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Additive Manufacturing of Ceramic Structures by Laser Engineered Net Shaping

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Abstract: Ceramic is an important material with outstanding physical properties whereas impurities and porosities generated by traditional manufacturing methods limits its further industrial applications. In order to solve this problem, direct fabrication of Al_2O_3 ceramic structures is conducted by laser engineered net shaping system and pure ceramic powders. Grain refinement strengthening method by doping ZrO_2 and dispersion strengthening method by doping SiC are proposed to suppress cracks in fabricating Al_2O_3 structure. Phase compositions, microstructures as well as mechanical properties of fabricated specimens are then analyzed. The results show that the proposed two methods are effective in suppressing cracks and structures of single-bead wall, arc and cylinder ring are successfully deposited. Stable phase of α-Al_2O_3 and t-ZrO_2 are obtained in the fabricated specimens. Micro-hardness higher than 1700 HV are also achieved for both Al_2O_3 and Al_2O_3/ZrO_2, which are resulted from fine directional crystals generated by the melting-solidification process. Results presented indicate that additive manufacturing is a very attractive technique for the production of high-performance ceramic structures in a single step.

Keywords: lasers, net shaping, alumina, ceramics, additive manufacturing

1 Introduction

Ceramic is an important inorganic non-metal material which has been widely utilized in many areas, such as mechanical, ocean, electrical and information engineering as well as the biomedical engineering due to its outstanding mechanical, physical and chemical properties[1–4]. Traditional methods for fabricating ceramic components usually consist of many processing steps(as shown in Fig. 1(a))[4]. Only simple structures could be shaped by the traditional methods and defects like low-purity, porosity along with the poor efficiency are hard to solve because many binders or sintering agent are usually used in the process which need removing and long-time solid state sintering.

In order to solve the limitations of traditional fabricating methods for ceramic components, two additive manufacturing methods, the direct and indirect methods, were developed for this area and have been successfully demonstrated for their advantages. The indirect method, as shown in Fig. 1(b), uses many additive manufacturing methods in the shaping steps by which complex structures could be obtained(as shown in Fig. 2)[5–9]. TANG, et al[5], developed a slurry-based selective laser sintering method to built rigid green block from alumina powder coated with water-insoluble polyvinyl alcohol. Alumina ceramic part with an average flexural strength of 363.5 MPa and a relative density of 98% was obtained after binder removing and sintering. ALLAHVERDI, et al[6], developed the fused deposition of ceramics based on FDM technology. A variaty of advanced green ceramic components were fabricated rapidly in a layer-by-layer fashion. LIU, et al[7], obtained a ceramic part with interconnected reticular porous by using the method of selective laser gelling(SLG). Freeze-form extrusion fabrication(FEF) is another indirect method for ceramics. This environmentally friendly process uses highly loaded aqueous ceramic paste with a small quantity of organic binder to fabricated ceramic green part at a temperature below the freezing point[8]. Generally, complex ceramic 3D structures can be achieved by indirect methods mentioned above, but binder removing and sintering steps are also needed because raw materials used in these indirect methods are usually mixed by ceramic powders and massive organic or inorganic binders as traditional methods. Consequently, limitations of low-purity and poor efficiency still exist because processing steps after shaping are the same as the traditional methods.

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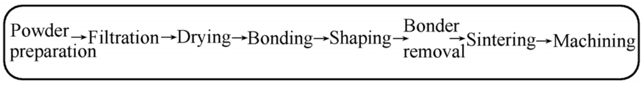
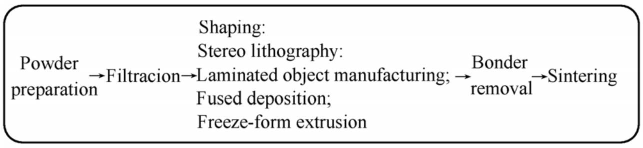
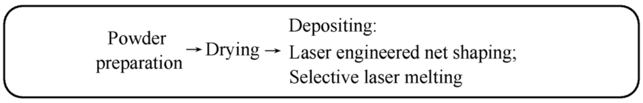


(a) Traditional methods



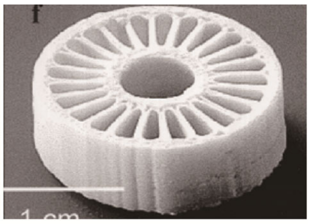
(b) Indirect AM methods



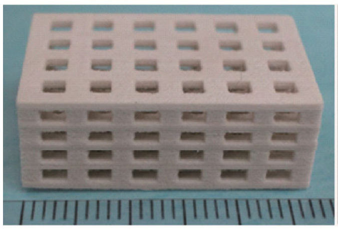
(c) Direct AM methods

Fig. 1. Ceramic fabricating methods

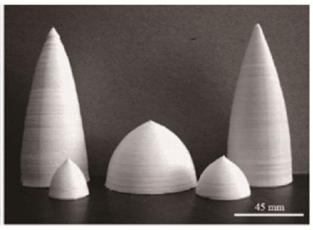
(a) Selective laser sintering(SLS) [5]



(b) Fused deposition(FD) [6]



(c) Selective laser gelling(SLG) [7]



(d) Freeze-form extrusion fabrication(FEF) [8]

Fig. 2. Ceramic structures fabricated by indirect AM methods

On the other hand, by the direct method, as shown in Fig. 1(c), pure ceramic powders can be melted directly by a high energy source without filtration or bonding and then the melt pool solidifies and deposits layer by layer to form certain structure. Due to the melting-solidification processes, fully densified ceramic structures with good performances could be produced more easily and rapidly. Because of the need for a high energy source, few direct processing methods have been developed. Laser engineered net shaping(LENS) and selective laser melting(SLM) are the only two methods which have been reported for direct fabricating ceramic structures in the last decade[10–17]. WILKES, et al[10–11], manufactured ceramic structures from ZrO_2/Al_2O_3 powders with an SLM system and obtained crack-free specimens with flexural strength higher than 500 MPa. BALLA, et al[12–13], used an LENS system to fabricate dense and net-shaped structures of Al_2O_3 and obtained cylindrical, cubic and gear-shaped structures which showed microstructural anisotropy with hardness of 1550 HV. BERTRAND, et al[14–15], applied SLM to manufacture net shaped parts from pure yttria-zirconia powders and demonstrated possibility of processing pure ceramic powders by SLM without doping. JUSTE, et al[16], added colloidal graphite within the alumina ceramic granules to increase the optical coupling between the laser beam and alumina during SLM. Large components with complex shapes and relative densities up to 90% were manufactured. LIU, et al[17], demonstrated the possibility of melting YSZ ceramic powder with IR fibre laser and obtained cubic samples with 88% relative density. The micro-hardness of fabricated cubic could reach 1209 ± 262 HV.

Although the direct AM methods perform well in properties and fabrication efficiency, there are also some technical challenges, such as shrinkage cavity or cracks control in the laser-aided processes due to the hard and brittle nature of ceramic material as well as large thermal gradients generated during laser radiation. Preheating was proposed by Wilkes and has been proven to be an effective method for suppressing cracks generated by the SLM process[10–11]. However, this method provided a limited preheating area and poor surface quality. Because the direct AM methods of ceramic structures have been developed in a relatively short time and only a few kinds of ceramics have been studied, they need to be further developed for industrial applications.

This paper applies LENS system to directly fabricate single-bead wall structures from pure Al_2O_3 powders firstly. Cracks characteristic, phase composition, microstructure as well as micro-hardness of the fabricated Al_2O_3 structures are then analyzed by XRD, SEM and Vickers respectively. Based on the analysis of Al_2O_3 deposition, two cracks suppressing methods, grain refinement strengthening and dispersion strengthening are proposed. Mixed powders of ZrO_2/Al_2O_3 and SiC/Al_2O_3 are then deposited respectively to verify the proposed methods for suppressing cracks.

2 Experimental Methods

The AM experiments were conducted using a LENS system as shown in Fig. 3. The system mainly includes a 1000 W continuous-wave Nd:YAG laser, a 3-container powder feeder with a coaxial nozzle, and a 3-axial NC machine. Pure argon is used to send ceramic powders to the coaxial nozzle and also used as a shielding gas for separating the fabricated part from atmosphere. The fabricated parts were deposited on a Ti-6Al-4V substrate of 6 mm thickness because of their good compatibility and the high laser absorptivity of the Ti-6Al-4V. For an efficient use of the sent powders, the nozzle was set on a certain position away from the substrate so as to make the focus powder stream accordance with the substrate surface.

Pure Al_2O_3, ZrO_2 and SiC ceramic powders of particle sizes in the range of 42–90 μm were used for all the direct fabricating experiments after drying at 100 ℃ for 4 h to eliminate moisture. The three ceramic powders were loaded into the respective different containers of the powder feeder.

Flow rate of the powders was independently controlled. Fabrication of pure Al_2O_3 ceramic single-bead wall structure of 15 mm length was conducted for fundamental research. During the deposition experiment, the coupled nozzle and laser assembly moved back and forth in the *x*-*y* plane to perform ceramic deposition. After each layer depositing, the substrate together with the already deposited layers moved away from the nozzle by one layer thickness along the *z* direction so as to maintain a constant focus position for depositing the next layer. The process was then repeated to finish the single-bead wall(as shown in Fig. 4(a)). Afterward, two phase ceramics Al_2O_3/ZrO_2 and Al_2O_3/SiC with different mixing ratios were then fabricated by the same deposition mode to verify their effects on suppressing cracks.

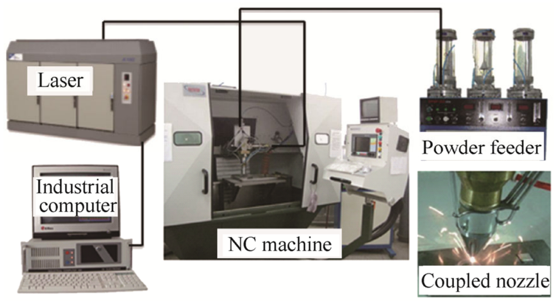


Fig. 3. LENS system

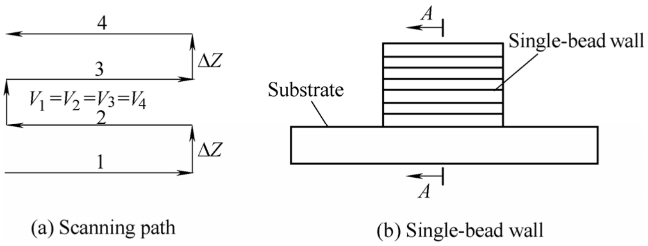


Fig. 4. Process of LENS and schematic of fabricated specimen

The fabricated ceramic parts were cut from the longitudinal cross-section A-A, as shown in Fig. 4(b), and then prepared by coating a thin Au to observe the microstructure in a SEM. For the Al_2O_3/ZrO_2 and Al_2O_3/YAG ceramic parts, their cross-sections were polished by a diamond disk and abrasive papers before being coated with Au. Phase compositions were analyzed by XRD and EDS. Vickers micro hardness measurements were made on the polished samples using a 1000 g load for 15 seconds, and an average value of 10 measurements on each sample was reported.

3 Results and Discussion

Fig. 5(a) shows a 50-layer single-bead wall structure of pure Al_2O_3. Vertical cracks along the deposition direction can be observed both at the two sides and middle of the wall, while there is no horizontal crack generating. Systematic experiments indicate that it is very hard to fabricate single-bead wall structure without cracks under the existing processing conditions. This may result from the intrinsic brittleness of Al_2O_3 ceramic and the extreme thermal gradients generated during the melting-solidification process[18]. Fig. 5(b) shows the micro-structure characteristics of the Al_2O_3 wall structure. The fabricated ceramic part is comprised of dense directional columnar crystals along the deposition direction with an intergranular space of about 10–15 μm. This directional crystal growth feature arises from directional heat dissipation through the colder metal substrate[19]. It is easy for cracks to propagate along the crystal boundaries in the deposition direction, whereas it may be much harder to propagate along the scanning direction because more energy is needed for crack propagation through a crystal. Consequently, it is more preferable for a crack to form and propagate in the deposition direction than in the other directions. For the above reasons, vertical cracks are preferably generated.

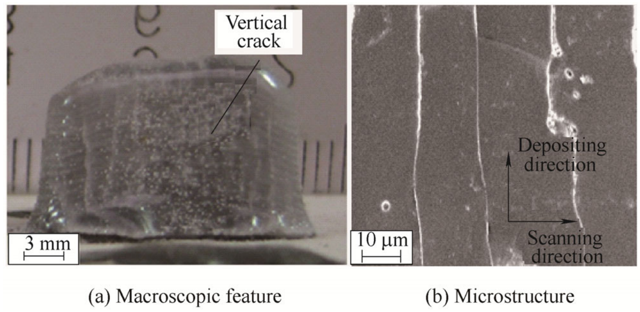


Fig. 5. Single-bead wall structure of Al_2O_3

From the XRD result in Fig. 6, it can be seen that the composition of the fabricated Al_2O_3 ceramic part is the stable phase α-Al_2O_3, which means solid-state phase transformation from γ- Al_2O_3 to α-Al_2O_3 has not happened for the material during solidification like the traditional fabricating methods.

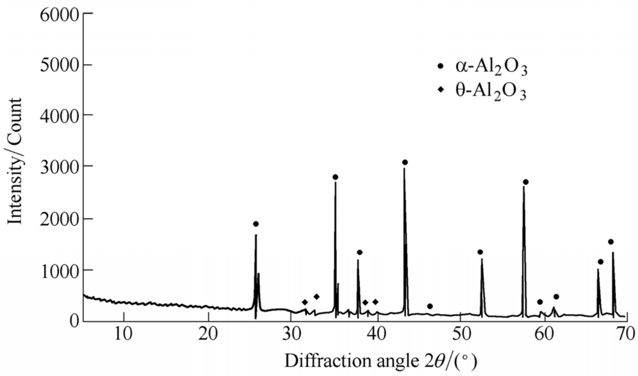


Fig. 6. XRD results of Al_2O_3 ceramic

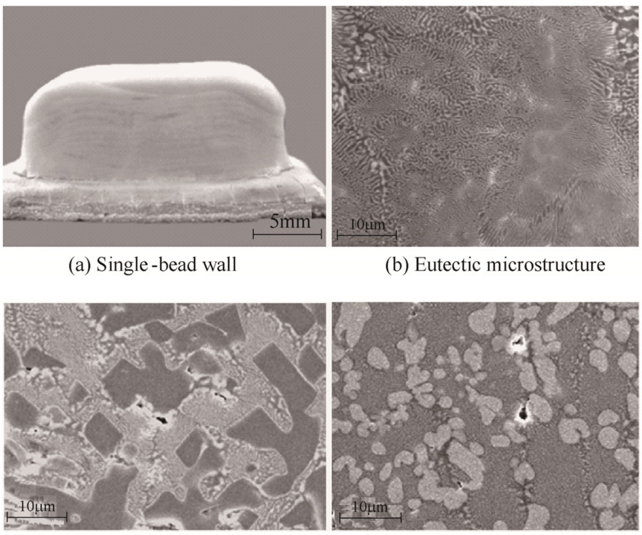
The deposited Al_2O_3 parts show high average micro-hardness 1800 HV, while traditional Al_2O_3 ceramic is only 1600 Hv. The high result essentially rest with the corundum structure of α-Al_2O_3 and its ionic(63%) and covalent(37%) bonding energies[20]. Compared with traditional solid phase sintering method, the melting- solidification process of

LENS in this study usually generates denser microstructure(as shown in Fig. 5(b)), which results in higher micro-hardness.

In order to suppress cracks generated during the fabrication process, different ratios of ZrO_2 was doped into the Al_2O_3 powders for deposition. 40 wt.%, 58 wt.% (eutectic ratio) and 70 wt.% ZrO_2 were mixed with Al_2O_3 powders and deposited respectively. Experiments results show that crack free single-bead wall parts can be successfully deposited by all the three mixed ZrO_2/Al_2O_3 (Fig. 7 (a)).

Typical microstructure of the fabricated part is shown in Fig. 7(b), from which we can see that nano-sized eutectics is formed due to the melting-solidification process of material with eutectic ratio[21]. The microstructure is comprised of a light phase and a dark phase. According to the EDS and XRD results, the light phase can be identified as tetragonal ZrO_2(t-ZrO_2) and the dark phase is α-Al_2O_3 which both are stable phase at normal atmospheric or high temperature(as shown in Fig. 8)[21]. The lamellar microstructure is the main body of the Al_2O_3-ZrO_2(Y_2O_3) eutectic ceramic, and accordingly a decisive aspect in determining the mechanical properties of the fabricated specimens. Around the colony are coarse granular microstructure formed by light disordered t-ZrO_2 embedding in the dark Al_2O_3 matrix. In this study, the eutectic spacing is about 100 nm(seen as Fig. 8(b)), which is finer than other same eutectics prepared by Bridgman[22], LFZ[23–25], EFG[26] and μ-PD[27–28]. This finer eutectic spacing may be a result of the high thermal gradient near the molten pool. On the other hand, conduction through the substrate is the dominated heat release mode, which result in the lamellar microstructure grows along the deposition direction.

On the other hand, Hypoeutectic and hypereutectic microstructure are formed respectively by depositing of 40 wt.% ZrO_2/60 wt.%Al_2O_3 and 70 wt.% ZrO_2/30 wt.%Al_2O_3. Little contraction cavities can be observed in them from Figs. 7(c) and (d)[29].



(a) Single-bead wall (b) Eutectic microstructure

(c) Hypoeutectic microstructure (d) Hypereutectic microstructure

Fig. 7. Specimen and microstructure of fabricated ZrO_2/Al_2O_3

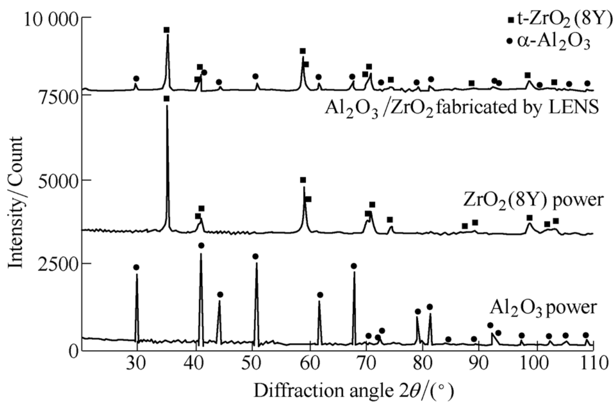
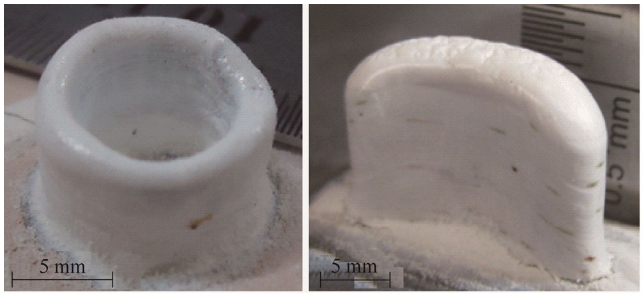


Fig. 8. XRD results of Al_2O_3-ZrO_2 (Y_2O_3) eutectic ceramic[23]

Similar to fabricated Al_2O_3, the deposited eutectic ZrO_2/Al_2O_3 ceramic also appears higher average micro-hardness(1715 HV) than the same ceramic fabricated by traditional solid phase sintering method(1200–1460 HV). The increment may be resulted from the finer microstructure formed by direct melting-solidification process of LENS[29].

Doping of ZrO_2 into the Al_2O_3 powders is proved as an effective method for refining microstructure and suppressing cracks during the LENS. Geometries of arc and cylinder ring without any cracks were successfully fabricated as shown in Fig. 9.

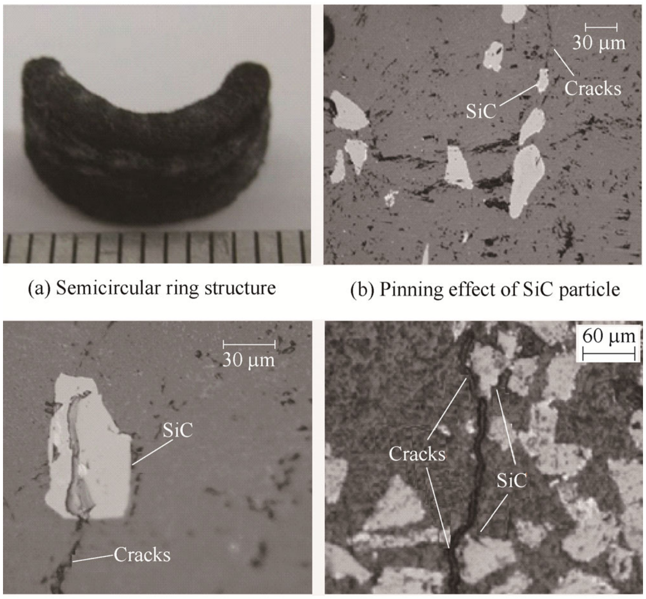


(a) Cylinder ring (b) Arc wall

Fig. 9. Al_2O_3-ZrO_2 (Y_2O_3) eutectic ceramic structures fabricated by LENS

Another method for suppressing cracks during LENS of ceramic structures is dispersion strengthening. In this study, dystectic SiC particles were doped into Al_2O_3 powders for depositing experiments. Through systematic optimization of process parameters, deposition by the mixed ratio of 10 wt.% SiC/90 wt. % Al_2O_3 shows fewer cracks than other mixed ratios[30]. Fig. 10(a) is a semicircular ring without any visible cracks deposited by 10 wt.% SiC/90 wt. % Al_2O_3.

From microstructure of the fabricated SiC/Al_2O_3 structures, it can be seen that the SiC particles are not melt totally, but dispersed in the Al_2O_3 matrix. This is because the melting point of SiC (3000 ℃) is obviously higher than that of Al_2O_3 (2050 ℃), which allows the laser beam to melt Al_2O_3 only. On the other hand, the atomic binding strength of SiC is also higher than Al_2O_3. Therefore, when a propagating crack encounters the SiC particles, more propagation energy will be consumed, and consequently the cracks will change direction or be restrained, as shown in Figs. 10(b), (c) and (d) [30].



(a) Semicircular ring structure (b) Pinning effect of SiC particle

(c) Trans-granular fracture (d) Crack deflection

Fig. 10. Specimen and microstructure of fabricated SiC/Al_2O_3

4 Conclusions

(1) Direct AM of Al_2O_3 by LENS is demonstrated as an effective method for fabricating high-performance ceramic structures, but cracks are hard to solve under the routine deposition conditions due to intrinsic brittleness of Al_2O_3 ceramic and the extreme thermal gradients generated during the melting-solidification process.

(2) Grain refinement strengthening method by doping ZrO_2 and dispersion strengthening method by doping SiC are verified by fabricating single-bead wall structure and proved as effective crack suppressing methods in direct AM ceramic.

(3) Crack-free geometries of arc, cylinder ring as well as semicircular ring are successfully achieved. The fabricated specimens all show dense microstructures and according high micro-hardness.

(4) XRD results indicate that the deposited ceramics are all composed of stable phases.

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