Original Research

Processing Capability of Apatite-Mullite Glass-Ceramic Materials for the Production of Dental Restorations

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Abstract

Introduction: There is currently significant interest in all-ceramic dental restorations. A potential non-metallic material for such restorations is an apatite-mullite glass-ceramic that could be processed to shape using different processing techniques. The aim of this study was to assess and evaluate the ability of an apatite-mullite glass-ceramic material to be processed using three routes: lost-wax casting, hot pressing and CAD-CAM milling. **Methods**: A batch of glass for an apatite-mullite glass-ceramic material was produced based on the formula (4.5SiO2-3Al2O3-1.5P2O5-3CaO-0.5 CaF 2). The batch was converted into glass by heating at 1050˚C/1450˚C for two hours. The final melt was quenched to obtain a glass frit. The glass was thermally treated based on the DTA data. The sequence of crystallization and their micro structural evaluation were analysed using X-ray diffraction (XRD) and scanning electron microscopy (SEM). An anatomically correct mould of an upper right first molar was selected to trial the three different manufacturing techniques. Empress II and VITA block mark II materials were used as control materials. **Results**: The apatite-mullite material being evaluated can be cast and milled to shape, but the ceramic form of the material is not capable of being hot pressed. **Conclusion**: The materials tested show great possibility as restorative materials and could be heat treated inside and outside the investment casting material to produce a crystalline microstructure of apatite and apatite-mullite. It is possible to produce acceptable restorations using the milling technique. Hot pressing the material is not recommended due to its high liquidus temperature.

Keywords: Apatite-Mullite, Glass-Ceramic, Processing, CAD-CAM, Hot Pressing, Lost-Wax.

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Introduction

Teeth preservation is considered to be a growing demand in patients. Conventional metal-based restorations have been reported to be unsafe, and there are concerns regarding the health hazards associated with mercury in dental amalgam (1, 2). Furthermore, reports have suggested that the levels of mercury released from amalgam fillings may lead to the Alzheimer's disease (3).

Materials that could be used in dental applications have several requirements. For instance, they should be biologically compatible within the oral environment, safe for the patient and dentist, aesthetic, highly strong, and have fracture toughness. Moreover, these materials should not be susceptible to cause damage as this may result in crack formation and tooth wear. These materials should not dissolve, erode or corrode and should have physical properties similar to natural enamel and dentine, whilst also being capable of forming to shape to the required dimensional tolerances, easily and economically.

Currently, there is growing interest in all-ceramic dental restorations among dentists and patients since they have good aesthetics, low thermal conductivity, high strength, durability, biocompatibility, and relative ease of manufacturing. There are recent development in dental porcelains which have resulted in the use of glass-ceramics, which have been employed commercially as synthetic tooth filling materials and produce aesthetic metal-free dental restorations in dental prostheses (4).

Apatite-mullite glass-ceramic is a potential non-metallic material for metal-free dental restorations, which could be cast to shape in order to produce crowns or inlays using conventional dental metal-casting techniques. In addition, it could potentially overcome the disadvantages of the current dental ceramics.

Apatites are found naturally in metamorphic, igneous, and sedimentary earth rocks. Recently, various forms of hydroxyapatite have been discovered on the surface of the Moon (5). Furthermore, apatite is the

major inorganic component that is naturally found in the hard tissues of vertebrates; therefore, it has a biological and clinical significance.

Several bioactive apatite-containing glass-ceramics have been developed for orthopedic applications, which could be classified based on the type of the secondary crystal phases present in glass-ceramic, apatite-wollastonite (commercially known as Cerabone®), apatite-fluoromica (commercially known as Bioverit®), and apatite-mullite.

Since the early 1970s, several types of glass-ceramics have been developed, which could crystallize to apatite phases under controlled heat treatment. A glass-ceramic material could be cast into complex shapes through lost-wax casting, which is often a simple, cost-effective process. Moreover, glass-ceramics could be processed through computer-aided design/computer aided manufacturing (CAD/CAM) or hot pressing. Apatite-mullite glass-ceramics could be manufactured via selective laser sintering (6).

Extensive research has been conducted, or are currently underway, aiming to improve or develop new dental materials with enhanced properties that could be processed using advanced technologies, such as CAD/CAM or 3-D printing (7). For instance, Hill. R et al. (4, 8) developed a castable glass-ceramic in a study on glass ionomers. After ceramming heat treatment, the apatite-mullite materials exhibited enhanced fracture toughness and flexural strength compared to the currently available dental ceramics (9, 10). The amount of fluoride plays a pivotal role in the formation of glass-ceramics. With the increased fluoride content, the glass transition temperature decreases. Conversely, low fluoride content results in inhibiting the nucleation of crystals and the subsequent crystal growth, thereby affecting the properties of glass-ceramics (11).

As core materials, glass-ceramics meet the standards of the International Organization for Standardization (BS EN ISO 6872:2008) (12, 11), and their favorable biocompatible properties have been confirmed (13). In this regard, Fathi et al. (11, 14) claimed that their produced apatite-mullite glass-ceramics could be used as core materials with a proper veneering ceramic, while they cannot be applied as stand-alone body ceramic.

Apatite-mullite glass-ceramics are reported to have good castability at the temperature of 1,450˚C using the lost-wax technique (15). Restorations could be rapidly processed, so that the material could be cast and cerammed within a workday (15). According to Gorman et al. (16, 17), these glass-ceramics could also be processed via hot pressing. The mentioned study also indicated that further heat treatment after the hot pressing of apatite-mullite increased crystallization, while improving fracture toughness and the strength of the glass-ceramics.

The present study aimed to assess the processing capability of glass-ceramics using three different routes, including lost-wax casting, hot pressing, and CAD-CAM milling.

Materials and Methods

The material used in the present study was a derivative of glass ionomer cement based on the general formula of $4.5SiO_2$-$3Al_2O_3$-$1.5P_2O_5$-3CaO-$0.5CaF_2$. The glass was referred to as an HG glass-ceramic and produced by the Department of Restorative Dentistry at the University of Sheffield. HG is a given name for the new composition of this type of glass, so that it could be distinguished from the LG glasses that were previously developed at the University of Limerick by Hill et al. (8) in 1991.

Glass Production and Characterization

A batch of glass composed of an apatite-mullite glass-ceramic material was produced based on the mentioned formula. After the accurate weighing of the raw materials, the components were completely mixed for 10 minutes. The mixture was packed into a lidded sillimanite crucible and preheated overnight at the rate of 2˚C/min-1 to the temperature of 1,050˚C. Afterwards, the crucibles were transferred to a high-heat furnace and heated to the temperature of 1,450˚C for two hours. The final melt was rapidly quenched by pouring into a mesh basket submerged in a bucket of water with room temperature, and the resultant frit was collected in the mesh basket (Figure 1-A, 1-B). The frit was dried in an oven at the temperature of 150˚C for two hours.

An anatomically correct mould of an upper-right first molar, which had been prepared for an occlusal onlay, was selected to trial the three different manufacturing techniques, including lost-wax casting, hot pressing, and milling.

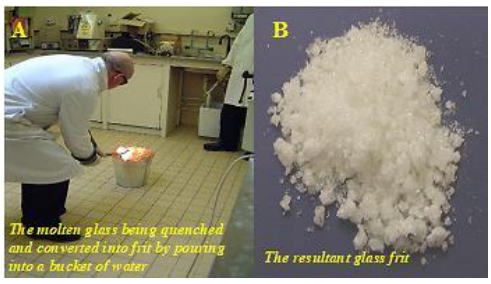

Figure 1. A-B. The resultant glass frit

Test Pattern

The samples were crushed using a percussion mortar, ground, and filtered through a 45-μm sieve.

Differential thermal analysis was carried out to evaluate the phase evolution in the glass and determine the heat treatments in terms of the production of glass-ceramics using a Stanton-Redcroft 6734. Fired alumina (Al_2O_3) powder was used as a reference phase, and X-ray powder diffraction (XRD) was performed using Philips PW1050.

Scanning electron microscopy (SEM) was carried out on the fracture site of the glass-ceramic materials using a high-resolution scanning electron microscope (JEOL JSM 6400) in order to examine the glass-ceramic microstructure. In addition, an anatomically correct mould of an upper-right first molar, which had been prepared for an occlusal onlay, was selected to trial the manufacturing techniques, including lost-wax casting, hot pressing, and milling.

Lost-Wax Casting

Hard inlay carving wax was used to produce the wax pattern, which was sprued using a three-millimeter wax sprue. To reduce the porosity of the cast glass, one-millimeter wax air risers were used. The wax pattern was invested inside a casting ring using a gypsum bonded investment material (Cristobalite, Whip-mix Corp, USA). The ring was preheated in a burnout furnace to the temperature of 700˚C at the rate of 13˚C per minute to dry and burn out the wax. Following that, the oven temperature was lowered to 520˚C, so that the glass would not crystallize within the mould during casting.

Glass frit of HG0.5 was melted in a mullite sillimanite crucible at the temperature of 1450˚C and centrifugally cast using a centrifugal casting machine (Degussa, TS3, Germany). A programmable furnace (UAF 15/5 Lenton Thermal Designs, UK) was also employed to ceram the mould to the temperature of 765˚C at the rate of 2˚C per minute and preserved for three hours, followed by another three hours at the rate of 2˚C per minute to the temperature of 1931˚C. At the next stage, the ring was cooled to room temperature within the furnace at the rate of 1˚C per minute. The investment material was removed though grit blasting with aluminum oxide particles (50 μm), and the sprue was removed using diamond burrs.

Hot Pressing

The Empress system (Ivoclar Vivadent, Liechtenstein) was used to evaluate the experiment material. A number of stages had to be programmed to apply a hot pressing machine, and each of the materials that was pressed using the hot pressing machine had a special setting.

Since the apatite-mullite glass-ceramic materials had not been pressed, proper operating conditions were required. To perform the pressing operation for the experimental material, a wax spiral test pattern was prepared in order to ensure that the experimental material would be hot pressed. The pattern was achieved using a three-millimeter diameter wax wire.

Empress II ‘speed’ investment material was used to invest the patterns. The invested moulds were preserved for one hour and placed into the burnout furnace at the temperature of 800˚C for one hour. The HG0.5 material was tested in the glass and apatite-mullite glass-ceramics. We also evaluated the selected temperatures between the glass-melting and nucleation temperatures, which were determined to be 1150˚C and 1200˚C, respectively based on the DTA analysis. The hot pressing programs used for the processing of the experimental HG0.5 material are presented in Table I.

Table I. Hot Pressing Programs for Processing of Experimental Materials to Form Glass Ingots

Program Setting	Program I	Program II
t	60°C	60°C
B	700°C	700°C
H	60 Min	60 Min
T	1150°C	1200°C
V_1	700°C	700°C
V_2	1150°C	1200°C
Pressing Pressure	5 Bar	5 Bar

Ingots of HG0.5 glass-ceramic were produced through lost-wax casting. Large and small ingots were reproduced in the spread wax and invested afterwards. Glass of HG0.5 was cast directly into the preheated mould (520˚C) as previously described. The ingots were in the form of cast (glass), and the other rings were placed into a programmable furnace (UAF 15/5 lenton Thermal Designs, UK) for the heat treatment of the glass ingots at the temperature of 765˚C and rate of 2˚C per minute for three hours, followed by another three hours at the temperature of 1031˚C in order to form the apatite-mullite and cooled at the rate of 1˚C per minute.

CAD-CAM Milling

In the current research, we used Cerec-Scan CAD-CAM unit (Sirona, Germany). To achieve an appetite-mullite glass-ceramic restoration using this method, a block of HG0.5 glass-ceramic was produced via the lost-wax processing route as described previously. Afterwards, the block was cerammed at the temperature of 765˚C for three hours at the rate of 2˚C per minute, followed by another three hours at the rate of 2˚C per minute and temperature of 1031˚C. Additionally, a copy of the onlay restoration was produced in the dental

stone, trimmed to size, and mounted on the scanning stand. At the next stage, the mould was covered in white powder using a titanium dioxide-based surface agent in order to enable a digital impression. A milling test was carried out using the VITABLOC Mark II, which provided the data on the block size required to mill the pattern to fabricate an apatite-mullite glass-ceramic restoration from the HG0.5 block.

Results

In the DTA analysis, various nucleation and growth temperatures of the heat treatments were determined based on the exotherms of the peak crystallization (apatite) and peak two crystallization (mullite). The nucleation and crystallization temperatures identified on the traces are depicted in Fig. 2.

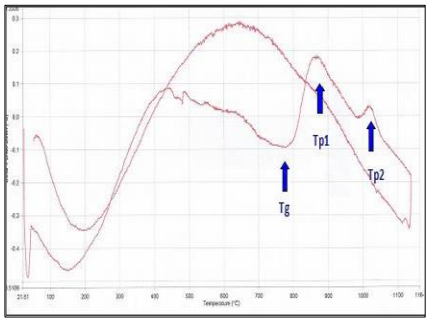

Figure 2. DTA trace of HG 0.5

The XRD traces for the glass-ceramics of HG0.5 are illustrated in Figures 3-A and 3-B. As can be seen, there was a difference in the structure of these samples in the comparison of the samples subjected to heat treatment to form the apatite with the samples that were subjected to heat treatment to form apatite-mullite. The SEM of a fracture surface demonstrated that interlocking needle-like crystals were present throughout the samples, showing that the material had been converted from a glassy, amorphous state into a ceramic during heat treatment (Fig. 4).

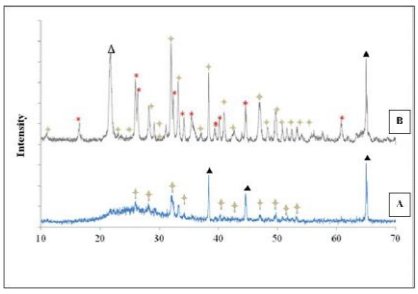

Figure 3. XRD of HG0.5 that were subjected to heat treatment (A) to form fluorapatite and (B) to form Apatite-mullite (▲Aluminium, ♦ Fluorapatite,* Mullite and Δ Cristobalite)

Figure 4. SEM of a fracture surface showing some needle-like crystals

In the present study, it was possible to effectively perform inlay casting for the lost-wax casting route. However, there were signs of porosity at the cusp tips although the air risers (vents) were incorporated. The fit of the crown to the original die was a precise fit with no observable gaps at the margins, which favorably compared to the same inlay of the same material that was produced using the CAD-CAM milling route. The completed crown is depicted in Fig. 5.

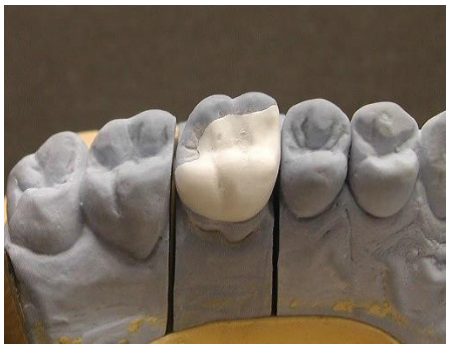

Figure 5. The cast and cerammed HG0.5 crown formed using the lost-wax casting technique

According to the findings of the current research, the VITAMARC II inlay produced via the hot-pressing route had clinically acceptable fits with no porosity. In addition, the HG0.5 glass ingots using the maximum pressing temperature capable by the Empress system (1200˚C) had only a slight movement into the sprue channel (Figs 6-A and 6-B).

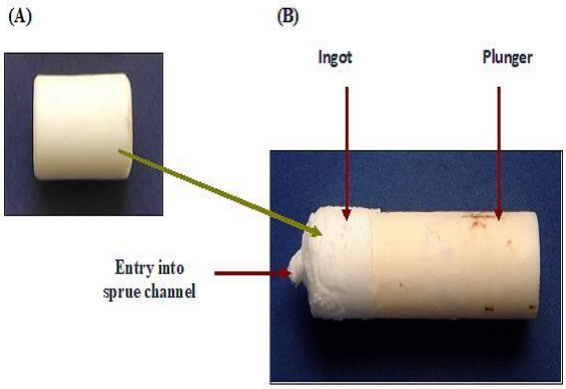

Figure. 6A-B. (A) - HG0.5 glass-ceramic ingot before pressing. (B) -The pressed glass ingot, subsequently cerammed, showing a small amount of pressed material with the plunger still attached

The casting temperature used during the lost-wax casting of the HG0.5 glass-ceramic material was 1450˚C, and the maximum temperature of the Empress II unit was 1200˚C, which was inadequate for producing a flow in the HG material. On the other hand, the HG0.5 glass-ceramic block produced by lost-wax casting for the milling process was accepted by the Cerec-Scan milling unit to have the correct size. The onlay was taken to the mill from the block in approximately 10 minutes with no differences in the milling duration of materials between HG0.5 and Vita II. Moreover, the HG0.5 onlay produced by the milling unit showed good marginal integrity and compared very favorably to the commercial material VITAMARC II (Figs. 7-A and 7-B).

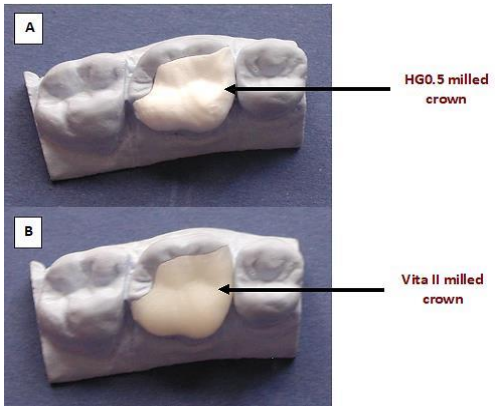

Figure 7A-B. The fit and occlusal surface of the HG0.5 milled (A) crown and the Vita Mark II milled crown (B)

Discussion

The present study aimed to assess the possible routes of placing apatite-mullite glass-ceramic materials into the mouth. Although the lost-wax casting technique was the most documented method, it demonstrated unpredictable results for the fabrication of ceramic dental castings in terms of porosity. Porosity is a common occurrence in metal castings (18), it may partly ignore due to the high strength of the metal. However, porosity in glass-ceramic materials may lead to severe consequences regarding strength.

The Empress hot pressing system has proven successful in dentistry (19). Unfortunately, this technique would not be a suitable for the formation of apatite-mullite materials in glassy or cerammed states as the maximum pressing temperature of the machine is 1200˚C, and the optimal casting temperature of the studied HG0.5 materials is 1450˚C. Therefore, it is difficult to recommend apatite-mullite as a suitable material for hot pressing unless the glass transition temperature of the material could be dramatically reduced through altering the chemical composition of the material (11).

CAD-CAM is expected to be the most efficient method for potential dental restoration production using a glass-ceramic system. In the present study, the Cerec-Scan unit was able to produce a crown that fitted the model accurately within a significantly limited timeframe compared to the casting or pressing routes. The findings of the current research confirmed that the experimental materials could be successfully produced through lost-wax casting and CAD-CAM milling. Both of these systems had compatible fits. Despite this success, the coloration of the milled restoration was not acceptable and remained a problem.

In order to produce acceptable colors for use in dental glass-ceramics, colorants are required with the capability for producing yellow to yellow-red in the crystallized product. In this regard, Weyl (20) and Grossman (21) stated that the combination of TiO2 and Ce in glass composition could import strong yellow colors in glazes and glasses .. Therefore, further development is required to improve the aesthetics, and it is also essential to develop formulations in accordance with the accepted dental standards. To date, no studies have been focused on the coloring of the glasses that were produced in the present study.

Conclusion

According to the results, glass-ionomer, cement-based glasses could be cast to shape successfully using electrical resistance melting and centrifugal casting forces. As such, it is possible to produce acceptable restoration from the tested materials via the CAD-CAM milling route. In addition, the hot pressing of the tested materials was not possible due to its high liquidus temperature. The experimental materials had a great possibility to be applied as restorative materials and could be heat-treate inside and outside the investment

casting material in order to produce a crystalline microstructure of apatite and apatite-mullite.

Conflict of interest

The authors declare that there is no conflict of interests regarding the publication of this paper.

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