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Properties of selective laser melted spodumene glass-ceramic

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ABSTRACT

In this article, we established the process conditions and characterised the resulting properties of additively manufactured spodumene important for selective laser melting of LAS–Al_2O_3. X-ray diffraction analyses revealed the as-printed samples printed with layer thickness of 50 μm were fully crystalline. Energy dispersive X-ray spectroscopy showed that the major elements before and after the printing process were present and in similar quantity. Micro-computerised tomography inspection also revealed layer thickness-dependent pore formation in all printed samples. In terms of mechanical properties, the highest flexural strength measured using the three-point bend test method was 4.33 MPa. More importantly, these results demonstrated that there is still potential in the direct laser melting of ceramics.

1. Introduction

Recent technological advancements have encouraged and allowed industries to complement existing manufacturing processes with additive manufacturing (AM). American Society for Testing and Materials (ASTM) F2792 defines AM as a manufacturing technique in which a three-dimensional (3D) part is created by adding materials in a layerwise fashion [1]. A unique advantage of AM is the substantially reduced amount of time and costs needed for creating near net-shape or functional end parts [2]. The AM method this study focuses on is selective laser melting (SLM) – a powder bed-based AM technique mainly used for processing metals [3–5]. As compared to selective laser sintering (SLS), this process uses a laser beam to fully melt the powder particles [6]. The advantage of this method is the ability to produce a relatively full dense part from fully melted particles [7,8]. However, the limited material range available for SLM has slowed down the progress of this technology despite its rapid adoption and implementation in industries. For instance, direct laser melting of ceramics remains to be a persistent challenge today.

Near net-shape fabrication requirement and high production costs further affirmed the importance of AM of ceramics nonetheless. According to Travitzky [9], the necessity of advanced ceramics is primarily dependent on the industrial demands for high-performance applications, such as that in the aerospace industry. For instance, ceramics surpass metals in withstanding higher temperatures and resisting corrosion [10]. However, the widespread usage of advanced ceramics is dependent on the ability to manufacture near net-shape 3D parts [11]. Current production of ceramic components involves machining of low-strength green components incurring high costs contributing up to 80% of manufacturing costs [12]. In addition, conventional production processes are not suitable for small-scale and prototype fabrication needed for redesign evaluation [13]. As such, it is important to improve existing methods to make AM of ceramics possible. Another challenge for direct printing of ceramics to be considered is part removal. The current method of removing built parts from the substrate plate is by electric wire cutting. However, most ceramics/glass-ceramics are not conductors of electricity. As such, optimal support structure design will be required to allow part removal without damaging the built parts. Gan and Wong [14] demonstrated that minimal material usage for support structures is possible, which is also potentially useful for ceramic printing.

In the area of SLM of ceramics, Wilkes, et al. [15] demonstrated that preheating the powder layer to > 1600 °C produced an Al_2O_3/ZrO_2 part with almost 100% relative density and flexural strength of > 500 MPa. Other studies on laser-based AM of binder-free ceramics include stereolithography (SLA) of Al_2O_3 by Chen, et al. [16] achieving sintered density of 98% and flexural strength of 476 MPa and direct selective laser sintering (SLS) by Gahler, et al. [17] achieving a low relative density of about 56%. In another laser sintering study, Regenfuss, et al. [18] reported achieving a relative

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density of >95% and flexural strength of 100 MPa for alumina-silica parts. In addition, Friedel, et al. [19] and Shahzad, et al. [20] have also shown AM's ability to fabricate complex ceramic parts by selective laser curing and indirect SLS.

In addition to AM of ceramics, there were also studies on laser AM of silica-based glasses and glass-ceramics. Glass-ceramics are attractive materials due to their low thermal expansion coefficient [21]. Due to the low expansion property, they are a potential candidate for dental applications [22]. Moreover, Chakraborty, et al. [23] reported that spodumene glass-ceramic is more durable in human body fluid. In one study, Gupta Manob, et al. [24] selectively laser melted amorphous lithium aluminosilicate (LAS) powder and obtained the relative density of approximately 60%. According to the results from this study, high energy densities caused preferential loss of silicon and oxygen. Furthermore, the printed glass darkened as a result of higher absorption caused by the formation of colour centres [25]. As opposed to using dry powder, Zocca, et al. [26] laser sintered LAS glass slurry. At appropriate process parameters, qualitative well-densified layers could be printed. While both studies reported amorphous structure after laser processing, none of them printed a proper 3D part for appropriate analysis. In another study of a similar material, F. Klocke, et al. [27] direct SLS borosilicate glass powder achieving a 3D part having a relative density of 54.4% after thermal post-processing. For low-cost material, J.L. Song, et al. [28] laser sintered silica sand to print patterns for casting. M. Fateri and A. Gebhardt [29] also reported a study on selective laser melted soda lime glass powder, obtaining parts with a high relative density of 93%. Furthermore, the surface hardness of the manufactured parts was similar to conventional fused silica. In another study, Gan and Wong [30] showed the possibility of producing continuous melt track from spodumene mineral through direct melting.

The attractiveness of lithium aluminosilicate (LAS) stems from its low coefficient of thermal expansion which gave it the thermal shock resistant property. Leveraging on this property allows the use of alumina in a temperature-fluctuating environment in the form of composite as reported in the following studies. Firstly, García-Moreno, et al. [31] demonstrated the favourable solid-state compatibility between Al_2O_3 and one of the phases of LAS (β-eucryptite) for designing of such new ultrastable material. Secondly, LAS–Al_2O_3 put under thermal shock conditions exhibited minimal strength deterioration as compared to their alumina counterpart although the overall strength was one-third lower [32]. Thirdly, a study by Latella, et al. [33] concluded that liquid-phase sintered (LPS) Al_2O_3 containing spodumene had properties similar to commercially available debased Al_2O_3 ceramics used for wear applications. Last but not least, García-Moreno, et al. [34] also demonstrated that spark plasma sintering of LAS–Al_2O_3 improves mechanical properties as compared to furnace sintered parts and can be used in oxidising conditions. Following these research works, we additively manufactured spodumene to establish the critical process conditions for SLM of LAS–Al_2O_3.

In this study, spodumene samples for the flexural test were additive manufactured using SLM and its properties are characterised before and after heat-treatment. From past studies on SLM of LAS glass-ceramics by Gupta Manob, et al. [24] and Zocca, et al. [26], a proper 3D part was not manufactured successfully. Furthermore, these studies used synthesised powder obtained from quenched glass melt containing Li_2O, Al_2O_3, SiO_2, and various nucleating agents like TiO_2 and ZrO_2 [35]. However, in a crystallisation and microstructural study of spodumene mineral, A. Nordmann and Y.B. Cheng [36] showed that it was possible to form LAS glass-ceramic parts by heat-treating spodumene mineral. Hence, this study aims to investigates the direct SLM of spodumene, followed by printability assessment and characterisation of material properties before and after post-process heat-treatment.

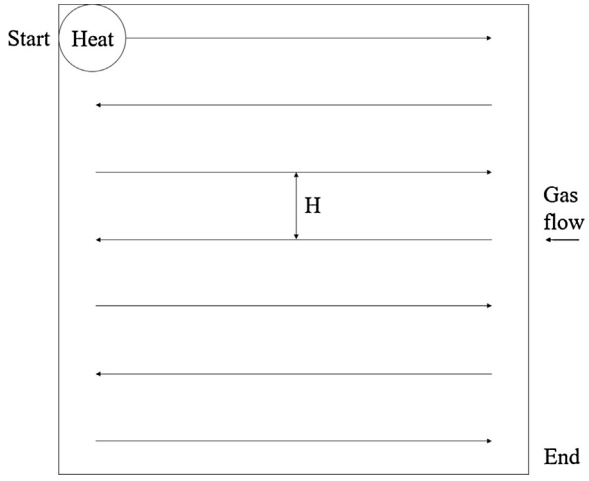

Fig. 1. Scan strategy used in this study.

2. Material and methods

2.1. Selective laser melting (SLM)

SLM of spodumene was carried out in a nitrogen gas-filled SLM machine equipped with a ytterbium fibre laser. In the chamber, a feed container deposits a layer of powder in the build area. Excess powder falls into the overflow and is recycled as the feed container moves across the build cylinder. The laser then heats the powder to temperatures exceeding the melting point to induce liquid phase fusion. The mirror scanner deflects the laser beam in the *x*-*y* axis to scan the cross-section corresponding to that layer. After scanning each layer, the build cylinder displaces by an amount equivalent to the layer thickness defined by the user (*z*-axis). A roller then spreads a new layer of powder in the build area and the process repeats. The scanning strategy used for this study is as shown in Fig. 1. The arrows indicate the path over which the laser moves and the hatch spacing (H) represents the distance between each path. In this study, the laser power and hatch spacing were kept constant. In addition, the scanning direction was parallel to the direction of the gas flow.

2.2. Sample preparation

The chemical composition of the spodumene powder used in this study is as shown in Table 1. Using the Ytterbium fibre laser equipped SLM machine, samples were built vertically along the *z*-axis (see Fig. 2) for two important reasons:

i) to reduce the risk of breakage during removal, and
ii) to measure the flexural strength along building direction.

Table 1
Composition of spodumene powder used.

Oxides	Wt%
Li_2O	7.1
Fe_2O_3	0.1
K_2O	0.25
Al_2O_3	25.4
SiO_2	65.7
P_2O_5	0.13
Na_2O	0.23
Loss on ignition	0.32

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