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The influence of various additions on a glass-ceramic matrix composition based on industrial waste

I. Rozenstrauha [a], D. Bajare [a], R. Cimdins [a], L. Berzina [a], J. Bossert [b], A.R. Boccaccini [c,*]

[a] *Department of Chemistry, Riga Technical University, Azenes St. 14/24, Riga LV-1002, Latvia*
[b] *Technical Institute: Material Science, Friedrich-Schiller University, D-07743 Jena, Germany*
[c] *Department of Materials, Imperial College London, Prince Consort Rd. London SW7 2BP, UK*

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Abstract

The influence of chamotte additions on the microstructure and mechanical properties of glass-ceramics made from a combination of silicate wastes is investigated, in order to obtain reinforced glass-ceramic composites by powder technology and viscous flow sintering. The base glass-ceramic matrix was formulated from fly ash, peat ash and clay from Latvian origin and, in once case, addition of waste glass. Chamotte particles obtained from clay in concentrations of 10, 20 and 30 wt.% were added to the starting silicate composition. The optimal concentration of chamotte additive was found to be 20 wt.%, which resulted in a material with high relative density (96% theoretical density), relatively high ultimate bending strength (72 MPa) and large sintering interval ($\Delta T > 50$ °C). The results confirmed that chamotte particles are useful and cost-effective additions as reinforcing phase to produce improved silicate glass-ceramics from wastes.

Keywords: A. Sintering; B. Porosity; D. Glass-ceramics; Chamotte; Silicate waste

1. Introduction

Previous studies of sintered glass and glass-ceramic matrix composites from silicate wastes have concentrated on the use of commercially available and relatively costly reinforcements, such as Al_2O_3 platelets [1,2], and SiC, TiC [3,4] or metallic particles [5]. The influence of the reinforcement addition on the microstructure and mechanical properties of waste derived glass-ceramics has been investigated and usually a better mechanical behaviour with increasing volume fraction of reinforcement addition has been achieved [1–5]. It is interesting therefore to investigate other kinds of reinforcing components, coming from less expensive sources, for production of waste derived glass-ceramic matrix composites. There are a few previous investigations where this approach has been explored, but mainly composites with glass matrices rather than with glass-ceramic matrices have been developed. These include an early experiment using coal fly ash from Polish power stations combined with a borosilicate glass to produce a glass matrix composite [6] and, more recently, the development of glass matrix composites from a combination of municipal solid waste incinerator fly ash and solid wastes from aluminium alloy production by an Italian group [7].

The preparation of glass matrix composite materials, where the matrix is composed of fly ash, peat ash and clay of Latvian origin, has been carried out in the past and commercially available Al_2O_3 platelets were used as reinforcement [1]. In contrast to the previous investigation, in the present study, chamotte was added to the silicate matrix as a reinforcing component. Chamotte made from the same clays has been described in the literature originated in Latvia [8] and Russia [9] as suitable reinforcing addition for producing stoneware materials and fine ceramics articles. The main objective of the present investigation was thus to

* Corresponding author. Tel.: +44 207 5946731; fax: +44 207 5843194.
E-mail address: a.bocccaccini@imperial.ac.uk (A.R. Boccaccini).

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optimise a powder technology route for production of improved glass-ceramics from silicate wastes of Latvian origin and chamotte as reinforcement. Process parameters were chosen in order to maximize the sintering temperature interval by keeping as high as possible the silicate waste content, highest sintered density as well as the highest possible mechanical strength of the materials obtained.

2. Experimental

The starting materials for the glass-ceramic matrix (labelled 3S0) are fly ash from the steel plant ''Liepajas metalurgs'' (Latvia) and peat ash from the Riga coal power station, as well as limeless clay, as reported elsewhere [1,10]. Clay was added as a binder to improve the bonding properties between particles during the pressing process. The waste materials contain as main chemical elements: Si, Ca, Al, Fe, Zn, Mg, Pb as well as trace amounts of Sr, Mn, Ni, Cu, Cd and Sn [11]. As reported in previous studies [11,12], the fly ash contains spinel ($ZnAl_2O_4$), sphalerite (ZnS), hematite (Fe_2O_3) and palmerite ($K_2Pb(SO_4)_2$), while peat ash contains calcite ($CaCO_3$), anhydrite ($CaSO_4$), corundum (Al_2O_3), albite ($(Na,K)AlSi_3O_8$) and quartz (SiO_2). The ecologically incompatible element lead, which is contained in the fly ash, has been found included in the palmerite phase. The relatively high SiO_2 content in the peat ash indicates the feasibility to use this waste composition to develop glass matrices for composite materials, and the nominal chemical composition of the optimal glass-ceramic matrix has been determined in previous studies [1,10,11]. As reinforcing addition, chamotte made from the mentioned clay was used. Limeless clay from deposit Liepa (Latvia) was thermally treated at 900 °C for 1 h and milled using a ball mill for 24 h up to an average particle size of 10 μm. The density of the powdered glass-ceramic matrix and the chamotte, determined by He pycnometry, are 2.923 g/cm^3 and 2.715 g/cm^3, respectively. From the starting glass-ceramic composition (labelled 3S0) two batches of composite mixtures were prepared by adding 20 and 30 wt.% of chamotte, these were labelled compositions 3S2 and 3S3, respectively. Combined compositions with 10 and 20 wt.% of chamotte and the addition of 10 wt.% of waste glass (from Valmiera Glass Fibre Plant, Latvia) were also investigated, these samples are labelled 3SV and 3SV2, respectively. The density of the waste glass was determined to be 2.267 g/cm^3. Mixtures in dry state were milled using agate mills for 20 min and subsequently water was added (8–12 wt.%). The humid powders were screened (screen aperture: 3 mm) by keeping the moisture content at a level of 12–14%. The sintering behaviour and thermal changes of the mixtures were determined by heating microscopy (Leica Wetzlar 38818) and differential thermal analysis (DTA) (STA 409C) in the temperature range 20–1300 °C. Cylindrical samples (diameter = 20 mm; height = 4 mm) were uniaxially pressed at room temperature using pressures of 50 MPa. The powder compacts were sintered in air, the heating rate was 8 °C/min and sintering time was 60 min. The sintering temperature was varied between 1000 and 1120 °C. Rectangular test bars (25 mm × 5 mm × 5 mm) were also fabricated by sintering at the optimum temperature for each composition. The sintered bars were used for bending strength tests, as described below.

The density and water uptake of the sintered samples were determined according to DIN EN 993-1, while the values of the theoretical density of the compacts were calculated based on their composition and density of constituents. The microstructure of sintered materials was studied by scanning electron microscopy (SEM) (LEICA S 440 I). The chemical identification of elements was carried out by EDX spot analysis (OXFORD 5431). Four-point bending test on rectangular bars (25 mm × 5 mm × 5 mm) was used to determine the ultimate fracture strength (INSTRON 430) according to DIN EN 100. At least five test bars were tested for each composition and the results were averaged.

3. Results and discussion

Differential thermal analysis (DTA) results of various compositions investigated are shown in Fig. 1. All DTA curves exhibit shallow endothermic effects in the temperatures range 570–575 °C, which indicates transformations of low-temperature quartz (SiO_2) to a high-temperature quartz modification (α-quartz to β-quartz), in agreement with the literature [13]. The next two exothermal effects occurred in the temperature ranges 720–730 °C and 930–970 °C. The first of them (720–730 °C) may be related to oxidation of Fe^{2+} and formation of spinel phase at the same time. The crystallization of a pyroxene solid solution takes place in the temperature range 920–970 °C, as observed on crystallization studies of iron-containing glass-ceramic [14,15]. In our case, a second exothermic effect in the range 930–

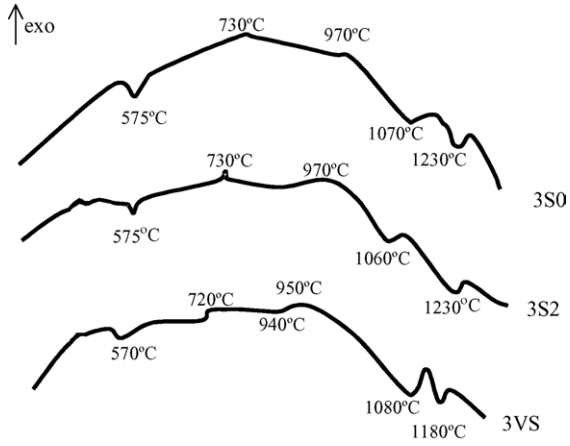

Fig. 1. DTA curves of powder samples of the different compositions investigated: unreinforced glass-ceramic matrix (3S0), composites with 20 wt.% of chamotte (3S2) and combined matrix with 10 wt.% waste glass and 10 wt.% chamotte (3VS).