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The effect of spark plasma sintering on lithium disilicate glass-ceramics

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ABSTRACT

Objectives. To evaluate the effects of spark plasma sintering (SPS) on the microstructure of lithium disilicate glass-ceramics.

Methods. IPS e.max CAD glass-ceramic samples were processed using spark plasma sintering (SPS) and conventionally sintered (CS) as a comparison. Specimens were sintered at varying temperatures (T1: 840 °C, T2: 820 °C, T3: 800 °C), heating rates (HR1: 150 °C/min, HR2: 300 °C/min, HR3: 500 °C/min) and pressures (P1: 15 MPa, P2: 50 MPa, P3: 70 MPa). IPS e.max Press glass powder samples were densified at 750 and 800 °C (50 or 200 MPa pressure). Samples were characterized using XRD, HTXRD, and SEM and quantitative image analysis.

Results. There was a significant increase in median crystal size (MCS) between the CS and the SPS T1 groups. A statistical difference ($p > 0.05$) in MCS between SPS T1 and SPS T2 groups was observed. The SPS HR3 sample produced a smaller MCS than the CS, SPS HR1 and HR2 groups ($p < 0.05$). The SPS P3 sample had a reduction in MCS compared with the CS group ($p < 0.05$). XRD of the SPS samples revealed major lithium disilicate/lithium metasilicate phases and minor lithium orthophosphate and cristobalite/quartz phases. Densified IPS e.max Press glass samples resulted in fine fibrils or graduated lithium disilicate crystals.

Significance. The effects of SPS were used to refine the microstructure of IPS e.max CAD lithium disilicate glass-ceramics. Densification by SPS of IPS e.max Press glass resulted in textured and fine nano-crystalline microstructures. SPS generated glass-ceramic microstructures may have unique properties and could be useful in the production of CAD/CAM materials for dentistry.

1. Introduction

Lithium disilicate glass-ceramics are now well established for the construction of dental appliances using heat extrusion and computer-aided design/computer-aided machining (CAD–CAM). Depending on the manufacturing process these materials are produced as ingots (IPS e.max Press, IPS Empress

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Table 1 – SPS heat-treatment schedules for IPS e.max CAD glass-ceramics.

IPS e.max CAD	Pressure (MPa)	Heat rate (°C/min)	Temperature (°C)	Hold (min)
SPS (T1)	50	90	840	7.0
SPS (T2)	50	90	820	7.0
SPS (T3)	50	90	800	7.0
SPS (HR1)	50	150	840	7.0
SPS (HR2)	50	300	840	7.0
SPS (HR3)	50	500	840	7.0
SPS (P1)	15	90	840	7.0
SPS (P2)	50	90	840	3.0
SPS (P3)	70	90	840	7.0

T, temperature; HR, heat rate; *P*, pressure.

2, Ivoclar-Vivadent, Schaan, Liechtenstein) or blocks (IPS e.max CAD, Ivoclar-Vivadent). A colored pressure cast block used for CAD/CAM applications is produced in a partially crystallized state with a 40% lithium metasilicate phase (Li_2SiO_3), to allow ease of machining and reduced tooling costs. A second heat treatment is given following the milling procedure at 840–850 °C to allow the formation of 70% lithium disilicate crystals ($Li_2Si_2O_5$) [1]. The ingots for heat extrusion are in contrast produced by the crystallization of the base glass to form the final lithium disilicate glass-ceramic which can be heat extruded at 920 °C. This also imparts the material a high flexural strength and fracture toughness of 360 MPa and 2.5 MPa $m^{1/2}$ for IPS e.max CAD [1], and 440 MPa and 2.75 MPa $m^{1/2}$ for IPS e.max Press [2,3].

These glass-ceramics are however synthesized by crystallizing a base glass using nucleation and crystal growth heat treatment schedules in a conventional furnace. This process is limited by the typical maximum rate at which a furnace can be ramped (typically 5–30 °C/min) and is carried out at atmospheric pressure. Heat extrusion of these glass-ceramics is also conducted at very low pressures (3–4 × 10^5 Pa) compared with more modern processing techniques. These manufacturing restrictions potentially limit the extent and scope of the microstructural and property optimization of these materials.

Spark plasma sintering (SPS) is a consolidation technique used to rapidly produce fully dense ceramics/glasses with fine grained microstructures at low temperatures. The SPS process involves the compaction of a powder into a graphite mold followed by applying a pulsed DC current, under uniaxial pressure and at high heating rates (up to 1000 °C/min) [4]. These rapid heating rates make it possible to avoid undesirable grain growth at the lower sintering rates used in conventional techniques like hot pressing [5]. This has resulted in dramatic reductions in grain size (0.5–0.6 μm) when compared with conventional sintering (30 μm) of alumina ceramics [6,7]. The applied pressure in this process also acts as a driving force for sintering resulting in higher densities at lower temperatures [7,8]. It is also thought that during SPS the production of a plasma acts to clean the powders surface of impurities (absorbed CO_2 and H_2O), enhancing grain boundary diffusion processes and densification [9]. SPS of ceramic materials results in improved transparency [10], reduced porosity and increased flexural strength [11,12]. SPS has demonstrated some success in producing dense optical materials including oxygen sensitive chalcogenide glasses and glass-ceramics for infrared applications [13]. This processing technique may therefore be useful in the preparation of dental materials to tailor/optimize the optical and mechanical properties. There may also be the opportunity for greater control of the crystallization, giving greater latitude for more precise phase/crystal size control [14]. Faster and more efficient densification of fine grained CAD–CAM ceramic materials compared with existing methods could therefore be achieved. Fully optimized SPS ceramic materials produced as blocks could be integrated with CAM to allow more cost effective machining/processing and retaining unique material properties. There is little known in the literature on the effect of SPS on lithium silicate glass-ceramics and glasses used in dentistry, including the effects of changes in temperature, heating rate and pressure. The aims of the study were to evaluate the effects of spark plasma sintering (SPS) on the microstructure of lithium disilicate glass-ceramics.

2. Materials and methods

The materials used in the current study comprised lithium disilicate glass-ceramics/glasses used in dentistry to produce either CAD–CAM restorations (IPS e.max CAD, Inlab LT A3/C14, LOT: P37665, Ivoclar-Vivadent, Schaan, Liechtenstein) or heat extruded restorations (IPS e.max Press glass LT A2, LOT: 603221, Ivoclar-Vivadent). These materials were selected as they provided the opportunity to evaluate the effects of SPS on glass-ceramics in this system in the partially crystallized state (IPS e.max CAD) and on that of the starting glass (IPS e.max Press) before the crystallization heat treatments.

The IPS e.max CAD glass-ceramic was used for the spark plasma sintering experiment in powder form at varying pressures, temperatures, and hold durations (Table 1). To produce the powder the glass-ceramic block was cut using a diamond disc (Skillbond, UK) and ground in to a powder using an alumina mortar and pestle (Milton Brook 3, UK) and screened to 45 μm. The IPS e.max Press glass was received as a powder (D_{50} = 8.25 μm) from the manufacturer.

2.1. *Differential scanning calorimetry*

The IPS e.max CAD and IPS e.max Press powders were characterized using differential scanning calorimetry (DSC) using a Stanton Redcroft DSC 1500 (Rheometric Scientific, Epsom, UK). A bulk sample (IPS e.max CAD) was also run to help identify the type of crystallization (e.g. surface or bulk). Alumina was