMAM of FGMs can be varied across material combinations of metal–metal [133,134,136,138,139], metal–ceramic, ceramic–ceramic [140], and polymer-based materials [141]. The FGMs developed using MMAM technology along with their potential application areas are listed in Table 3.

As observed in Table 3, the potential application areas of additively manufactured functionally graded multi-materials vary in aerospace, military, biomedical, and automotive industries. Certain examples of the developed functionally graded MMAM components are illustrated in Fig. 12.

In addition to these applications, certain parts of die-mold applications can be produced by MMAM [139]. Although FG materials were initially designed for heat-resistant applications, they can be employed to control the deformation, pressure, wear, and corrosion, as well as to replace sharp material transitions in which high stresses are generated. Traditionally, their application was confined to the development of ceramic coatings and composite materials [139], and the grading of metallic materials has not received considerable attention. However, with the development

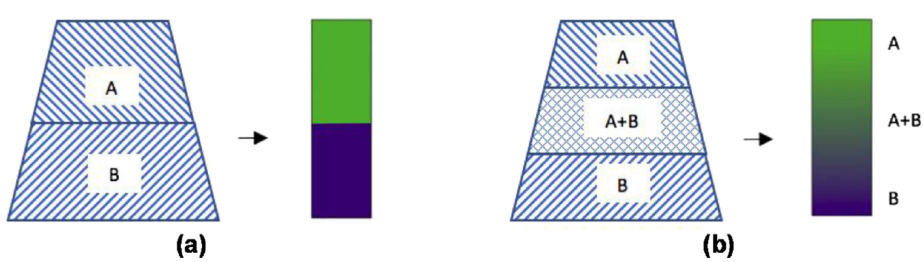


**Fig. 11.** (a) Conventional multi-material additive manufacturing *vs.* (b) functionally graded MMAM [17].

**Table 3**
Literature review of functionally graded MMAM studies.

| Reference | Materials/Products | AM Process | Potential Application Areas |
|---|---|---|---|
| Pelz et al. [140] | SiC-$B_4C$ ceramics | Direct ink writing (DIW) | Ballistic applications |
| Lee et al. [141] | Biopolymer composites | Extrusion based | Biomedical applications, Tissue engineering |
| Carroll et al. [133] | AISI 304L-Inconel 625 | DED | Aerospace, Nuclear power generation |
| Ren et al. [134] | Ti6Al4V-Ti6.5Al3.5Mo1.5Zr0.3Si alloys | DED | Compressor blisks |
| Xu et al. [138] | AISI 316L-Ti6Al4V | DED | Aerospace industry |
| Hengsbach et al. [137] | AISI 316L-H13 | SLM | Aerospace and Biomedical industries |
| Hasanov et al. [45] | Carbon fiber reinforced ABS | FFF | Aerospace and Automotive |
| Markandan et al. [142] | Cu-PE foam | Selective heat melting | Electromagnetic field devices, Graded index lenses |
| Li et al. [136] | AISI 316L-Ti6Al4V | DED | Aerospace, Nuclear, and Chemical industries |
| Ostolaza et al. [139] | AISI 316L-H13 | DED | Die and Mould industries |

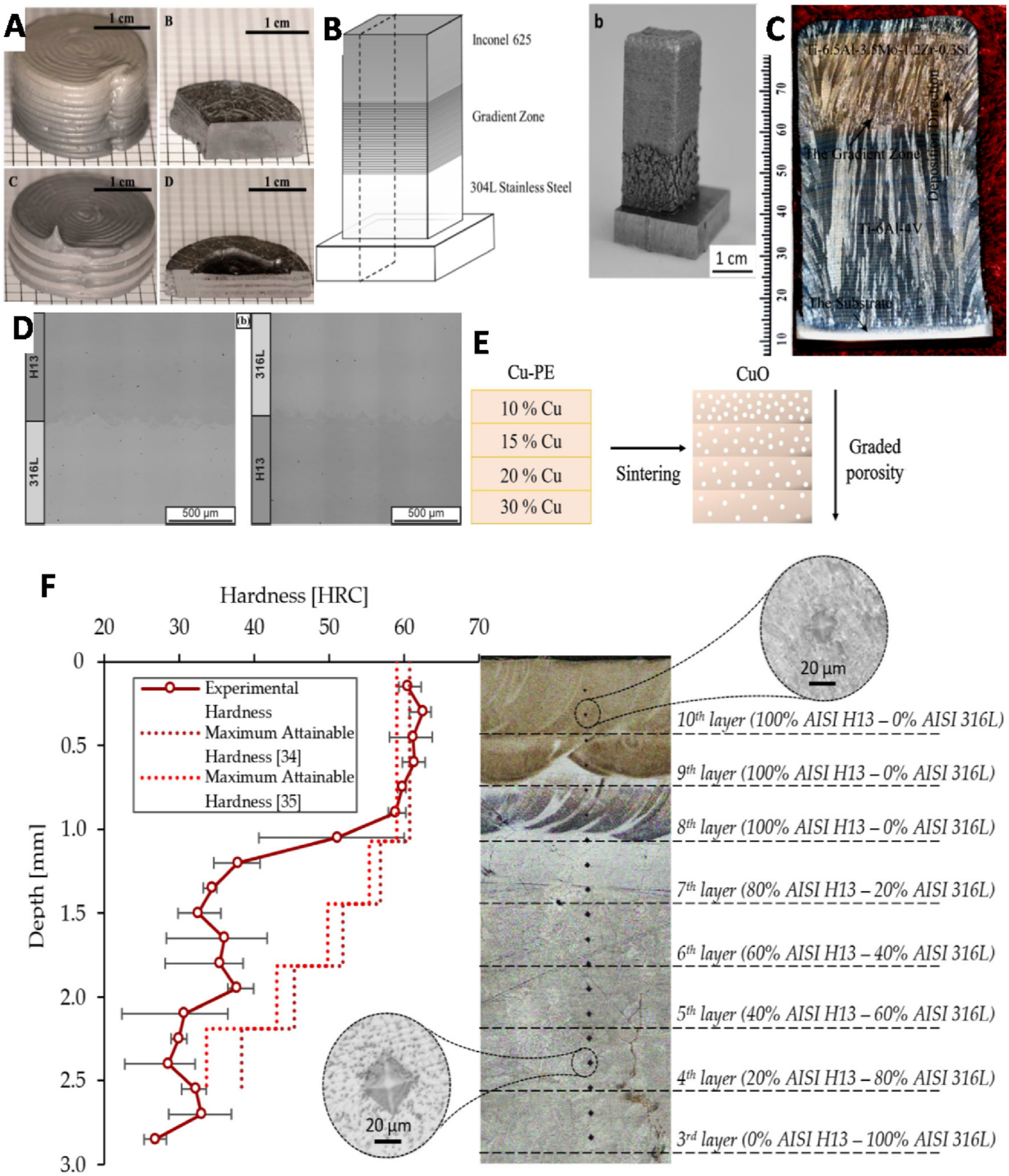


**Fig. 12.** (a) a continuous FG carbide-based sample produced via DIW [140], (b) deposited stainless steel-superalloy sample [133], (c) titanium-based FG deposit [134], (d) two variations of 316L-H13 deposition [137], (e) CuO foam fabrication with graded porosity [142], (f) hardness distribution in a 316L-H13 FGM [139]

of AM processes such as the aforementioned L-DED [139], the FGMs of complex metallic gradients of dissimilar alloys have emerged. Obtaining good metallurgical bonds between various alloys is not trivial. Complete solubility over all compositions and temperatures is uncommon in binary alloys, and it is much more complex in ternary or multicomponent alloys [139]. This issue might be addressed by introducing compositional gradients. Aremu et al. [143] used AM to develop functionally graded lattice structures with variations of cell properties in the structure. Nonetheless, several challenges existed, such as controlling numerous variables, fluctuations in thermal fields, and AM process optimization for the fabrication of FGMs. Although the development of artificial intelligence techniques, design and development of AM process diagnostic methods, and creation of thermodynamic databases have resolved several of these aforementioned issues, several challenges must be addressed.

### 3.6. Properties studied

From the literature reviewed thus far, we have identified that only a limited number of materials in each category (Fig. 4) can be utilized for MMAM. Therefore, a customized and optimized object cannot achieve the required characteristics if it is manufactured using either a single material or a combination of unsuitable materials. Thus, the MM additive manufacturability of a wide range of materials (under each category of materials in Fig. 4) is crucial for achieving the complete benefit of the products fabricated using MMs.

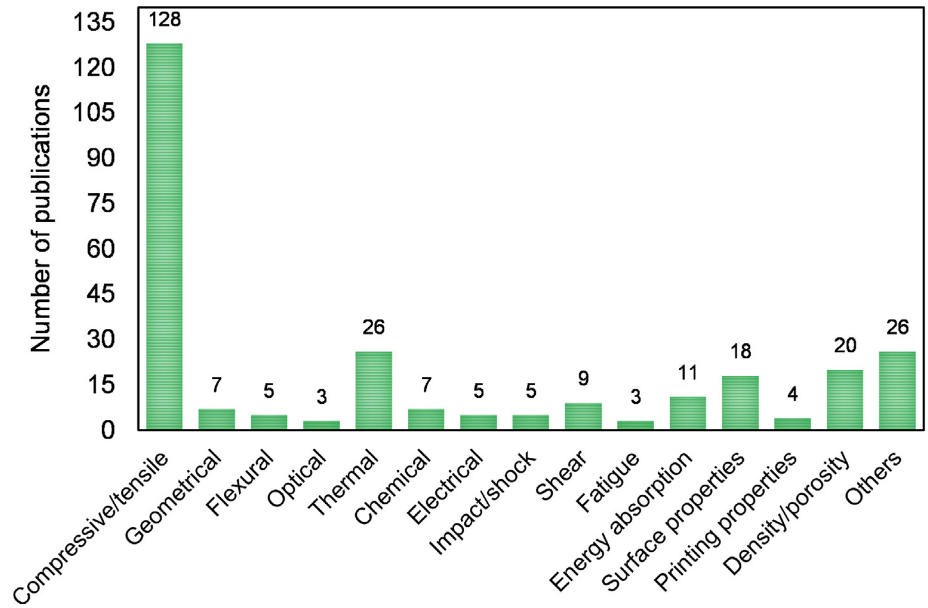


**Fig. 13.** Properties of multi-material components/samples investigated from analyzed literature.

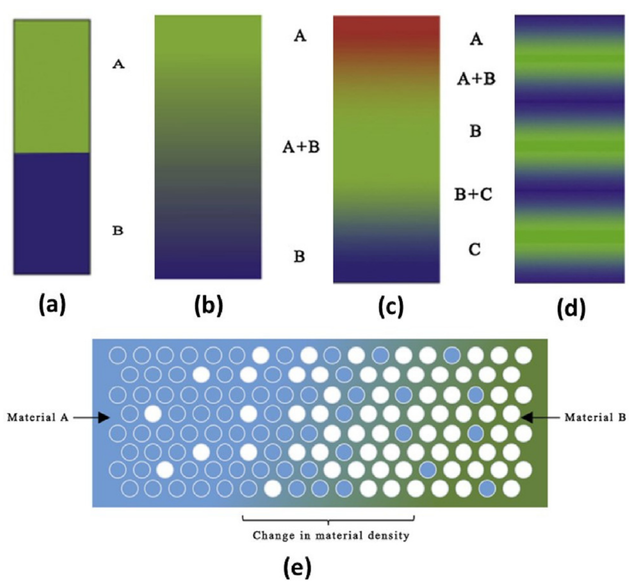


**Fig. 14.** Design combinations of multi-material compositions. (a) Transition between two materials. (b) Heterogeneous compositional transitional between two materials. (c) Transition between three materials. (d) Switched composition between various locations. (e) Combination of two materials in terms of density and compositional gradation. [17]

Various properties of multi-material additive manufactured components are presented in Fig. 13. Most studies have focused only on the compressive and tensile properties (compressive/tensile stress, strain, etc.) of multi-material additive manufactured components/samples. Although these components might possess customized properties such as high functionality, a standardized design of the experiment has not been employed to investigate the aforementioned (Fig. 13) properties. Owing to this issue, the results and methodologies of existing literature cannot be feasibly compared. As the majority of the existing literature has focused only on basic properties, the essential properties of fatigue, 3D printing, optical, impact/shock, flexural, and shear of MMAM components still require further investigation.

## 4. Multi-material components design and analysis

### 4.1. Design and modelling

Multi-material AM can be referred to as multi-material functionally graded additive manufacturing (MMFGAM) that considers the aspect of multi-material gradients and/or complex morphologies. The arrangements of material and/or morphology gradience govern the properties and functionalities of multi-material components. In MMAM components, distinct boundaries can be eliminated using a heterogeneous compositional transition or smooth concentration gradients. The implementation of diverse materials in integrated structures has improved functionality, reduced weight, and adjusted the manufacturing processes by combining assembly and production into a single processing step [144]. In summary, the required properties can be designed by combining multiple materials into a single integrated component.

The design of MM components can be classified based on several materials combined into a single piece, as depicted in Fig. 14 [17]. According to the desired properties and functionality, the material in a MM component can be distributed in several manners (Fig. 14(a–e)). Qiu and Langrana [145] have developed a computer-aided design (CAD) system to provide high-quality tool parts during the multi-material AM process. In another study, Bhashyam et al. [146] developed a CAD system with libraries of planning algorithms and material compositions, which can be utilized for designing the compositions and geometries of multi-material objects.

Multi-material workflow is similar to the AM workflow for single material. However, the primary difference is that the MM process prioritizes the definition and allocation of the materials (of every layer and/or every voxel) within the designed MM component [17]. In a recent study, Yao et al. [147] proposed a complete framework (Fig. 15a) for designing the multi-material components that can be realized using AM process. Their design framework comprises four integrated modules, including the identification of design requirements, primary material selection, AM method selection, multi-material composition, and determination of component geometry. Furthermore, they compiled a database covering the rules and guidelines, constraints, and capabilities of AM

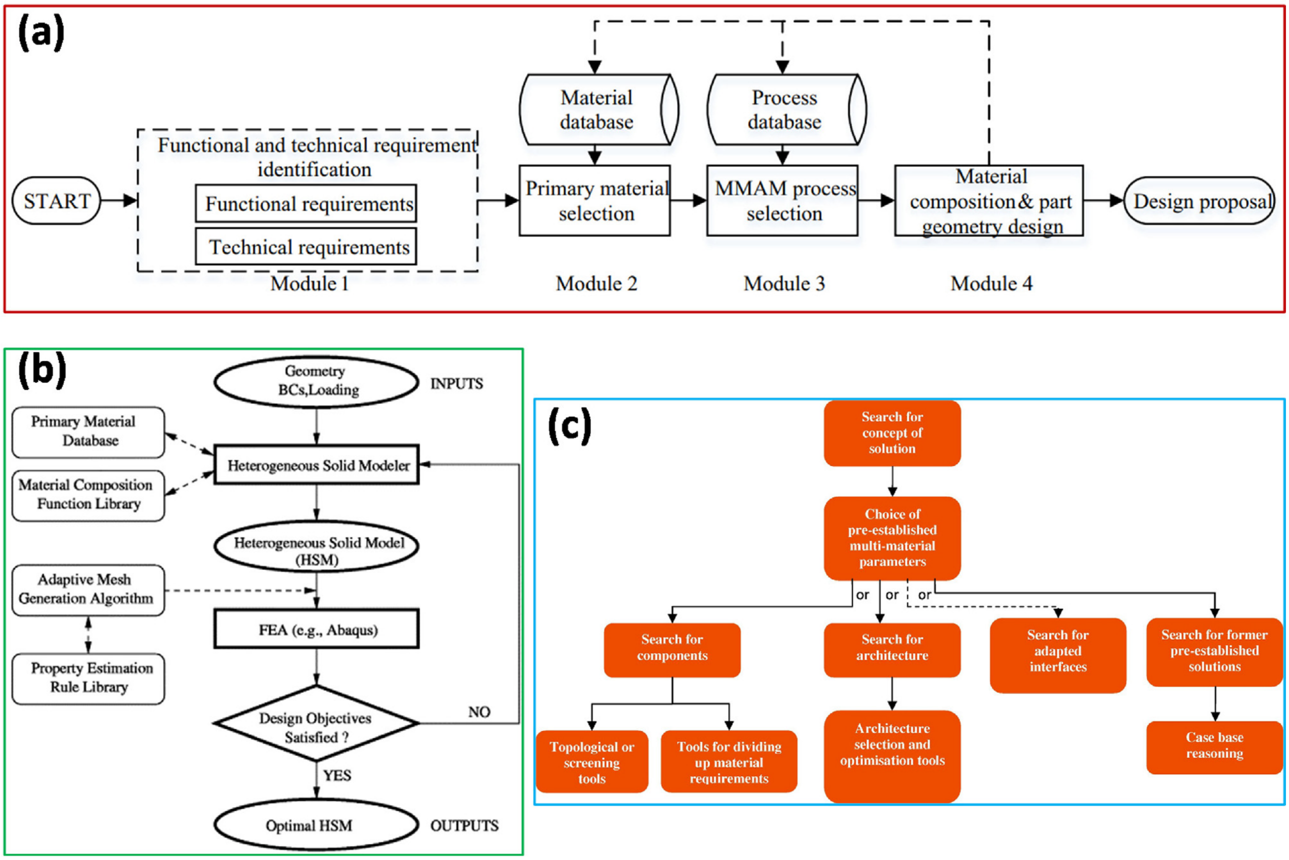


**Fig. 15.** Flowcharts of several designs for MMAM frameworks. (a) Design for MMAM procedure proposed by Yao et al. [147]. (b) Variant design workflow by Shin et al. [148]. (c) Material searching procedure and tools for multi-material solutions by Wargnier et al. [150].

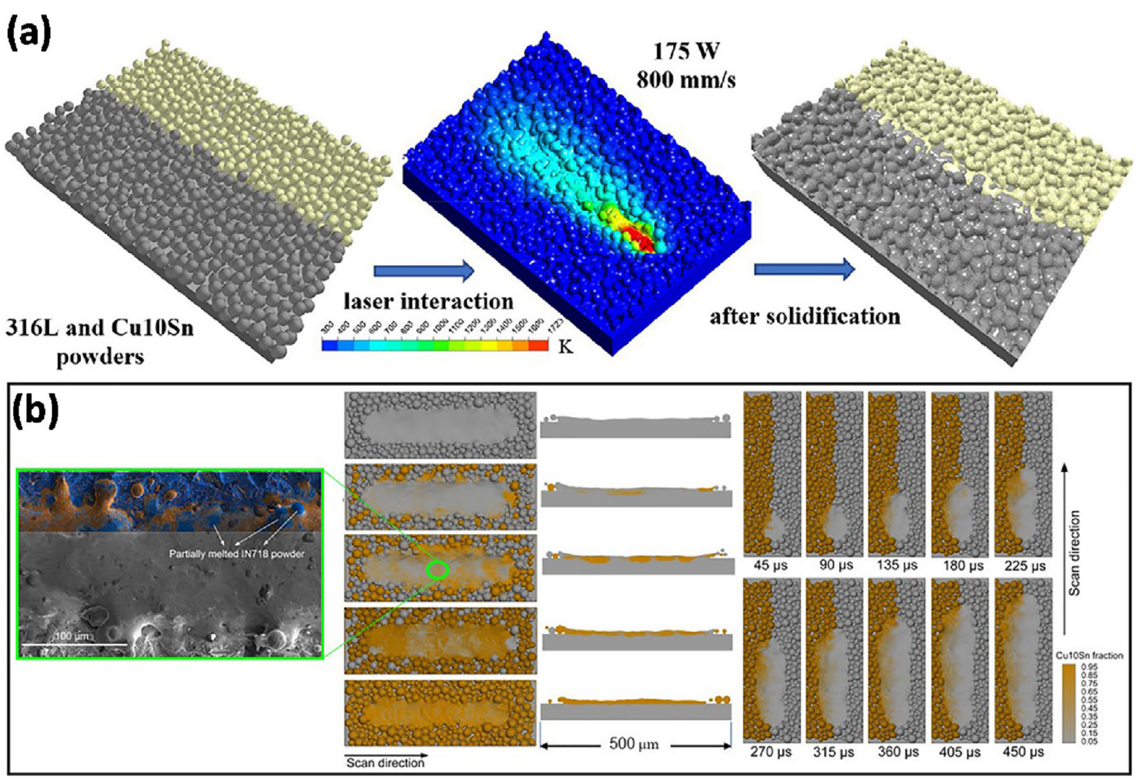


**Fig. 16.** Numerical simulations for MMAM designs. (a) integrated DEM-CFD approach for 316 L/Cu10Sn deposition in PBF process [156], (b) DEM–CFD approach for IN718/Cu10Sn in PBF process [157].

processes, which assisted in decision-making in several MM product design phases. Their method can be a critical guide for designers working in the MMAM research area. Shin et al. [148] proposed two methods for designing the MM objects. The first method involves the generative design approach in which the material distribution and geometry of the object were optimized using homogenization design algorithms [149]. In the second approach, they employed a knowledge-based variant approach (Fig. 15b) in which the designers could use their personal experience and creative thinking to design a MM object. In another study, Wargnier et al. [150] developed a conceptual procedure for designing MM objects by following material-searching approach (Fig. 15c) according to the functional requirements of the designed components.

However, multi-material design rules for each AM process and various materials still require to be studied. In addition, in-depth studies should be conducted on the MM component design for each AM process. Furthermore, CAD software packages capable of design, multidisciplinary optimization, and simulation methods for MM components require further investigation.

### 4.2. Advances in numerical simulations

In the MMAM design procedure, numerical analyses are essential for improving the reliability of components, prediction of stresses, and parameter processing optimization. Owing to the complexity of fusing MMs (shape of the melt pool, interface morphology, and microstructures), study, observation, and simulation of MMAM processes is difficult, computationally expensive, and time-consuming, especially the fusion of multimaterials involving metals [151]. Owing to the development of high-performance computers and simulation workstations, the simulations can be applied to predict and solve the MM-related problems, either based on the material fusion mechanics or fabrication processes [152]. In the DED and PBF processes, numerical simulations, including the finite element method (FEM) [153], discrete element method (DEM), computational fluid dynamics (CFD) [154], and molecular dynamics (MD) were implemented for MMAM designs to explain and predict the melt-pool phenomena such as convection, surface tension gradients, recoil pressure, vaporization, and momentum losses in mushy zones [155]. The rack morphology obtained after the solidification and elemental distribution of the multi-material combination of 316 L/Cu10Sn and IN718/Cu10Sn by DEM-CFD approach is depicted in Fig. 16a & b. Prior to experiments, the MMAM design simulation simulates the inhomogeneous temperature distribution owing to the mismatch in thermophysical properties. Furthermore, powder fraction can be conveniently adjusted *via* simulation instead of repeated experiments for MMAM design.

However, most simulation methods encounter several challenges, such as large computational power requirements and difficulty in acquiring multimaterial data. Among the available software packages, only a few have features for designing and optimizing MMs. Moreover, the existing FEM packages (Fig. 17) offer limited functionality for MMs. Currently, no software package has been specifically developed for the design, optimization, and simulations of MM-related studies. In the analyzed literature, most researchers employed a general software for design, optimization, and finite element analysis (Fig. 17), e.g., ABAQUS [31,40,158-160], ANSYS [30,161,162], level set model [163], COMSOL Multiphysics [7,164,165], Rhino [117,118,166], and MATLAB[98,167-171]. Notably, these software are not specifically designed for MM-related research and development. Thus, significant development is required to apply these packages for MM-related investigations.

### 4.3. Multi-material lattice structures

The realization of an optimized and application-specific structure using AM [172-174] is possible only if their 3D models can be created digitally using a software. Nevertheless, most commercially available software can design models with limited material information, which is critical if the part is designed for fabrication

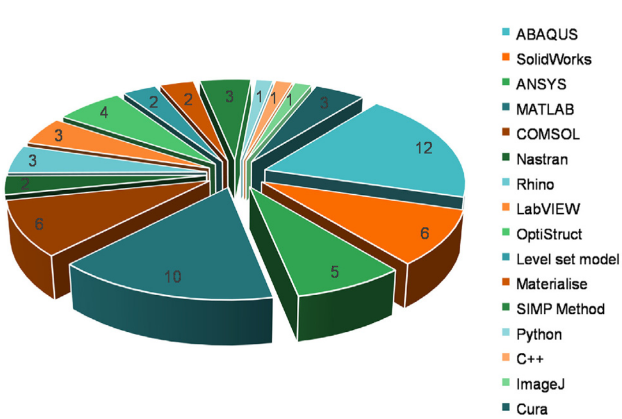


**Fig. 17.** Software packages used for design and finite element analysis of multi-materials components investigated in analyzed literature are as follows: ABAQUS, 12; SolidWorks, 6; ANSYS, 5; MATLAB, 10; COMSOL, 6; Nastran, 2; Rhino, 3; LabVIEW, 3; OptiStruct, 4; Level set model, 2; Materialise, 2; SIMP Method, 3; Python, 1; C++, 1; ImageJ, 1; Cura, 3.

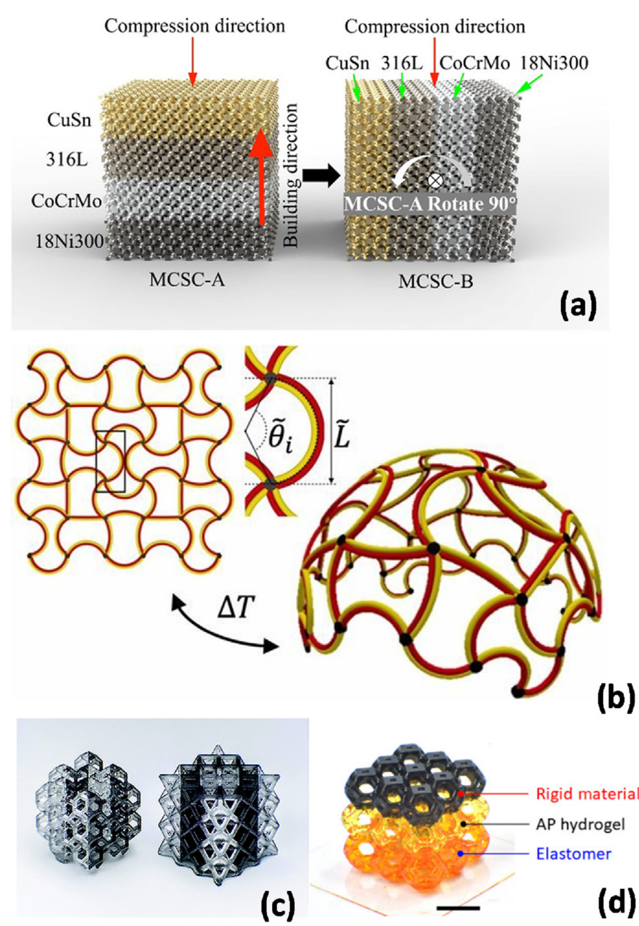


**Fig. 18.** (a) Compression model of lattice structures designed using four distinct materials [176]. (b) Multi-materials 4D printable two-dimensional lattice structure that can morph into the shape of a spherical cap [177]. (c) Veroblack and Veroclear materials are 3D printed using a DLP system [178]. (d) Multi-material designed Kelvin lattice structure comprising three distinct materials [179].

with MMs. This is a formidable task if the 3D models require to be designed with the incorporation of lattice structures [175]. Wei et al. [82] presented a manual data preparation procedure for designing multi-material structures, although their method requires a significant amount of manual work. Thus, their approach cannot be employed for extensive production and industrial applications. Therefore, developing an advanced data interface and 3D file is crucial for integrating the geometric and multi-materials information, while simultaneously connecting to the AM system for the fabrication of the MM structures [17].

Several researchers have studied the 3D-printed lattice structures of MMs using various materials. Zhang et al. [176] studied the MM porous structures fabricated using SLM of four distinct materials (CuSn, 316L, CoCrMo, 18Ni300) in a single specimen, as depicted in Fig. 18a. They concluded that the compression behavior of lattice structures can be improved by combining high-strength brittle material and low-strength plastic material. Boley et al. [177] studied 4D printing of heterogeneous morphable lattices (Fig. 18b) to investigate design and fabrication issues. These structures with multi-material 3D printing can transform a diverse range of applications. Recently, strut-based lattice structures were fabricated using photosensitive materials [178,179] to realize elastomeric and rigid material properties in a single component.

However, the design procedures of MM structures are very limited. Thus, they require new methods of computational modeling that contain detailed geometric (availability of a wide range of strut, surface, and stochastic-based lattice structures) and materials information while managing material distribution at the voxel level. Generally, voxel-level material distribution and modeling of structures are computationally expensive. Therefore, to reduce the amount of computation, Richards [180] proposed a modeling system for representing material–geometry–topology using volumetric texture maps. This system can enable the designer to perform modifications at the voxel level. Thus, the functional representation-based modeling methods effectively describe multi-materials and incorporate complex lattice structures.

## 5. Multi-material additive manufacturing processes

The high level of complexity of design and material distribution in MM components creates insurmountable challenges for fabrication using traditional manufacturing processes. This limitation motivated certain researchers to explore MMAM technologies in the late 1990s [13]. Consequently, the continuous advancements in AM processes enabled almost all AM processes to fabricate components with multiple materials in a single component. As depicted in Fig. 19, nearly all AM processes can fabricate multi-materials. In particular, materials extrusion, PBF, and material-jetting methods are most commonly utilized. Furthermore, the vat-photopolymerization process, commonly referred to as SLA, was the first AM technology and has been widely used for manufacturing MMs. A single AM technology encounters several challenges during the combined printing of multiple materials. Thus, hybrid AM technologies [94,117] have gained considerable research attention. In a recent study [43], researchers have integrated four AM technologies, namely, fused filament fabrication (FFF), direct ink writing (DIW), aerosol jetting (AJ), and inkjet (IJ), for developing a hybrid 3D printer that can rapidly fabricate complex multiple materials with a range of functionalities.

Several commercial and laboratory-developed multi-materials 3D printers commonly used for fabricating multi-materials components and complex lattice structures in the reported literature are listed in Table 4, which is a valuable reference for readers.

In general, a MMAM system includes the delivery of multiple materials and layer bonding systems [183]. To construct multi-

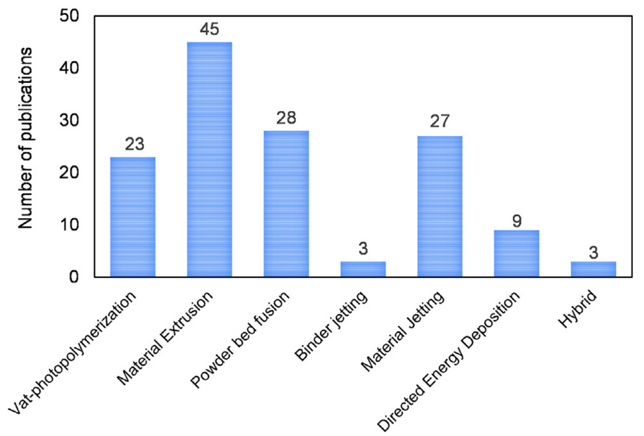


**Fig. 19.** Additive manufacturing technologies used for fabricating multi-material components.

functional MMAM components, the selection of AM method for the fabrication is critical to ensure strong bonding between various types and compositions of materials. Thus, the success of producing such components primarily depends on the applied AM technology and the optimization of AM process parameters. Each AM method offers its unique advantages and limitations for multi-material processing. The advantages and limitations of each AM method for multi-material fabrication are examined in the following subsections and summarized in Table 5.

Each AM method is capable of MM fabrication with a recently designed powder feeding system or powder mixing process. However, MMAM processes are still in progress to overcome the challenges caused by the selection of dissimilar materials. In principle, the MMAM process bonds dissimilar materials to enhance the performance of the structure, considering location-specific properties. However, the bonding quality remains a major issue for the success of MMAM processes. Lumpe et al. investigated MM interfaces and determined that bonding quality relies on the material combination [184], thereby rendering the selection of MM for MMAM a critical consideration. Otherwise, the failure at the interface zones can be detected [116]. Lately, studies have focused on the gradient transformation in the material composition for MMAM processes to increase the interface bonding [45,132,185,186]. However, the effect of the material transition zone and gradient composition remains unclarified. Accordingly, the properties of the transition zone with respect to the combining multi-materials require further research [187].

### 5.1. Extrusion-based systems

Extrusion-based AM method is one of the most well-known AM processes (Fig. 19) that displaces the material feedstock through a nozzle or multiple nozzles, enabling the construction of a 3D structure with multiple materials [188]. This process is capable of printing various materials, including thermoplastics [189], metal-filled thermoplastics, composites [190], flexible elastomers [41], and blended multi-material powder-based feedstock [132,185]. Similar to the BJ method, the compatibility of the binder with the feedstock material is crucial for the success of the process considering unintended interactions/reactions during extrusion or sintering. To ensure structural integrity, the components produced by the ME process require post-AM de-binding and sintering. Moreover, as the binder is mixed with the material instead of being extruded on a powder bed, the powder recycling/reuse process is not required, thereby saving both time and cost. Nonetheless, the predominant disadvantage of this technology is the printing resolu-

**Table 4**
Prevalent 3D printers for multi-material additive manufacturing.

| Technology with refs. | 3D printer | Company/institute (country) | Multi-materials |
|---|---|---|---|
| DLP [56] | D7 Plus | Wanhao, China | Urethane acrylates |
| Extrusion [95] | Lab developed | Ulsan National Institute of Science and Technology (UNIST), South Korea | BiSbTe materials, TE inks |
| SLS [60] | HORI H1 | Beijing Huitianwei Technology, China | TPE |
| PJ [115,117] | Objet 500 Connex 3 | Stratasys, USA | VeroPlus, agilus, wood, polymer |
| DLP [181] | Lab developed | Tsinghua University Beijing, China | Different compositions of Kaolin ceramic materials and ceramic–metal MM |
| Extrusion [67] | BioX 3D printer | Cellink, USA | Alginate and methylcellulose – Sodium alginate/ Methylcellulose |
| Extrusion [68] | A custom-built 3D printer system | McMaster University Hamilton, Canada | UV curable silicone inks |
| MJ [73,114] | Stratasys J750 | Stratasys, USA | TangoPlus (FLX980, FLX935) and the rigid VeroClear (RGD810, RGD852). |
| Two-photon lithography [64] | Lab developed | University of Nottingham, Nottingham, United Kingdom | Photopolymers |
| Hybrid [43] | m4 3D printer | 3D systems, USA | Soft elastomer resin, Conductive ink, ABS, photocurable acrylate-based polymer |
| DMLS [83] | ProX200 | 3D Systems, USA | AlSi12 powder: aluminum and silicon |
| Hybrid [98] | Custom-built multimaterial 3D printer | Imperial College London, United Kingdom | Metal, rubber, and plastic |
| Extrusion [166] | Objet1000 | Stratasys, USA | Biodegradable hydrogel composites (natural hydrogels, such as chitosan and sodium alginate) |
| VP [182] | Lab developed | Johns Hopkins University Baltimore, USA | Hydrogel and gelatin methacryloyl (GelMA) |
| FDM [28] | Commercial FDM printer | 3D systems | Multi-blended and hybrid-blended PLA matrix |
| FDM [44] | CL1720 3D printer | KAIROS | ABS and polycarbonate (PC) |
| Hybrid [118] | Objet Connex 3 260 | Stratasys, USA | Polypropylene, glass fibre, carbon fibre or Kevlar, and low-density polymer |
| FFF [29] | Ultimaker 3 dual-extruder | Ultimaker, USA | Acrylonitrile Styrene Acrylate (ASA), PLA, polyethylene terephthalate glycol (PETG) |

**Table 5**
MMAM processes and their general advantages and limitations.

| **Process** | **Advantages** | **Limitations** |
|---|---|---|
| Powder bed fusion | Dimensional accuracy<br>Variety in material choice | Powder feeding system<br>Powder recycling and reuse |
| Direct energy deposition | Multi-material powder feeding<br>Printable part size | Dimensional accuracy<br>Surface roughness |
| Sheet lamination | Low-cost<br>Time-efficient | Delamination<br>Inhomogeneity |
| Binder jetting | Low-temperature process | Post-processing<br>Binder compatibility<br>Powder recycling and reuse |
| Material extrusion | Variety in material choice | Post-processing<br>Binder compatibility |
| Material jetting | High-speed process<br>Variety of material choice | Clogging problem<br>Low viscosity |
| Vat-photopolymerization | High accuracy | Material waste<br>Only photopolymers |

tion that depends on the powder size and nozzle diameter. Moreover, products with a large aspect ratio and overhanging designs are challenging to manufacture and require a consistent flow of feedstock and fast solidification. To this end, a multi-material feedstock design is crucial.

In extrusion-based MMAM systems, the novel hybrid metal extrusion and bonding method has been considered to fabricate various multi-material systems in which Al-based components were highly studied, enable to produce continuous extrusion and good quality bonding by utilizing friction stir welding [191,192]. However, fused filament fabrication (FFF) is one of the most commonly used extrusion-based AM systems (several extrusion-based systems are listed in Table 4). Among other AM systems, the FFF is the most adopted system. In particular, it uses thermoplastic-based feedstock filaments that transition through several thermal events, including heating and melting in a hot extruder, and are extruded through a nozzle on the build platform in a layer-by-layer fashion. The FFF system often uses multiple extrusion heads to deposit discrete thermoplastic materials and fabricate monolithic parts, as depicted in Fig. 20. Conventionally, the primary material being extruded through an extrusion head is used as a model material, and the secondary material being extruded through another head is used as a support material. However, a MM structure can be fabricated by utilizing dual extrusion heads, and a secondary extrusion head can be employed for another thermoplastic or various materials (e.g., carbon fiber) to produce composite components. A representative example of a typical multi-material FFF system is presented in Fig. 20A. The ability of the

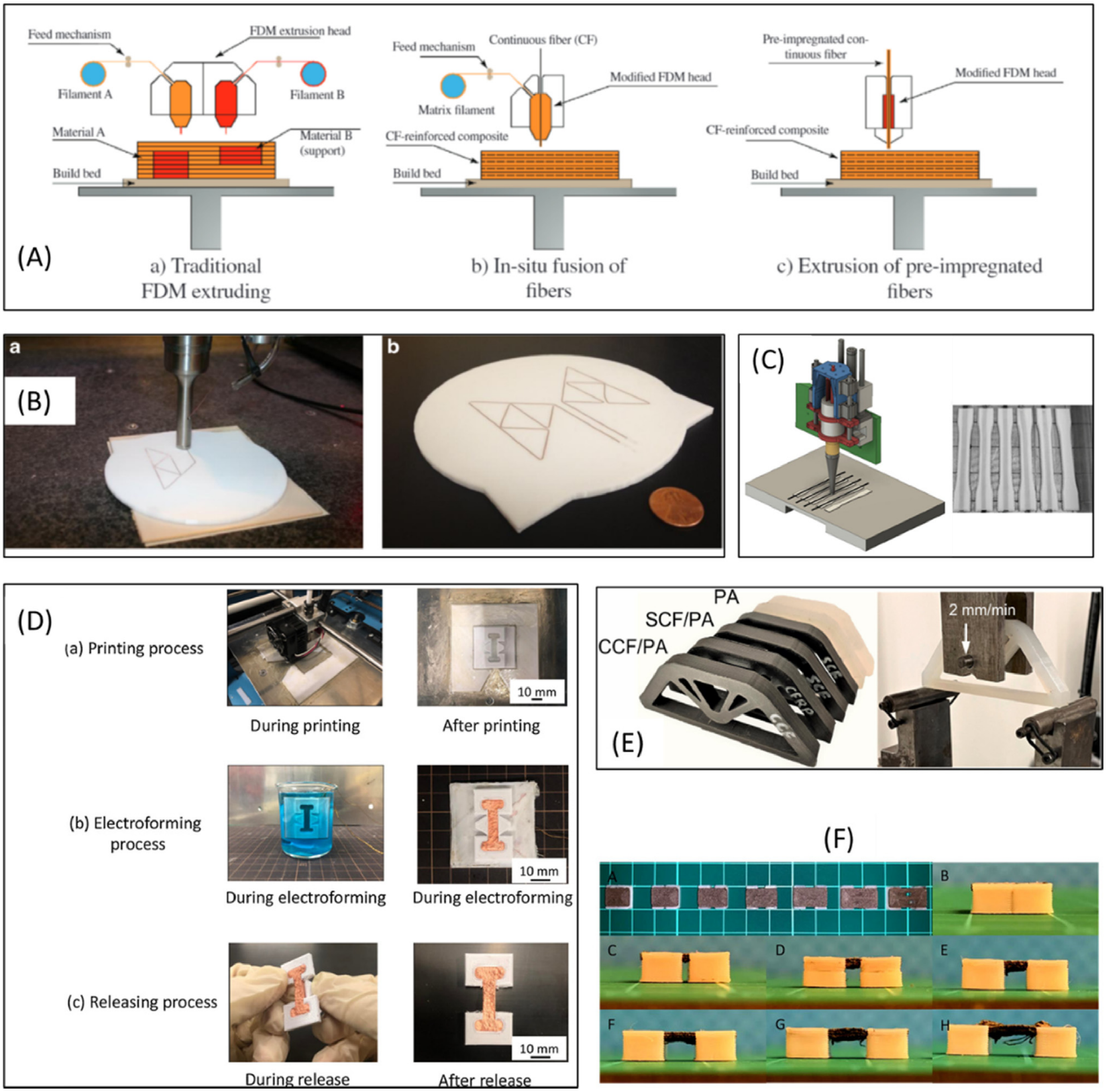


**Fig. 20.** (A–F) Examples of FFF-based MMAM system. Lower case letters represent the steps of individual research work. (A) Representative FFF system with dual material, *in-situ* fusion of fibers with molten thermoplastic in nozzle, and extrusion of pre-impregnated fibers [132]; (B) example of embedded copper wires in ULTEM 9085 and PC substrates using an ultrasonic embedding tool for embedded electronics [197]; (C) continuous carbon fiber embedding in PC using ultrasonic energy [198]; (D) multi-material electroforming parts in FFF system [97]; (E) short and continuous fiber-reinforcement in FFF system [201]; (F) Multi-material bridge span was printed with LDPE–Cu–Sn 35 vol% metal-filled composites. Simultaneously, the pillars were composed of printed polyester thermoplastic elastomer to demonstrate support-free printing and sensor applications [202].

FFF system to fabricate continuous fiber, filament, and wires offers a tremendous opportunity for the fabrication of multifunctional parts, including embedded sensors [193-196], electrical circuits [197], reinforced composite parts [198], heating elements [199,200], and functionally gradient structures [132].

Regardless of the types of thermoplastics, several challenges are associated with the FFF multimaterial system. Similar to the polymer–polymer MMAM system, the FFF system poses limitations in component design, process development, and materials. For instance, the deposition of fiber material within the thermoplastic material is restricted to a certain volume. A higher volume percentage of the fiber content reduces the matrix material within the component and increases the voids, thereby affecting the mechanical properties and increasing the anisotropy [203]. Another example of the part design limitation is the accessibility of the fabricated component. The embedded electronics fabrication with an FFF system is a unique area of application that enables monolithic yet multifunctional device manufacturing. However, research describing the method of assembly and disassembly for servicing has not been established. Therefore, the reliability of the fabricated components should be adequately high to increase the end-user's confidence in rapidly replacing the traditional components and deploying them for multifunctional utilization. In the FFF MMAM system, several processes are developed and maintained under the patent, e.g., wire coextrusion systems [201], wire embedding [204], fiber embedding [205], and fiber impregnation [206].

Although each process improved the capability of MMAM, each process poses a limitation on the fiber placement, control, maintenance of volume fraction, or the initial placement of the continuous fiber/wire. Moreover, a mismatch of the CTE of the discrete material limits the process development of MMAM systems. Typically, the thermal conductivity and CTE of the thermoplastic-based matrix materials considerably reduce the continuous fiber, including carbon fiber, glass fiber, and any conductive and resistance wires.

### 5.2. Powder bed fusion

The principle of the PBF process is to create a layer of metal powder on a substrate with a powder feeding system, such as rolling, blade, or brush, and selectively melting or sintering the area determined according to the computer-aided design (CAD) of the desired component using a laser (L-PBF) or electron beam (E-PBF) as a heat source [207]. This process is repeated for the subsequent layer by descending the build stage, feeding the powder on

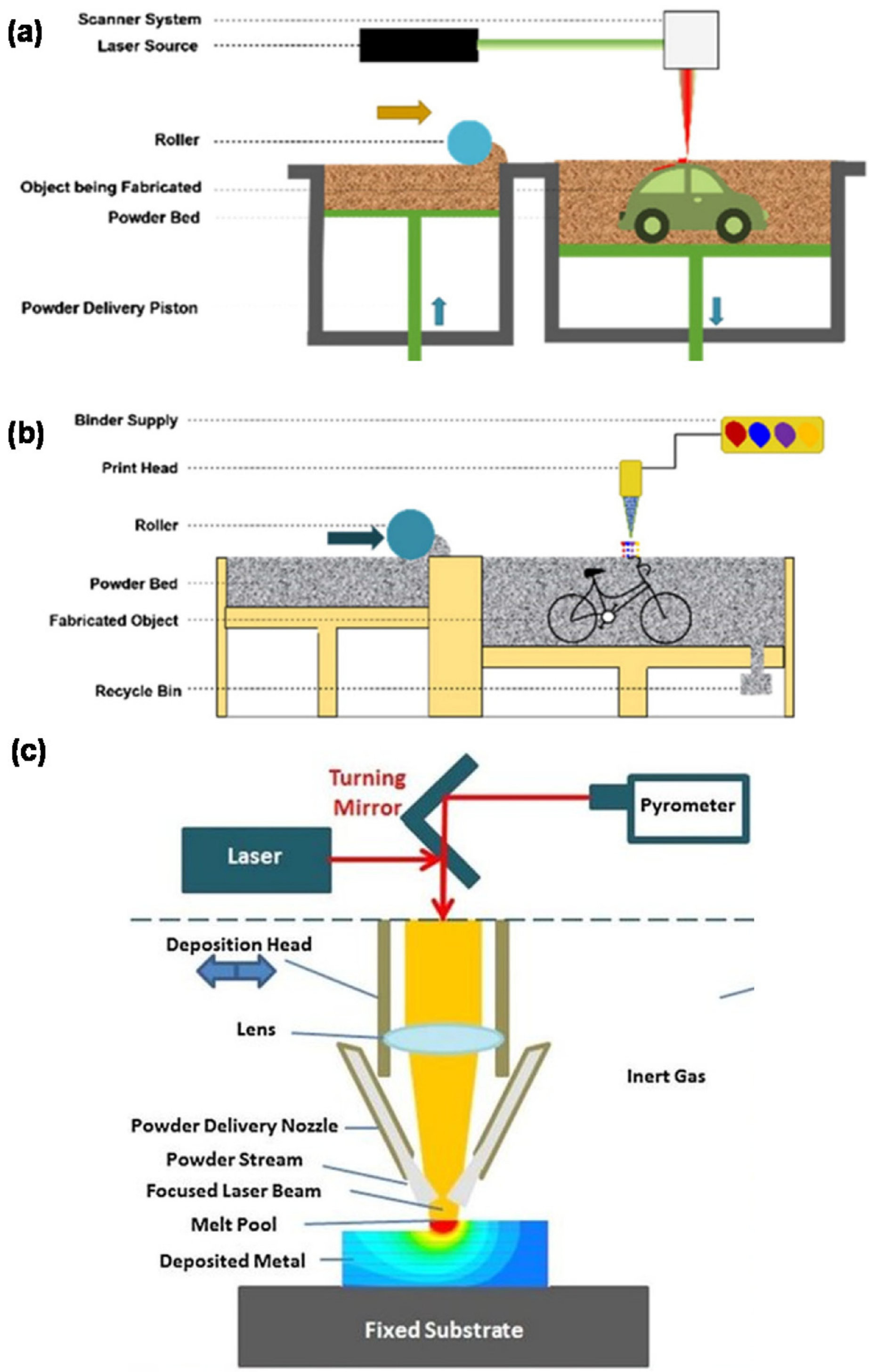


**Fig. 21.** Illustrations of metal AM processes [207,216] (a) Power bed fusion process, (b) binder jetting process, (c) direct energy deposition process.

the previous layer, and melting or sintering the current layer along with the previous layer(s) (Fig. 21a) [188]. However, this technology is commonly utilized for single-metal powder production with one powder-bed dispensing unit [208]. Specifically, the single-powder delivery system limits the fabrication of MM components, and accordingly, a mixture of metal–metal or metal–ceramic powders can be printed even with the single-powder delivery system to resolve this limitation. Thus, using blended materials, multi-material PBF (Fig. 19) has been achieved for various material combinations [119,209].

To realize the complete potential of the intended multi-material system, several factors should be considered to achieve a strong and long-lasting bond between dissimilar materials [210]. Primary concerns for the success of the fabrication involve the optimization of the PBF process parameters that allow sufficient re-melting into the previous layers and strengthening of the bond between the layers [211], which might be affected by the powder characteristics [212]. During optimization of the process parameters to avoid undesirable defects, the component is susceptible to exhibiting a lack of fusion owing to the low-energy density of the heat source, wherein high-melting-point alloying elements are located or the applied energy density of the porosity remains constant because of the existing low-melting-point elements. Otherwise, the cracks in between dissimilar materials are caused by differences in thermal properties [213,214]. An approach to using smaller powder sizes for materials with high melting point has been proposed to increase their surface area and the amount of energy absorption during the PBF process [212,215].

Thus, the fabrication of multi-material components by PBF systems requires comprehensive consideration for process parameter optimization and powder characteristics. Furthermore, post-process heat treatment might be required even after a successful fabrication to ensure the desired compositional homogeneity. Furthermore, PBF methods are required to provide an improved system for recycling and reusing the powder in the build stage, which is a limiting factor for PBF systems as an emergent technology for MMAM compared to DED systems.

### 5.3. Jetting-based systems

Similar to PBF, binder jetting (BJ) requires a powder bed, but it differs in terms of bonding methodology. Although the PBF solidifies the powder by thermal energy, the BJ bonds the powders with an adhesive to form the structure at low temperatures (Fig. 21b) [188]. This technique requires a compatible binder with the metal powder, such as polymer binders, metal inks, and metallic slat compounds [217]. The application of bonding on a powder bed follows the same procedure as that in the PBF. Concurrently, the BJ process exhibits similar challenges as the PBF to manufacture MM components, such as multiple powder feeding and efficient powder recycling systems. BJ-processed components that require post-AM heat treatment for structural integrity do not experience residual stress and differ from the PBF process. However, post-AM heat treatment causes structural shrinkage that can be detrimental to the structural application, considering the variations in the thermal properties of multiple materials [218].

Material jetting (MJ) or inkjet 3D printing system is appropriate for MMAM owing to its applicability for multiple jetting heads. Polyjet (PJ) [115,117] is an excellent example of MJ technology that has been widely used for multicolor AM, as noted in Table 4 and depicted in Fig. 19. However, the limited availability of materials [219] and measurement of material rheology [220] during printing are the challenges of the MJ technology.

### 5.4. Directed energy deposition

In the DED process, the material in the form of powder or wire is fed through a nozzle that can perform multiaxial displacement based on the component design, and the material is melted using the heat source (Fig. 21c) [221]. The ability of complex design and dimensional accuracy is compromised to increase the deposition rate using wire and arc additive manufacturing (WAAM) [222]. Moreover, WAAM process is highly implemented for multi-material component fabrications owing to its powder feeding design making it easy for MMAM approach [223]. However, there are challenges regarding final product performance fabricated by WAAM, which are excessive grain growth due to heat accumulation, resulting a decrease in mechanical properties [224].

Although high dimensional accuracy and low surface roughness is required for near-net shape AM products, powder-based DED techniques, laser-engineering net shaping (LENS), and direct laser/energy/metal deposition (DLD/DED/DMD), such as WAAM, are established as the best method for realizing these requirements by allowing multi-material delivery to the heat source to yield multifunctional components [225]. The most critical challenge for DED and the other similar AM techniques is to form a strong bond between dissimilar materials. Although powder feeding is comparably more efficient than PBF processes, the mismatches in material characteristics such as thermal expansion coefficients, thermal conductivity, laser absorption, and melting points are certain material-specific challenges that cause residual stress and metallurgical defects during DED fabrications.

### 5.5. Hybrid MMAM systems

A hybrid process merging both additive and subtractive manufacturing can maintain dimensional accuracy and surface finish as well as improve the productivity through thick layers. Hybrid manufacturing can avoid the weaknesses of each process while exploiting the strengths to achieve a system with capabilities beyond each individual process [226]. The advantages of the hybrid process have been extensively exploited in producing metal components, with complex geometry being produced with AM and areas requiring precise dimensions or surface finish being machined [226]. In addition, hybrid systems have been used to produce polymer components and components comprising multiple materials [227].

More recently, the combination of metal AM with metal forming has been developed to improve the shape of the deposited material layers *via* local plastic deformation, while providing improved stiffness and wear resistance to the built components. Currently, this process is evolving and has expanded rapidly by encompassing new concepts from sheet and bulk metal forming processes. The initial developments of hybrid metal AM systems aimed to utilize multiple thermal energy sources and apply a combination of metal AM with metal cutting to improve the productivity and quality of the built components. These developments resulted in the commercialization of the first hybrid metal AM systems in the mid-2010s [228,229]. The notable examples of commercialized hybrid-metal additive manufacturing systems/companies include Optomec, Hybrid Manufacturing Technologies, Concurrent Technologies Corporation, etc.

Hybrid multi-material AM (HMMAM) technology has been used to fabricate several MM components, including metal–metal as well as metal–polymer material couples [226]. Weflen and Frank proposed a method for producing metal–polymer components using hybrid manufacturing systems. First, the performance of the polymer extrusion AM tool is assessed, and the process parameters are integrated into a machining center. To evaluate polymer extrusion thermal characteristics in the machining center, the relationship between component cooling and geometry is studied. Thereafter, the strength of the component is baselined on various cooling periods and material flow rates, which allow the comparative analysis of the structural performances of the extruded polymer and the metal–polymer interface [226].

Recently, hybrid MMAM machines and setups in metal AM (additive/subtractive) have been further developed [230]. Muguruza et al. [231] developed a hybrid system to produce ceramic particle-reinforced photosensitive resins; their system is composed of digital light processing (DLP) technology and a 2D drop-on-demand inkjet printing system. The DLP technology enables the construction of 3D objects using the photopolymerization of photosensitive resins, whereas the inkjet printing system deposits tiny drops of conductive inks. In a similar system, as observed in Fig. 22, functional materials (e.g., printed circuits) for electronic devices have been developed. In another study, Ma [215] developed a novel technique named "hybrid deposition manufacturing" by combining the FDM and MJ AM processes to fabricate MM components for robotics and mechatronics-related applications. Their method considerably reduced the manufacturing time, required manual labor, amount of waste material, and complexity of assembly compared to alternative processes [232]. Combining the DED process with a multi-axis machining center enables the fabrication of new products with MM geometries and surface finishes that cannot be achieved *via* additive processes alone. Consequently, hybrid DED systems have successfully demonstrated the implantation of a Bluetooth active sensor within a high-strength steel component [233] fabricated using pH 13–8 stainless steel and Invar 36 alloy multi-material deposition.

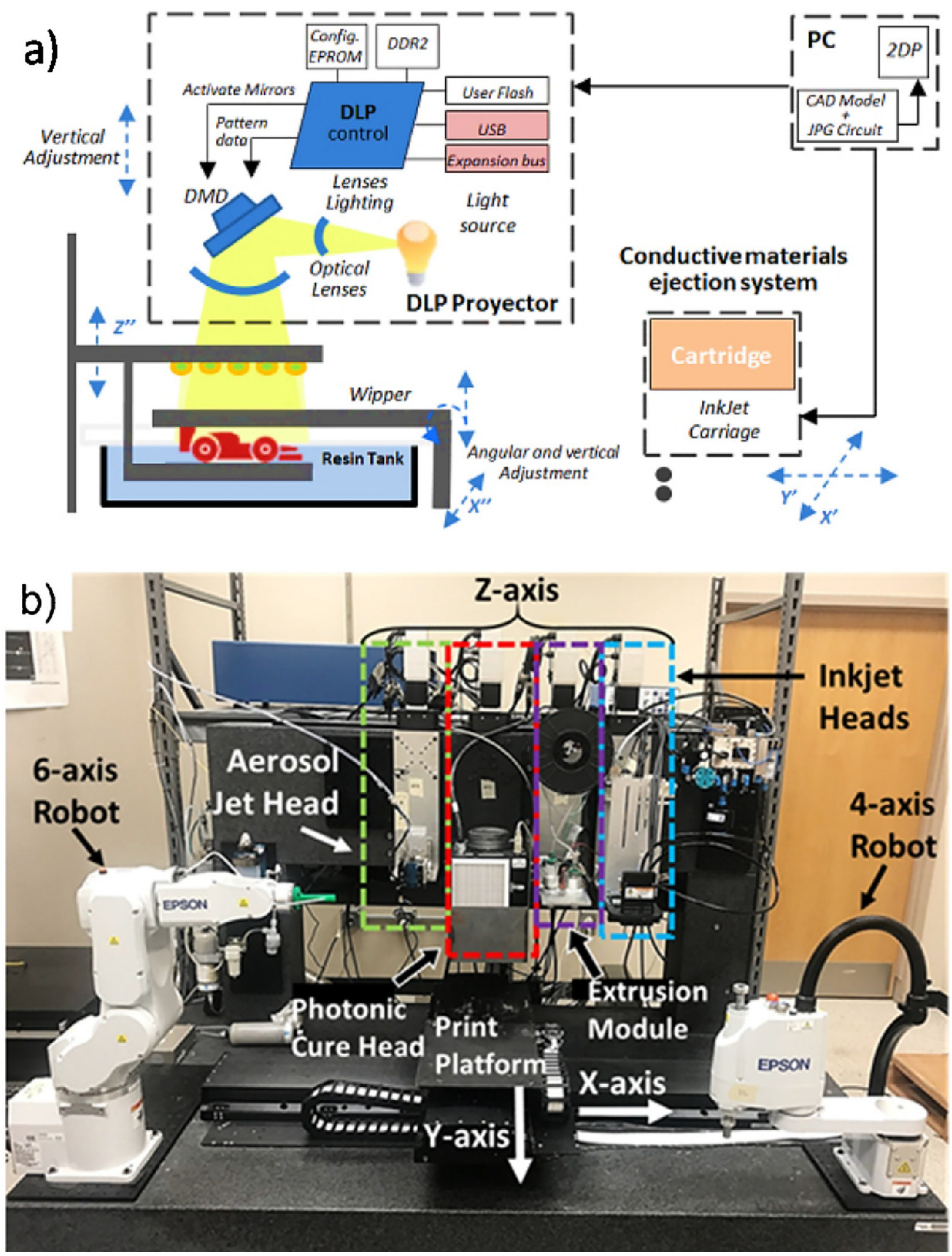


**Fig. 22.** (a) Developed DLP system (used for 3D printing) and inkjet system (for circuits) [231]. (b) m4 3D printer, a hybrid multi-material multi-method AM system for fabricating intricate structures [43].

Despite the benefits in terms of increased productivity, surface quality, and dimensional precision [16], hybrid MMAM systems pose several challenges and limitations. For instance, as DED is a fusion-based process, there are potential structural problems of dissimilar metal welding, namely, the formation of undesirable intermetallic phases [23,133]. With the implementation of the hybrid approach, certain technological challenges still remain (e.g., process planning, decision planning, use of cutting fluids, and need for post-processing). As novel hardware technologies emerge, supporting software tools remain incompetent to fulfill the potential of hybrid systems. Considering the gap in the existing literature, more research effort is required to develop comprehensive hybrid-AM software solutions [234]. Dilberoglu et al. [234] proposed a hybrid manufacturing simulator that enables users to interpret G-codes, which aids in visualizing the tool paths and shapes of the manufactured objects. Therefore, addressing the aforementioned limitations would ultimately enhance the capabilities of hybrid systems, especially for multimaterial fabrication.

## 6. Post-processing for MMAM

Post-processing of MMAM components differs from those fabricated using a single material. In case of thermal post-processing, temperature and other parameters should be adjusted considering the thermal properties of both materials. Therefore, a new post-processing method should be developed based on the compatibility with all materials in combination instead of accounting for only a single material. Post-processing confirms that the quality in terms of mechanical properties, surface properties, geometric accuracy, tolerance, and aesthetics, among other factors of AM components, satisfies the standards and design specifications. In general, the post-processing processes of AM components include but are not limited to laser micromachining, chemical processing, micromachining, manual finishing operations, electroplating, machining, tumbling, electro-polishing [235]. Several post-processing techniques for complex geometries are presented in Fig. 23, and several processes were used for application-specific components, e.g., Goh et al. [12] have discussed various post-processing treatments for 3D printed multi-material and multilayered electronics in a recently published review.

Among the AM methods, certain technologies are highly capable of manufacturing various materials in combination in a continuous and gradual process. For instance, DED and LENS AM processes can realize MMs with continuous and/or discrete material gradient with and across the layers in comparison to PBF-based technologies [241], which can fabricate only discrete material gradient. However, the high capability of the abovementioned processes for MMAM comes with enormous post-processing requirements to ensure dimensional accuracy and precise shape.

Commercially available post-processing technologies can manage components printed using homogeneous material, designed with limited complexity, and realized using only a single material for the entire component. Therefore, to progress toward achieving fully functional components for several applications, a novel system must be developed for the post-processing of MMAM or functionally graded AM components. In addition, the post-processing aspect of MM components must be considered at the design stage to minimize the requirement for this null-value-addition process [147]. Furthermore, effective post-processing, including consideration during part design, post-heat treatment, and support material removal, will enhance production efficiency as well as minimize the overall cost of the component.

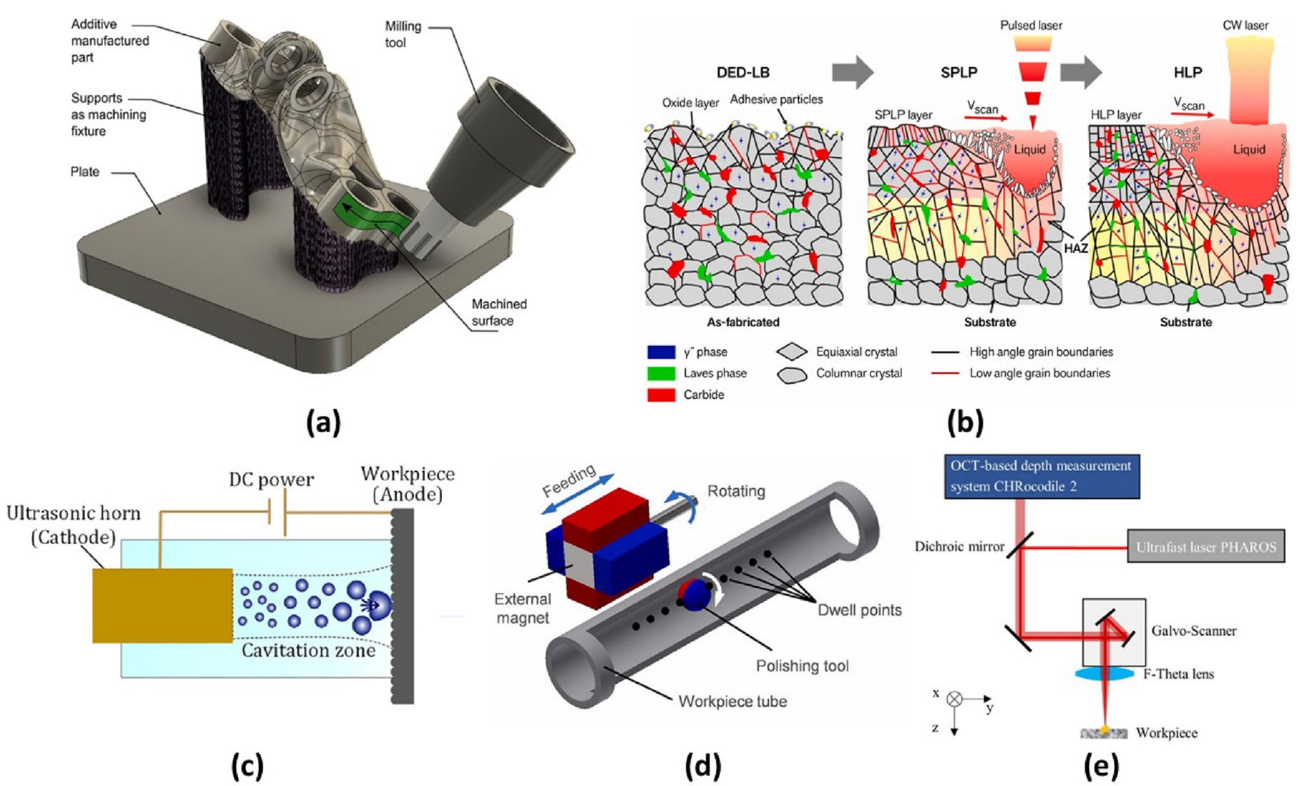


**Fig. 23.** Several processes used in post-processing of additive manufactured parts. (a) Milling machining process used for post-processing of complex AM components. Support structures used as a clamping device [236]. (b) Single-pulsed laser polishing and hybrid laser polishing processes enhance surface characteristics [237]. (c) Hybrid processes such as electrochemical polishing and cavitation peening [238]. (d) Design of magnetic polishing tool [239]. (e) Working principle of laser surface structuring and laser micromachining using ultrashort laser pulses [240].

## 7. Applications and opportunities

New studies and technical breakthroughs concentrated on rising multifunctionality in diverse applications caused by MM products providing significant benefits to the worldwide industrial and research communities. Compared to conventional structures, the ability to fabricate specialized MM structures utilizing 3D printing technology permitted particular material choices and improved various attributes of target components [242,243].

The development of MMAM has considerably aided several high-tech engineering fields, especially the aerospace industry (Fig. 24). In addition, the significance of creating and using multi-material components has been recognized, because they aid in creating lightweight designs and enable tooling testing in space. The material selection, e.g., metal powder, ceramics, polymer, and reinforced composites, and its performance is crucial for manufacturing MM components used in any aircraft or space mission [32]. Moreover, technological breakthroughs related to MMAM have resulted in great advances in medicine. Significant progress has been achieved in bio-inspired fabrications of tissue engineering structures for delicate human-body parts, including the use of biodegradable polymers for cell encapsulation and drug delivery systems [247]. Furthermore, 4D printing of various components and shape-memory polymers are imperative achievements of MMAM in the medical field [248]. Thus, the significant applications of MMAM in several industries have been elaborated in the following subsections.

### 7.1. MMAM of electronic components

MMAM has enabled the integration of electrically distinct materials, including conductors, semiconductors, and dielectrics, which are essential components in the direct production of three-dimensional electronic devices. Several electronic components can be fabricated using MMAM, such as pressure sensors, smart sensor integration, and microelectromechanical systems. Hainsworth et al. [249] introduced a malleable soft-robot actuator with embedded sensors and integrated it into a robotic arm's end-effector grasping system to aid human-occupied processes, which was impossible if not performed by 3D printing technology because of the labor of assembly. Several MMAM techniques can utilize multiple materials to fabricate electronic devices or components, such as FDM that can fabricate embedded sensors, and multi-material extrusion technology to create pressure sensors [250].

The capability of electronic components fabricated *via* single-layer deposition (traditional technique) poses several limitations and renders it unsuitable for high-performance electronic applications. However, the multilayered capacitors and inductors can be fabricated (Fig. 25a), which eliminates the previous design restriction and creates new possibilities for high-performance electronics. Although the manufacturing of components with multilayered MM eliminates several commonly required processes in conventional manufacturing [251], multilayered circuit fabrication encounters several restrictions owing to the availability of limited options in printing techniques. A particular study has successfully demonstrated the use of AM methods for fabricating various types of completely printed multilayered circuits [252]. A stretchable multilayer strain and pressure sensor are displayed in Fig. 25b. The active components include certain core elements for logic circuits that are widely employed in sophisticated electronics, e.g., transistors and diodes considered active components (Fig. 25c). These components often exhibit multilayered designs and require various materials, including semiconductors, conductors, and dielectric materials [253]. The functional components of the OLED display comprised six layers that were 3D-printed with MMs, as portrayed in Fig. 25d. The capability of MMAM 3D printing to incorporate various materials in the intricate design of LEDs is

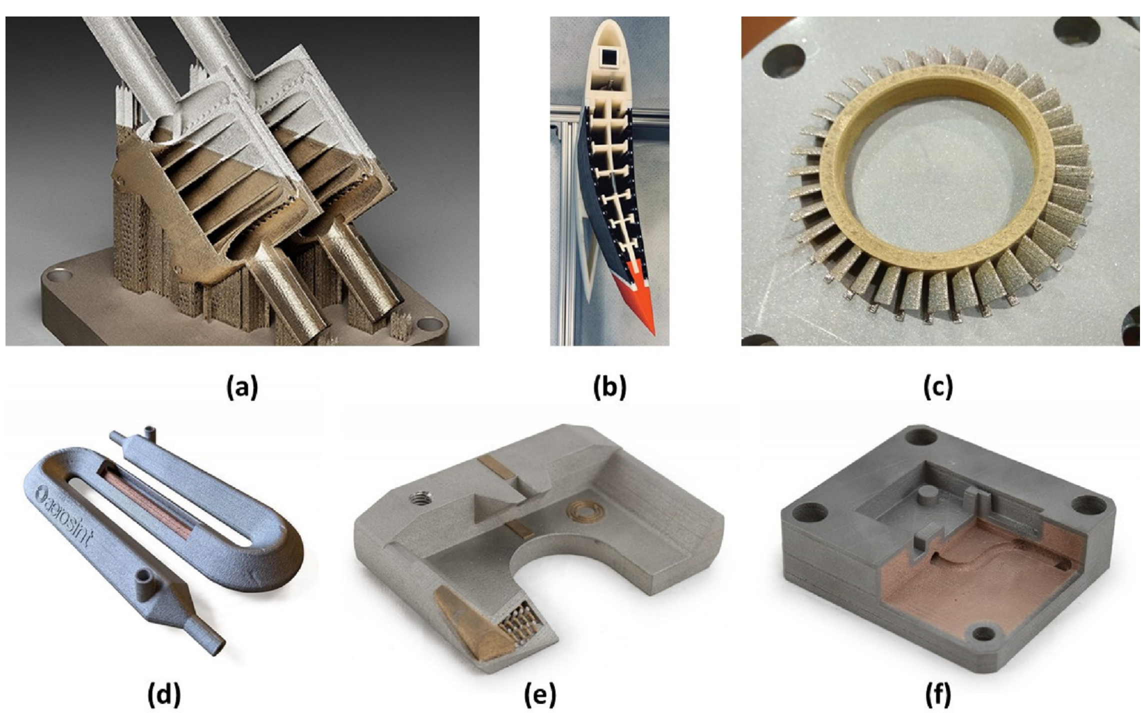


**Fig. 24.** (a) Aerospace heat exchanger AM using stainless steel SS 316 and Inconel 718 (Image copyright: Netherlands Aerospace Centre). (b) MMAM wing section inspired by fishbone to adjust wing curvature [244]. (c) MMAM aircraft engine disk fabricated using 316L stainless steel and Cu10Sn materials [245]. (d–f) Dual-metal components fabricated using copper on Aerosint recoater metal 3D printer, photo by Aerosint [246].

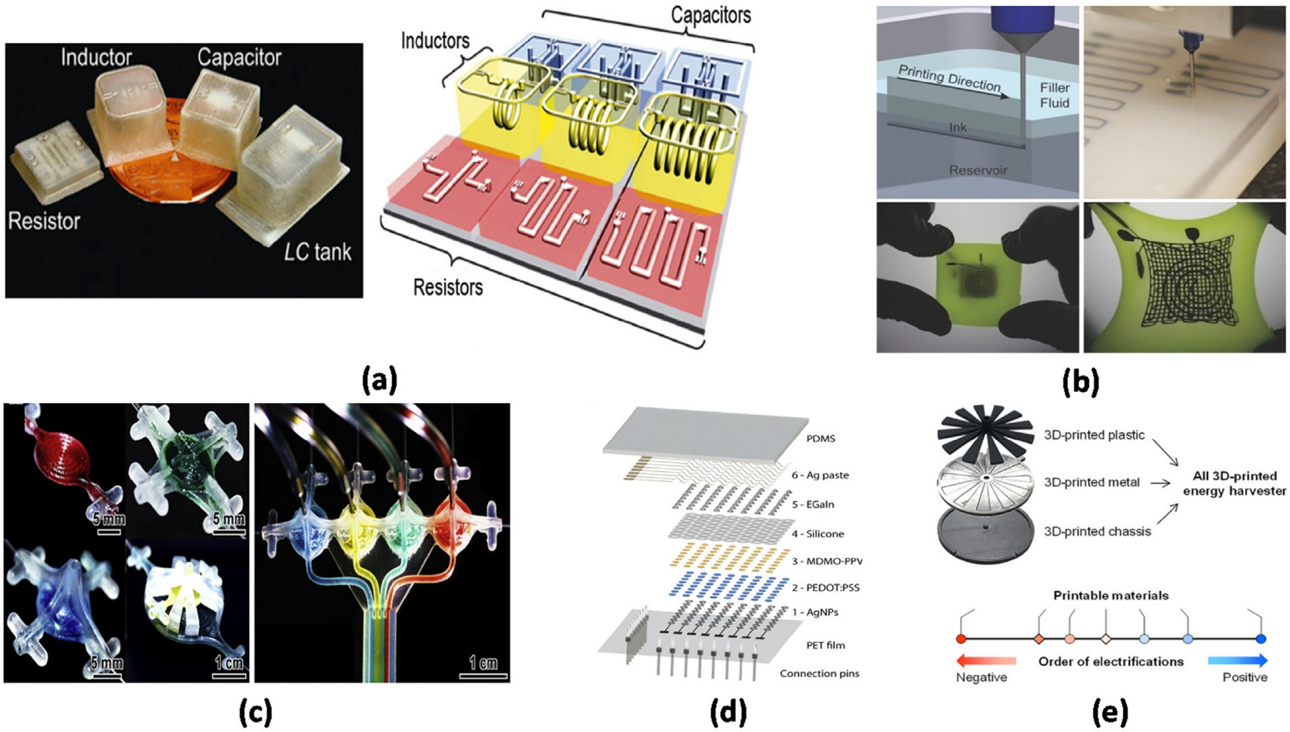


**Fig. 25.** 3D printed electronic components (a) 3D-printed passive electronic components [254] (b) Stretchable multilayer strain and pressure sensor [255] (c) 3D-printed microfluidic capacitor, diode, and two transistors and a multiflow controller [256] (d) Exploded view of OLED display demonstrating its layer-by-layer structure. Layers 1–6 are 3D-printed components [257] (e) Design and operation principle of the 3D-printed energy harvester [258].

promising for producing innovative electronics, thus, being highly researched to replace the conventional approach for electronic component fabrications.

Compared to other electronic parts and gadgets, 3D-printed energy devices are comparatively new and form a developing research field. Energy devices can be categorized into energy harvesting and energy storage devices. A schematic of the energy harvester device using metal–plastic MMs is illustrated in Fig. 25e [258,259].

### 7.2. MMAM sustainability

In the realms of rapid manufacturing, personalized design, and structural applications, the realization of MMAM will be a watershed moment. To leverage the combined or hybridized qualities of several materials, multi-material 3D printing can be potentially used in structural engineering applications. In addition, it enables the rapid production of durable, high-quality structures with the qualities of all combined materials [231]. Compared to single-material AM, MM printing is more eco-sustainable and capable of creating new innovative 4D structures that can provide particular shapes and characteristics. In addition, the components fabricated using MMAM exhibit superior tensile strength, mechanical characteristics, and durability [260].

Prospective space missions will be heavily dependent on manufacturing, as it will reduce the launch costs and offer on-demand tools for extended operations without requiring replenishment from Earth. Recently, the International Space Station (ISS) used the FDM process with ABS material to print in microgravity, but the performance of ABS in microgravity is still unclarified. This is the initial stage in manufacturing advanced materials and more intricate metal structures in space. To ensure self-sustainability, the primary objective of in-space manufacturing is to achieve meaningful space exploration that is less dependent on Earth [261]. MMAM has reduced waste materials, which is essential for in-space manufacturing applications, because self-sustenance in space is crucial. Earlier, only plastic components were capable of being recycled and reused. However, researchers plan to construct a fully integrated fabrication system, which will be a completely multi-material recycler that can process metals as well. As reported, MMAM can potentially provide diverse advantages related to sustainability. Owing to the advancements in 4D printing and smart MMAM, a wide range of industries will be impacted by technology, specifically for continuous deep-space exploration and self-sufficiency.

### 7.3. 4D printing and MMAM for soft robotics

Soft robotics involves using soft, flexible materials for robotics applications instead of the stiff, hard joints used in conventional robots. Owing to greater degrees of freedom, they offer a variety of applications in the bio-robotics, medicine, industry, and aerospace industries. The primary objective of soft robotics is to develop robots that are flexible and resemble natural movement [262]. MMAM technology offers enormous potential in developing soft robotics and actuators in various fields. The widespread use of AM technologies for the creation of soft robots is possible because of the recent advancements in the 3D printing of soft materials such as thermoplastic silicone or polyurethane-based materials [263], biologically inspired materials [264], shape-memory materials (SMMs) [265], stimulus–response materials, and stimuli-responsive materials [266]. The major AM techniques for soft robotics fabrication include FFF, direct ink writing, DLP, and SLA processes.

The actuation is the transformation of energy into mechanical work, which may be considered the backbone of a soft robot. Therefore, it influences several factors, including the motions executed by soft robots and the required manufacturing process

(Fig. 26a & b). Several actuation systems have been developed, such as flexible fluidic actuation (FFA), cable-driven actuation, shape-memory materials (SMMs), electroactive polymer (EAP), less-used actuation systems, and hybrid actuation. A Harvard research team created an AM-based pneumatic network (PneuNet) actuator to increase the wall thickness and number of chambers of PneuNets based on an FFA actuation system, which revolutionized the field of soft robotics [267]. A schematic of soft pneumatic actuators is presented in Fig. 26c & d. In addition to FFA soft robots, Shintake et al. [268] developed an innovative soft actuator based on a hybrid actuation system by utilizing a 3D printed PLA mold.

Soft robots are growing in popularity owing to their versatility in diverse applications, especially for jobs requiring safety, dexterity, and conformal deformation. Soft robotics are vital for numerous applications, including the investigation of newly uncharted regions such as biomedical (robotic muscles, climbing robots, edible robots, wearable robots, and prosthetic robots; Fig. 27), aerospace, electronics, biomimetic, food and agriculture, e-textiles, manipulation of objects, and automation. Given the present rate of advancement in the field of soft robotics, such robots may soon be realized as MMAM for significantly enhancing this development.

Despite all the advancements in materials science and system design, significant gaps are still present in this area in terms of conventional production methods, which concerns the manufacturing of several materials and the adhesion between the components for producing soft robots. In soft robotics, however, MMAM technologies are gradually assuming an increasingly essential role Owing to their inherent qualities which are ideally suited to the needs of soft robot production, such as the scarcity of soft materials on the market and commercial 3D printers suitable for soft materials. Thus, researchers should focus on enhancing the use of MMAM in soft robotics, enhancing the process parameters of unconventional materials, and printing techniques to resolve several printing issues that negatively impact 3D-printed soft robots.

### 7.4. Multi-material bioprinting

The term "bio-printing" refers to the use of the "material transfer process" in producing biological materials such as cells, tissues, and organs employing cells, biological molecules, and chemicals, as depicted in Fig. 28a. Owing to an upsurge in organ transplants but limited availability of organs for transplant including the present limits of tissue engineering, the creation of a 3D or 4D bio-printing engineering branch is highly useful [275]. Single-material-based 3D bio-printing typically results in restricted diversity in the physical and chemical characteristics of products. Therefore, a rapidly growing interest in the modification and diversification of generic printing materials by combining them with other materials exhibiting special properties to create a 3D printable composite with excellent performance, such as ideal mechanical properties, desired biocompatibility, improved biomimicking of tissues structures, and appropriate fidelity promotes the application of MMAM in the bioprinting field [276].

The objective of MM bio-printing is to resolve the issues related to single-material and imitate the intricate architecture of biological tissue and organs in scaffolds [277]. Multi-material 3D bio-printing integrates diverse qualities into a single project and delivers useful items with vital strength and performance [278]. Technological breakthroughs in MMAM technology have resulted in great advances in medicine. Significant progress has been achieved in bio-inspired fabrications of tissue engineering [279] structures for delicate human body parts and the use of biodegradable polymers for cell encapsulation and drug delivery systems. This has enabled the construction of multiscale, fully functioning, heterogeneous, and multicellular hepatic structures that can deliver effi-

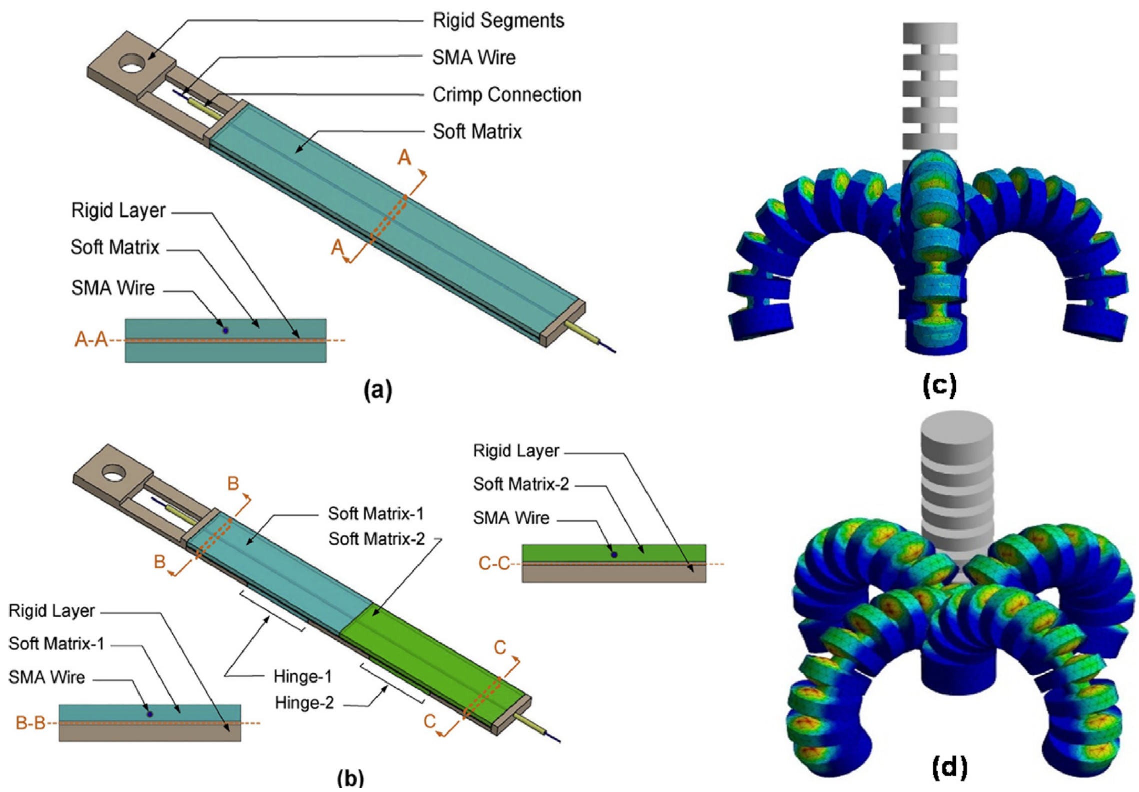


**Fig. 26.** Design of 3D-printed soft MM bending actuators; (a) bending actuator with a uniform cross-section over entire effective length of actuator; (b) bending actuator with multiple segments of equal lengths [269]; innovative soft pneumatic actuators with (c) three and (d) four chambers can bend in any direction [270].

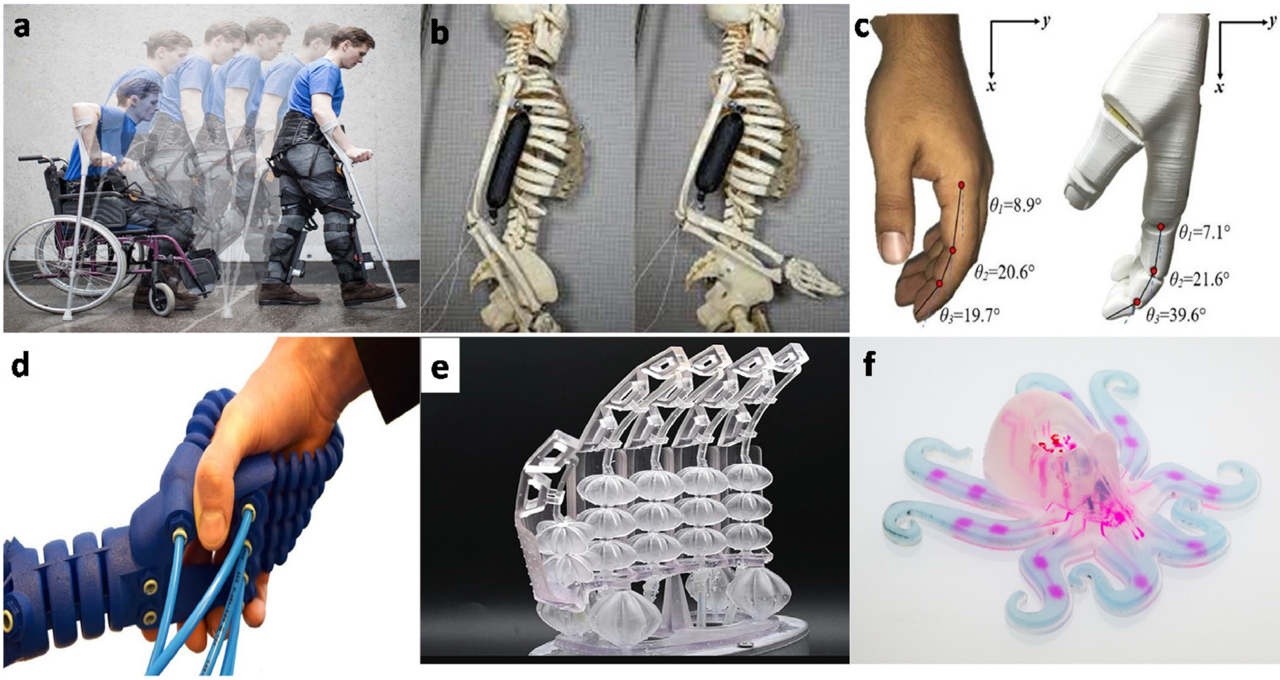


**Fig. 27.** 3D-printed different applications of soft robotics: (a) wearable robotics for assistance and rehabilitation [271]; (b) synthetic muscle mimics natural muscle that is three times stronger [272]; (c) natural-pose joint angles for human hand and 3D-printed prosthetic hand [273]. (d) Soft robotics hands respond to human grip [249]; (e) 3D-printed biomimetic artificial muscles using soft actuators that contract and elongate [274]. (f) Octobot: a self-powered soft 3D-printed robot [260].

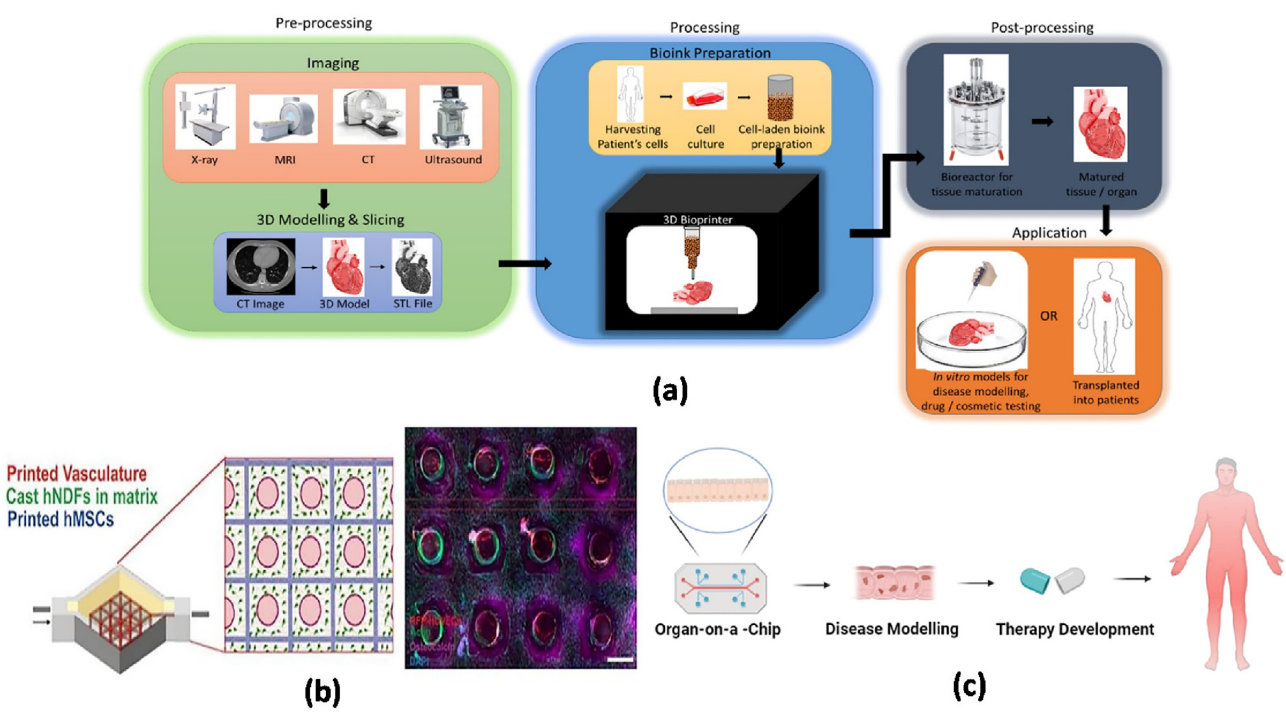


**Fig. 28.** (a) Diagrammatic representation of 3D bioprinting process [282]. (b) Vascularized tissues fabricated by simultaneously printing cell-laden bio-ink and sacrificial bio-ink [283]. (c) Organ-on-a-chip development for disease modeling and therapy development for precision modeling [280].

cient and expedient performance. The schematic of vascularized tissues fabricated by simultaneously printing cell-laden bio-ink and sacrificial bio-ink is illustrated in Fig. 28b.

In addition, the co-printing of hydrogel and sacrificial materials allows the utilization of MM bio-printing for fabricating physiologically appropriate vasculature, which made possible with multi-material approach. Furthermore, organ-on-a-chip is another technique for creating biological tissues. For biological investigations, tissue/disease modeling and drug screening models have been used to simulate human organs, as depicted in Fig. 28c [280].

Moreover, MM bio-printing offers the potential to produce more accurate biomimetic tumor models, because it can replicate the intricate 3D microenvironments of tumors. Based on its dynamics, bio-printed heterogeneous tumor models facilitate the process of tumor growth [281].

In the past few years, 3D bioprinting has achieved remarkable development, spanning from straightforward proof-of-concept prints to intricate, tissue-like structures created from various materials. The techniques for 3D bioprinting promise immense potential for development. However, certain issues are still required to be resolved in further investigations into bioprinting methods, cellular sources, biomaterial choices, etc. Compared to real tissues/organs, the lack of accuracy is among the most significant disadvantages of existing bioprinting methods. Mostly, tissue/organ architectures are extremely delicate for existing bioprinting technologies. Additionally, slow printing speed for complicated structures is problematic regarding the biofabrication of several multi-materials in sequence [277].

### *7.5. MMAM for architecture and construction*

The obstacles encountered by construction industry results in excessive material and energy consumption and nearly stagnant productivity growth [284]. The current construction techniques pose several drawbacks, including high construction limitations and the work required to align the components in accordance with them, as well as a high vulnerability to construction errors caused by inadequate communication between the design and execution. A significant portion of construction work is conducted in an ineffective and labor-intensive manner [285]. To address these issues, AM integrated with multiple materials can significantly alter the current manufacturing and construction operations. MMAM can fabricate objects with varying material qualities without additional assembly and multi-material architecture design. This modification can manage inefficiencies in manufacturing and construction by reducing the number of required production processes as well as providing solutions to challenges connected with the connection of specific materials or elements [286].

Although most AM procedures can use various materials, limited research has been conducted to apply these techniques to the architecture and construction field. Concrete material delivers acceptable qualities for construction at a moderate cost. Depending on the diverse characteristics of the mixture and the specific additives or fillers in concrete, as such, MMAM enables to produce this mixture, therefore, mixed concrete offers a wide range of qualities and uses compared to conventional applications. In context, numerous approaches can be employed to alter these qualities by adjusting the concrete mixture composition, thereby rendering concrete as an ideal material for use in MMAM. The most prevalent strategy in MMAM of concrete is to alter the material density. The MIT Mediated Matter group studied aluminum powder as a foaming agent for concrete, also known as variable density concrete. In presence of aluminum powder and uncured concrete, the evolution of hydrogen gas results in the foaming of the concrete mix that reduces the overall weight and consumption [287]. Additionally, an extensive investigation of additive-based lightweight concrete demonstrated that additives significantly influence the overall performance of concrete mixture [288]. Alternatively, MMAM can be utilized with concrete with the addition of fibers, which can improve both the tensile strength and ductility of the material.

MMAM aims to alleviate more widespread problems that plague the construction industry; for instance, high consumption of materials, limited adaptability of components to varying environmental conditions and structural requirements, and an overload of complexity in the assembly and construction procedures currently in use.

## 8. Limitations and challenges

MMAM does not indicate that the advantages of each material can be necessarily employed for desirable applications. In certain cases, researchers have exploited the disadvantages of the joining MMs, such as insufficient bi-material bonding and residual stress at the interface. Controlling the AM process is challenging owing to the miscibility and wetting constraints of various materials and the dissimilarities in their properties (e.g., thermal conductivities and expansions, melting points) [155]. Mismatches in coefficients rely on the similar or dissimilar material selection, bi-material systems such as metals–plastics, metals–ceramics plastics–ceramics can create huge mismatches, resulting in defects such as cracks, pores, residual stresses that affect the integrity of the components, dimensional stability, crack growth resistance, and mechanical properties in real-life applications [23]. Therefore, future research should focus on the material selection, design, and manufacturing aspects of MMAM parts in particular the residual and surface stresses generated during AM of metallic parts [289]. Specifically, understanding the composition and optimal distribution of appropriate materials, mechanisms of reaction kinetics, bonding, residual stresses, and cracking mechanics are essential for designing MMs. As such, nonuniform properties in the designed MM components are complex; efforts in materials designs, characteristics, chemical compositions, properties, and manufacturing constraints are necessary prior to MMAM processing [290].

Furthermore, for the specific process, alloy formation at the interface of bi-material *in-situ* is far from an equilibrium state and difficult to characterize [155]. The defects at the grain boundaries, e.g., microcracks, usually occur in the metal material PBF process. In addition, other outstanding challenges such as low production throughput, poor scalability and surface finish, low interfacial bonding, and high cross-contamination should be overcome [291]. Apart from the basic understanding, technical breakthroughs such as the design-related concern of the development of CAD programs, design protocols, and procedures should be developed [290].

AM of a complex MM component may require adjusting the materials thousands of times. Thus, MMAM processes are more gradual in comparison to single-material AM processes. Furthermore, varying materials does not mean that swapping the ink cartridge/spool, which requires various printing process parameters that vary with the jetting nozzles in extrusion-based processes. Thus, most of the AM processes designed for fabricating single-material components face enormous challenges while employing them for manufacturing MM components. Furthermore, defining and distributing multi-materials in a single component requires extensive knowledge of 3D printable materials, their chemical compositions, optimal printing parameters, and manufacturing constraints [180,292]. Owing to several limitations such as a limited selection of 3D printable materials [293], scanty design guidelines exist on material compatibility and multi-material 3D printability, which is a major limitation in realizing MMAM component fabrication for the end-user.

As depicted in Fig. 29, most analyzed existing research applied empirical methodologies to investigate the properties of multi-material AM components (Fig. 13). However, the majority of them did not conduct validation studies of experimental results using FEM. As indicated in Fig. 29, approximately 140 studies fabricated MM samples, whereas only 23 studies included finite element analysis validation of their empirical results. In addition, a few researchers have used optimization algorithms to improve their multi-material designs or fabrication methodology. Overall, a negligible amount of literature has focused on software development for MMAM. Therefore, MMAM software requires a significant

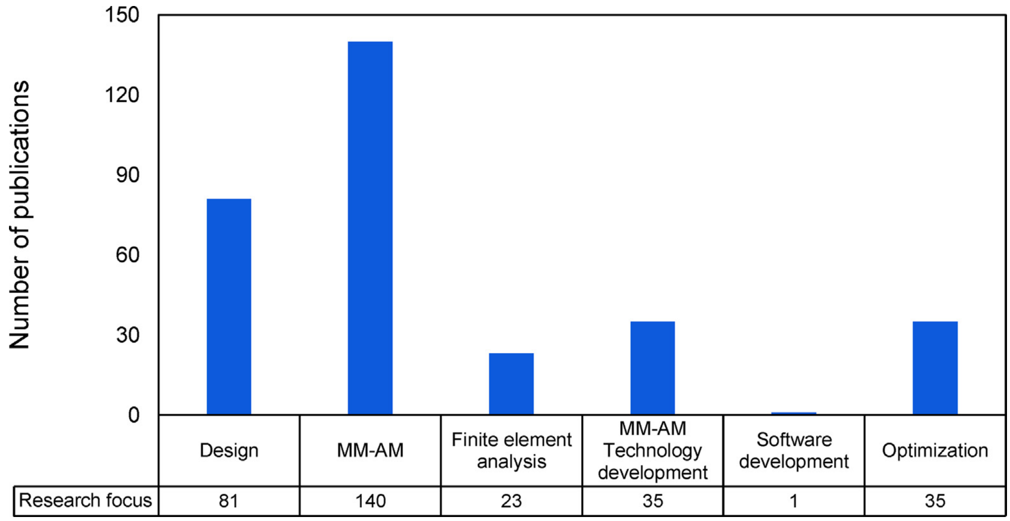


**Fig. 29.** Analyzed research literature of MMAM classified into six categories.

amount of research and development to utilize the full potential of MM research. Another important aspect of the software for consideration is the slicing of MM components and appropriate AM process selection [294]. Although Hascoet [295] attempted to formulate the slicing of multi-material gradients, novel techniques for preparation, analysis, and slicing of multi-material and/or functionally graded parts are still required.

Certain research groups have focused on AM processes with MM manufacturing capabilities and new internal structures based on high performance computers and optimization tools. Metamaterials and advanced lattice structures can be applied in flexible materials and have potential in various disciplines, such as aerospace, civil, textile, and tissue engineering applications. Using micro/nano multi-material AM with metamaterials and lattice structures can result in innovative functional components.

## 9. Summary and future recommendations

This article summarizes the progress of several aspects of MM research, including various types of MM compositions, component design, modelling, and their analysis strategies. Furthermore, applications, post-processing, technologies challenges, and potential research gaps are significantly discussed and reviewed. A summary of findings, challenges, issues, and research gaps for future research is stated as follows:

• MMAM opens a range of new opportunities for the design, complexity, and functionality of highly personalized and high-value products with improved properties. The integration of materials at various scales can customize the thermal, electrical, mechanical, optical, and multifunctional properties of the components.

• Majority of the researchers have focused on certain selected multi-materials of polymer and metal based. However, several alternative materials, e.g., elastomers and hydrogels, have not been studied significantly. For metal MMs, process- and material-related challenges such as dimensional accuracies and post-processing requirements form the major challenges for consideration.

• To date, no significant research has been conducted regarding FFF-based MMAM, focusing on the relationship between printing conditions and dissimilar materials interface quality. The lack of information concerning multi-material extrusion-based 3D printing persists to be the area that remains unexplored, specifically the problems such as equipment, component design, and printing materials.

• According to this review, the majority of the analyzed literature studied only the compressive and tensile properties of MM components. Thus, other properties—chemical, fatigue, impact, etc.—require further investigation. In addition, multi-objective investigations should be performed to explore the complex characteristics of MMs.

• The standardized design of the experiment has not been employed to investigate MM properties. Therefore, the results and methodologies of existing literature cannot be feasibly compared for analysis. Most existing literature has focused only on basic properties, and most characteristics such as fatigue, 3D printing properties, optical, impact/shock, flexural, and shear properties of MMAM components require considerable investigation.

• The research on designing MM components is very limited, and the design rules for each AM process and material requires further study. In particular, in-depth studies should be conducted on MM component design for each AM process.

• Only a few software packages are available with features for designing and optimizing MMs. Furthermore, the existing packages do not offer full-range finite element analysis of MM. In particular, no software package is specifically developed for the design, optimization, and simulations of MM-related studies. Thus, significant development is required in this research field. With the development of new technologies (e.g., big data, machine learning, digital twin), new methods using these new techniques of MM design for AM can be explored in the future.

• Lastly, it is revealed that commercially available post-processing technologies still handle components printed using homogeneous material, designed with limited complexity, and realized using only a single material for the entire component. Therefore, a novel system must be developed for the post-processing of MMAM or functionally graded AM components to progress towards using fully functional parts for several applications.

This review will provide insights to researchers and engineers for designing and manufacturing complex nature-inspired objects by incorporating MMs.

## Data availability

Data will be made available on request.

## 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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