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REVIEW ARTICLE

Development of ceramic additive manufacturing: process and materials technology

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Abstract
Ceramic additive manufacturing (C-AM) is highlighted as a technology that can overcome the inherent limitations of ceramics such as processability and formability. This process creates a structure by slicing a 3D model and stacking ceramic materials layer-by-layer without mold or machining. C-AM is a technology suitable for the era of multiple low-volume because it is more flexible than conventional methods for shape complexity and design modification. However, many barriers to practical use remain due to process speed, defects, and lack of knowledge. This review focuses on studies to overcome the limitations of C-AM in terms of process and materials. The C-AM process has been advanced through various studies such as model/equation-based parameter control and high-speed sintering using external energy. Besides, by improving and fusing existing technologies, high-precision high-speed printing technology has been improved. A variety of material studies have been made of manufacturing ceramic structures with superior properties using preceramic polymers and composite materials. Through these studies, C-AM has been applied to various fields such as medicine, energy, environment, machinery, and architecture. These continued growths and diverse results demonstrate the importance and potential of C-AM based ceramic manufacturing technology.

Keywords Ceramic additive manufacturing · 3D printing · Printing process · Printing materials

1 Introduction

Ceramic additive manufacturing(C-AM) stacking materials layer by layer (bottom-up approach) is fundamentally different from subtractive manufacturing in conventional methods [1]. This process consists of four steps: model design and slicing, printing, post-treatment, and heat treatment. (1) The desired design for C-AM is transformed into data that is divided into several thin layers through a slicing process. (2) The transformed data is transmitted to a 3D printer, and a three-dimensional green body is created through the process of stacking materials layer by layer. AM process time varies greatly depending on the setting values such as the thickness of the layer and the lifting speed. The green body is made of the desired final material through a (3) post-treatment such as washing and removal of the support, (4) followed by a heat treatment process of debinding and sintering. C-AM is advantageous for the production of personalized products over conventional techniques.

As shown in Fig. 1, AM materials can be classified into three types of liquid-based, solid-based, and powder-based systems according to the supplied state [2]. The liquid-based systems include photo-polymerization, material jetting, extrusion method. Photo-polymerization is a method of forming an object through the polymerization of a photocurable material through the irradiation of light [3]. Material jetting (MJ) is a method of making an object by spraying a liquid material and curing it with ultraviolet light. The extrusion method is a method of continuously pushing a heated solid material or ink to create a three-dimensional object, and direct ink writing (DIW) belongs to the liquid-based system among them [4]. Photopolymerization technology is the mainstream in liquid-based systems and 3D Ceram and Lithoz are leading this field worldwide. The solid-based systems include the fused deposition modeling(FDM) method pushing the heated solid material [5], and the sheet lamination (SL) method that forms the object by processing-adhesive-laminating [6]. The field has remained focused on SME-oriented development such as Unfold (FDM),

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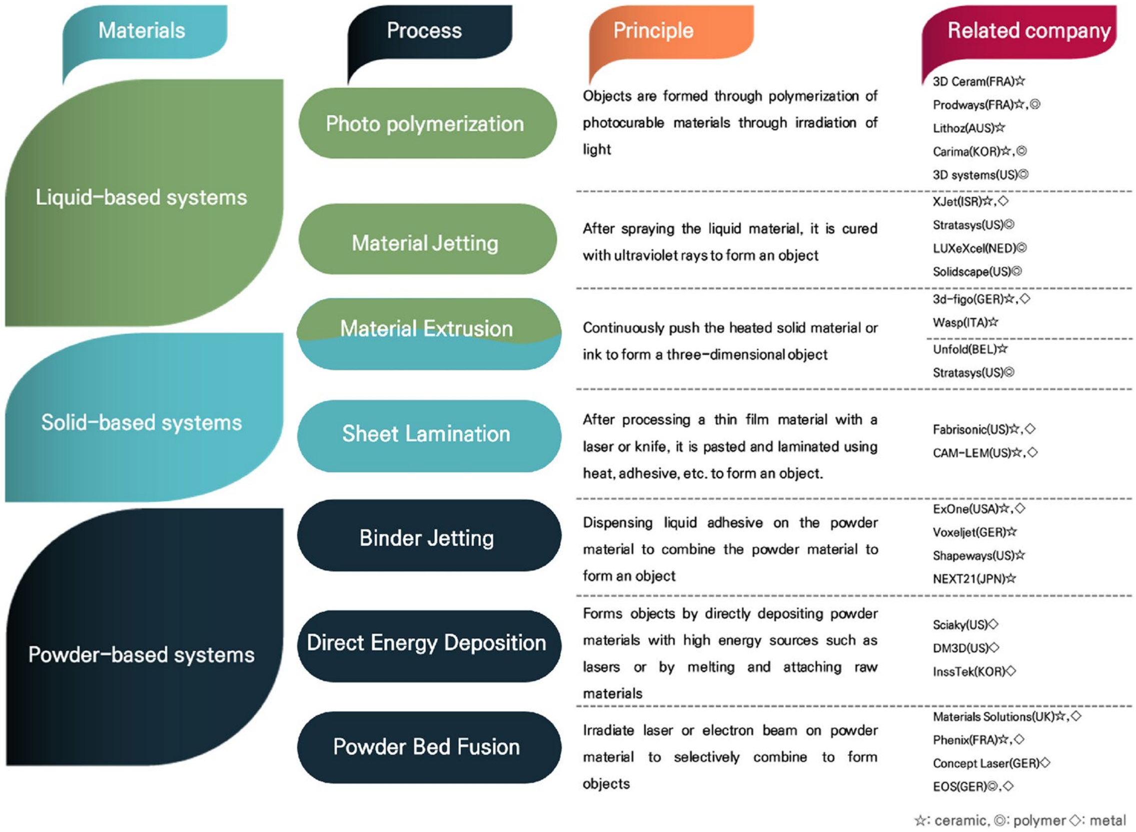

Fig. 1 Classification of additive manufacturing by material and method

Fabrisonic, and CAM-LEM (SL). Finally, powder-based systems include binder jetting (BJ), direct energy deposition (DED), and powder bed fusion (PBF). Binder jetting is a method of dispensing a liquid adhesive to a powder material to bond the powder material to form an object [7]. Direct energy deposition (DED) is a method of forming an object by directly depositing a powder material on a high energy source such as a laser or by dissolving and attaching a raw material [8]. Powder bed fusion is a method of forming an object by selectively combining a powder material with a laser or an electron beam [9]. The development of binder jetting technology is dominant in powder-systems, and leading companies in binder jetting printers such as ExOne and Voxeljet are entering the ceramic field.

C-AM is a key process for producing more complex ceramics by solving the inherent limitations of ceramics such as machinability and formability [10]. However, despite these expectations, there are many challenges to be solved for C-AM to become key technologies leading the fourth industrial revolution. First, there are limitations in slow production speed and product size. It takes a lot of time to stack numerous layers, and the size of the product is usually limited by the size of the 3D printer. Second, defects causing unstable bonding between lamination surfaces, non-uniformity of materials are great problems in printing quality. In particular, even very small defects can grow during the heat treatment process after printing, greatly affecting the overall properties of the molding. Therefore, it is urgent to develop a technology capable of more effectively controlling defects. Finally, C-AM materials are very limited, and sporadic technology development is progressing in case-by-case depending on the printer.

This review focuses on studies to overcome the limitations of C-AM in terms of process and materials. In the C-AM process, model/formula-based research, high-speed sintering research, and technology fusion research have been conducted. Also, research on precursor ceramics and composite materials and various applications using these technologies were covered.

2 Process development of ceramic additive manufacturing

2.1 Printing process

The early try-and-error C-AM study was time-consuming and limited to the application of case-by-case. Therefore, the development of C-AM was very slow and there were problems to be solved. Recently, various fundamental studies on modeling and formula-based printing process control have been reported to solve these problems. This method has had the effect of preventing failure and improving process efficiency in advance.

The Michel Bellet group reported a study on three-dimensional finite element thermomechanical modeling according to process parameters such as laser distance, speed, and layer thickness in the alumina selective laser melting (SLM) method [11]. Due to the rapid heating and cooling by the laser applied to the ceramic in SLM, it is difficult to optimize the printing process parameters through experimental studies. As shown in Fig. 2, the author studied the thermodynamic phenomena in a single/continuous pass of ceramic through FEM. The effects of liquid viscosity and surface tension were also investigated, and the effects of process parameters such as temperature distribution, bead morphology, and melt pool profile were described. Through this result, it is possible to make high-quality ceramic products by suppressing thermal stress in SLM.

Bae et al. [12, 13] reported a study on the correlation between design-material-process variables by designing a computational model to solve the segregation problem of the ceramic powder generated during the stereolithography (SLA) process (Fig. 3). The powder used as a ceramic mold (> 50 μm) prevents the dimensional change of the mold during casting. However, the coarse particles settle rapidly during the SLA process causing separation of particles in the layer. To prevent such layer separation, the printing speed in the layer should be faster than the segregation speed of the ceramic particles. Based on this, the authors derived separation parameters from SLA and presented segregation regions according to parameters such as separation parameters, layer thickness, surface area, laser power, and particle size(Fig. 3a, b). Also, the segregation prevention region through variable control was derived, and the experiment proved the applicability of the computational model to effectively suppress the layer separation defects caused by segregation (Fig. 3c, d) [14]. The authors had successfully manufactured defect-free ICCM (Integrated Cored Ceramic Mold) with a complex structure for superalloy turbine airfoil using this model [15].

2.2 Sintering process

The sintering process, which is essential for the production of ceramic products, acts as a bottle-neck for the production

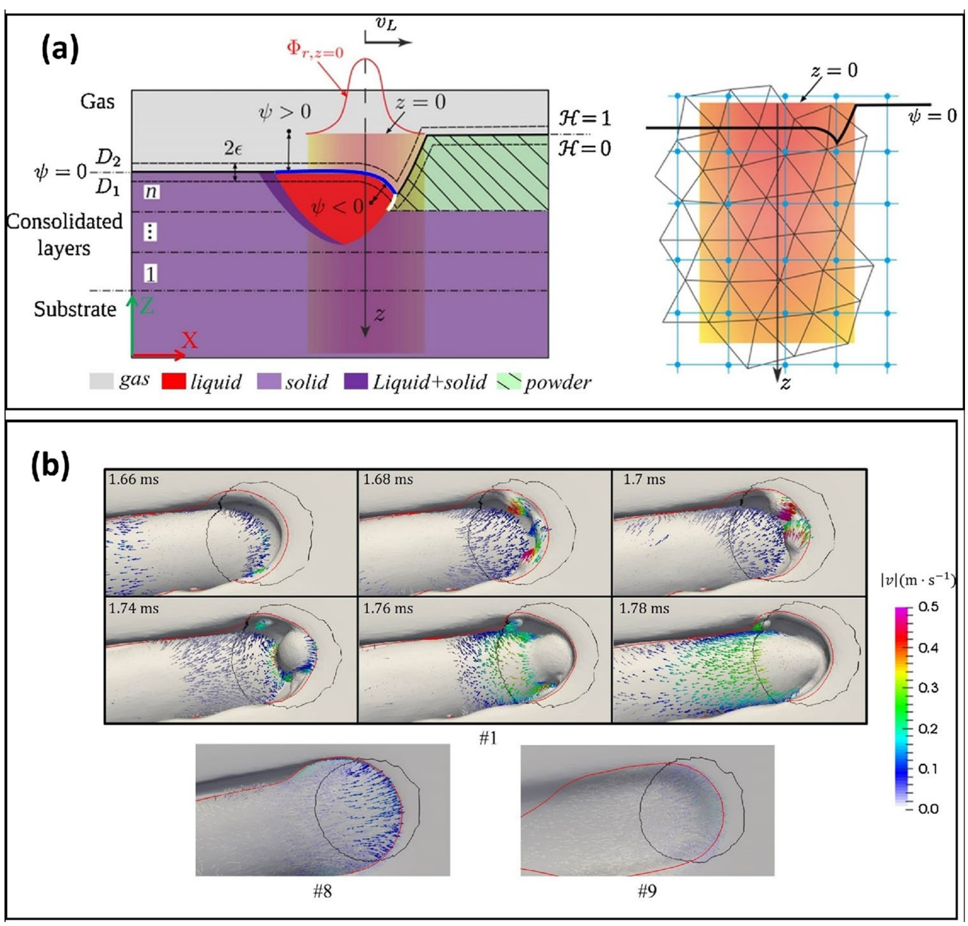

Fig. 2 Schematic diagram of **a** C-AM process modeling and analysis method for heat source, **b** melt pool analysis according to surface tension

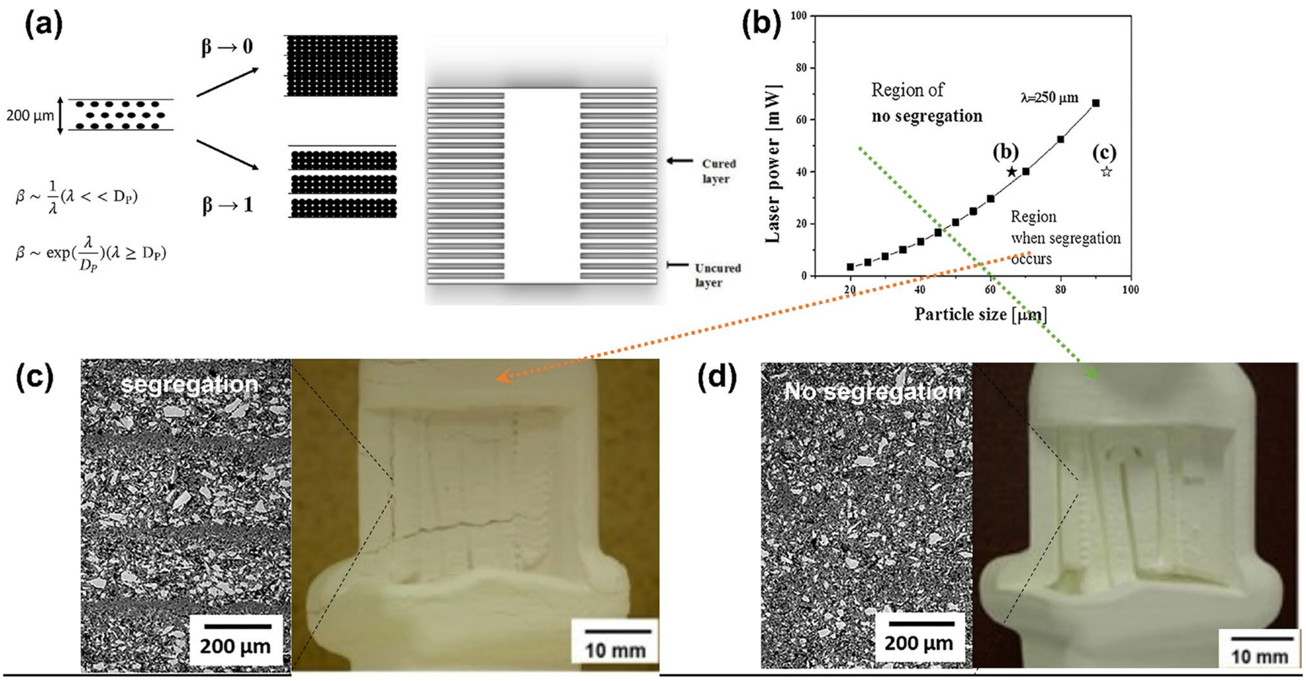

Fig. 3 A study on the control of layer separation defects by establishing and applying a layer separation phenomenon calculation model **a** segregation state with segregation parameter and Comb-shaped object to detect segregation in a layer. **b** Effect of particle size distribution on intralayer separation. SEM and digital images of 3d printed integrally cored ceramic mold (ICCM) in **c** no segregation region and **d** segregation region

of ceramic products due to the long process time. As one of the methods to reduce the sintering time, studies on applying other external energy such as microwave sintering have been reported. Microwave sintering is economical and energy-efficient because it sinters the ceramic at a very fast rate at low temperatures. However, since the sintering speed is too fast, there are problems in uniformity and heating rate control. Manière et al. [16] tried to understand microwave conditions through multiphysics simulation. The authors investigated the electric field distribution in heating and densification through electromagnetic-thermal–mechanical (EMTM) simulation (Fig. 4a). The EMTM model was obtained based on modeling of the pressure-less sintering, microwave heating, and boundary conditions. As shown in Fig. 4b, the 35 mm ZrO_2 ceramic gear was experimentally sintered and compared with the simulation. In the simulation, the temperature difference in the range of 1000-1080 °C was 40-50 K, which is believed to be the cause of the experimentally obtained distortion. Through this, the author produced a ZrO_2 sintered body with limited particle growth in the low temperature (< 1080 °C) region. Curto et al. [17] also applied microwave sintering for the rapid sintering of alumina ceramics. The sample printed by mixing coarse particle 60 wt% and fine particle 40 wt% showed a more uniform and smaller microstructure than convention sintering. This grain growth limitation is believed to be due to the fast heating rate of alumina (250 °C min). Microwave sintering is 75% faster than traditional methods and 30% faster when considering the entire process. Such microwave sintering could be used as one of the ways to dramatically increase the production speed of ceramic 3D printing products.

2.3 Printing equipment

The development of equipment for ceramic 3D printers has been actively conducted by companies. As shown in Fig. 5 , XJet introduced NanoParticle Jetting (NPJ) technology that forms a three-dimensional object through rapid evaporation of the dispersion through heating (250 °C) and bonding between nanoparticles after spraying a suspension containing nanoparticles [18]. This technology increases the degree of freedom of design by filling all the spaces by spraying the build material and the support material together, and manufactured high-precision ceramic parts through the introduction of nanoparticles.

In the development of equipment for ceramic 3D printers, a new trend has recently been attempted to fuse different types of printing technology. As shown in Fig. 6, EnvisionTec has developed 3SP (Scan, Spin, and Selectively Photocure) technology that combines the advantages of the existing DLP and SLA methods [19]. 3SP is a technology that irradiates the laser in 2D form using a field-programmable gate array (FPGA) control board that can turn the laser on and off very quickly in the Y scan direction. As shown in

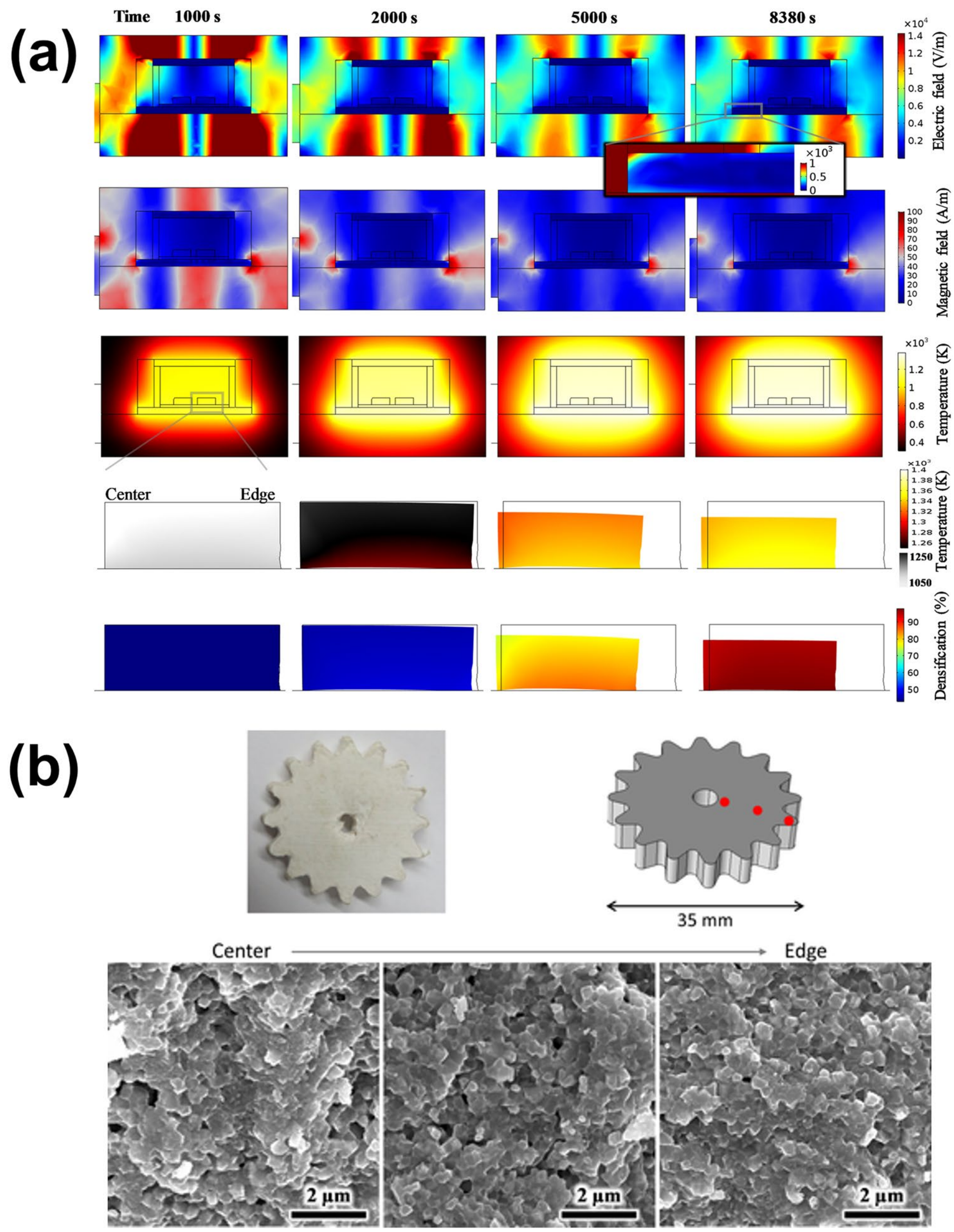

Fig. 4 **a** Electromagnetic-thermal–mechanical (EMTM) simulation model and **b** sintered ZrO_2 ceramics

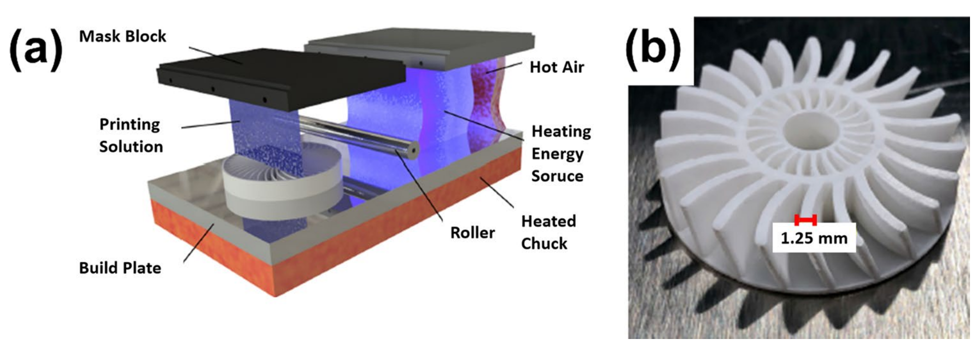

Fig. 5 **a** Schematic diagram of nanoparticle jetting process and **b** printed zirconia part

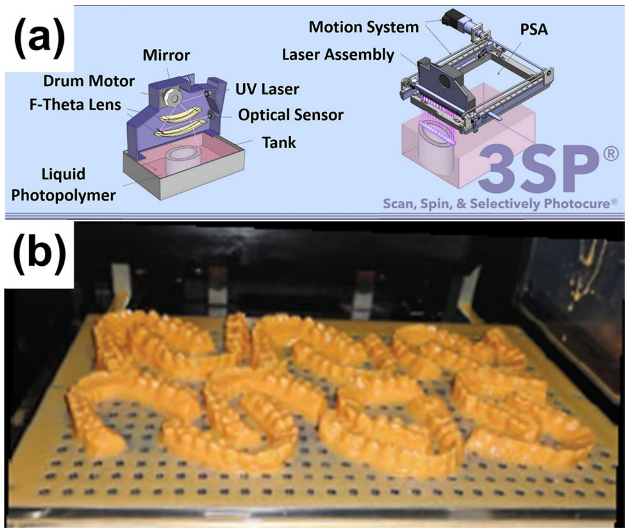

Fig. 6 **a** Schematic diagram of 3SP (Spcan, spin, selectively photocure) process, and **b** printed dental impression

Fig. 7, HP has developed a Multi Jet Fusion (MJF) printer through a fusion technology using powder bed fusion technology and binder jetting technology [20]. The technology is a method of irradiating energy after spraying different types of ink with different functionalities. After spraying a fusing agent on the area to be selectively combined, and a detailing agent around the contour, more precise printing quality can be obtained through energy irradiation.

3 Material development of ceramic additive manufacturing

In C-AM products, material selection is significant because material properties affect not only the printing process but also the heat-treatment process. More specialized materials technology is required for C-AM materials development because the heat treatment process such as binder-burn out and sintering can causes defects and distortions of the structure affecting the reliability of the final product. Due to these limitations, the studies are still in the basic stages, and there are many tasks to be solved compared to other materials. Recently, as the limitations of material have been solved due to the rapid development of equipment/process, interest in materials development has increased. The development has mainly been conducted in studies that control the particle size and surface properties of ceramic materials and studies that identify and control the interaction between binders and dispersants.

3.1 Preceramic polymers

Researches on material conversion technology using ceramic precursors and the application of composite materials are actively underway. The preceramic polymer can efficiently create a complex structure without problems such as inhomogeneity and sedimentation compared to the conventional powder procedure and is transformed into a desired ceramic product by heat treatment.

As shown in Fig. 8, Eckel et al. [21] reported that (mercaptopropyl) methyl siloxane and vinyl methoxy siloxane were used to print the desired complex shape in the form of a pre-ceramic polymer. Printing using a preceramic polymer is made into a ceramic product through the following three steps: (1) mixing of photoinitiator and curable ceramic monomer (2) formation of a preceramic polymer through photopolymerization printing (3) conversion to ceramic through pyrolysis. The SiOC structure was produced by thermal decomposition at 1000 °C. In argon after printing. After heat treatment, 42% of mass loss and 30% of shrinkage occurred, and there was no special structural distortion. The printed SiOC had a heat resistance of 1700 °C and 10 times higher strength than the commercial ceramic foam.

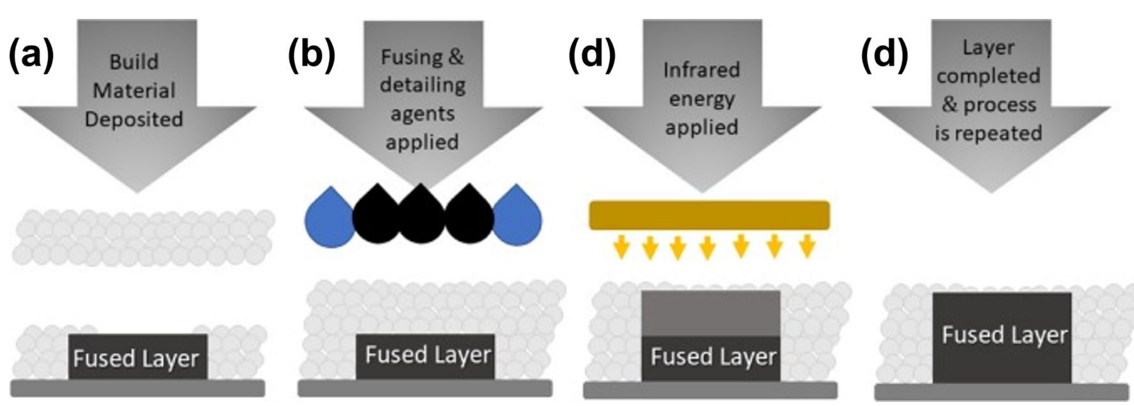

Fig. 7 Schematic diagram of Multi Jet Fusion. **a** Build material deposited, **b** fusing and detailing agent applied, **c** infrared energy applied, **d** layer completed and process is repeated

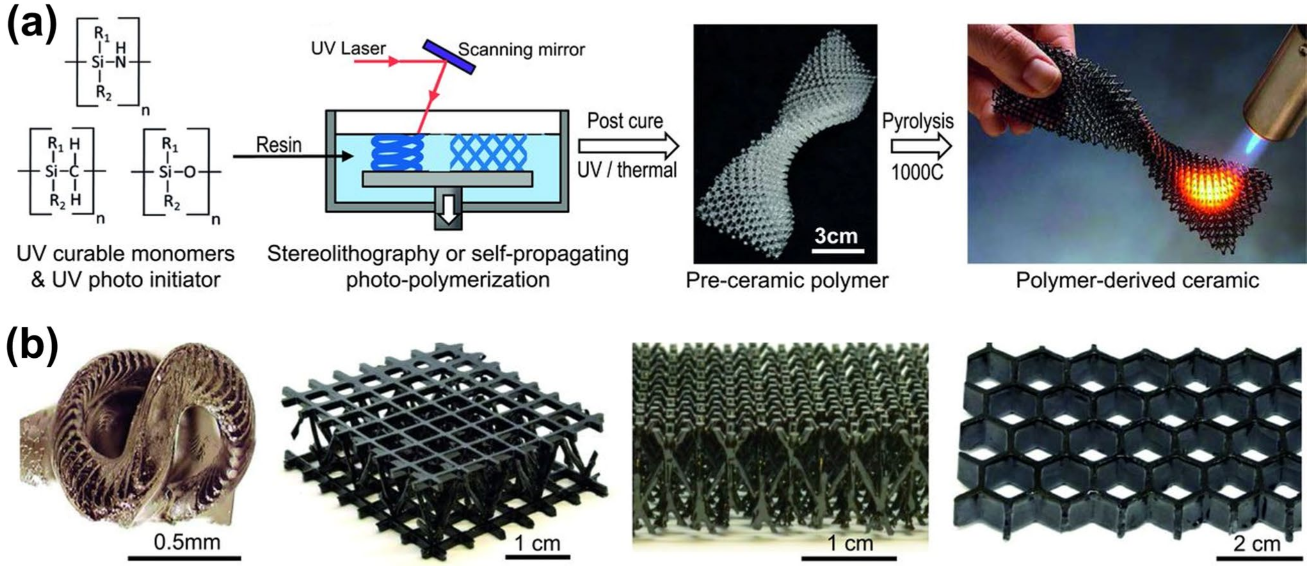

Fig. 8 **a** Printing process of SiOC material using preceramic polymer and **b** printed ceramic structures

The printing method using the preceramic polymer is particularly effective for ceramic products such as SiC, Si_3N_4, and SiOC, which are difficult to sinter. With these results, this group has signed a research contract with DAPPA on ceramic products that withstand heat when the spacecraft enters the atmosphere.

However, to commercialize the material conversion technology using a precursor, it is necessary to solve problems such as defects due to volume change during ceramic conversion. Raj et al. [22] conducted a study on the fracture that occurs when a preceramic polymer is converted to a ceramic phase. Pre-ceramics generate microcracks due to shrinkage during sintering depending on the heating rate and the thickness of the specimen. As shown in Fig. 9, the authors presented a processing map to suppress these microcracks and investigated the crack initiation temperature (750 °C) due to gas evolution. Above 750 °C, as the concentration of =Si–CH_2 · radicals increases, preceramic is converted to ceramics with the release of hydrogen. To suppress micro-defects promoted by a hydrogen concentration gradient, the authors proposed a pyrolysis process in a high-pressure hydrogen/methane gas environment.

3.2 Composite materials

Research on the application of composite materials to modify customized microstructures and material properties for advanced ceramic technologies has also been actively reported. Muth et al. [23] prepared a ceramic ink of particle-stabilized foams surrounded by colloidal particles to produce

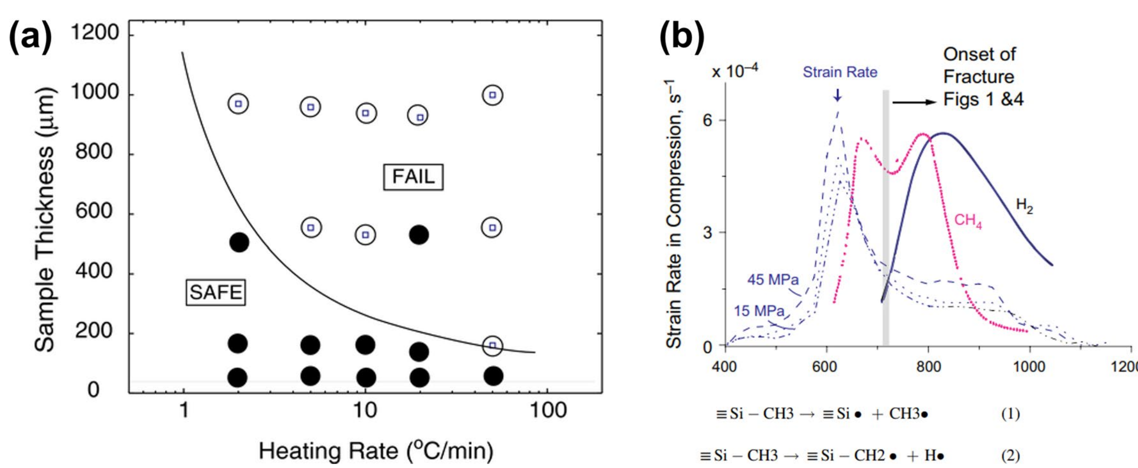

Fig. 9 **a** Processing map to suppress these microcracks and **b** one set fracture temperature correlated with H_2 evolution

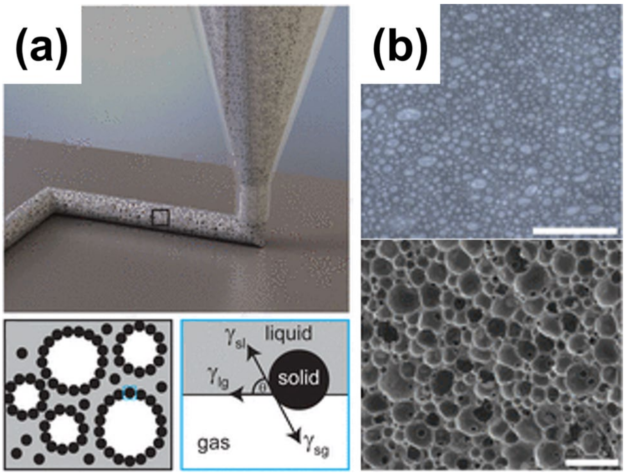

Fig. 10 **a** Schematic diagram of direct foam writing of porous filamentary features and **b** microstruetures of the printed part

a hierarchical cellular structure (Fig. 10). The bubble in the ink was stabilized by controlling the contact angle of the colloidal particles. After sintering, the porous structure consisting of tens-nm sized pores is obtained with a porosity of 85%. The authors made a ceramic honeycomb structure with greater specific strength (> 107 Pa/kg/m^3) than the conventional AM method through microstructure and shape control. This cellular ceramic manufacturing technology is scalable to applications such as lightweight construction and insulation.

As shown in Fig. 11, Roh et al. [24] proposed a PDMS/water capillary ink method as a complex shape production method using poly dimethyl siloxane (PDMS), which can be used in biomedicine and soft robotics. Mixing of microbeads and liquid PDMS improves the printability of PDMS by controlling the rheological properties of the ink. When the microbead and liquid PDMS are mixed in water, the liquid PDMS coats the microbead. This liquid shell gels the ink by forming an interparticle bridge by capillary effect. The gelled ink is printed through a nozzle and heat cured at 85 °C for 2 h to form a PDMS complex structure. The ink can be cured in air and water, and the cured structure shows superior elasticity and elasticity. Due to the biocompatibility of PDMS, it can be used to produce 3D printed structures such as bioscaffolds.

4 Applications

C-AM technology that can overcome the inherent difficulty of ceramics has been applied to various fields including industrial machinery, aerospace, electronics, and biomedical [25]. Especially, the biomedical field is the most actively applied to solve problems of metal and polymer materials such as toxicity and biocompatibility. As a typical example of medical C-AM, personal customization medical product is dominant such as dental implants, artificial bone, and orthopedic products. Low-temperature manufacturing is particularly important for biomaterials containing bioactive molecules or drugs. As shown in Fig. 12a, Lin

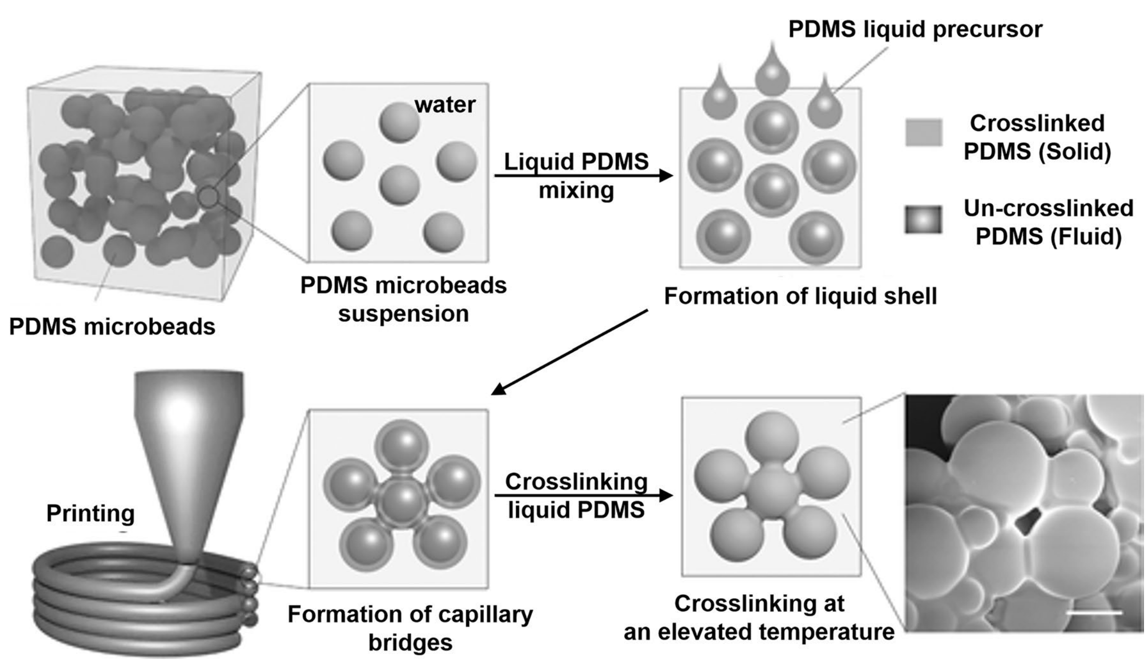

Fig. 11 Schematic diagram of multiphase silicone/water capillary inks process

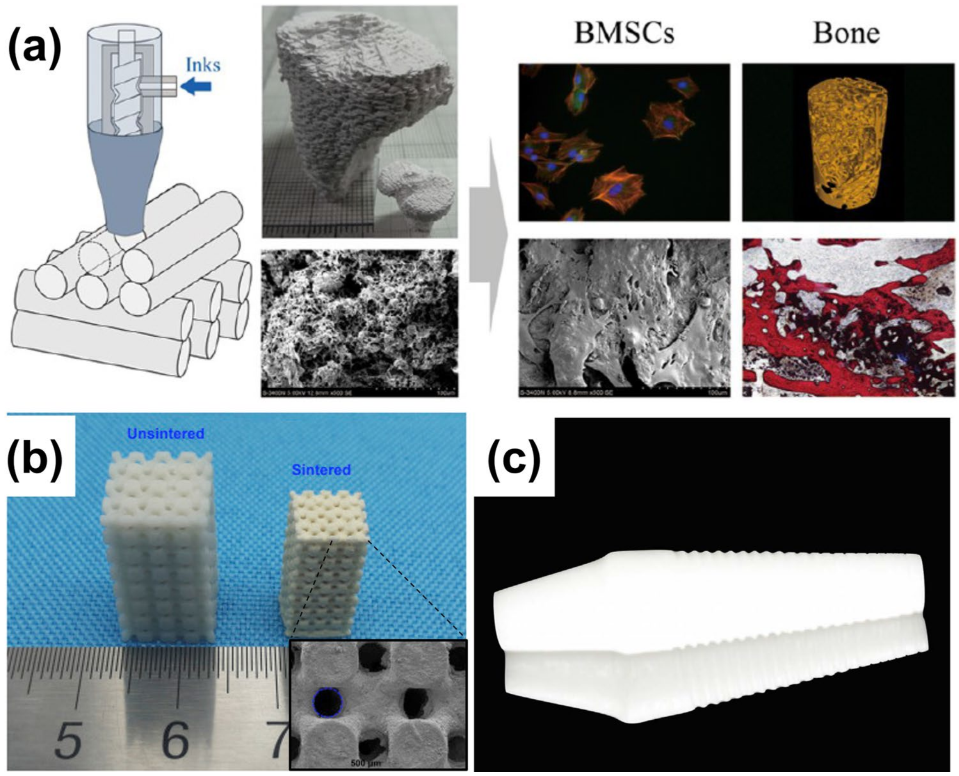

Fig. 12 Applications of ceramic additive manufacturing in the biomedical field. **a** HA/collagen scaffold printed by DIW method at low-temperature, **b** HA bone scaffold printed and **c** zirconia dental implant printed by DLP method

et al. [26] proposed the low-temperature DIW method of hydroxyapatite(HA)/collagen scaffolds for bone recovery. To print the HA/collagen scaffold at low temperature (4 °C), they controlled rheology and wettability of the materials with the acetic acid solution. After printing the structure, the solvent was completely removed using the freeze-drying method to increase biocompatibility. The fabricated bone scaffold was shown to promote new bone growth in the histological analysis of new bone formation. This technique has shown the possibility of 3D printing of bone scaffold with different drugs or bioactive materials. Liu et al. [27] also fabricated HA bone scaffolds using the DLP method with improved ceramic slurry and process (Fig. 12b). Printed HA scaffolds showed a flexural strength of 41.3 MPa and biocompatibility for orthopedic application. As shown in Fig. 12c, Osman et al. [28] effectively printed the implant structure using ZrO_2 using C-AM. The ZrO_2 implant printed using the DLP method exhibited a roughness of 1.59 µm and flexural strength of 943 MPa. The mechanical strength was similar to that of the traditional ceramic manufacturing method (800–1000 MPa) and showed high dimensional accuracy. However, technology improvement such as two-step sintering or low-speed sintering was required to suppress the occurrence of microcracks due to the sintering process.

C-AM is also actively applied to energy/environment ceramics. Particularly, in the energy field, research is being conducted to design a complex electrode structure capable of fastly moving lithium ions to overcome a correlation between high capacity and rapid charging. As shown in Fig. 13a, Park et al. [29] developed a high energy densified battery ink that can be applied to AM and suggested the possibility of manufacturing electrodes in various shapes. Also, Sun et al. [30] developed microbatteries that can be used in various fields such as medical and sensor using AM (Fig. 13b). C-AM is also widely used in the field of industrial machinery requiring mechanical properties. As shown in Fig. 13c, Costakis et al. [31] used B_4C materials with direct ink writing to print complex shapes that were difficult in the conventional methods. This complex B_4C structure is expected to be applicable in defense and numerous industries such as bulletproof vests, tanks, engines. Also, C-AM can create a miniature before building it and use it for building plans or output the building directly to an extra-large 3D printer (Fig. 13d) [32].

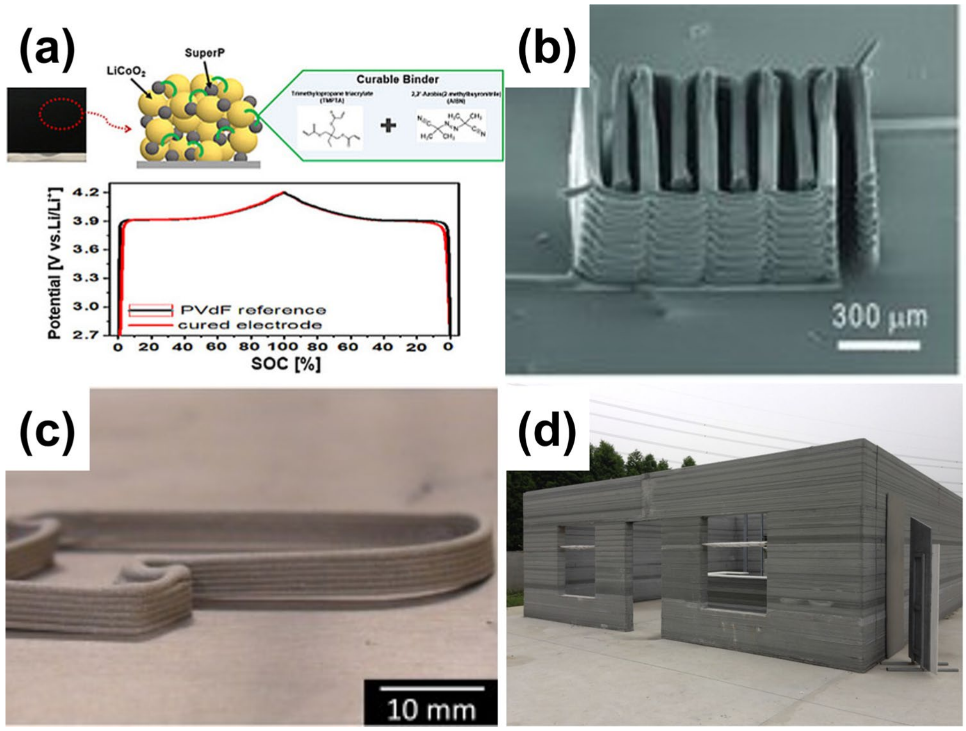

Fig. 13 Applications of ceramic additive manufacturing in other fields. **a** 3D printable lithium-ion battery ink, **b** interdigitated LTO-LFP electrode, **c** B_4C structure, and **d** construction printed by DIW method

5 Conclusions

C-AM technology is expected to overcome the limitations of ceramics and become a technology that can further expand and deepen the ceramic industry. However, C-AM has technical problems such as slow production rate, defects, and limited printing materials. To overcome these limitations, basic studies on modeling and formula-based printing process control have been actively conducted. Also, numerous studies have been reported that complement the technical limitations by fusion between printing technologies. In the C-AM process, model/formula-based research, high-speed sintering research, and technology fusion research have been conducted. The model/formula-based process is a key factor in improving process efficiency and accuracy. This effectively suppresses micro-defects and prevents failures in advance. The sintering process following printing has a great influence on the processing speed. To overcome these limitations, a high-speed sintering process using external energy has been studied, and one of them is microwave sintering. In addition to the process, the fusion between printing technologies has enabled ultra-high-speed, high-precision printing. The material has been actively developed only recently compared to other elements.

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Compliance with ethical standards

Conflict of interest All the authors declare no conflict of interest.

Ethical approval This article does not contain any studies with human participants or animals performed by any of the authors.

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