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Crystallisation of nepheline-based glass frits through fast-firing process

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ABSTRACT

The aim of this research was to study the feasibility of producing sintered nepheline glass-ceramic through a fast firing route. The thermal behaviour of the original glass was analyzed by mean of differential scanning calorimetry (DSC) and X-ray diffractometry (XRD). The microstructural analysis of glass-ceramic materials was carried out by field-emission scanning electron microscopy (FE-SEM). The results showed that the studied wastes are able to produce a glass and glass-ceramic materials through a sinter-crystallisation process, 100 μm being the critical glass particle size. Glass-ceramics are composed of residual glassy phase and crystalline phases such as nepheline (NaAlSiO₄), augite (Ca, Na)(Mg, Fe, Al)(Si, Al)₂O₄ and a solid solution belonging to the melilite group (Ca, Na)(Al, Mg, Fe, Si, Al)₂O₇. In a first evaluation, water absorption (0.02%) and bending strength (71 MPa) of glass-ceramic achieved after thermal treatment at 1100 °C/5 min suggest that sintered glass-ceramics can be easily produced from coal fly ash and metallurgical slag wastes by a fast-firing cycle and they are extremely serviceable for outdoor flooring and wall cladding.

1. Introduction

Glass-ceramics are multi-phases materials composed by at least one glassy phase acting as matrix and a finely dispersed crystalline phase uniformly embedded in the matrix. Glass-ceramics are developed through controlled devitrification (nucleation and crystal growth) of a parent glass. This parent glass is melted and subsequently it undergoes a thermal process aiming to precipitate one or more crystalline phases. By controlling internal crystallisation, efficient nucleation step conducts to the development of randomly oriented fine crystals and glass-ceramics materials free of defect like micro-cracks or porosity. In most of cases, nucleating agents are added to the parent glass for improving the nucleation process [1]. However, glass-ceramic materials could be also prepared by sintering-crystallisation process [2]. In this case, crystallisation takes place on interfaces between glass particles. The major advantage of this process is the absence of nucleating agents since particles boundaries act as nucleating sites but its disadvantage is the final residual porosity. Generally, glass-ceramics present so many benefits in relation to parent glasses such as higher strength, wear resistance, thermo-physical properties, etc., which make them attractive materials for structural applications.

In the design of glass-ceramics, the two most important factors are composition and microstructure [1] since their properties fundamentally depend on the type and appearance of the crystalline phase. The

major crystalline components of glass-ceramics are silicates. Among this, nepheline-based glass-ceramics are characterised by high chemical and mechanical strength and impact resistance. In the last decades glass-ceramics containing nepheline as crystalline phase had been developed for several applications, such as microwave ovens [3] and dental applications [4,5].

Nepheline glass-ceramics are usually prepared from bulk glasses of the Na₂O-Al₂O₃-SiO₂ system and the addition of different nucleating agents, such as TiO₂, Cr₂O₃, ZrO₂ or LiF is mandatory to promote crystallisation [6,7]. Recently, Martín et al. [8,9] and Andreola et al. [10] reported on the viability of the production of a wide variety of forsterite-nepheline glass-ceramics by a sinter-crystallisation process from a SiO₂-Al₂O₃-MgO-Na₂O base glass. Tiling and cladding was suggested as potential applications for these materials given that sintered nepheline materials showed similar bending strength values than the commercial glass-ceramics tile Neoparies® and water absorption < 0.5%, which allowed to classify these materials into the Group BIa characteristic of high sintered ceramic tiles according to European Standard rule.

In last years, fine-grained nepheline glass-ceramics were also produced by vitrification of coal fly ashes and subsequent controlled heat treatment of the parent glasses. The results showed that glass ceramics exhibit attractive mechanical properties if compared with building ceramic tiles [11–13]. Nevertheless, in these investigations

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glass-ceramics were prepared from bulk glass samples and devitrification was normally carried out at slow heating rate and long dwell times (120 min) at the crystallisation temperature. Such conditions unfit the fast-firing thermal cycle used in the manufacture of ceramic tiles (60–90 min cold-to-cold and temperature rates up to 60 °C/min) result in glass-ceramic materials from fly ashes that are economically unviable due to high energy costs associated with processing.

The present manuscript studies the feasibility of producing sintered nepheline glass-ceramic through a fast firing route, so that they can be easily adapted to the cycle used in the manufacture of ceramic tiles and therefore, to obtain a new marketable product to be used as material for the construction industry.

2. Experimental procedure

2.1. Raw materials

Fly ash and metallurgical slag were used as raw materials to obtain a glass formulation; moreover, Na₂CO₃ was added to the glass composition to assist the melting process.

The fly ash used in this investigation was originated from Petacalco coal-fired power plant, located in La Unión, Guerrero State, Mexico. The ash was used as-received without any prior conditioning stage. The metallurgical slag proceeds from ArcelorMittal steelmaking plant located in Lázaro Cárdenas, Michoacán State, México. In this case, prior to its use, the slag was grinded by high energy milling and sieved to a particle size < 100 µm.

2.2. Materials preparation

A glass (hereafter designated CEC glass) in the SiO₂-Al₂O₃-CaO-Na₂O base system was formulated by using the fly ash as silica source and the metallurgical slag as alkali oxides supplier. A previous vitrification study evidenced that the optimum composition to maximise the use of fly ash and slag, while producing a melt with an acceptable viscosity was 58.5 wt% fly ash + 31.5 wt% Arcelor Mittal metallurgical slag + 10.0 wt% Na₂O [14]. The batch components were mixed for 15 min in a blender (TURBULA) to get a homogeneous mixture. The batch was placed in an aluminosilicate crucible and heated at 15 °C/min in an electric furnace up to 1450 °C. After a holding time of 60 min at the melting temperature, the melt was quenched by pouring into water producing a glass frit. For performing the crystallisation study, the original glass was sieved to a particle size < 100 µm and the glass powders were moistened by spraying with distilled water (5 wt%) and pressed into 15 × 15 × 8 mm compacts by using a laboratory uniaxial hydraulic press (Mignon-S Nanetti) at a pressure of 40 MPa. Moreover, test pieces of 50 × 15 × 5 mm were prepared for bending strength measurements. The green compacts were sintered at 750–1130 °C and soaked for 5 to 30 min at the sintering temperature. The samples were placed in a furnace at the temperature required for the heat treatment and removed as soon as the exposure time had expired.

2.3. Materials characterization

The chemical composition of the CEC glass was determined by X-ray fluorescence (XRF) (Bruker model S8 Tiger equipped with the software package SPECTRA^{PLUS}). To evaluate the thermal behaviour of CEC glass, differential scanning calorimetry (DSC) was performed using a Setaram (Labsys) TG-DTA/DSC unit in air atmosphere. The analyses were carried out at a heating rate of 50 °C/min, in platinum crucibles and calcined alumina as reference. DSC curves were recorded on samples with particle sizes ranging from 63 µm to 1 mm. The temperature precision given by the equipment is ± 0.1 °C. Each measurement was reproduced three times in order to estimate experimental errors. The activation energy (E) can be estimated using Ozawa [15] and Kissinger

[16] analyses, according to Eqs. (1) and (2) respectively:

$$\ln \varphi = -\frac{E}{RT_x} + \text{cte} \quad (1)$$

$$\ln \frac{\varphi}{T_c^2} = -\frac{E}{RT_c} + \text{cte} \quad (2)$$

where φ is the heating rate, T_x is the onset crystallisation temperature, T_c is the peak temperature of crystallisation peak, cte is a constant and R is the gas constant. The evaluation of E was carried out from the DSC scans on CEC glass with particle size < 100 µm recorded at different heating rates (10, 20, 30, 40 and 50 °C/min).

The evaluation of the amorphous nature of CEC glass after melting, and the mineralogical study of the crystalline phases devitrified after thermal treatment was performed by X-ray diffraction (XRD) (Bruker model D8 Advance) with Ni-filtered Cu K α radiation operating at 30 mA and 40 kV. Data were recorded in the 5–60° 2 θ range (step size 0.019732° and 0.5 s counting time for each step). The microstructural analysis of glass-ceramic materials was performed by field emission scanning electron microscopy (FE-SEM) (Hitachi model S-4800) using an accelerate voltage of 20 kV. Semi-quantitative analysis of different phases were obtained by energy dispersive X-ray spectroscopy (EDS) (Bruker model QUANTAX Esprit 1.9) provided by a beryllium (Be) window. SEM observation were performed on different samples; viz., fresh, fracture; polished specimens using 6, 3 and 1 µm diamond pastes after grinding with silicon carbide paper and water; and polished surfaces etched in 5% HF solution, washed ultrasonically with distilled water and ethylic alcohol. All the samples were Au-Pd coated in a sputter (Balzers SCD 050).

The density of selected glass-ceramic materials was measured using Archimedes' principle with distilled water as an immersion liquid. The error in this method has been determined based on repetitive measurements. The errors in density were found to be about 0.1%. Water absorption and bending strength of a selected glass-ceramic material was measure according to ASTM C329-88 and UNE-EN 841-1, respectively. Bending strength tests were performed in an electronic universal tester (Servosis, ME402101 model) by a three point loading test with a span of 36 mm and a crosshead speed of 1 mm/min. The tests were carried out on 6 test pieces of 50 × 15 × 5 mm. The errors in bending strength were found to be about 0.3%.

3. Results

Table 1 summarizes the chemical composition of the original CEC glass, determined by XRF. Fig. 1 shows the DSC curves recorded on both bulk (~3 × 3 × 4 mm) and fine (powder without granulometric control) CEC glass samples. The DSC curve of the powder sample exhibits clearly the glass transition temperature (T_g) at 663 °C. After T_g , an exothermic peak at 868 °C (T_m) indicates the formation of crystalline phases and finally an endothermic peak at 1148 °C points out the formation of a liquid phase. Regarding the bulk sample, it exhibits T_g and T_m at the same temperatures than the powder sample. In order to

Table 1
Chemical composition (wt%) of the original CEC glass. The errors in the experimental composition are approximately ± 0.2 wt%.

Oxide	Percentage	Oxide	Percentage
SiO ₂	42.88	SO ₃	0.28
Al ₂ O ₃	18.10	P ₂ O ₅	0.23
CaO	14.12	Rb ₂ O	0.06
Na ₂ O	11.60	Cr ₂ O ₃	0.06
Fe ₂ O ₃	4.98	ZrO ₂	0.05
MgO	4.94	SrO	0.05
TiO ₂	0.97	PdO	0.03
MnO	0.84	NiO	0.01
K ₂ O	0.83	ZnO	0.01

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