Study on Preparation, SLS and Post-Process of ZrO Coated Powder

Xia, Sijie; Liu, Kai

2014

Xia, S., & Liu, K. (2014). Study on Preparation, SLS and Post-Process of ZrO Coated Powder. Proceedings of the 1st International Conference on Progress in Additive Manufacturing (Pro-AM 2014), 202-206.

<https://hdl.handle.net/10356/84405>

https://doi.org/10.3850/978-981-09-0446-3_118

© 2014 by Research Publishing Services.

Downloaded on 26 Apr 2023 18:11:04 SGT

STUDY ON PREPARATION, SLS AND POST-PROCESS OF ZrO₂ COATED POWDER

XIA SIJIE

*School of Material Science and Engineering, Huazhong University of Science and Technology,
School of Material Science and Engineering
Wuhan, 430074, China*

LIU KAI

*State Key Laboratory of Material Processing and Die&Mould Technology, School of Material
Science and Engineering
Wuhan, 430074, China*

ABSTRACT: A novel method named dissolution-precipitation process was used to prepare zirconia (ZrO₂) ceramic composite powder. Nylon 12 was used as a binder to be coated on the surface of ZrO₂ powder. The coated powder obtained by dissolution-precipitation process is more uniform and suitable for selective laser sintering. SLS optimized process parameters were determined as follows: laser power 5.5W, scanning speed 2000mm/s, scanning space 0.1mm, and layer thickness 0.1mm. The law of properties of SLS parts with pressure of cold isostatic pressure was studied. The reasonable post-process of cold isostatic pressing/debinding/furnace sintering was carried out. Bending strength and relative density of SLS parts were greatly enhanced after the post-process.

Keywords: Zirconia; Composite; Selective laser sintering; Bending strength; Relative density

INTRODUCTION

Selective Laser Sintering (SLS) is a kind of Additive Manufacturing (AM) technology which is mature. Rapid manufacturing of complicated shape ceramic parts by selective laser sintering has broad application prospects. Zirconia (ZrO₂) ceramic is inorganic non-metallic material of high temperature resistance, wear resistance and corrosion resistance which will meet more and more particular demands in fields such as aerospace, oil, etc. But it is hard for traditional methods to shape it, especially a part with complex shape because of its high melting point and hardness.

AM is regarded as the primary ceramic fabricating technology in future, S. Blackburn (2005). SLS of ceramic parts can be divided to direct and indirect. Y. Hagedorn et al. (2010) tried to directly melt Al₂O₃-ZrO₂ powder by Selective Laser Melting (SLM) technique to get full dense Al₂O₃-ZrO₂ parts, and a high preheating temperature of above 1600°C was required to avoid ceramic cracks. Although it worked, the surface roughness and accuracy were very poor, and the height of parts was limited to 3 mm. J. Ebert et al. (2009) used three-dimensional printing (3DP) to fabricate dental prosthesis of zirconia ceramics. This study demonstrated the feasibility of building up dense three-dimensional parts of a dental crown out of high viscosity zirconia slurry by 3DP. But it was impossible to shape complex parts with an internal structure and its surface accuracy was also very poor.

To solve these problems, indirect SLS was adopted to form green bodies by melting polymer binders in composite powder. Fig.1 shows the schematic illustration of the SLS system. Composite powder can be prepared by mechanical mixing, spray drying or thermally induced phase

separation (TIPS) technology, Khuram Shahzad et al. (2012). Nealkent et al. (2004) proved that the green bodies obtained by the polymer coated ceramic powder had higher strength than those just mixed two different kinds of powders mechanically; they also had better forming precision and mechanical properties of final products. Because the green bodies obtained by the coated powder had more uniform distribution of ceramic particle and binder, the shrinkage deformation in post-process was relatively small and the microstructure of the parts was more uniform. Khuram Shahzad et al. used TIPS to produce spherical polypropylene–zirconia composite powder for SLS. Pressure infiltration and warm isostatic pressing were applied to increase the green density of the ZrO₂–PP SLSed parts. However, some parts produced by PP-ZrO₂ were very fragile to be post-processed, Khuram Shahzad et al. (2014).

Figure 1. Schematic illustration of the SLS system

EXPERIMENTAL DETAILS

Powder preparation

We used dissolution-precipitation method process to prepare ZrO₂-PA composite powder. Nylon is a kind of anti-solvent resin, it is difficult to dissolve in ordinary solvents under the condition of normal temperature, but it could dissolve in a suitable solvent at high temperature. So we chose ethanol as solvent, adding the nylon powder, antioxidant and ZrO₂ powder in it. The nylon dissolved at high temperature. Then during gradual cooling process, the nylon will preferentially precipitated in the surface of ZrO₂ powder because of the heterogeneous nucleation mechanism, so coated ZrO₂ powder was prepared.

Because the polymer powder has large specific surface area, it is prone to oxidative degradation in SLS process, resulting in poor performance, so it is necessary to add antioxidant to reduce oxidative degradation during SLS forming process and aging in the use of the part. The silane coupling agent (APTS) was used to improve the adhesive strength and durability of nylon 12 and

zirconia surface. The zirconia surface treatment by silane coupling agent adds compatibility in zirconia and nylon 12, improves the interface bonding between zirconia and nylon 12 so as to facilitate the zirconia dispersed uniformly in solvent precipitation process, Chunze Yan et al. (2009).

Specific process of dissolution-precipitation method was carried out as follows: (1) ZrO_2 powder ($\sim 80\mu\text{m}$, 5 mol% yttria-stabilized, NanBo, China) surface treatment by APTS. Then the treated ZrO_2 powder, nylon 12 (PA12, Degussa), antioxidant 1098 (0.2 wt.% of composite powder) were put into the stainless steel reaction kettle (10L) with sufficient solvent, sealed the reaction kettle. The solvent was ethanol (analytical pure); (2) The reaction kettle temperature gradually rises to 142°C at a speed of $1\sim 2^\circ\text{C}/\text{min}$, mechanical stirring at the same time, the nylon completely dissolved in the solvent, and ZrO_2 is dispersed in the alcohol solution of PA12, temperature and pressure were held for 1h ~ 2h; (3) The reaction kettle temperature was set to cool to room temperature slowly, the nylon gradually to ZrO_2 powder was coated on the surface of nucleation, crystallization, the formation of nylon coated ZrO_2 powder suspension. (4) The coated powder suspension was removed from the reaction kettle, it would precipitate naturally. We got the composite powder aggregates, after the ethanol was recovered. About 90% of ethanol solvent can be recycled. (5) The aggregates obtained was put in a vacuum drying box, after that, the blocked dry powder was sieved (150 mesh) after grinding. Then the PA/ ZrO_2 composite powder materials needed in the experiment was obtained. The coated ZrO_2 powder with 20 wt.% nylon content was prepared in this experiment. Fig.2 is micrograph took by SEM (JSM-7600F, Japan).

Figure 2. SEM micrograph of the ZrO_2 powder (a) and coated ZrO_2 powder (b)

SLS and post-process

The machine used for SLS process was a HRPS-IV SLS system made by Huazhong University of Science and Technology (HUST) in China, equipped with a 55 W CO_2 laser with a wavelength of $10.6\ \mu\text{m}$ and a laser beam diameter of $300\mu\text{m}$. Considering the forming efficiency and cost factors, the optimized SLS process parameters were selected as follows: preheating temperature 130°C , laser power 5.5W, scanning speed 2000mm/s , scanning space 0.1mm , and layer thickness 0.1mm .

After SLS process, post-process of the green part including cold isostatic pressing, debinding and furnace sintering was carried out to improve the performance of the final part. Isostatic pressing

refers to take the powder or specimen in a plastic airtight set, and put the set in liquid. Uniform forming of the ceramic green body is achieved by the liquid pressure with characteristics of uniform pressure in all directions, isostatic pressing method used for molding specimens at room temperature is called cold isostatic pressing (CIP).

The purpose of debinding is to remove the binder in the green body, preparing for the furnace sintering, if debinding is incomplete, the residual binder will become the impurity of the part and reduce strength, mechanical properties of it. Debinding was carried out in a muffle furnace. According to the thermal gravity (TG) curve of PA12, the heating rates were set, respectively, as 4°C/min, 1.6°C/min and 4°C/min before 250°C, 250°C~350°C and 350~470°C. Furthermore, it was held for 1h at 350°C, 2h at 470°C and 1h at 1100°C. Finally, it was allowed to cool naturally, Chunze Yan et al. (2009).

Furnace sintering (FS) is designed to make mechanical engagement by pressing into grain boundary atomic bonding between powder particles. So debinded parts were sintered in silicon molybdenum rod sintering furnace from room temperature to 1500°C and held for 2 h.

RESULTS AND DISCUSSION

The law of relative density and bending strength of SLS parts with pressure of cold isostatic pressure was studied, and the part properties of before and after post-process under different pressure were compared. We used molding instead of cold isostatic pressing to find the law, because the trend is similar. Cylinder specimens (d25mm×5mm) and cuboid specimens (30mm×7mm×7mm) were molded for testing relative density and bending strength respectively. Table 1 is relative density comparison, as the molding pressure increases, specimen relative densities are increased, and the density after post-process is much higher than before. It shows that high pressure is favor densification of zirconia ceramics. Table 2 is bending strength comparison and the bending strength after post-process is far greater than before. So the part properties greatly improved after post-process.

Table 1. Relative density comparison before and after post-process under different pressure.

Molding pressure /MPa	Relative density / %	
	Before post-process	After post-process
167	55	96
233	56	97
300	57	98

Table 2. Bending strength comparison before and after post-process under different pressure.

Molding pressure /MPa	Bending strength / MPa	
	Before post-process	After post-process
100	6.19	196.00
133	6.58	142.69
167	7.67	136.56

CONCLUSION

The dissolution-precipitation method process was used to prepare ZrO₂-PA composite powder. The coated powder obtained by dissolution-precipitation process is more uniform and suitable for SLS. SLS process parameters were optimized to fabricate. The SLS green body had the best quality when laser power, scanning speed, scanning space and layer thickness were 5.5W, 2000 mm/s, 0.1mm and 0.1mm respectively. The law of bending strength and relative density of SLS parts with pressure before and after post-process was studied. Properties of SLS parts were greatly enhanced after post-process. This research had developed a promising approach to fabricate high density ceramic parts with complex shape via indirect SLS/CIP/FS processes.

ACKNOWLEDGMENTS

This study is financially supported by the National Natural Science Foundation of China (Grant No. 51109087), National Key Technology R&D Program of the Ministry of Science and Technology (2012BAF08B03).

REFERENCES

- Chunze Yan, Yusheng Shi, Jingsong Yang & Jinhui Liu (2009). "Preparation and selective laser sintering of nylon-12 coated metal powders and post processing". *Journal of Materials Processing Technology*, 209, 5785-5792.
- HAN Zhao, CAO Wen-Bin, LIN Zhi-Ming, LI Jiang-Tao & FENG Tao (2004). "Progress on Rapid Prototyping of ceramic parts by Selective Laser Sintering". *Journal of Inorganic Materials*, 19(4), 705-713.
- J. Ebert, E. Ozkol, A. Zeichner, K. Uibel, O. Weiss, U. Koops, R. Telle & H. Fischer (2009). "Direct inkjet printing of dental prostheses made of zirconia". *Journal of Dental Research*, 88(7), 673-676.
- Khuram Shahzad, Jan Deckers, Stijn Boury, Bram Neirinck, Jean-Pierre Kruth & Jef Vleugels (2012). "Preparation and indirect selective laser sintering of alumina/PA microspheres". *Ceramics International*, 38, 1241-1247.
- Khuram Shahzad, Jan Deckers, Zhongying Zhang, Jean-Pierre Kruth & Jef Vleugels (2014). "Additive manufacturing of zirconia parts by indirect selective laser sintering". *Journal of the European Ceramic Society*, 34, 81-89.
- S. Blackburn (2005). "New processes or old: complex shape processing of advanced ceramics". *Advances in Applied Ceramics*, 104(3), 97-102.
- Y. Hagedorn, J. Wilkes, W. Meiners, K. Wissenbach & R. Poprawe (2010). "Net shaped high performance oxide ceramic parts by selective laser melting". *Physics Procedia*, 5, 587-594.