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MULLITE CERAMIC FABRICATION BY 3D PRINTING

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ABSTRACT
This study is the mullite (porcelainite) specimens for mechanical properties evaluation fabricated by additive manufacturing technology. A slurry-based and selective laser sintering (SLS) 3D printing (3DP) machine was designed and developed to create these ceramic work pieces. Alumina powder, silica powder and silica sol was blended in adequate wt% to prepare the slurry. The slurry was paved layer by layer in the 3D Printer and laser selectively radiated the slurry to form ceramic layer. Thus, the ceramic green parts were created and post heat treatment to synthesize mullite ceramic. The bending and shrinkage test, SEM surface microstructure of these mullite samples were examined. This SLS process needs only low laser power (< 20W) to build ceramic parts. Therefore, a distortion due to post sintering process is avoidable. Hence, this process can produce more precise ceramic prototypes. This process has potential to fabricate the inner complex ceramic components for industrial applications.

Keywords: 3D printing, ceramics, mullite, alumina, silica.

INTRODUCTION
Recently, the Alumina (Al_2O_3) and silica (SiO_2) are the two familiar and elementary compositions not only applied in conventional ceramic but also in advanced ceramic applications. Mixing Alumina and silica powder with appropriate wt % and heat treatment at specific temperature, a composite ceramic Mullite (porcelainite) can be form in two stoichiometric forms $3Al_2O_32SiO_2$ or $2Al_2O_3SiO_2$. The widely application of mullite is because of its favorable mechanical properties.

Traditionally, mullite can be synthesized in three main methods: a) sinter-mullite, b) fused-mullite and c) chemical-mullite (Hamano, 1986; Sacks, 1982; Schneider, 1994; Han, 2011; Saruhan, 1996). The conventional manufacturing process of mullite mostly is difficult to produce complex structure and free-form shape. In order to overcome these issues, the 3D Printing (3DP) also called rapid prototyping (RP) technologies are suitable for generating the porous and complex shape parts are easily to be built in this way (Okada, 1990; Lee, 1994).

3DP techniques offer completely methods to construct physical objects from three dimensional Computer Aided Design (CAD) data, it allows designers to quickly create original prototypes rather than two-dimensional images. General advantages are faster building speed, rapidly generating complex-shaped prototype from a concept.

In this paper, the slurry-base ceramic was used in the selective laser sintering (SLS) technology (Chang, 2015). The slurry was prepared by mixing alumina powder, silica powder and silica sol for laser printing green parts of complex ceramic parts. The home-developed 3D printer can create green part of ceramics (Lee, 2014). These green parts were post heat treatment at specific temperature and complex mullite workpieces can be produced.

MATERIALS

For fabricating a green part by 3D Printing machine and synthesize mullite by post heat treatment, the slurry-based materials must be prepared with well dispersed, suitable viscosity and appropriate mixture of silica and alumina. The ceramic slurry was constituted of alumina (Al_2O_3) powder, silica (SiO_2) powder and silica sol. In this study, a CO_2 laser is employed to sinter the ceramic slurry mixed with alumina powders and silica slurry in a proportion of 45: 55 wt.% for subsequent experiment. The content of alumina and silica is listed in Table 1.

Table 1 - Content of Alumina and silica

Content	Al_2O_3 powder	SiO_2 slurry
Ratio	45 wt.%	55 wt.%

SELECTIVE LASER SINTERING

When the ceramic slurry is scanned by a laser beam, the sintering effect occur, then the silica sol links together to bond the alumina and silica particles to form a solid body, while the other un-scanned portion remained in slurry state.

According to above method, an SLS process was developed for forming a ceramic green part. The process was carried out as shown in Figure 1, this process involves following steps: (a) the alumina powder, silica powder and silica sol are blended as a raw material in ceramic slurry, (b) the slurry is paved by a scraper onto the elevator to from a slurry thin layer, (c) the paved slurry layer is selective sintered via a CO_2 laser beam according to the sliced 2D patterns from a 3D CAD mold, (d) the working platform is lowered to the thickness of one layer, (e) repeating steps (b) to (d), a multi-layer 3D ceramic part is obtained (Lee, 2014).

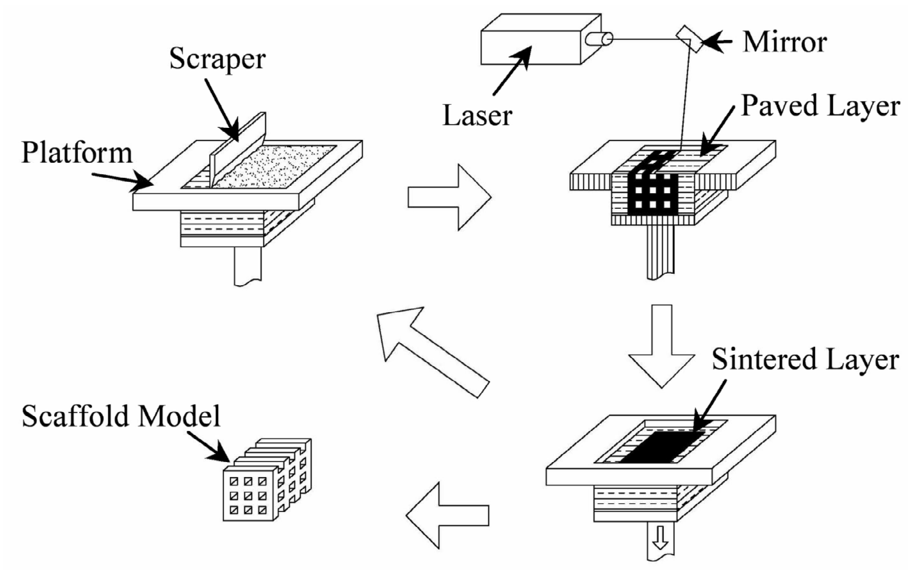

Fig. 1 - Schematic of a ceramic manufacturing processes of selective laser sintering.

EXPERIMENTAL RESULTS

LASER PARAMETERS

In this work, sintering the alumina-silica ceramic slurry needs only low laser power below 20 W. The laser power was adjusted from 5 to 20 W to build three specimens with a size of 25 mm×25 mm×5 mm. The laser power against the laser scan speed is indicated in Figure 2. When the laser power was less than 5 W, the sintered structures in the slurries were difficult to form due to insufficient laser power density for solidifying alumina-silica ceramic slurry. As laser power was increased from 5 to 20 W, the laser scan speed increased from 50 to 200 mm/min. That is the laser power increased with laser scanning speed. When the laser power exceeded 20 W, the solidified ceramic layer could not be formed because the laser power density was too large for building ceramic layer.

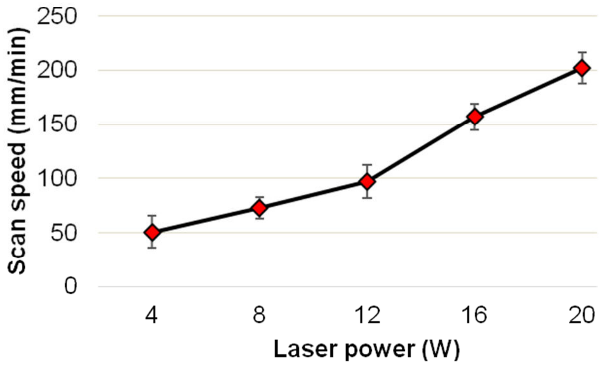

Fig. 2 - Laser power vs. laser scanning speed for forming the mullite ceramic specimens.

From our experimental results, the ceramic slurry can acts as a support material. Therefore, a ceramic part with inner channel and porous structure could be built shown in Figure 3. The bottom of the inner channel has no sagged deflection due to the support effect of ceramic slurry.

In order to present the advantage of the ceramic slurry, a mullite ceramic part with inner channel structure which has a rectangular hole was produced shown in Figure 3. The size of the ceramic is 8 mm×8 mm×4 mm, the length of the overhanging is 2 mm, the width of the rectangular hole is 2 mm. The inner channel structure of ceramic is hard to be built by using machining processes. The process parameters for building the inner channel structure are as follows: a laser power of 12 W, a laser scan speed of 150mm/s, a laser scan hatch of 0.15 mm, a layer thickness of 0.1mm.

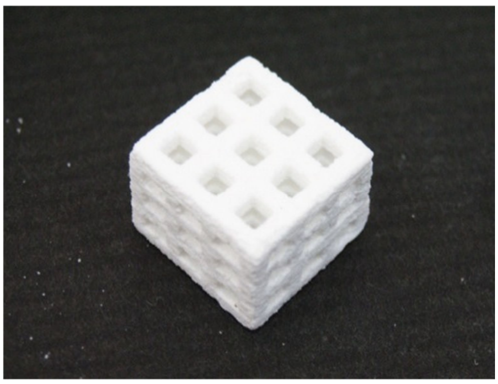

Fig. 3 - Photo-micrograph of a mullite ceramic part with inner channel structure.

MECHANICAL PROPERTIES

Alumina and silica slurry was used in a 3D printer to create specimens of green part (35 mm×4 mm×3 mm). These green parts were applied post heat treatment at 1200, 1300, 1400, 1500, and 1600 ºC to investigate mechanical properties. The shrinkage of mullite ceramic at different sintering temperature was displayed on Fig. 4. As treatment temperature was increased from 1200 to 1600 ºC, the shrinkage of alumina-silica ceramic increased from 0.6 to 2.0 %.

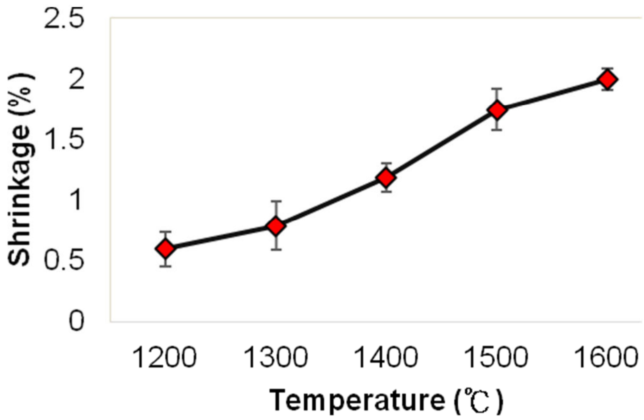

Fig. 4 Shrinkage of mullite ceramic at different sintering temperature

The strength of ceramic components is essential to be obtained in industrial applications. The test samples with a size of 45mm×4mm×3mm was fabricated and measured by the three point bend method. The bending strength of the tested samples is shown in Figure 5. When the treatment temperature is increased from 1200 to 1600ºC, the bending strength is ranged from 20 to 65 MPa. When the temperature equals 1500ºC, the bend strength has a maximum value of 65 Mpa. As temperature increases to 1600ºC, the bend strength reduces to 55 Mpa.

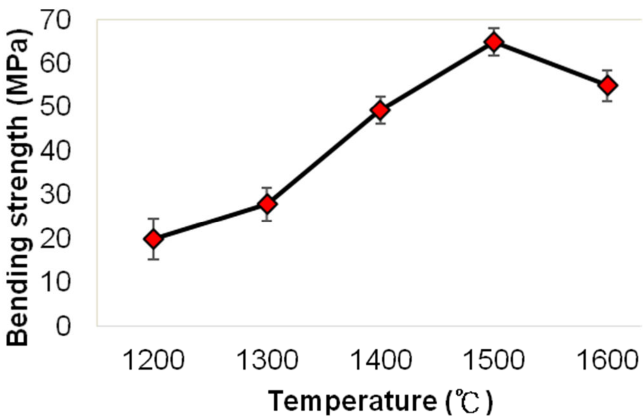

Fig. 5 The relation between bending strength and different sintering temperature.

SURFACE MORPHOLOGY

Surface Microstructure on these mullite ceramic samples were examined and analyzed by scanning electron microscope (SEM). Fig. 6 displays the SEM Micrographs (JEOL JSM 6500F) of the surface of specimen deposited by 3D printing.

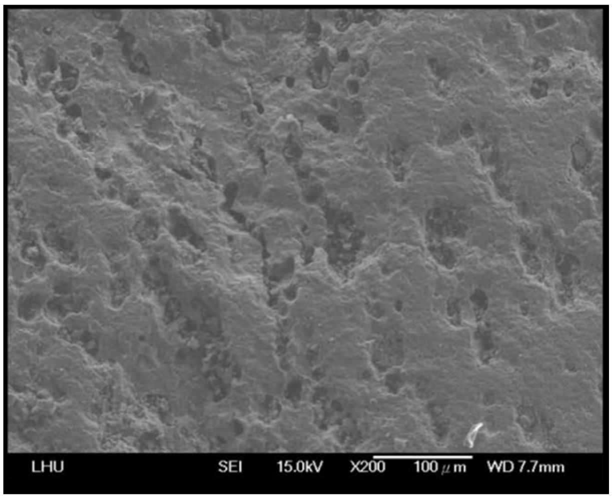

Fig. 6 - SEM Micrograph of a mullite ceramic sintered at 1600ºC

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The samples were sintered at 1300ºC, some crack networks was appeared on the surface and its bending strength was not good at all. Samples sintered at 1500ºC, all alumina and silica particles fuse together well but with some small pores. Sintering up to 1600ºC, the melting point of silica, some silica was melt, filled into void between particles and solidified as cooling and then more and bigger pores was produced inside the specimens. Because sample sintered at 1600ºC, alumina and silica particles fused together well but with some larger pores in the surface so that its bending strength was lower and porosity was bigger than those of sintered at 1500ºC.

CONCLUSIONS

A home developed 3D printer was succeeded to build mullite green parts using the slurry of blending alumina and silica powder and silica sol as binder. The better mechanical properties can be achieved by constituted of the alumina and silica powder mixed in 45:55 wt% with appropriate binder silica sol for 3D printing the green parts which then were heat treatment at 1500□. The bending strength and shrinkage can be reached 65 MPa and 2 % respectively. Furthermore, this laser sintering process needs only low laser power (< 20W) to build ceramic parts, the deflection and shrinkage of each ceramic layer can be decreased, also a distortion due to post sintering process is avoidable, Hence, this process can produce more precise ceramic parts. In particular, the inner channel parts. This process has potential to fabricate the inner complex ceramic components for industrial applications.

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