A review on the ceramic additive manufacturing technologies and availability of equipment and materials

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Abstract

Ceramic additive manufacturing allows the fabrication of small series of complex parts without the high costs of molds usually associated with traditional ceramic processing. Although research into ceramic 3D printing by all technologies started back in the 90s, its industrial application is still quite restricted when compared to polymers and metals, which is related to the limited availability and costs of equipment and materials for such applications. This review examined the advantages and limitations of each process (binder jetting, direct ink writing, directed energy deposition, fused deposition, material jetting, selective laser sintering, selective laser melting, and vat photopolymerization), discussing their particularities. It also summarized the commercially available 3D printers and raw materials for ceramic processing, pointing out to trends and challenges of each technology.

Keywords: 3D printing, additive manufacturing, ceramics, digital light processing, stereolithography, viscosity.

CERAMIC ADDITIVE MANUFACTURING

3D printing or additive manufacturing (AM) is a set of processes that fabricate parts by adding materials layer by layer. After the great development of additive manufacturing of polymers and metals, developments of this technique applied to ceramic materials have gained prominence in recent years [1, 2]. Ceramic AM enables the fabrication of customized complex 3D parts without molds [3-5], reducing costs and lead times [6]. Ceramic parts can be produced by a variety of AM technologies [5], choosing one of them for a given application should consider their strengths, weaknesses, and commercial availability of equipment and feedstock. Although research into ceramic 3D printing by all technologies started back in the 90s [7-12], its industrial application is still quite restricted when compared to polymers and metals [6, 13, 14]. The widespread of ceramic AM depends on technological availability [15]. Thus, proper feedstock [13] and equipment [15] availability have been an issue. Moreover, many technologies present high-priced raw materials, and their supply is linked to the equipment supplier. Several companies have launched their ceramic 3D printing solutions in the last 5 years. To the best of our knowledge, this is the first review focused on commercially available raw materials and 3D-printing systems for each technology of ceramic AM. This paper also discusses the technologies' capabilities and limitations, and the main trends and challenges of ceramic additive manufacturing.

TECHNOLOGIES

According to ISO/ASTM 52900:2015 standard [16], additive manufacturing technologies can be divided into two types. In the multi-step (or indirect [17]) processes, two or more operations are needed to reach the final part. On the other hand, the single-step (or direct [17]) processes achieve the final shape and properties in a single operation. For ceramic AM, multi-step processes are the most common. Additives and binders are used to create a green body that is subsequently treated for debinding (to eliminate the organics) and sintered (to increase density) [5, 13, 18]. The debinding is a critical step to successfully obtaining the ceramic parts and the heating rates must be suitable to avoid cracks and/or delamination [5, 19, 20]. Also, debinding becomes more difficult with increasing wall thickness [5], and some AM technologies that use a high amount of organic material, such as vat photopolymerization and fused deposition, have the maximum wall thickness limited [6]. Table I shows a summary of multi-step ceramic AM technologies, their main characteristics, equipment suppliers, and commercially available feedstock. All these technologies already have commercial solutions for printing advanced ceramics such as aluminum oxide. Although all these technologies are layerwise processes, they differ in the way each layer is formed, which provides very different products for each technology.

Binder jetting, direct ink writing, and selective laser sintering are limited to manufacturing porous parts with poor surface finish. On the other hand, these technologies are capable of producing large parts. Direct ink writing is a low-cost technology with paste feedstock indicated for parts with smaller geometrical complexity. On the other hand,

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Table I - Ceramic additive manufacturing technologies characteristics and commercially available feedstock and equipment suppliers.

Technology	Advantage	Disadvantage	Best suited to	Raw material	Ceramic particle size (μm)	Equipment supplier	Commercially available material
Binder jetting	Scalability, throughput, complex part, overhanging structure	Low density, poor surface finish, accuracy	Porous ceramic, tissue engineering	Powder, binder (bonding agent)	>30 [21]	Cometrue, Voxeljet	Al_2O_3, ZrO_2, SiC
Direct ink writing (robocasting)	Low cost, fast manufacturing	Poor resolution, surface finish, smaller geometrical complexity	Porous ceramic, tissue engineering	Ceramic slurry	0.03-2.20 [15]	3D Potter, Cerambot, Duraprinter, Rapidia, StoneFlower, Vorm vrij, WASP	Al_2O_3, cordeirite, zircon, porcelain, clay
Fused deposition	Widely known process (for polymer)	Surface finish, staircase effect	Structural ceramic	Thermoplastic polymer, dispersed ceramic powder	0.1-1.0 [15, 22-25]	3D-Figo, Pollen, Nanoe, Uprise 3D, Xerion	Al_2O_3, ZrO_2, Si_3N_4, B_4C
Material jetting	Surface finish, resolution	Low productivity, coffee stain	Compact part	Powder, solvent	0.2-0.6 [6, 26-30]	XJET3D, Tritone	Al_2O_3, ZrO_2
Selective laser sintering	Overhanging structure without secondary support	Low density, poor surface finish, accuracy	Porous ceramic, tissue engineering	Powder, agglomerate, binder	10-100 [17]	Wuhan Huake 3D Technol.	-
Vat photo-polymerization	Precision, surface finish	Expensive feedstock, limited wall thickness	Structural ceramic	Photo-sensitive polymer, powder	0.1-0.5 [31]	3DCeram, 10dim Tech, Admatec, AON, Carima, Fortify, Lithoz, Photocentric, Prodways, SK Fine, Soosolid, Tethon 3D	Al_2O_3, ZrO_2, Si_3N_4, AlN, SiO_2, cordierite, mullite, TCP, hydroxyapatite

binder jetting and selective laser sintering are powder bed technologies capable of producing complex parts with overhanging structures without secondary supports. Among them, binder jetting stands out for its high scalability and throughput. Fused deposition and vat photopolymerization are the processes indicated to produce dense structural parts with adequate mechanical properties. However, both technologies present limited wall thickness [6] due to the high amount of organic associated with the processes. Vat photopolymerization stands out for having excellent resolution and surface finish [1, 6, 32, 33]. It is the most well-established technology having an industrial readiness level [6] with a wide range of commercially available advanced ceramic feedstock and more than 10 equipment suppliers. Finally, material jetting is a suspension-based technology still little explored in ceramic manufacturing. Considering that it is a low productivity process, it is indicated for very compact applications such as the manufacture of cathodes and electrolytes. All indirect technologies are discussed in detail in their sections.

On the other hand, the single-step processes for ceramic additive manufacturing are directed energy deposition (DED) and selective laser melting (SLM). These technologies can become the fastest way to produce ceramics by AM since they do not include the time-consuming debinding and sintering steps [6, 14]. However, ceramic materials have a high melting point and limited thermal shock resistance [6, 14]. Thus, thermal gradients, an intrinsic feature of these technologies, are generated by inducing thermal stresses, which induce delamination and cracks [6, 13, 14]. Accordingly, their application is still restricted to research, not much development has been achieved [1], and no dedicated system is commercially available.

Binder jetting

Binder jetting is an AM technology in which a liquid bonding agent is selectively deposited to join powder materials [16]. Fig. 1 illustrates the schematic of the binder jetting technology with two different powder feeding approaches. In both cases, after a new layer of powder is spread in the powder bed, a print head jets the liquid binder into the powder, creating layers with a predefined 2D pattern [21]. Fig. 1a illustrates the use of a hopper feeding system. The hopper deposits the powder from its reservoir and subsequently, it is spread by a roller [21]. On the other hand, Fig. 1b shows a binder jetting with two chambers: the powder feed supplier and the build chamber. In this case, the roller performs the transport of the powder and recoating [34]. This technology is the most suitable for large parts, having scalability and throughput [6, 14]. Also, it is capable of producing complex parts with overhanging structures due to its self-supporting powder-bed [6]. On the other hand, the ceramic particles should be large (>30 μm [21]) to ensure flowability in the layer spreading which decreases the density of the final part and provides a poor surface finish [1, 5, 6, 13], as shown in Fig. 2. Thus, binder jetting is best suited to porous parts, not being adequate for structural parts [5]. The research group from Texas A&M University led by Profs. Pei and Ma is the leading research group on ceramic binder jetting. Their research is related to the improvement of sinterability and part strength by improving the feedstock (particle coating [36], granulation of nanopowders [37], etc.), having already manufactured alumina [38], porcelain [39], and silicon carbide [40]. In addition, some other groups are researching this technology, some of them having studied its applicability in support of heterogeneous catalysis [41] and bone tissue [42].

The low density of the sintered parts is an important issue for ceramic binder jetting [38] and the relative density usually ranges between 50% and 75% [37, 38, 42-48]. On the other hand, infiltration has been used to improve sintered parts properties. For example, Vogt et al. [49] reached alumina samples with densities above 90% and improved bending strength from 60 to 145 MPa with infiltration of nanometric alumina powder. Also, Maleksaeedi et al. [50] showed that infiltration can also improve surface quality, reducing surface roughness (Ra) from 13.2 to 0.9 μm. Ceramic binder jetting has several parameters that have a significant influence on the final parts. Such parameters have been used in the following ranges in related works [37, 41, 42, 44-47, 49, 51-55]: layer thickness: 30-150 μm; recoater speed: 50-80 mm/s; roller recoating speed: 100-500 rpm; and binder saturation or fraction of pore space filled with the binder: 50-100%. Also, the binder must be compatible with the process with viscosity around 10 mPa.s and surface tension from 23 to 30 mN/m [41]. It is usually water-based with some additives for surface tension and viscosity adjustment [41] as isopropyl alcohol [41, 45, 48], diethylene-glycol [48], polyvinyl alcohol [56], and glycerol [45]. Also, some related works [38, 44, 49, 51] used a commercial aqueous binder (BA005, ExOne). Table II shows the additive manufacturing systems available for binder jetting of ceramics. While the M10 Ceramic 3D printer (ComeTrue)

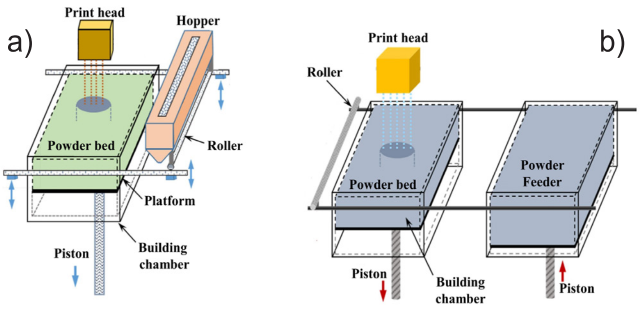

Figure 1: Schematics of the binder jetting technology with two different powder feeding approaches (adapted from [34]): a) powder is supplied using a hopper and oscillation and subsequently spread using a roller; and b) powder is swiped from the powder feeder and spread in the powder bed by the roller.

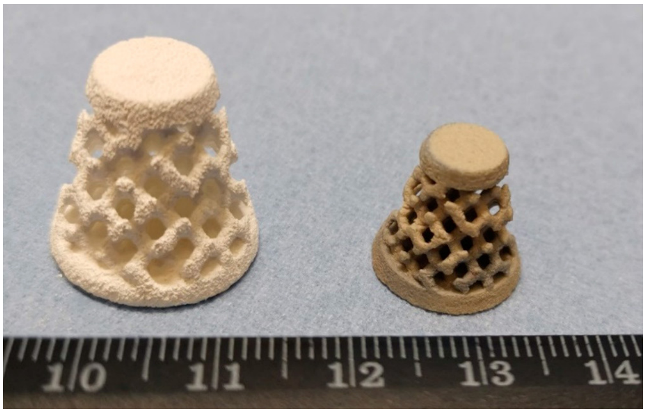

Figure 2: Barium titanate samples with lattice structures fabricated by binder jetting [35].

Table II - Binder jetting of ceramics: equipment suppliers.

Manufacturer	Headquarter	System	Indicated layer thickness (μm)	Maximum build size (mm^3)	Commercially available material
Cometrue [57]	Taiwan	M10 Ceramic 3D printer	120	200x160x150	Traditional ceramic for ceramic craft and education
Voxeljet [58-60]	Germany	VX200	300	300x200x150	Al_2O_3, ZrO_2, SiC
		VX1000	300	1000x600x500	-

is focused on the additive manufacturing of traditional ceramics [57], Voxeljet has two systems capable of working with advanced ceramics, being VX200 [59] suitable for research and development and VX1000 [60] an industrial printer capable of producing large parts with a building volume of 1000x600x500 mm^3. Moreover, ExOne company has been working on the development of ceramic feedstock for binder jetting presenting advanced ceramics aluminum oxide, zirconium oxide, silicon carbide, and boron nitride under development [61]. Also, some researchers have used non-dedicated ExOne 3D printers to process ceramics [38, 44, 49].

Direct ink writing

Direct ink writing (DIW), also called robocasting is an AM based on material extrusion in which a ceramic slurry (paste) is selectively dispensed through a nozzle (Fig. 3). The paste is forced through the nozzle by a piston, which can be operated in two ways: pneumatic or mechanic. A detailed review of the different extruders available for this technology has already been published [62]. DIW is a low-cost [6, 62] and fast additive manufacturing process which produces parts with limited geometrical complexity, poor resolution, and surface finish [6, 13], as illustrated in Fig. 4. A research group from the University of Aveiro led by Prof. Ferreira has made important contributions to the manufacture of scaffolds/lattices by ceramic DIW, developing suspensions with particles (zirconia [64, 65], bioactive glass [66], piezoelectric ceramics [67], etc.) and aqueous solution with dispersant, binder, and coagulant agent. Moreover, the Department of Industrial Engineering from the University of Padua also stands out with research on ceramic direct ink writing. Profs. Bernardo and Colombo have been leading research in the area, mostly using preceramic silicone as the binder [68-74] to create scaffolds made of a variety of ceramic and glass materials with potential application in bone tissue engineering applications.

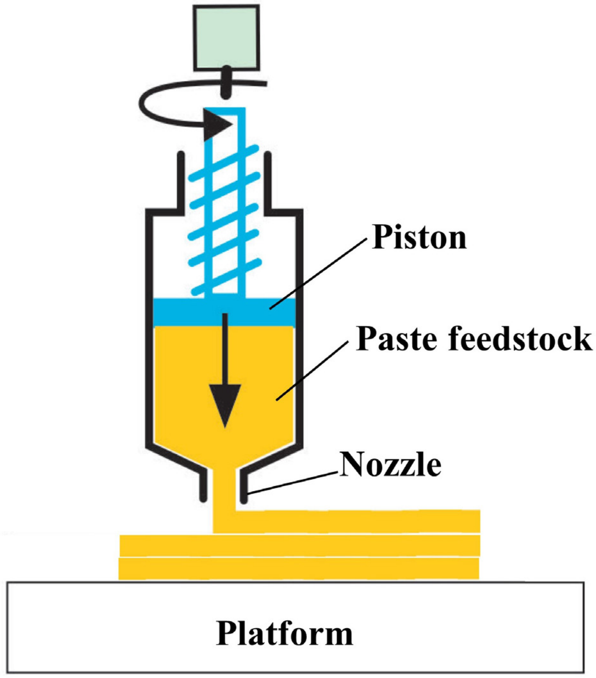

Figure 3: Schematic of the direct ink writing technology (adapted from [62]).

Ceramic direct ink writing has been shown to be suitable for the fabrication of highly porous ceramic scaffolds and foams with porosity, which can exceed 80% [72]. Gaddam et al. [65] produced zirconia scaffolds with macroporosity of about 70% and average compressive strength of ~236 MPa. For this process, the reported shrinkage usually varies between 17% and 24% [72, 75, 76]. The surface quality of this process is a major issue and roughness (RA) above 20 μm has been reported [77]. Furthermore, the side surface may present such a distorted surface that roughness can not even be measured [78]. In this process, the characteristics of the ceramic slurry, usually composed of ceramic powder, deionized water, and additives, are very important [2, 79]. The high-loaded ceramic slurries (typically 40-50 vol%) must have adequate rheological behavior and should be smoothly extruded through a narrow nozzle without clogging. Besides, it should be self-supporting (to avoid collapsing) and be able to retain its shape [79]. Thus, suitable additives must be chosen. Hydroxypropyl methylcellulose and polyethyleneimine have been extensively used as viscosifying and coagulant agents, respectively [64, 65, 67,

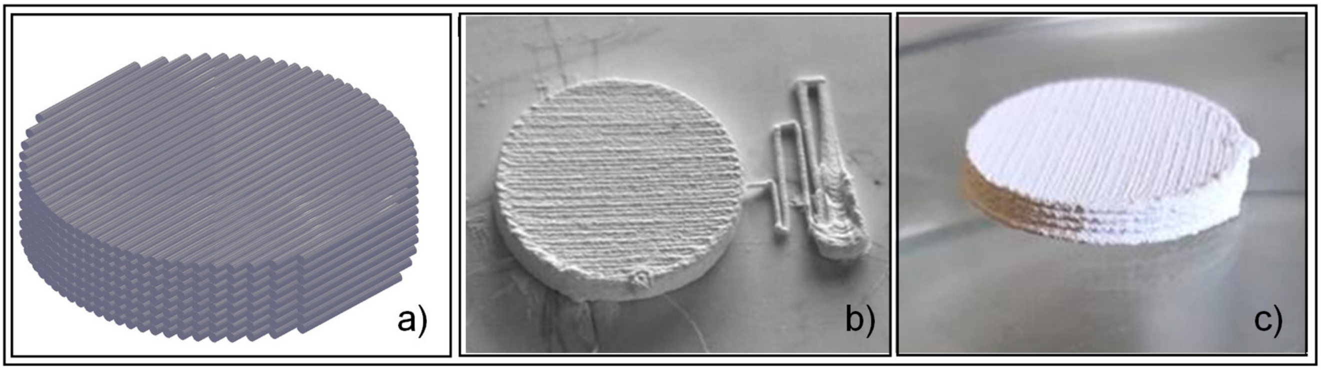

Figure 4: Al_2O_3-Y_2O_3 sample fabricated by DIW [63]: a) schematic representation of layers deposition; b) 3D printed sample; and c) 3D printed sample after drying.

75, 80, 81]. Also, a variety of commercial dispersants have been employed, depending on the selected ceramic powder. However, details of their compositions are not disclosed by the manufacturer. In addition, the choice of nozzle opening diameter must be made properly to avoid choking the nozzle or sudden release of the slurry [2, 82]. The nozzle should have at least 15 times the size of the largest particle size [2, 82] and the optimum diameter is typically between 400 to 800 µm [2]. In addition, the printing speed is a parameter that should be selected carefully. The printing speed should be adequate to form a continuous strut and to achieve inter-layer bonding and small modification of this parameter may

Table III - Direct ink writing of ceramics: equipment suppliers.

Manufacturer	Headquarter	System	Indicated layer thickness (µm)	Maximum build size (mm^3)	Commercially available material
3D Potter [87, 88]	USA	3D PotterBot Micro 10	N/S	280x265x305	Clay
		3D PotterBot 10 Pro	N/S	415x405x500	
		3D PotterBot Super 10	N/S	415x405x500	
		3D PotterBot 10 XL	N/S	490x490x710	
		Scara Mini V1-329	N/S	Ø1473x584	Cementitious material
		Scara Standard	N/S	Ø1829x1143	
		Scara Elite	N/S	Ø2845x2134	
		Scara Heavy Duty	N/S	Ø3658x2743	
Cerambot [89, 90]	China	Eazao Zero	400-1000	150x150x240	Clay, porcelain
		Eazao Mega 5	400-2000	370x390x470	
Duraprinter 3D [91]	Brazil	E01	>100	Ø145x180	Clay
		E02	>100	Ø200x360	
		E03	>150	Ø300x450	
Lynxter [92]	France	S600D+PAS11	N/S	Ø380x600	Al_2O_3, cordierite, zircon, porcelain, clay
Rapidia [93, 94]	Canada	ExOne Metal Designlab	N/S	200x280x200	Al_2O_3, ZrO_2 (in development)
StoneFlower [95]	Germany	StoneFlower 4.0	300-4000	480x550x500	Clay, porcelain
WASP [96-98]	Italy	Delta WASP 2040 Clay	>500	Ø200x400	Clay, stoneware, earthenware, porcelain
		Delta WASP 3MT Concrete	N/S	Ø1000x1000	Concrete mortar (particle size ≤1 mm), fiber reinforced concrete, earth-based mixture + natural fiber
		Crane WASP	>9000	Ø6300x3000	Earth-based material, concrete mortar, geopolymer
Vorm vrij [99]	Netherlands	Lutum 4m Clay Printer	500-3000	250x250x400	Clay
		Lutum 4.6 Clay Printer	500-3000	430x460x500	
		Lutum 5M Clay Printer[a]	500-3000	250x250x400	
		Lutum 5 Clay Printer[a]	500-3000	400x460x500	
		Brutum[b]	-	800x600x800	

[a]: professional; [b]: industrial.

result in vastly different outcomes [78]. On the other hand, a systematic study to optimize this parameter has not yet been reported and related works have reported very varied printing speeds from 2.5 to 70 mm/s [77, 78, 83-86]. Table III shows the additive manufacturing systems available for DIW of ceramics, evidencing that most 3D printers of this technology are indicated just to traditional ceramics as clay and porcelain [87, 89-91, 95, 96, 99]. Lynxter is the only manufacturer to date to have a commercially available solution for advanced ceramics [92] and Rapidia is developing feedstocks of aluminum oxide and zirconium oxide [93]. Lastly, systems focused on building and structures have emerged. 3DPotter has a variety of 3D printers for cementitious materials [88], and WASP has systems able to deal with concrete mortar with a printing volume of Ø6.3x3 m^3 [97, 98].

Fused deposition

Fig. 5 illustrates the schematic of the fused deposition technology with two types of raw material: filament and granule. In both cases, the feedstock is composed of the ceramic powder (usually around 40-50 vol%) and multicomponent binders usually containing a thermoplastic (ethylene vinyl acetate [100-102], polylactic acid [103], polyvinyl butyral [22, 104], polyolefin [105, 106], low-density polyethylene [107]), surfactant (stearic acid [102, 104, 107, 108]), and plasticizer (polyethylene glycol [22, 103, 104]). This material is heated to just above the melting point of the polymer. So the material is selectively extruded through a nozzle, being deposited layer-by-layer according to a pre-defined path [109, 110]. Fig. 5a shows the fused filament fabrication operation, which uses a ram extruder, with the filament pushing the softened material out of the nozzle. Conversely, the granule-based 3D printers use a screw extruder for pushing the material to be deposited (Fig. 5b) [110].

In the fused deposition of ceramics, the nozzle diameter usually varies between 0.4 and 0.6 mm [22, 23, 101, 102, 105, 106, 108, 111-113] and the layer thickness from 100-300 µm [22-24, 103-105, 107, 108, 111-114]. Also, nozzle temperature and printing speed are important parameters and presented a great variation in related works reported in the literature [22-25, 100-108, 111-114] (130-260 °C and 1-95 mm/s, respectively). Such a difference is explained by the dependence of the operation range on the material and equipment used. This technology is capable of producing dense structural ceramic parts. However, the surface finish is limited and the staircase effect is an issue [1], as illustrated in Fig. 6. Also, there is a limitation on the maximum wall thickness that can be produced without the formation of cracks during the subsequent heat treatment [6]. Efforts to produce ceramics by fused deposition started in the late 1990s [8, 9]. On the other hand, equipment suppliers and available systems (Table IV) have raised in the last 3 years. 3D printers for filament and granules are available and the difference between these feedstocks is discussed next.

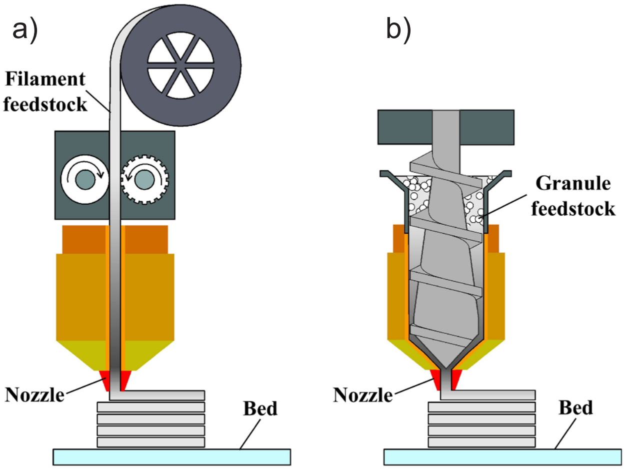

Figure 5: Schematics of the fused deposition technology [110]: a) fused filament fabrication; and b) fused granules.

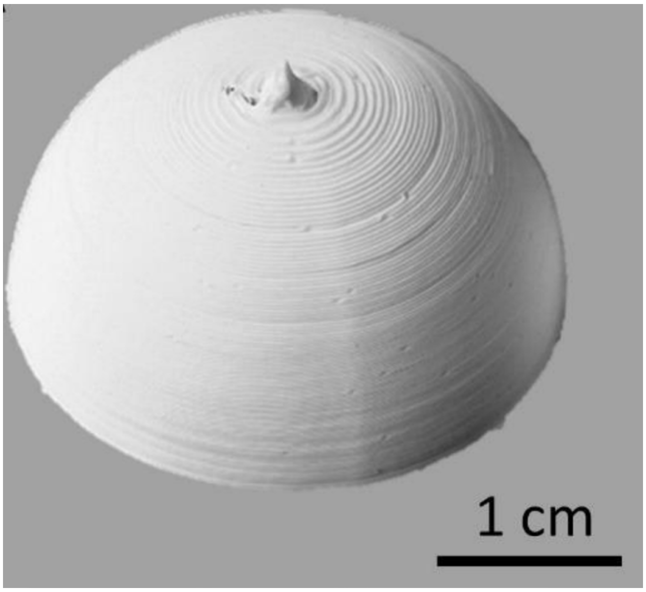

Figure 6: Alumina parts fabricated by fused deposition [115].

Fused filament fabrication: using filaments could benefit from the extensive use of FDM (fused deposition modeling) already applied to polymers, and recent studies [101, 102, 105, 106, 109, 111, 129] have indicated that common FDM 3D printers, which are low-cost and easy to operate [24], could deal with ceramic filaments. Oppositely, creating filaments with well-dispersed high ceramic loading is a big challenge [13], and they are commonly brittle and difficult to handle [23, 109, 110]. Moreover, nozzle blockages can occur due to particle agglomeration [109, 130]. Research about ceramic fused filament fabrication has increased in recent years. The research group from Empa (Switzerland) led by Dr. Clemens is the leading research group on this subject. They have shown that is possible to process different ceramic materials (alumina [101, 102], mullite [100, 131], barium titanate [132], and PZT [132]) in ordinary commercial printers. Fused filament fabrication proved to be capable to produce dense ceramic parts (~99%) [102, 107, 114] and the

Table IV - Fused deposition of ceramics: equipment suppliers.

Type of raw material	Manufacturer	Headquarter	System	Typical layer thickness (µm)	Maximum build size (mm^3)	Commercially available material
Granule	3D-Figo	Germany	FFD150H	300 [22]	150x150x120 [116]	Al_2O_3, ZrO_2, Si_3N_4 [117]
Granule	Pollen	France	Pam Series MC	200 [118]	Ø300x300 [119]	Al_2O_3, ZrO_2, Si_3N_4, B_4C [120]
Granule	Uprise 3D	China	UPS-240D	50-300 [121]	210x240x180 [121]	Al_2O_3, ZrO_2 [122, 123]
			UPS-250	50-300 [124]	250x250x250 [124]	
			UPS-556	50-1000 [125]	500x500x600 [125]	
Filament	Nanoe	France	Raise3D Pro2	150-200 [126]	305x305x300 [127]	Al_2O_3, ZrO_2 [126]
Filament	Xerion	Germany	Fusion Factory	N/S	245x230x200 [128]	Al_2O_3, ZrO_2 [128]

firing shrinkage usually ranges between 17% and 23% [101, 102, 104, 107, 111]. A surface roughness (Ra) of around 10 µm can be reached with the process [111, 112]. Conversely, the mechanical strength of ceramic parts produced by this technology has still been little explored. Xerion offers the Fusion Factory, an integrated additive manufacturing solution that includes 3D printing, debinding, and sintering in one system, focused on industrial production [128]. On the other hand, Nanoe provides a lower-cost 3D printer [127] and ceramic filaments (aluminum oxide and zirconium oxide [126]) compatible with common FDM 3D printers.

Fused granules: using granules favors the application of fused deposition of ceramics on a larger scale since granules feedstock is already available [22] due to the mature technology of ceramic injection molding [23]. Also, it can reach higher ceramic solid loading than filaments [22, 23, 110] and are easier to be prepared [23, 104]. 3D-Figo [117] and Pollen [119] offers 3D printers able to work with granules used for injection molding. Few studies about ceramic fused granules have been carried out so far [23-25]. Even so, dense zirconia parts (~99%) with flexural strength comparable to that made by conventional methods (890 MPa) have already been obtained [24]. In this work, the reported shrinkage was about 20%.

Material jetting

Fig. 7 presents the schematic of the material jetting technology. Small droplets of ceramic suspension (build-up material) and support material are dispensed by hundreds of nozzles contained in the printheads, creating the predefined cross-section [133, 134]. The build plate (substrate) may be heated to begin to evaporate the solvent of the printed ink [135]. In order to complete the solvent evaporation, the drying device moves over the printed layers [133]. Compared with the other ceramic additive manufacturing technologies, very little study has been done regarding parameter optimization. In this process, layer thickness has been used at around 10 µm [133, 135]. This technology is best suited for compact parts [1], having excellent resolution (~20 µm [6, 135]), as shown in Fig. 8. It stands out for the ability to deposit droplets of multiple materials simultaneously [6]. In contrast, material jetting has low productivity (~1 mm heigh per hour) [6]. Also, 3D printed parts by material jetting may present 'coffee stains' formed in the drying step, in which solid particles segregate from the center to the edge of the printed patterns [14, 136].

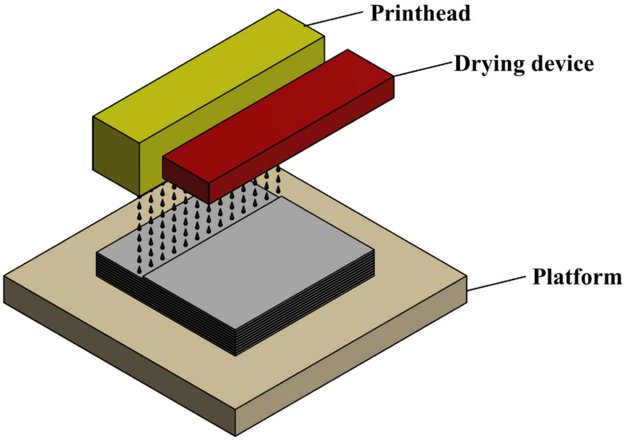

Figure 7: Schematic of the material jetting technology.

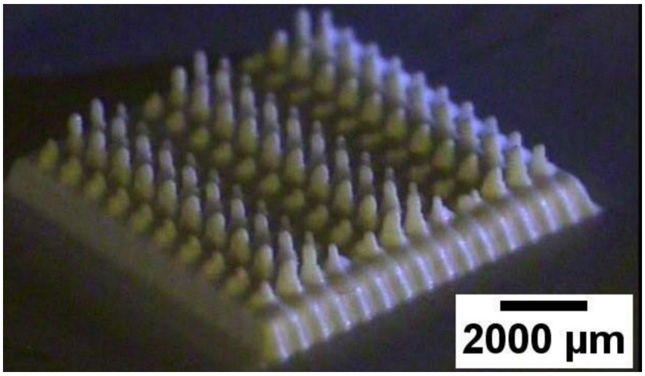

Figure 8: Alumina cooling element fabricated by material jetting [137].

Table V - Material jetting of ceramics: equipment suppliers.

Manufacturer	Headquarter	System	Indicated layer thickness (μm)	Maximum build size (mm^3)	Commercially available material
XJET3D [142, 143]	Israel	Carmel 11400C	N/S	500x140x200	Al_2O_3, ZrO_2
Tritone [144, 145]	Israel	Dominant	40-200	400x240x120	Al_2O_3

Ceramic material jetting has still been little explored. Around the early 2010s, a research group from RWTH Aachen University (Germany), led by Prof. Telle, investigated the development of ceramic inks and their application in the material jetting process [26, 134, 138-140]. However, few articles have been recently published on the topic. Usually, very thin ceramic (thickness smaller than 1 mm) bodies were fabricated [30, 133, 141], aiming to produce cathodes/electrolytes for solid oxide fuel cells [30, 141] or dielectric resonator antenna [135]. There is very little information about the properties of parts produced by ceramic material jetting. Willems et al. [133] reported having obtained zirconia parts by material jetting with a density of 99.7%, surface roughness (Ra) of about 7 μm, and flexural strength greater than 1000 MPa. This work also reported firing shrinkage between 16.8% and 17.6%.

For this process, the feedstock is a suspension with well-dispersed ceramic particles in the liquid solvent, with suitable stability, viscosity, and surface tensions [1]. Unlike the pastes used in DIW technology, the material jetting's feedstock is ink-based, ejecting a low-viscosity (~20 mPa.s) fluid [135] with surface tension not exceeding 60 mN/m [30]. To avoid clogging and blockage, the diameter of the nozzles should be 100 times bigger than the particle size [29, 30]. Consequently, nanoparticles are more desirable in formulating inks, since the nozzle diameters usually range between 20 and 30 μm [30, 134, 141]. On the other hand, smaller particles are more likely to agglomerate [1]. In addition to deionized water, some additives such as dispersants (triethanolamine [141], polyethylene glycol [141], polyacrylic acid [30]), glycerol [30, 134, 141], and ethanol [134, 141] are added to the suspension to adjust the ink properties and performance [30, 141]. Material jetting presents just two additive manufacturing systems for ceramics, as shown in Table V, and aluminum oxide and zirconium oxide are the only feedstock commercially available.

Selective laser sintering

Selective laser sintering (SLS) is a powder bed additive manufacturing technology in which the binder is melted by the scanning laser and binds to the ceramic powder. Fig. 9 illustrates the schematic of the SLS technology. A typical SLS 3D printer has two chambers: the powder feed supplier and the build chamber. The transport of recoating of the powder in the build chamber is performed by the powder recoater [146]. A laser beam is steered by an X-Y scanning mirror (usually a galvano scanner), locally heating and sintering the raw material creating layers according to predefined geometries [146, 147]. Although a variety of lasers may be used in SLS 3D printers, the CO_2 type is the most common [148]. This laser is suitable for processing oxide ceramics due to its high optical absorptivity in its wavelength (λ=10.6 μm) [149, 150]. SLS is best suited to porous applications that do not require high geometrical accuracy and surface roughness such as scaffolds for tissue engineering [1, 13, 14]. Fig. 10 shows an alumina part fabricated by SLS. The process commonly uses coarse ceramic powders (10-100 μm [17]) to guarantee flowability and the formation of uniform powder layers [6, 14]. The main process parameters to be considered are laser beam power, laser scan speed, scan spacing, and layer thickness [146-148] and an orthogonal method may be performed for optimization [151-153]. In recent work [151-160], the parameters have varied in the

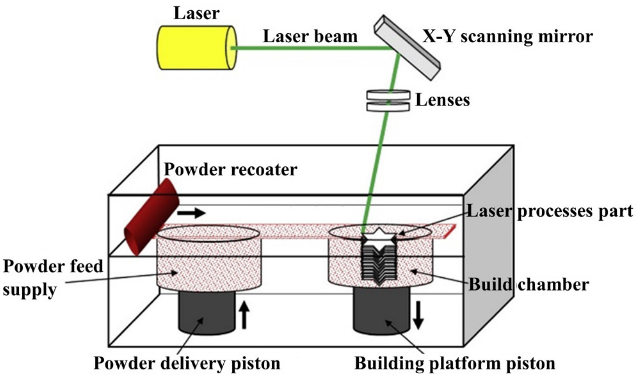

Figure 9: Schematic of the SLS technology [146].

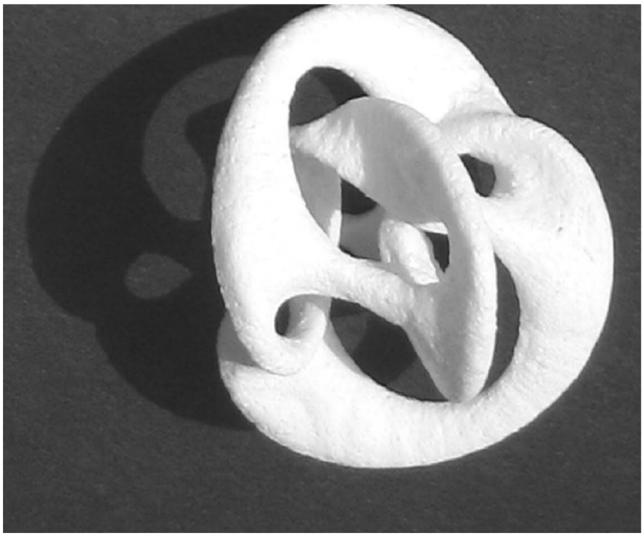

Figure 10: Alumina part fabricated by selective laser sintering [17].

Table VI - Selective laser sintering of ceramics: equipment supplier.

Manufacturer	Headquarter	System	Maximum build size (mm^3)
Wuhan Huake 3D Technol. [162]	China	HK C250	250x250x250 [158]

following range: laser power: 6-12 W; scanning speed: 1500-2500 mm/s; scan spacing: 50-150 μm; and layer thickness: 100-200 μm. Also, a pre-heating of the powder bed can reduce thermal stresses and thus help prevent crack formation in sintered parts [147]. Such parameter depends on the selected binder. For example, the most used in recent work has been the epoxy resin (E12), and a pre-heating of 45 °C has been reported [151, 158, 159]. On the other hand, polyamides may require a higher pre-heating temperature [156].

Most recent research on ceramic SLS has been done by the State Key Laboratory of Material Processing and Die & Mould Technology from Huazhong University of Science and Technology (Wuhan, China) [151-160]. Their research focuses on improving the ceramic parts produced by SLS by considering adding additives ($CaSiO_3$ [154], Al [157]), adjusting process parameters [151-153] (laser power, scanning speed, scanning space), sintering temperature [155, 156], and particle size distribution of the raw materials [158, 159], and by combining SLS with other processes such as vacuum infiltration [152]. Such works have shown the potential use of this technology to produce ceramic cores (to fabricate turbine engines and gas turbine hollow blades) and high porous ceramics (used for thermal insulators, filters, catalyst supports, separation membranes, etc.). The SLS technology can be used to fabricate parts with low firing shrinkage (<5% [155, 156, 158]) due to the low content of binders (which typically ranges from 7.5 to 15 wt% [152-158]). The sintered bodies typically have low relative density, with porosity usually greater than 50% [151, 154-157, 160], and may exceed 85% [155, 156]. Efforts have been made to increase mechanical strength. For example, the compressive strength of alumina parts increased almost 30 times with the addition of a sintering additive ($CaSiO_3$), reaching 8.39 MPa for a porosity of 68.16% [154]. Also, vacuum infiltration remarkably improved the flexural strength of silica-based parts, reaching 15.04 MPa, being compatible with the requirements for ceramic cores for manufacturing hollow blades [152]. Currently, there is a variety of SLS for metals and polymers. However, adapting these pieces of equipment for ceramic manufacturing requires considerable modifications and research [161]. There is just one commercially available SLS 3D printer for ceramics, as shown in Table VI.

Vat photopolymerization

Fig. 11 illustrates the schematic of the vat photopolymerization technology. In this additive manufacturing process, a liquid photosensitive raw material contained in a vat is selectively cured by light-activated polymerization [16]. The layers can be formed by scanning a laser (stereolithography, SLA) or by projecting the entire layer at once (digital light processing, DLP) [31, 163, 164]. This technology is best suited for small ceramic components for high-precision applications [6]. It can produce dense parts with desirable mechanical performance [1, 165, 166] and excellent resolution (~40 μm [167-170]) and surface finish [1, 6, 32, 33], as shown in Fig. 12. However, ceramic vat photopolymerization cannot produce monolithic large parts due to the high amount of organic material associated with the process and that would lead to cracks in the debinding of thick sections [6, 171]. Suppliers specify 10 mm as the maximum wall thickness to be produced [171, 172].

Vat photopolymerization has two main parameters: the layer thickness (usually varying between 25 and 100 μm) and the light exposure energy that determines the cure depth. This key factor should be higher than the layer thickness to ensure layer integration [20, 166, 175, 176]. Conversely, if cure depth is too high, the accuracy is reduced [166, 175]. Thus, the cure depth should be between 1.10 and 1.35 times the layer thickness [175] and the exposure energy must be defined empirically for each photosensitive feedstock to satisfy such condition. Moreover, the photosensitive slurry (feedstock) have several requirements concerning

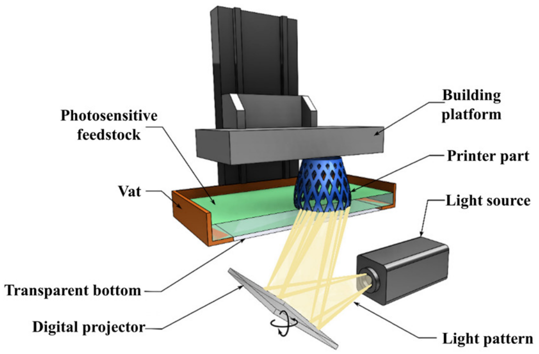

Figure 11: Schematic of the vat photopolymerization technology (adapted from [173]).

Figure 12: Zirconia part fabricated by vat photopolymerization [174].

Table VII - Potential applications and materials used in articles related to vat photopolymerization.

Potential application	Material used
Ceramic membrane	Al_2O_3 [200-203]
Bone repair/tissue engineering	Hydroxyapatite [204-216], TCP [217-228], Al_2O_3 [229], biosilicate [230], baghdadite [231-233], wollastonite-diopside [234], Mg-substituted wollastonite [235, 236], hydroxyapatite+TCP [237-242], hydroxyapatite+akermanite [243], calcium phosphate+bioglass [54, 244, 245], TCP+calcium pyrophosphate [246], calcium silicate+hydroxyapatite [247], Al_2O_3+ZrO_2 [248]
Catalytic application (automotive, hydrogen production, biocatalyst in continuous industrial process, etc.)	Al_2O_3 [249-253], ZrO_2 [254], zeolite [255], illite [256], TiO_2+MgO [257]
Ceramic core for investment casting	Al_2O_3 [258-262], SiO_2 [263, 264], zircon+silica [265, 266]
Piezoceramic (transducer, actuator, etc.)	$BaTiO_3$ [267-272], PZT [273, 274], alkaline niobate [275]
Dental restoration (crown, bridge, etc.)	ZrO_2 [276-287], Al_2O_3 [288-290], Al_2O_3+ZrO_2 [291], fluorapatite [292], $Li_2Si_2O_5$ [293], Li_2O+SiO_2 [294]
Solid oxide fuel cell	Y_2O_3-stabilized ZrO_2 [175, 181, 295], Sc_2O_3-stabilized ZrO_2 [296]
Heat exchanger/dissipation	Al_2O_3 [170], AlN [297-299]
Tool with optimized design (inner cooling structure)	Al_2O_3 [180], cemented carbide [300]
Optical application (lens, armored/sensor window, etc.)	Al_2O_3 [301], $MgAl_2O_4$ spinel [302], SiC [303], Nd-doped YAG [304], Yb-doped YAG [305]
Antenna	Al_2O_3+glass [306], $Ba_{1-x}Sr_xZn_2Si_2O_7$ [307], $MgTiO_3$+$CaTiO_3$ [308]
Nuclear fusion	Li_2TiO_3 [309], Li_2SiO_3 [310]

ceramic loading (>40 vol% [177, 178]), proper rheological behavior (<3 Pa.s [4, 177, 179]), stability, etc. Hence, a large number of studies have been carried out about the formulation of photosensitive ceramic suspensions for vat photopolymerization. Such slurry is composed of monomers, photoinitiators, ceramic powders, and additives such as dispersants, diluents, defoamers, plasticizers, and light absorbers. Due to the huge number of components already used, such a study is beyond the scope of this paper and can be found in a review dedicated to the topic [31]. Vat photopolymerization (VP) of ceramics can produce dense parts [4, 166, 175, 179-191] (>99%) with mechanical strength comparable to those of conventional methods. For example, some works have reported zirconia with flexural strength greater than 700 MPa [166, 182, 183, 189] and may exceed 1000 MPa [188, 192]. The reported shrinkage shows great variation (from less than 15% [193] to more than 30% [194, 195]), being usually around between 19% and 24% [3, 166, 182, 196, 197]. Such characteristic depends closely on the solid loading and sintering temperature [198]. Finally, this process presents outstanding surface quality and roughness (Ra) below 0.5 μm have been reported [4, 166, 188, 199]. Research about ceramic VP has become widespread in recent years and over 30 research institutions published at least 10 papers indexed by the Web of Science in the last 5 years. Ceramic VP may be applied in the most diverse areas and Table VII presents potential applications and ceramic materials used in recent articles. For more in-depth information about this technology, a few review articles are recommended [31, 311, 312].

Ceramic vat photopolymerization is the most mature technology with industrial readiness level [6] and several commercial machines [1] (Table VIII). Although most related companies emerged in the last 5 years, there are well-established companies with years in the market as Lithoz [334] and 3DCeram [313] with a variety of 3D printing systems and feedstock commercially available. Even systems capable of working with multi-materials have emerged in recent years [314, 319, 342]. Contrastingly, producing carbides and borides with this technology remains challenging due to the high refractive index (RI) of these materials, which would cause light scattering due to the RI mismatch with the usual photosensitive materials, leading to poor resolution and reducing the photopolymerization reaction [6, 13, 14]. Such materials are not yet commercially available. One alternative approach to deal with this issue is the use of polymer-derived ceramics [13], which are converted into ceramics without the addition of ceramic particles [31, 345-349]. It proved capable of manufacturing SiOC [346, 347, 350-352], SiC [350], SiCN [350, 353], and SiBCN [348] by ceramic vat photopolymerization. This AM technology is relatively expensive [5]. The raw materials are costly because they depend on high-priced photosensitive materials [15]. Efforts have been made to reduce feedstock

Table VIII - Vat photopolymerization of ceramics: equipment suppliers.

Manufacturer	Headquarter	System	Type	Indicated layer thickness (μm)	Maximum build size (mm^3)	Commercially available material
3DCeram [313-318]	France	C100 Easy Lab	SLA	10-125	100x100x150	Al_2O_3, ZrO_2, SiO_2, Al_2O_3-toughened ZrO_2, cordierite, TCP, hydroxyapatite, AlN, Si_3N_4
		C100 Easy Lab		10-125	100x100x150	
		C900 Flex		10-125	300x300x100	
		C900 Hybrid		10-125	300x300x100	
		C3600 Ultimate		25-125	600x600x300	
10dim Tech [319-325]	China	Autocera-R	DLP	25-100	96x54x100	Al_2O_3, SiO_2, β-TCP, art ceramic
		Autocera-U	DLP	50-100	136x76x200	
		Autocera-L	DLP	10-500	136x76x200	
		Autocera-X	SLA	50-100	350x350x500	
		Autocera-Multi	DLP	25-100	96x54x100	
ADMATEC [172, 326, 327]	Netherlands	Admaflex 130	DLP	10-200	96x54x100 to 160x100x400	Al_2O_3, ZrO_2, SiO_2, hydroxyapatite
		Admaflex 300		10-200	260x220x500	
AON [328, 329]	South Korea	ZIPRO Dental	DLP	N/S	77x43x75	ZrO_2
		ZIPRO Industrial		25-100	107x60x150	
Carima [330, 331]	South Korea	IMC	DLP	25-100	110x61x130	Porcelain, material for investment casting
Fortify [332, 333]	USA	Flux Core	DLP	25-250	204x114x330	Al_2O_3
Lithoz [334, 335]	Austria	CeraFab LAB L30	DLP	25-100	76x43x170	Al_2O_3, ZrO_2, SiO_2, β-TCP, hydroxyapatite, AlN, Si_3N_4
		CeraFab System S25		10-100	64x40x320	
		CeraFab System S65		10-100	102x64x320	
		CeraFab System S230		25-100	192x120x320	
		CeraFab Multi 2M30		10-100	76x43x170	
Photocentric [336]	UK	Ceramet 1	DLP	N/E	310x83x95	Al_2O_3 (in development), ZrO_2, SiO_2, hydroxyapatite, SiC
Prodways [337-339]	France	ProMaker V6000	Moving-light DLP	25-150	120x150x150	Al_2O_3, ZrO_2, TCP, hydroxyapatite
					120x350x150	
					120x500x150	
SK Fine [340, 341]	Japan	SZ-1000	SLA	5-100	300x300x50	Al_2O_3
		SZ-1100		10-200	100x100x50	
		SZ-2500		50-200	250x250x250	
Soonsolid [342, 343]	China	CeraRay TC-1	DLP	10-100	64x40x200	Al_2O_3, ZrO_2, SiO_2, hydroxyapatite, clay
		CeraRay CR-2		10-100	64x40x200	
Tethon 3D [332, 344]	USA	Bison 1000	DLP	25-100	110x60x138	Al_2O_3, cordierite, hydroxyapatite, porcelain, mullite

waste. Thus, machines that work with a small amount of material (60 mL) [316] and studies about recycling raw materials [286, 354] have emerged. Also, the ceramic vat photopolymerization system made by the main suppliers (3DCeram, Admatec, Lithoz) surpasses US$100,000. These systems have a dedicated recoating system [4, 355, 356] which allows the spreading of constant and homogeneous micrometric layers even for high viscosity feedstock as the high ceramic loading photosensitive suspensions [357]. On the other hand, research using home-built prototypes [174, 189, 357, 358] and low-cost 3D printers [198, 359, 360], commonly used in the manufacturing of polymers, have emerged and may be a suitable solution for the use of the technique in laboratories and small businesses.

CHALLENGES AND TRENDS

Ceramic parts produced by additive manufacturing still have to improve performance (resolution, surface quality, mechanical properties), reduce the associated costs (equipment and feedstock), and increase productivity to become more competitive with the traditional fabrication technologies [6, 14]. Although ceramic AM enables the fabrication of customized complex parts with reduced lead times [3-6], 3D printing of large and dense ceramic pieces is still not feasible. In addition to the low density associated with most ceramic additive manufacturing technologies, parts produced by these processes may have defects that impair mechanical properties, and process control is a critical factor to be considered [1, 14]. The feedstock design is another key point for improvement, and it requires multidisciplinary efforts. The raw materials usually include a variety of materials and they should fit the process requirements that can range from the most common as flowability/rheological behavior, to specific for each technology as photosensitive parameters to vat photopolymerization.

Powder bed processes (binder jetting and selective laser sintering) usually provide low-density parts due to the coarse ceramic particles used to provide the proper flowability of the feedstock. Research about combining different particle sizes [361] and using granulation of nanoparticles, with high sinterability, into micron-sized granules [37] might alleviate the problem. Vat photopolymerization and fused deposition have the potential to produce dense ceramic parts, but their wall thickness is limited [6] due to the high amount of organic associated with the feedstock that is necessary to maintain proper rheological behavior [31], and improvements in post-processing can be key to this issue.

The post-processing of ceramic additive manufacturing has been the subject of study such as adding additional post-processing steps (vacuum freeze drying [278], different types of infiltration [278, 362, 363], isostatic pressing [1, 364] and sanding [169]), or optimizing the required step: drying [180, 365], debinding (heating rate [366], special atmosphere [188, 191, 272, 366], using vacuum [367], multiple-steps [180, 190, 195, 365, 368, 369]), and sintering [198, 274, 369, 370]. Finally, ceramic additive manufacturing is expected to follow the general 3D printing concern with sustainability [371] and expand studies on its environmental implications (considering materials savings in the life cycle and energy consumption [372]). There are already related studies about the reusability/recycling of feedstock [354], the use of recycled glass as raw materials [373, 374], and steel dust waste as an additive in ceramic 3D printing [375]. Also, care must be taken about the environmentally hazardous materials involved in 3D printing and post-processing [15].

CONCLUSIONS

The performance of the ceramic parts made by additive manufacturing is still the main issue to its widespread in industry. Most ceramic additive manufacturing technologies are not able suited to produce dense parts. On the other hand, vat photopolymerization, which is the most well-established ceramic 3D printing process and can produce structural parts, has limited wall thickness due to the high amount of organic materials associated with this process that has to be eliminated during the debinding. Moreover, costs and availability of equipment and feedstock are still a matter for ceramic additive manufacturing. Several companies have launched their ceramic 3D printing solutions in the last 5 years. On the other hand, much research on equipment, material, 3D printing, and post-processing has yet to be carried out to allow ceramic additive manufacturing to have an extensive industrial application.

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