CERAMIC ADDITIVE MANUFACTURING: A REVIEW OF CURRENT STATUS AND CHALLENGES

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Abstract

In recent years, various additive manufacturing (AM) technologies that are capable of processing ceramic materials have been demonstrated. On one hand, many of the AM ceramic technologies have demonstrated geometry freedom capability and broad range of material flexibility. In some of the ceramic AM processes the part accuracies have also been favorably demonstrated. On the other hand, when reviewing the requirements of ceramic structures from applications perspective, there still appears to exist a misalignment between the demonstrated capability of ceramic AM and the required performance. The lack of critical microstructural characteristic and performance evaluation results are likely setting significant barriers for the broader adoption of ceramic AM, which should be addressed via close collaborations between academia and industries.

Keywords: Ceramics, additive manufacturing, review, applications

Introduction

Compared to both polymers and metals, ceramic materials possess many attractive physical and mechanical properties, most notably the generally superior mechanical properties (e.g. modulus, hardness, strength) at elevated temperatures and many unique physical properties such as electrical and thermal conductivities that can be tailored at broad ranges for different applications [1]. In many application areas such as aerospace, automobile and energy, many structures and components are often subjected to high operation temperatures, such as the gas turbines, engines, batteries and heat exchangers, for which ceramic materials appear to be most suitable [2-5]. However, the structures for these applications are often complex due to various performance and assembly needs, which presents significant challenges to the ceramic materials. It is well known that ceramic materials generally possess low toughness, low ductility and high crack damage sensitivity, which, combined with the high hardness and high melting temperatures, make them difficult to work with during the manufacturing [6-8]. Furthermore, due to the intrinsic material characteristics of ceramics, multi-material systems are often needed in order to develop functional components, which further adds challenges to manufacturing [9-13].

A vast body of literature exists about the manufacturing of ceramic materials, and it is rather difficult to establish a rigorous classification system to clearly distinguish all these manufacturing processes. In one of the recent works, it was proposed that these technologies can be classified into five categories [14]:

1. Casting/solidification process: Involves liquid-solid state change of the starting materials, which is often accompanied by volumetric change;
2. Deformation process: Involves the shaping of ceramic structures via plastic deformation

3. Machining/Material removal: Involves the material removal of a ceramic workpiece using subtractive methods;
4. Joining: Assembly of ceramic components via the creation of interfacial bonding;
5. Solid freeforming: involves additively manufacture the ceramics.

Most of the traditional ceramic processing technologies fall into the first four categories. The casting/solidification processing encompasses a large number of processes including the suspension and gel based casting processes, the injection molding processes, and reaction based processes. Due to the high melting temperatures of ceramic materials, the casting and molding of ceramics are usually achieved via the phase transformation of the "carrier" phases, which can be either solvent solutions or matrix materials. In addition, most of the casting/solidification processes utilize defined surfaces (e.g. mold cavities, containers) to facilitate the shaping of the ceramic geometries. Therefore, in general these processes face multiple challenges. First of all, due to the state change of the carrier phases during the fabrication processes, the ceramic structures are often prone to defects such as voids and cracks [15-17]. Secondly, the loss of the carrier phases (e.g. evaporation) causes the potential loss of the geometrical accuracy [18, 19]. Thirdly, the use of molds introduce significant limitations with geometrical design of the structures and render certain designs such as internal features infeasible [20, 21]. The deformation processing technologies include processes such as extrusion, gel foaming and superplastic forming, which mostly deal with ceramic suspensions and slurries that have higher viscosities, which enables them to be processed in similar ways as typical polymer materials [22, 23]. On the other hand, due to the extensive effects of shear, macro-phase segregation might occur, which in certain cases could even cause catastrophic part failure [24]. In addition, for feedstocks with non-spherical ceramic phases, the shear flow could also introduce additional anisotropy in the final material properties by aligning the ceramic phases preferably along the shear flow direction [24]. Compared to the first two categories, machining/material removal processing is relatively less broadly employed. This is largely due to the low damage tolerance and high crack sensitivity of the ceramic materials upon the machining. In many traditional ceramic material removal processes such as grinding and machining, damages such as pulverization and micro-cracking occur [25]. On the other hand, non-contact machining methods such as ultrasonic machining possess capability to process hard ceramic materials with minimum surface damage at the cost of processing speed, while laser machining suffers from the thermal residual stress due to the induced heat during the machining [25, 26]. A common approach to alleviate the machining damage of ceramics is to process the ceramic parts in their green states. However, even for the machining of the green ceramic parts, micro-cracking is still likely to occur due to the loss of mechanical strength of exposed green part surfaces, which could only be partially alleviated even with the introduction of additional binder phases or pre-sintering processes [25, 27]. Due to the limitations with geometrical flexibility for processes of the first three categories, the joining processes such as ultrasonic welding, co-sintering and diffusion bonding are often employed. The use of joining for complex ceramic structures such as porous scaffolds [28], multi-material composites [29] and functionally graded materials [30, 31] have been demonstrated, which often involve rather extensive and labor-intensive process chains.

Solid freeforming, on the other hand, refers to a series of relatively new processing routes that take advantage of additive manufacturing (AM) technologies. Compared to the processes of the other categories, AM technologies possess the unique capability to realize complex geometries and structural architectures directly from digital model designs. The use of AM for direct and indirect ceramic structure fabrication have been demonstrated in various literature using processes including material extrusion, stereolithography, binder jetting, ink jet printing, powder bed fusion and sheet lamination. However, unlike the AM polymer and metal materials that have been widely used in various applications, there have been very limited use of AM ceramics due to various reasons. In this paper the authors attempt to perform a review with the current status of the AM ceramic fabrication in the attempt to reveal some of the critical developmental gaps in this area.

Current applications of ceramics

While there exist various applications for ceramics, not all of them are readily justifiable for AM adoption. Many of these applications exploit only single attributes of the ceramic materials and do not require complex material/structural design. Such examples include the refractory containers, grinding wheels and chemical containers. With these applications, the benefit of AM is currently marginal. On the other hand, for the areas in which multi-objective designs or integrated designs could bring about improved performance, such argument is more easily made. These structures often favor multi-material designs or structures with complex architectures that renders traditional manufacturing approaches problematic. With this in mind, three areas of applications were reviewed as promising future directions, which are the engine and propulsion, the dentistry and the electronics.

Engine and propulsion

One of the most commonly exploited attributes of ceramic materials is their high-temperature mechanical properties, which allow them to survive the operations in extreme environments more reliably. Engine and propulsion components for aerospace, automobile and energy are among the most extensively investigation applications [32-35]. In these applications, the push for ever-higher operation temperatures in the attempt to improve the performance efficiency has stretched the traditional superalloys to their limit [36]. For example, for the next generation gas turbine engines with operation temperatures of over 1200°C or so, ceramics are considered to be the only suitable material options [35]. In these applications, ceramics are primarily used in two ways: either as thermal barrier coating (TBC) or as primary structural materials. [35, 37, 38].

The typical microstructural architectures of the TBC is shown in Fig.1, which clearly shows the TBC, the underlying superalloy substrate and the intermediate thermally grown oxide (TGO) layer in between [38]. The TBC topcoats usually have thickness of 100-2000μm and possess very low thermal conductivity across the entire operational temperature ranges in order to avoid the softening or melting of the superalloy substrates [38]. In addition, due to the frequent thermal cycles with large temperature ranges (room temperature to ~1300°C), the TBCs are also subjected to cyclic thermally-induced stress without fracture or delamination from the substrate, which usually have different coefficients of thermal expansion [35]. Such demanding requirements with both thermal and mechanical properties makes it difficult to find the suitable material candidates. Currently the most commonly utilized ceramic material for the TBC is the yttria stabilized zirconia (YSZ), although some other potential candidates such as pyrochlores ($A_2^{3+}B_2^{4+}O_7$) and fluorite oxides have also been investigated [35, 37-40]. The 7%wt yttria stabilized zirconia (7YSZ) is made into porous structures with microstructural defects to introduce low thermal conductivity and high compliance [36]. The ferroelastic toughening mechanism unique to this composition of YSZ also makes it mechanically robust [37]. However, YSZ-based TBCs are not without drawbacks. For example, at elevated temperature the oxygen diffusion from YSZ to the underlying superalloy substrates could significantly impact the performance of the structure, which necessitates the introduction of the TGO layer. The TGO layer usually consists of Al_2O_3 or mullite, which has very low oxygen diffusivity and bonds well with both the Ni-base superalloy and the TBCs [37, 38]. In order to overcome some of the other limitations encountered by YSZ-based TBCs such as the limited melting temperature (~1300°C) [36], the degradation of compliance due to the infiltration of the calcium-magnesium alumino-silicate (CMAS) that is formed from the ingested environmental dust [36, 41, 42], and the insufficiently low thermal conductivity at even higher service temperatures [36], multi-layer surface coatings have been investigated, which aims to further enhance the performance of the TBCs via combining the advantages of properties from multiple layers [40, 41]. For example, due to its higher temperature stability and low susceptibility to the CMAS infiltration, the $Gd_2Zr_2O_7$ (GZ) can be used as the topcoat, which is backed by the traditional YSZ that provides both toughness and good bonding with the TGO layer as shown in Fig.2 [40].

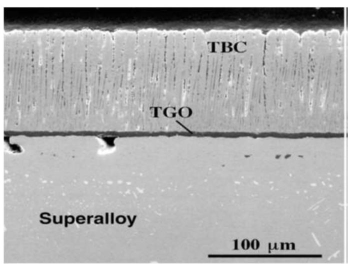


Fig.1 Cross section of a YSZ TBC deposited on a superalloy [38]

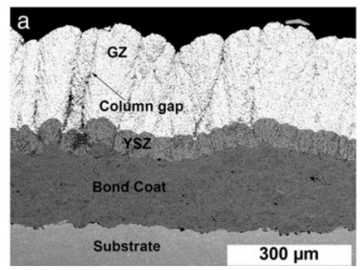
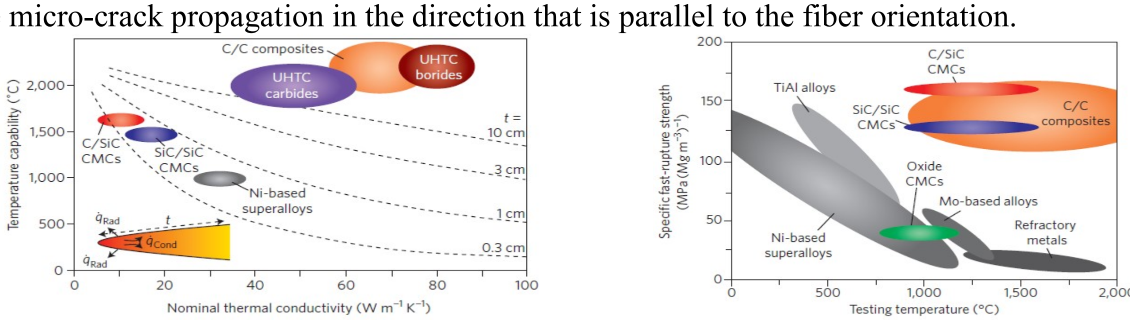


Fig.2 Cross section of a multi-layer GdZrO2/YSZ TBC on Hastelloy-X substrate [40]

Currently the TBCs are usually applied to the substrate via various surface deposition methods such as electron beam physical vapor deposition [39, 40], suspension plasma spray [41, 43], suspension high velocity oxy-fuel spray [43] and air plasma spray [44]. Some of the common objectives for all the processes include the introduction of controlled porosities and porosity distributions [44, 45], and the control of microstructure of the TBC layers [46, 47], which is often achieved via both process parameter adjustment and substrate preparation. Due to the layerwise structure and the need for controlled porosity for the fabrication of the TBCs, it appears that there exist potentials for the utilization of AM technologies. For example, it is known that the powder bed based AM processes possess the potential to produce samples with varying levels of porosity, which is often considered as defects but could be intentionally taken advantage of for the TBC fabrication. On the other hand, since the coatings must be applied to existing surfaces, the only AM technology currently capable of the coating of non-flat substrate is the directed energy deposition process, which has some significant drawback for this particular application, as it generally lacks porosity control and also introduces large in-process thermal gradients that could cause horizontal cracks that are detrimental to the performance [43, 48].

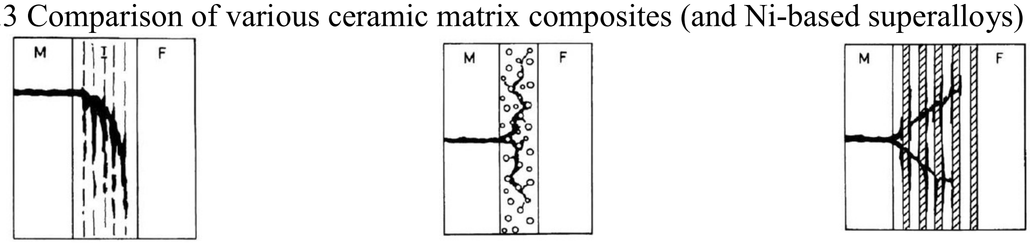
When even more stringent temperature requirement is imposed, the superalloy material could no longer meet the performance requirement, and the primary structures must also be made of ceramic materials. In most of these applications, monolithic oxide and carbide ceramics are usually not used due to their intrinsic crack sensitivity and brittle fracture modes [33], although some of these materials such as HfB_2 and ZrB_2 have been investigated for ultra-high temperature applications (>1500°C) such as the leading edge for hypersonic aircraft, in which the extremely-high temperature oxidation resistance and thermal cycling lifetime become of critical importance

[34]. Currently the ceramic matrix composites (CMC) are most employed for these applications due to their potentials to combine the thermal properties of ceramics with the mechanical properties of the reinforcement phases, which are usually in fiber form in order to provide the maximum reinforcement effects [49]. This is necessary since beside the temperature requirements, these components are also subjected to various mechanical requirements such as creep resistance [33], damage tolerance [36], high temperature strength retaining [32], impact resistance [50] and lightweighting [51]. Due to the exposure to the liquid propellants and other contaminants, chemical resistance is also a critical requirement [52]. Typical CMCs used in engine propulsion comprise SiC fibers embedded in SiC matrix, although other combinations such as ZrB_2/SiC [4], C/SiC [52] and Al_2O_3/Al_2O_3-$LaPO_4$ [53] have also been reported. Fig.3 shows the comparison of mechanical and thermal capabilities of various ceramic matrix composites. As a matrix material, SiC possesses several attractive characteristics for such applications, including high melting point (~2500°C), good mechanical properties (hardness, strength, creep resistance) at high temperatures, relatively good oxidation resistance, and relatively high thermal conductivity [51, 54]. However, since the oxidation resistance of SiC originates from the formation of the surface oxide SiO_2 layer, the material is prone to oxygen diffusion-induced oxidation failure, which could take place either through the matrix-reinforcement interphases or via the cracks upon damage [54, 55]. This issue imposes an important limiting factor during the design of both the reinforcement and interphase materials and the manufacturing processes. For example, although carbon fiber could theoretically offer maximum high-temperature mechanical strength benefits, due to its intrinsic tendency of oxidation, it would introduce significant oxygen diffusion pathway to the SiC matrix [36, 54]. Similarly, in the design of the interphase, the bonding between the interphase and the fiber surface should be strong in order to avoid oxygen diffusion through the reinforcement fiber [5.9]. On the other hand, the interphases are often designed to intentionally introduce controlled crack propagation mechanisms in order to enhance the toughness and damage tolerance of the ceramic matrix, which is usually relatively limited [54]. As shown in Fig.4, the interlayers are often designed to have laminated micro-architectures or highly porous microstructure in order to direct the micro-crack propagation in the direction that is parallel to the fiber orientation.



a. Temperature capability b. Mechanical strength

Fig.3 Comparison of various ceramic matrix composites (and Ni-based superalloys) [36]



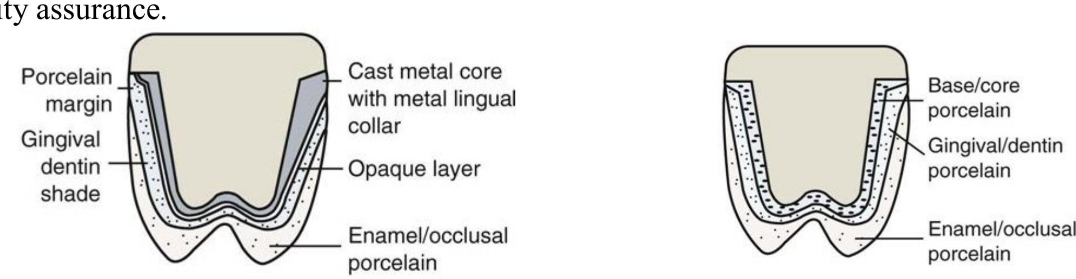
a. Single-layer pyrocarbon or hexagonal BN b. Porous SiC single layer c. Multilayer X-Y (X: pyrocarbon, Y: hexagonal BN)

Fig.4 Microstructural designs of interphases for ceramic matrix composites [54]

Multiple manufacturing processes are commonly employed to manufacture ceramic matrix composites, including liquid polymer infiltration [32], chemical vapor infiltration [33, 51], polymer impregnation and pyrolysis [33, 51], reactive melt infiltration [51], and impregnation-sintering [51]. In general the process routes for the ceramic matrix composites are similar to those for the other composites and are very complex, which makes the control of process quality a challenging task. This might have contributed to the very limited application of ceramic matrix composites in propulsion despite decades of intensive research and development. However, based on both the material and the structural requirements, it does not appear that AM currently has the capability to handle these materials, as there is a complete lack of literature with this subject. Multiple AM systems that are capable of fabricating fiber-reinforced composite structures have been developed, but they still lack the capability to deal with ceramic matrix materials [56, 57]. Due to the layerwise fabrication characteristics or AM, fiber reinforcements could only be oriented in the X-Y plane that is normal to the build direction. On the other hand, in theory a pick-and-place mechanism can be used to place fibers into the newly formed surface, which can be subsequently embedded into the matrix via either in-situ bonding (e.g. melting, photopolymerization) or post-process treatment (e.g. pyrolysis, sintering, infiltration) [58, 59]. To partially offset the challenge of processing ceramic materials directly, it has been suggested that the potential of polymer precursors could potentially be taken further advantage of [58], which was demonstrated in a recent work of AM ceramic matrix composite by 3Dynamics [60]. In this work, siloxane-based polymers (e.g. (mercaptopropyl)methylsiloxane and vinylmethoxysiloxane) and silazanes, which in combination with UV free-radical photoinitiator, free-radical inhibitor and UV absorber, were used as the photopolymer resin in the stereolithography-based process to fabricate precursor green parts. Further adding ceramic microfibers allows for the fabrication of CMC precursor, which after pyrolysis at 1000°C-1300°C in argon transforms to the CMC component based on silicon oxycarbonitride (SiOCN) matrix and ceramic microfiber reinforcement. Additives such as boron, zirconium were considered for the enhancement of high temperature capabilities of the structure, and additive such as silane could reduce the oxygen in the ceramic and pushes the ceramic composition towards SiC. However, for AM ceramic matrix composites, porosity and surface crack control remain significant challenges. Due to the anisotropy of the matrix structures as a result of the layerwise process, uniform shrinkage is yet to be achieved for pyrolysis. In addition, this work only demonstrated the manufacturing of ceramic matrix composites with microfiber reinforcements, while long-fiber reinforcements remain out of limit.

Dentistry

Ceramics have been widely used in dental applications for restorations of veneers, crowns and bridges [61, 62]. The most commonly investigated ceramic materials include alumina, leucite porcelain, zirconia and lithium disilicate. These materials are chosen largely due to their natural-looking characteristics, which are determined by various optical attributes such as color, opacity and translucency [63]. The dental ceramic materials are either used in combination with metals or in all-ceramic restorations, as illustrated in Fig.5 using dental crown restoration as an example. In veneer restorations, porcelain glass is among the first ceramics introduced into such applications, although it was not widely used before the concept of alumina (Al_2O_3) addition was introduced later in 1960s [62]. Another type of glass ceramic is the leucite reinforced glass ceramics. Leucite ($KAlSi_2O_6$) is a potassium alumino-silicate that has a tetragonal crystalline structure at room temperature and a cubic crystalline structure at temperature above 625°C. As a result, during the phase transformation, Leucite undergoes large volume change. Therefore, in the Leucite reinforced glass ceramic systems, the SiO_2-Al_2O_3-K_2O system develops internal compressive stress upon cooling from the sintering/casting temperature, resulted in a strengthening effect that acts to inhibit the crack propagation within the ceramic structures [61, 64]. In comparison, for the all-ceramic systems, due to the intrinsic crack sensitivity of ceramic materials, the overall performance is still yet to match that of the metal-ceramic systems [65]. Among the all-ceramic materials, zirconia is currently most broadly used due to its combination of high toughness, low thermal conductivity, low corrosion potential, high biocompatibility and good radiopacity [62, 66, 67]. Yttria stabilized zirconia (YSZ) is most often used in order to improve the material processibility and to enhance the toughness. Despite the promising properties, in clinical practice it has been found that dental parts made by zirconia or a combination of zirconia and layering glass ceramic have a higher incidence of fracture compared to prostheses fabricated exclusively out of dental metal alloys. One major cause of the failure is the occlusal overload due to bruxism, which causes crack at the cementation surface that propagates subsequently radically towards the surface and eventually causes the fracture of the entire restorations [65]. Also, there are other common minor complications that are related to the repeated loading of the restorations. Another issue with zirconia and other oxide ceramic materials is the low translucency and therefore undesired aesthetic properties. In order to overcome this, a top layer made from porcelain glass is often applied to the oxide ceramic restorations, adding another level of complexity to both process and quality assurance.



a. Ceramic-on-metal b. All-ceramic

Fig.5 Dental ceramic material systems for crown restoration [68]

The zirconia ceramic restoration parts are commonly fabricated by slip casting or machining [61]. In recent years, machining based approach becomes increasingly adopted, in which the CAD/CAM technology is employed to produce ceramic parts with accurate shapes [27, 69]. In order to avoid introducing excessive surface damages to the ceramics, partially sintered ceramics or green ceramics can be machined, which will be subsequently sintered to achieve full mechanical strength. A process flow for this CAD/CAM process method is shown in Fig.6. After the scanning of the patients' anatomy, the digital teeth models will be re-constructed, and a digital design of the dental restorations will be created. Based on the model, the CNC machining toolpath will be generated by the CAM software, and pre-sintered ceramic blocks will be machined to the shape of the restorations with up to 25% of scaling-up factor. Afterwards, the machined green ceramics will be sintered in a furnace to achieve the final mechanical strength. Due to the digitalization of dental restoration design procedures and the ease of machining with the green zirconia, this process route can readily achieve high production speed (~25-30min/part) and is reasonably cheap [7.8]. However, even with the green ceramic parts, micro-cracking is still likely to occur due to the loss of mechanical strength of exposed green part surfaces [25, 27]. Although this could be alleviate by adding binder into the green part compacts or by pre-sintering of the parts, the micro-crack issue could not completely eliminated [25]. This creates potential concerns with the long-term serviceability of the restoration parts and likely contributed partly to the higher failure rate of all-ceramic zirconia restorations.

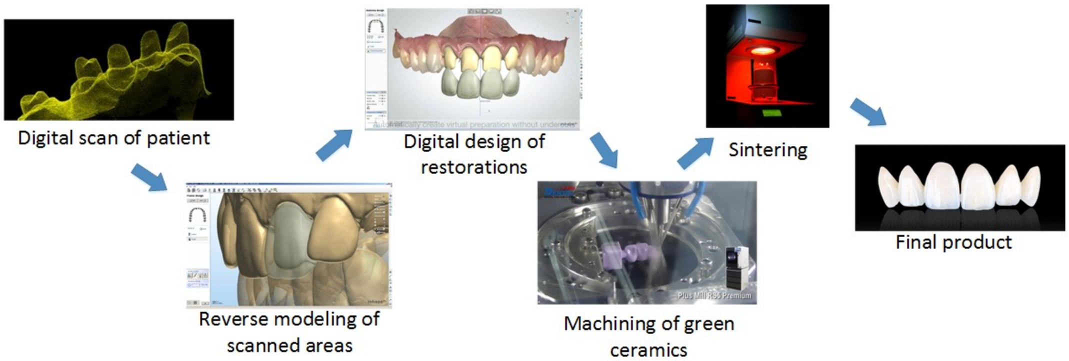
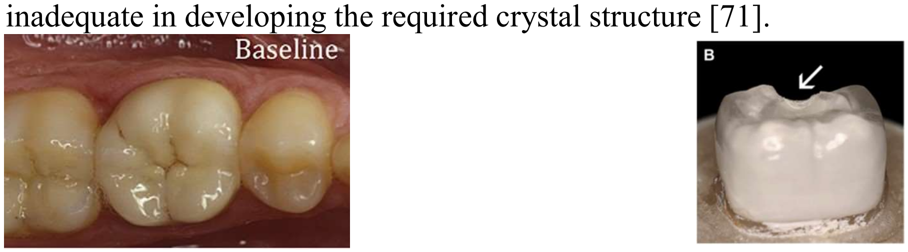


Fig.6 CAD/CAM for dental ceramic restoration manufacturing

Another relatively new ceramic dental material is the lithium disilicate ($Li_2Si_2O_5$). Due it its relatively high degree of crystallization (~70% vol.), lithium disilicate possesses significantly higher mechanical strength and biocompatibility compared to the other glass ceramics [62, 64]. Lithium disilicate is usually made by precipitation from the SiO_2-Li_2O-K_2O-ZnO-P_2O_5-Al_2O_3-La_2O_3 system, and the mismatch of the coefficient of thermal expansion between the lithium disilicate and the glass matrix is largely contributing to the improved fracture toughness of the material. With good clinical survival rate and high aesthetic quality, lithium disilicate possess potentials to be widely used in applications in inlays, onlays, crowns and other restorations. Fig.7 shows the comparison of a crown made by lithium disilicate and zirconia, and the difference in aesthetics is significant. Lithium disilicate dental parts are fabricated by either investment casting process or CAD/CAM process using the pre-made lithium disilicate feedstock, as in-process alloying is inadequate in developing the required crystal structure [71].



a. Lithium disilicate b. Zirconia

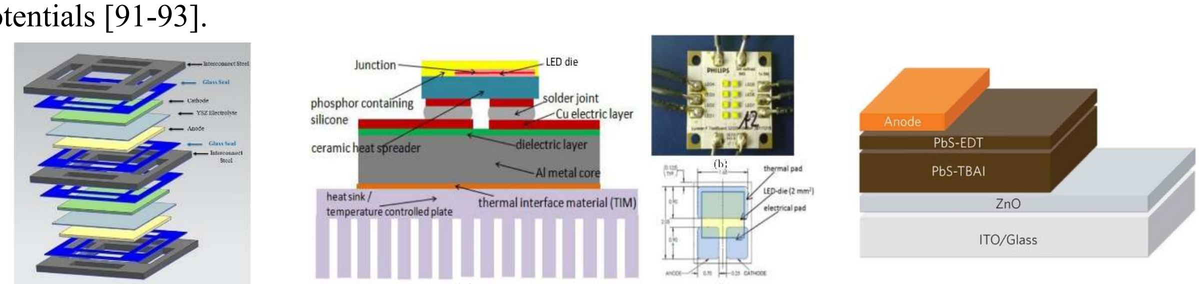
Fig.7 Crown parts made by two ceramic materials [62]

The use of AM in dental ceramic fabrication has been reported using several processes, although various issues remain unsolved [70, 72-75]. These issues including the lack of geometrical accuracy, staircase surface effects, and high porosities. For many dental prosthetic applications the marginal tolerance of the prostheses needs to be <0.1mm, which is difficult to achieve for many AM processes especially when material density and strength are also required [74]. In addition, due to the need of post-printing sintering processes with many AM processes, the porosity control and sintering distortion remain challenging. Compared to the CAD/CAM process, currently AM does not appear to have significant advantage with production speed, therefore for zirconia dental prostheses the justification might need to be established via performance enhancement in the future. Furthermore, as previously mentioned, one of the unique requirements for dental ceramics is the need of specific optical characteristics. Although the fabrication of glass using AM with somewhat controlled optical properties has been demonstrated

[76-79], so far no technical ground appear to exist for the design of AM process or materials for tailored optical properties. As for the near future, such issues will likely have to be handled via either the use of specific materials (e.g. lithium dilisicate) with pre-defined optical characteristics or post-printing surface treatment processes (e.g. coating).

Electronics

The use of ceramics in electronics applications is widespread. It was estimated that the global market for electronic ceramics was around $9 billion in 2014 [80]. In these applications, ceramic materials are used for their intrinsic dielectric properties and tailorable electrical and electromagnetic properties when doped. Ceramics are used as insulators, conductors, capacitors, piezoelectric sensors and actuators, semiconductors, optical and electromagnetic films and many other applications [81]. Therefore, the requirements of ceramic materials for this application category are most diverse. In many of these applications, a myriad of studies have elucidated the importance of microstructure including phases, grain sizes, grain boundary characteristics, contaminants/impurities, point defects (vacancy and interstitialcy) and phase morphologies, to name a few [82-87]. It is beneficial to design for small-size ceramic electronic components since this reduces the tendency of defect generation during the manufacturing processes [88], although the requirement of small size also comes from the demand of high performance efficiency and lightweight for many applications [89]. This implies that the manufacturing processes for these applications must address both the microstructural control and high-resolution geometry control while ensuring acceptable production rate, which is quite challenging for the current AM technologies. In addition, many of the ceramic electronics components and systems exhibit rather complex architecture as shown in Fig.8, which often involve the use of multiple materials that might cross categories (e.g. metals + ceramics). As multi-material fabrication is still a relatively new area for AM, it might take considerable research efforts for the AM technology to become competitive in comparison to the traditional micro/nano fabrication technologies. On the other hand, due to the extensive employment of layerwise micro-fabrication processes such as photolithography, sputtering and vapor deposition, the current design architecture for these electronic devices could theoretically allow for smooth transition towards AM if the multi-scale manufacturing issues can be solved. It could also be envisioned that for AM processes that are capable of voxel-based fabrication on non-flat surfaces such as the aerosol jet direct write technology [90], the capability to accommodate 3D integrated electronics design could potentially bring about disruptive application opportunities, as currently such research and development in such areas are significantly hindered by the existing fabrication processes despite the promising potentials [91-93].



a. Solid oxide fuel cell [94]

b. LED module package [95]

c. Quantum dot solar cell [96]

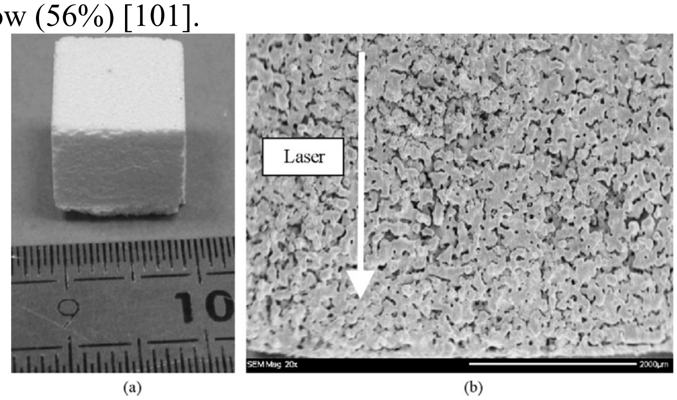
Fig.8 Architectures of some ceramic electronics

Current AM ceramics: state of the art

Almost all types of AM technologies except for material jetting have been employed for the fabrication of ceramic parts and structures. Most of the existing literature focus on the demonstration of AM ceramic fabrication capabilities with specific emphasis on geometry creations. In this paper, the emphasis was focused on the powder bed fusion, binder jetting, stereolithography and sheet lamination AM processes.

Powder bed fusion

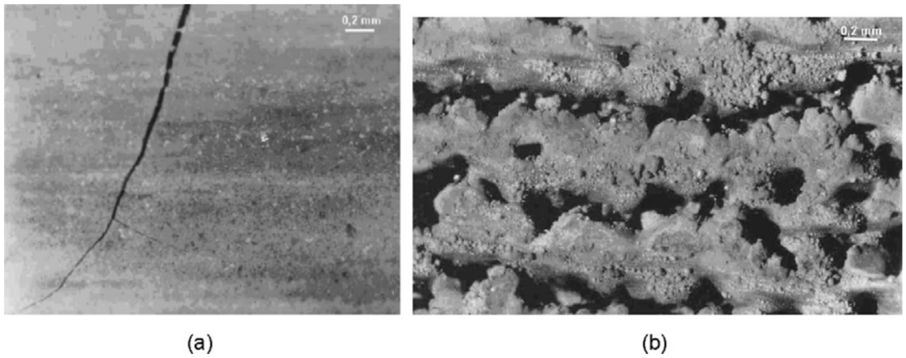
Powder bed fusion (PBF) processes was among the first AM processes that were employed for ceramic fabrication. The PBF processes that utilize electron beam as a heat source have received little attention from the researchers for the fabrication of ceramic materials likely due to the thermally induced challenges such as cracking and structural distortion [97, 98]. On the other hand, the applicability of the PBF processes with a laser beam heating supply have been studied by different researchers. Silica sand components was fabricated by laser melting in an attempt to produce casting molds [99, 100]. Fabrication yttria–zirconia powders via laser sintering/melting for dental applications was also experimentally investigated, although the resulting density of the structure was rather low (56%) [101].



a. Part view b. Top view

Fig.15 Zirconia cube fabricated by laser sintering/melting [101]

Yttria stabilized zirconia (YSZ) is among the few ceramic materials that have been investigated extensively using PBF processes, likely due to its relatively favorable phase stability after solidification and its high toughess. Shishkovsky et al. investigated the process of the mixture of alumina and ZST using laser melting PBF using different atmospheric conditions [102]. The mechanical performance of the fabricated ceramic parts was sensitive to the process parameters due to the formation of the porosities. Porosities of above 100 μm was observed, along with the occurrence of cracks as shown in Fig.15.1. The cracks were likely caused by the accumulation of heat during the process and is not common for laser melting PBF of this type of material and could be alleviated by preheating of the powder bed. Fig.15.2 shows a YSZ-Al_2O_3 dental bridge fabricated by laser melting, in which the powder was preheated to >1600°C [103]. The poor surface quality of the fabricated sample is attributed to the large melt pool size facilitated by high preheating temperatures as well as surface powder sintering. The authors reported that the fabricated parts were of almost 100% density with flexural strengths of above 500 MPa, and macroscale crack was largely prevented by the preheating. On the other hand, due to the thermal shocks imposed during the spreading the cold ceramic powder, presence of fine cracks on the surface of fabricated components were observed.



a. Crack of YSZ-Al_2O_3 parts –in air (x50) b. Porosity – in argon (x20)

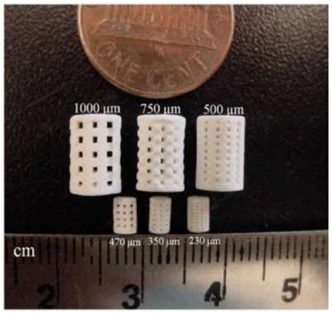
Fig.15.1 Characteristics of the laser melted YSZ-Al_2O_3 [102]

Fig.15.2 Dental bridge restoration printed via laser melting PBF [103]

In general, for the powder bed fusion processes, fabrication of ceramic parts seems to be more challenging compared to other AM process, and most studies have focused to assess the feasibility of PBF technology for ceramic material systems. With the successful demonstration of high quality fabrication of ceramic parts using the other AM technologies, the PBF does not draw extensive attention in this area.

Binder jetting

Binder jetting (BJ) has been extensively utilized for ceramic fabrication due to its unique process characteristics. Due to the use of binder as the geometry shaping mechanism, BJ process eliminates thermal inputs and could therefore process a broad range of materials including ceramics [104]. There exist an abundance of literature that demonstrated the use of BJ in the fabrication of various ceramics such as Al_2O_3, SiC, zirconia toughened alumina (ZTA), hydroxyapatite and calcium phosphate [105-122]. BJ has been investigated particularly for its potentials with bio-ceramics in biomedical applications [113-122]. Fig.16 shows two examples of the BJ ceramic structures for such applications. This is largely because that in many biomedical applications porosity is required for the structures, which is one of the characteristics of powder based ceramic structures. Since the BJ process is essentially a green-part fabrication process, in many applications it can be used to readily substitute the traditional powder-based ceramic processes to directly fabricate porous ceramic structures. These porous ceramic structures are either used directly or subjected to further post-processing that can be guided by essentially the same guidelines developed for traditional powder-based green ceramic components.



a. Calcium phosphate scaffold [122]   b. tri-calcium phosphate cranial segment [116]

Fig.16 Bioceramic structures by BJ

On the other hand, the BJ processes are subjected to similar constraints to the traditional powder based ceramic processes. Due to the powder spreadability requirement, the BJ processes often require that the powder feedstock exhibits good flowability [123]. This imposes significant constraints, as many ceramic powders are prepared via ball milling processes or chemical reaction based processes, and therefore often assume irregular particle morphology that exhibit low flowability and large aggregation [124]. Low flowability not only causes low packing density of the powder bed but also introduces significant spreading defects. In some works the ceramic powder was pre-treated with coating in the attempt to improve their flowaiblity. However, such approach inevitably introduces contamination phases that might be unwanted in applications [125, 126]. Another approach that might possess promising potential is the use of ceramic slurries as feedstock [127-130]. Such approach not only allows for the fabrication of low-flowability but also fine ceramic particles that are too small to be spread in dry status. On the other hand, since the green part printing resolution and accuracy of the BJ process is largely controlled by the binder liquid permeation process, the introduce of additional liquid phase will likely cause new issues with process control and optimization. In addition, manufacturing issues related to the use of ceramic slurry such as shrinkage crack and distortion must also be considered.

Various post processing techniques have been employed to achieve good density for BJ green ceramic parts, such as liquid phase sintering, infiltration, and isostatic pressing [131-136]. The liquid phase sintering has been reported as an effective method in increasing the part density of BJ green ceramic parts [131-133]. Works have been reported for the infiltration of BJ alumina components by lanthanum-alumino-silicate glass and copper alloys, which resulted in the significant increase in the part density [134, 135], and in some cases up to 450% in flexural strength increase was observed in the specimens infiltrated with epoxy [136]. For the applications where the compositional purity of the fabricated samples is required, traditional ceramic processing technologies such as warm/cold isostatic pressing might be helpful in reaching high densities of components with relatively simple geometries [107, 137].

Stereolithography

A lot of the recent development efforts with AM ceramics focus on the use of stereolithography (SLA) based technologies. SLA is known for its high process resolution, and is therefore considered a promising technology for AM ceramic fabrication development considering the fact that post-processing of ceramics tend to introduce damages. Although the need for photopolymer feedstock poses significant constraints with the material development, various monolithic materials such as ZrO_2, Al_2O_3, SiC, SiOC, SiO_2 and TiC have been fabricated using SLA with two primary forms of material feedstock: (1) ceramic suspension, and (2) ceramic-derivable polymer precursors. There exist an abundance of literature about the development of ceramic suspensions for SLA. Such developmental work is not trivial, as the control of rheological and photocuring characteristics is of critical importance in the control of the SLA part qualities and more so the ceramic SLA [138-140]. For ceramic suspensions, these properties of the ceramic suspensions are influenced by various factors such as ceramic solid contents, dispersant and diluent concentrations, photoinitiator concentration and temperatures [141-145]. This is also closely related to the configuration of the SLA systems. For the conventional top-down configuration, in which the part is submerged in the photopolymer resin during the process, the resin is spread over uneven surfaces during the recoating. Therefore, the large variability of the liquid thickness imposes constraints to the development of the feedstock resin recipes [140]. With bottom-up configuration, the part is placed upside-down with only the top layer of the part in contact with the resin, such recoating issue is alleviated, although the liquid phase separation and recoating upon each additional layer becomes an issue [146]. Lower viscosity is generally desired for the ceramic suspension [147], although this is often difficult to achieve due to the other considerations. To ensure effective sintering with the brown parts, it is suggested that a minimum solid content of 50% should be used for the ceramic suspensions [140, 148]. However, when the solid content exceeds 50%, the viscosity of the fluid increases drastically [149]. Solid contents of up to 60% has been reported, although porosities are still present in the final parts after sintering [141, 145, 150].

Beside the rheological property issues, laser irradiation-related issues must also be considered for the development of ceramic suspension [140, 143, 144]. As shown in Fig.20, depending on the refractive index contrast, the resin feedstock could exhibit very different curing profiles under the UV irradiation due to scattering, which in turn influences the process planning and part qualities. In addition, the modulation of light intensity during different stages of the irradiation could also affect both the geometry and mechanical quality of the parts [151]. Also, while excessive photocuring could reduce the part integrity and property, sufficient crosslinking is needed since the existence of residual monomer in the green ceramic parts could introduce cracking due to the thermally activated polymerization [152].

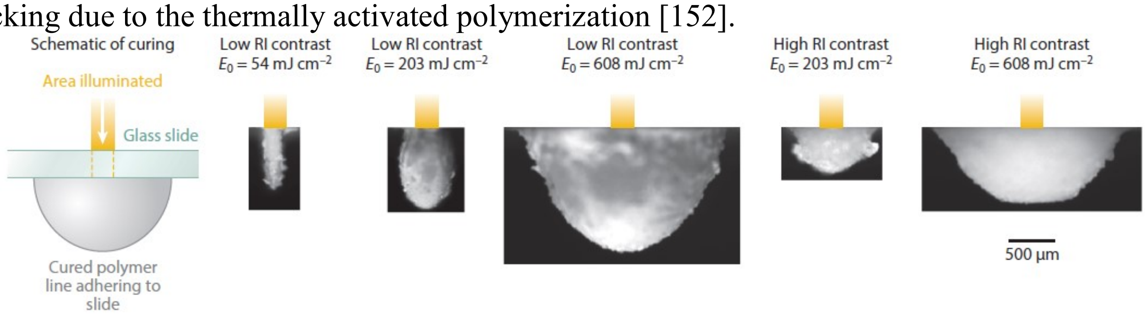


Fig.20 Shape of the curing profile for SiO_2 suspensions with varying refractive index values [140]

Such dilemma between processibility and final part property for ceramic suspension based SLA may have prompted the development of the ceramic-derivable photopolymer as the feedstock. The principle of this approach is to utilize polymers that could decompose into ceramic products upon sintering/ pyrolysis for the green part fabrication. Since the ceramic-derivable polymer precursors can be tailored to exhibit a broad range of electrical, optical, chemical and mechanical properties, the material development for SLA based ceramic systems appear to exhibit more potentials [153]. Additional compositional control could also be realized by adjusting the precursor recipes, which was demonstrated for the fabrication of SiC/SiOCN and SiC/SiO_xC_y composites for engine turbine blades [60, 154]. In addition, since the pyrolysis temperatures of the polymer-derived-ceramics are relatively low (<1300°C), it also requires less energy for the sintering processes. Microfiber reinforced ceramic matrix has been fabricated, although the resulting structures still exhibit various defects such as surface striation and internal porosities. Recently, the use of microfiber and short fiber reinforcements in ceramic-derivable photopolymer was also demonstrated [60, 155]. The fabricated structures exhibit various defects such as surface striation and internal porosities, and the increased viscosity of the photopolymer due to the fiber addition could potentially become an issue. However, the increased viscosity could also be utilized to introduce shear-induced fiber alignment via micro-oscillation, as shown in Fig. 21 [155].

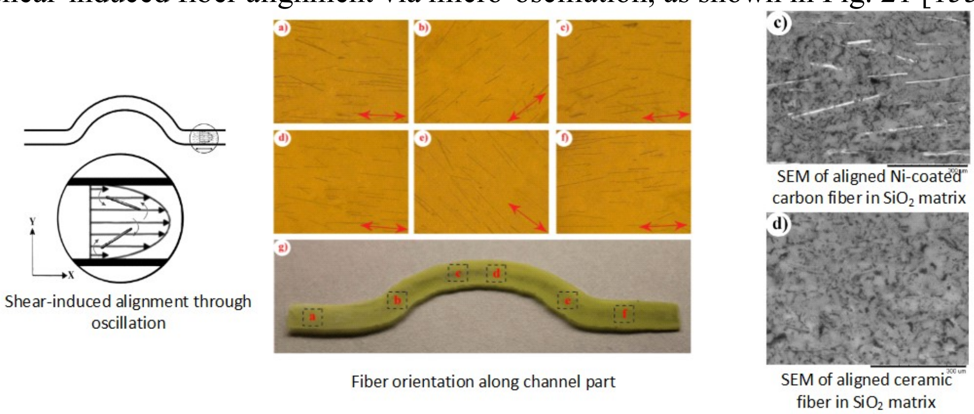
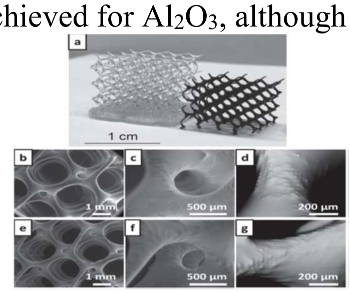


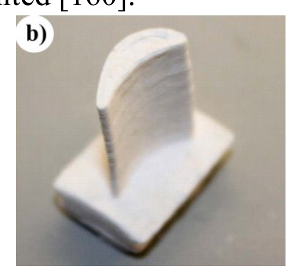
Fig.21 Shear-induced fiber alignment in ceramic matrix composites, adapted from [155]

Various literature has demonstrated the capability of SLA in the fabrication of complex ceramic geometries, such as the ceramic cellular structures [140, 153, 156] as shown in Fig.22a-b, turbine blades [60, 155] as shown in Fig.22c, investment casting cores [140] as shown in Fig.22d, dental crowns [74] as shown in Fig.22e and blood pump [157] as shown in Fig.22f. With vector and mask projection based SLA, minimum feature resolutions of 100-125µm were reported [150, 153, 158, 159]. For two-photo lithography based systems a minimum feature of 1.2µm was achieved for Al_2O_3, although the information about part quality is limited [160].

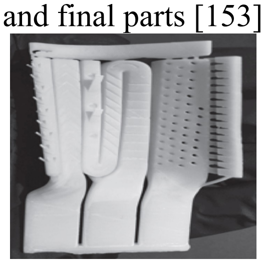


a. SiOC cellular parts - green and final parts [153]

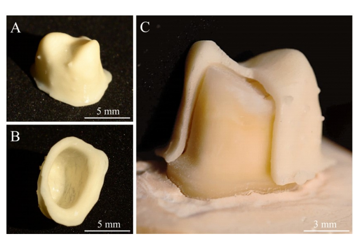
b. SiO_2 spring array [140]



c. SiO_2 blades [166]



d. SiO_2 investment core [140]



e. Al_2O_3 dental crown [74]

f. Ceramic blood pump [157]

Fig.22 Ceramic geometries fabricated by SLA

On the other hand, porosity issue is still prominent with SLA ceramics, which often exhibit porosities ranging from 3-7%. The inevitable volumetric change resulted from the post-fabrication sintering process remains the main contributor to the existence of porosities. Some interesting attempts have been reported in remedy of this. In [161] the zirconia-toughened alumina (ZTA) was fabricated via stereolithography. The structure exhibit 99.5% of theoretical density and toughness value comparable to that of conventional processes. It was suggested that a careful two-step debinding process that combines vacuum pyrolysis and air debinding was largely responsible for the elimination of part defects [162]. In another work by the same group, a liquid precursor infiltration process that serves to introduce additives or dopants to the printed ceramic parts was investigated [163]. The infiltration process was carried out after the debinding process, which improves the density and microstructure of the final parts after sintering. Such method was used on binder jetting processed samples as well [164]. The use of non-reactive plasticizing agent was also studied for the reduction of debinding defects, which reduces the amount of evaporated phase generated during the debinding [165].

Sheet lamination

Sheet lamination (SL) utilizes sheets of materials as the feedstock for structure fabrication, which implies that the microstructure and quality of the material within a single layer can be well controlled. The sheets used for part fabrication can be either cut and then stacked (Bond-then-Form) or stacked and then cut (Form-then-Bond) [166]. Although originally limited to paper feedstock, the SL principles have been successfully applied to the fabrication of metal, ceramic, and composite materials. In this case, ceramic tapes instead of paper sheets were used as build material for green part fabrication typically followed by high-temperature post-fabrication processing to debind and sinter the structures. Griffin et al fabricated ceramic components using alumina tapes via laminated object manufacturing (LOM) process, which exhibit comparable mechanical characteristics to pressed samples [167]. Further improvement in the bending strength of the fabricated alumina was accomplished by modifying the microstructure to an alternating composite structure of Al_2O_3 and Ce–ZrO_2 layers from 311MPa for Al_2O_3 up to 688MPa for the alternating composite [168]. The use of binding agent is common for the SL with paper feedstock, however, when using a binding agent between successive ceramic tapes, unsuccessful debinding process might result in anisotropic mechanical properties in the fabricated components [167-169]. The applicability of SL technology for processing LiO_2–ZrO_2–SiO_2–Al_2O_3 (LZSA) parent glass was investigated in various studies [170, 171], and 3D components have been successfully fabricated using LOM process as shown in Fig.23. However, high porosity values (greater than 10%) located at the lamination interfaces after post-processing limits the mechanical performance of the parts [170]. Other material systems such as glass-fiber reinforced polymers, SiC, SiC-fiber/SiC, AlN, Si_3N_4, HA, and TiC/Ni have also been examined as build material in sheet lamination technology [169, 172-175]. Due to the use of ceramic tapes, the SL processes usually only produce green parts, and consequent debinding and sintering must be accompanied by pressure in order to ensure the elimination of the porosities, and shrinkage along the layer stacking direction of less than 7% was observed due to the high green part densities [172]. The Reaction bonding Infiltration of LOM parts after sintering can be used to improve the density of the printed component [172]. The process flow of such approach is shown in Fig.24a. Successful demonstrate of SiC fiber reinforced SiC ceramic matrix composites was provided by the same group, which used alternating lay-up for consecutive ceramic layers to minimize fiber abrasion. However, it was observed that the interfaces between the SiC fibe and the SiC matrix were not well formed

(Fig.24b), which was caused by the lack of interfacial protection from oxidation during the sintering.

The full potential of SL appears to be yet exploited, since this technology is among the few AM technologies that are readily capable of multi-material fabrication. The capability of the SL with non-flat surface also implies that the process might be applicable to the fabrication of complex ceramic laminates on the surfaces of existing structures [172], which might be a promising alternative for the fabrication of TBCs. On the other hand, the difficulty in interfacial bonding quality control remain a challenge for SL to be employed for high-performance ceramic composites and TBCs. On the other hand, the z-direction resolution of the SL parts might be subjected to more constraints as the ceramic tape-pegs are required to possess minimum strength to be processed without damage. With the reported layer thickness of 0.25mm [173], the SL ceramic structures will be unsuitable for applications that require high geometrical accuracies such as dentistry.

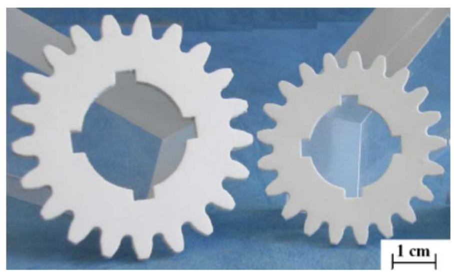
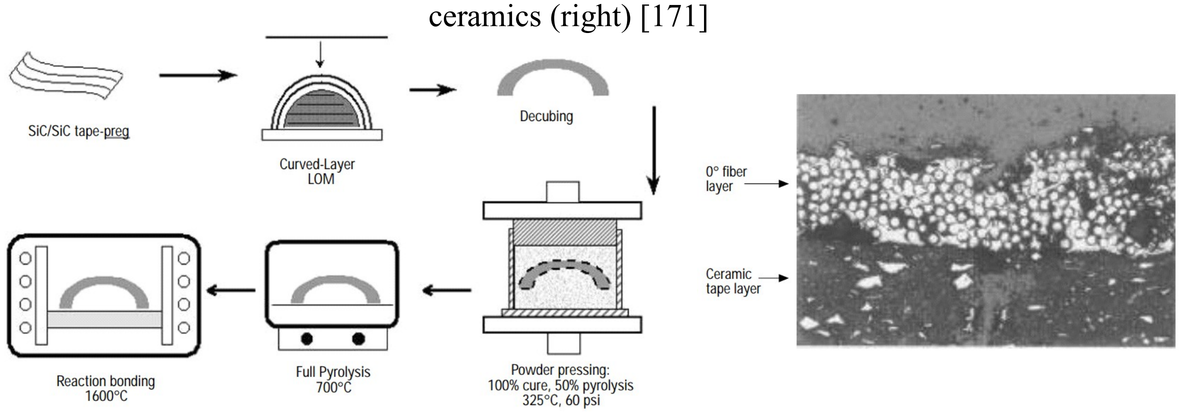


Fig.23 Gear wheel geometries: green laminate (left); sintered Li_2O–ZrO_2–SiO_2–Al_2O_3 glass–ceramics (right) [171]



a. Process flow chart b. Interfaces of SiC/SiC composite

Fig.24 Fabrication of SiC/SiC fiber reinforced composite using SL + reaction bonding [172]

Discussions and conclusions

It appears that currently there exist various misalignments between the demonstrated AM ceramic capabilities and the needs for particular applications in the identified areas. While AM is generally capable of realizing relative complex geometries [176], such advantage is significantly compromised by the lack of microstructural quality control with the ceramic parts. Various issues such as porosity, purity, micro-defects and interfacial defects commonly exist with AM ceramic structures, which still require extensive efforts to overcome [137]. Due to the intrinsic staircase effect with the AM processes, the notch sensitivity issue of the ceramic parts is signified [27]. The need for post processes such as debinding and sintering implies that many of the challenges with the ceramic geometry designs that the traditional ceramic manufacturing face are also applicable to AM ceramics, such as the limitation of feature thickness and the debinding/sintering distortion, which imposes geometry design limitations that greatly compromise the advantages offered by AM. In addition, from traditional powder based ceramic manufacturing it is known that finer ceramic particles are preferred for achieving higher densities during the sintering densification. However, refined particles tend to exhibit lower flowability and becomes difficult to spread using the powder bed AM approach. On the other hand, with the powder suspension feedstock based AM, due to the requirements with the rheological properties, the ceramic solid content is limited, which in turn limits the achievable density of the densified parts and necessitates more complex sintering strategies such as liquid phase sintering or HIP.

Due to the need of post processes that are similar to the traditional ceramic manufacturing processes, AM could considerably benefit from the existing expertise within ceramic industries. For example, for slurry and suspension based ceramic AM technologies, there exist extensive knowledge base in this subject that has yet to be explored within the context of AM. For example, various literatures have provided excellent reviews about the key consideration factors for the development of the ceramic pastes during the extrusion based shaping processes. The flow rheology of the viscous ceramic pastes can be modeled, and phase separation at macro-scale must be avoided as it could lead to defects such as cracks [177]. During the material extrusion processes, flow-induced alignment of the ceramic particles could cause additional anisotropy in the structures, which can be potentially utilized to create textured microstructure [178]. The paste must be free of large aggregates in order to produce defect-free structures. In addition, during the debinding of the green ceramic parts made of powder injection molding, broader binder decomposition temperature ranges, which can be characterized by either thermogravimetric analysis or digital scanning calorimetry, benefit the reduction of the crack formation in the de-binded brown parts, since the escape process of the evaporated gas phase from the binder is more gradual [178]. Slower binder decomposition is also beneficial for the retaining of the part shape. These knowledge will likely facilitate the development of new binder and slurry systems for the AM ceramics, which is an area that does not appear to be explored yet.

In terms of ceramic material availability, the existing AM technologies already demonstrated excellent material compatibility. New materials such as boron carbide (B_4C) and titanium boride (TiB_2) might also possess promising potentials for AM adoption due to their potentials in high value-added ceramic armor applications [179, 180]. In addition, the capability of AM in applying materials on a voxel-by-voxel basis could be potentially utilized for the doping of ceramics, which is an important method in altering the material characteristics of ceramics [181].

In the near term, in applications where functional ceramic structures are needed that use material feedstock already exhibiting the intended performance, the geometry realization potential of the AM could be better utilized [182]. For example, the fabrication of piezoelectric ceramic devices using high-piezoelectric coefficient ceramics could yield parts that are comparable to the traditional ones [182]. In addition, in specific applications where exotic geometries such as cellular structures are desired, AM could also find immediate use. For example, it was claimed that the printed HA implants have the required porosity for osseointegration while achieving mechanical strength 3-5 times better compared to the conventional implants with the same material and porosity [183]. In the long term, ceramic AM will likely benefit from close collaborative research and development efforts between the academia, system manufacturers and industrial users, as application-oriented development works with the platform, the materials and the process optimizations are all needed.

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