

Research Article

# Optimizing the Processing of Porous Melt-Derived Bioactive Glass Scaffolds

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**Abstract** We were able to produce the first ever melt-derived bioactive glass scaffold that met the requirement of an ideal bone scaffold by using the gel-cast foaming process and a new glass composition. However, initially a small fraction of sodium potassium sulfate crystals formed during processing. The purpose of this work was to study and prohibit the growth of those crystals, which was the result of the reaction between water, the glass and the initiator during the gel-casting foaming process of the scaffold. The degree of crystallinity was reduced by increasing the drying temperature and decreasing the initiator volume used.

**Keywords** melt-derived bioactive glass; gel-casting; porous scaffold; bone regeneration; sintering

## 1 Introduction

Bioglass® 45S5 was the first material that bonded closely to the host bone. It is available in powder form and has been clinically used for more than 20 years. However, Bioglass® cannot be made into porous scaffolds without crystallizing during sintering. Gel-casting is a widely applied ceramic production technique [1]; it was adapted by Sepulveda et al. to produce porous hydroxyapatite foams [2]. We have produced melt-derived bioactive glass scaffolds by foaming and gel-casting a glass powder of a novel composition, which is able to sinter without crystallizing. There was only one side effect, which was the formation of small amount of sodium potassium sulphate crystals during processing. The aim was to modify the process (the drying and sintering of the green body in particular) in order to prohibit the crystallization.

## 2 Materials and methods

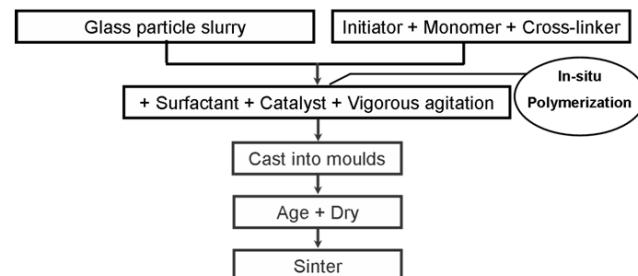
The glass used (ICIE 16M) was a modified composition of Bioglass® (49.26% SiO<sub>2</sub>, 27.27% CaO, 6.6% Na<sub>2</sub>O, 1.07% P<sub>2</sub>O<sub>5</sub>, 3% SrO, 3% MgO, 3% ZnO and 6.6% K<sub>2</sub>O, all in mol%). Additional components were added, in order to expand the sintering window and to optimize the glass

properties. Each component is harmless to human body with some of them are beneficial for osteogenesis.

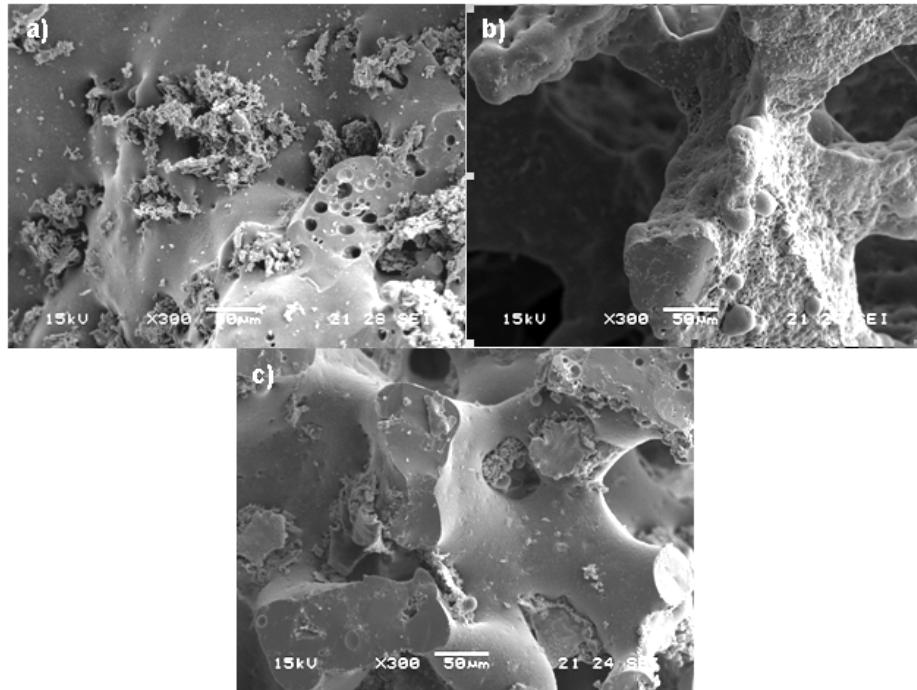
Porous glass foams were produced using the gel-cast foaming process, Figure 1 and Table 1 explains the flow of the process and the process variables involved in detail.

Glass powder was mixed with ultra-purified water, the monomer and the cross linker to produce a slurry, it was then vigorously agitated to produce a foam with the help of the surfactant. The disperser was added to help the dispersion of glass powder in the solution. Polymerization was initiated by addition of the initiator (APS solution) and the catalyst (TEMED), which react with each other and release a free radical particle. This free radical then reacts with both the monomer (acrylamide) and the crosslinker (bis); forming a large polymer network. Just before the polymerization occurs, the foam was cast into moulds and by gelation the green body of the scaffold was produced. It was then dried and sintered to burn out the organic parts, leaving the porous glass network as the final form.

The gelled foam was dried at different temperatures: 100 °C, 125 °C and 150 °C, followed by a two stage sintering, first ramp to 350 °C at 2 °C/min and hold for 1 h; then ramp again to different temperatures of 680 °C, 700 °C, 710 °C and 730 °C at 2 °C/min, hold for 1 h; and then furnace cooled to room temperature.



**Figure 1:** Gel-casting process flow chart.



**Figure 2:** Scaffolds dried at (a) 100 °C, (b) 125 °C and (c) 150 °C and sintered at 700 °C for 1 h.

Order	Components	Role	Amount
1	Glass powder (< 38 μm)		20 g
2	Ultra-purified water	Medium	18 mL
3	Methacrylamide	Monomer	6 g
4	N,N'-methylene bisacrylamide	Crosslinker	3 g
5	Ammonium Persulfate (APS) solution	Initiator	4 mL
6	Dispex	Disperser	2 drops
7	Triton X100	Surfactant	0.1 mL
8	Tetramethylethylene diamine (TEMED)	Catalyst	4 mL

**Table 1:** Gel-cast foaming process variables.

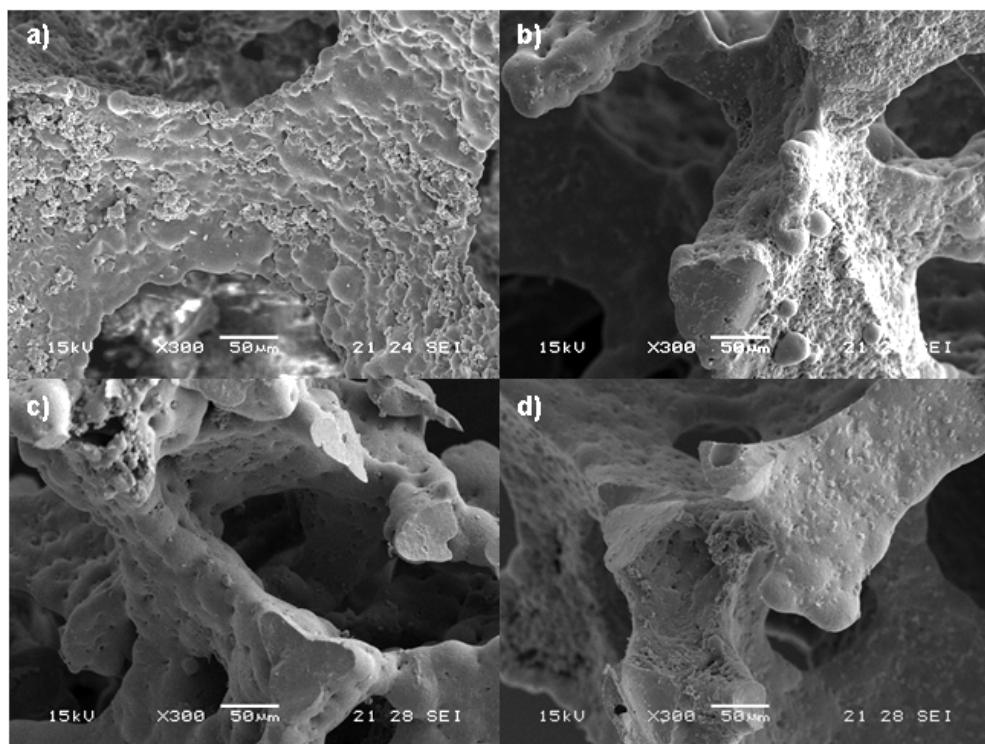
### 3 Results and discussion

Small crystals were observed at the surface of the glass when drying at 100 °C and 150 °C (Figures 2a and 2c, respectively), however the size of the crystals was significantly reduced when dried at 150 °C than at 100 °C. When dried at 125 °C there was minimum crystallization (Figure 2b), therefore the optimum drying temperature is between 125 °C and 150 °C.

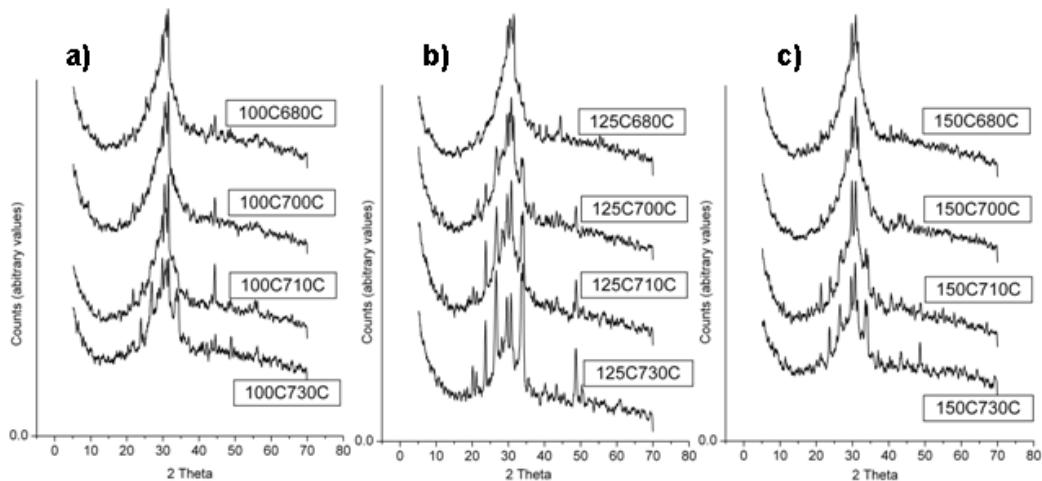
As the sintering temperature increased (Figure 3), the amount of sodium potassium sulphate crystals present was reduced.

The samples in Figure 3 were ground into powders and tested under XRD. Figure 4 shows that the degree of crystallization of the glass increased as the sintering temperature increased, and that 150 °C was the optimum drying temperature and 700 °C was the optimum sintering temperature, in order to prevent the glass crystallizing but obtaining efficient sintering (Figure 3) at the same time. After sintering at 730 °C, the glass crystallized significantly, as a result its bioactivity was greatly reduced. Therefore the optimum sintering temperature was determined to be between 700 °C and 710 °C with a drying temperature of 125 °C.

As the scaffold was sintered, the organic part was burnt out, leaving the porous glass structure behind. However the sulphate from the initiator was also left behind, it sat in between the glass particles and at the surface of the glass, and it would attract the sodium and potassium ions from the glass itself, when it was added to water, causing nucleation of crystals. The amount of water molecules present increased their rate of formation, and the raising temperature encouraged their growth. This crystallization of sodium potassium sulphate is a complicated interaction between the water, the glass and the sulphate. Increasing the drying temperature increases the rate of evaporation of the water in the gel, but it also increases the ion diffusion rate and lowers the crystallization temperature of the glass. A too high a drying temperature would also disrupt the polymer network before the sintering took place, which would result in a misshaped scaffold being produced.



**Figure 3:** Scaffolds dried at 125 °C, followed by sintering at (a) 680 °C, (b) 700 °C, (c) 710 °C, (d) 730 °C for 1 h.

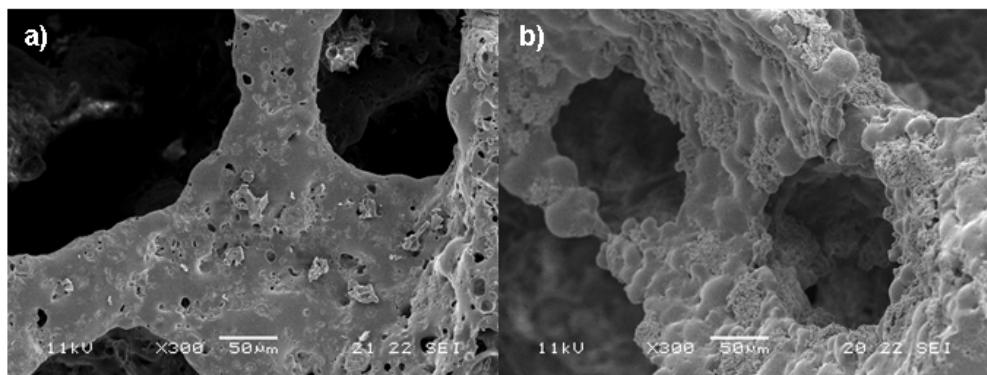


**Figure 4:** XRD spectra of scaffolds as a function of sintering temperatures at different drying temperatures, (a) 100 °C, (b) 125 °C and (c) 150 °C.

Other than varying the drying and sintering temperature, the amount of initiator present in the system was also investigated. In Figure 5 it is clear that as the initiator volume decreased, sodium potassium sulphate crystals were not formed as there was less sulphate available. However applying 1ml of the initiator caused the collapse of the green body due to insufficient gelation, therefore 2 mL was the optimum amount of initiator to be applied in the gel-casting process.

#### 4 Conclusions

The gel-casting foaming process for creating porous melt-derived bioactive glass scaffolds was optimized to maintain an amorphous structure. The optimum drying temperature was determined to be between 125 °C and 150 °C whereas the optimum sintering temperature was between 700 °C and 710 °C, with an initiator volume decreased from 4 mL to



**Figure 5:** SEM images of scaffolds produced using (a) 1 mL and (b) 4 mL of initiator, both dried at 125 °C and sintered at 680 °C.

2 mL. Open porous structures were successfully produced, with an average porosity of 75%–80%, interconnect pore size in excess of 100 µm and a compressive strength of 12 MPa.

## References

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