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Production of glass-ceramic from high frequency induction melted cordierite glass

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ABSTRACT

Cold crucible induction melting (CCIM) is a special method which can be used for producing high temperature materials. In this research, a cordierite glass was melted by CCIM at approximately 2200 °C. For determining the temperature which the glass adsorbs the induction currents, the electrical resistivity of the glass was measured in the range of 1100 to 1550 °C. Nucleation and crystallization temperatures of the glasses were determined by differential thermal analysis (DTA) and their microstructures were evaluated by scanning electron microscopy (SEM). The mechanical strength was measured by using the 3-point bending strength test and the dielectric constants were measured in the 1–10 MHz frequency range. For comparison, all of the mentioned tests were applied to the melted glass in an alumina crucible. The temperature which the glass starts to absorb the induction currents was 1500 °C with the electrical resistivity of 10 Ω.cm. The glass transition and crystallization temperatures were different for the alumina and CCIM samples. The bending strength of the CCIM sample was higher than the alumina sample. Cordierite was the major phase in all samples. The dielectric constant of the CCIM sample was the same of the alumina crucible sample.

1. Introduction

Ceramics and glasses generally are electric insulators so they cannot absorb induced currents and thus, cannot be melted by electromagnetic induction [1]. The electrical resistivity of ceramics and glasses decreases when temperature increases. In the molten state, glasses and ceramics have the lowest electrical resistivity [2]. But even in the molten state, glasses and ceramics do not absorb induced currents. Beside electrical resistivity, the frequency of the induction current is another important factor for induction melting. By increasing the frequency, absorption of the induced current will increase. In the theory of induction heating, more than 1000 GHz will be needed for melting an ordinary solid glass. Current technology is not able to make this frequency in the induction generators. Therefore, some researchers found a trick and invented a new method. They made an initial melt by using a susceptor (e.g. graphite). The initial melt can also be made by another heating source (e.g. a plasma torch). The initially melted area absorbs electromagnetic current if the frequency is in the MHz range and continues to melt its surrounding material until the whole glass specimen is melted. It should be noticed that the frequency of the induction current should be in the MHz range. In induction melting the crucible is usually made of ceramic materials. The problems of using ceramics as the crucible is the temperature limit and its corrosion by the glass melt. Cold crucible induction melting (CCIM) is a special method for melting high temperature materials. By combining CCIM and a high frequency induction generator most ceramics and glasses with high melting temperature can be melted. Also using CCIM, temperatures as high as 3000 °C can be achieved [1].Cordierite glass-ceramics were the first industrial glass-ceramics with many applications in different industries such as in the electronics, optics and refractories [3–6]. For example, cordierite glass ceramic was used as radomes for 50 years. Besides radomes, other applications for the new harder and tougher cordierite glass-ceramics are envisioned. Engine components are a possible area of application [3]. Cordierite is an orthorhombic pseudohexagonal with space group Cccm. The hexagonal rings are formed of six tetrahedra, of which five are silicon and one is aluminum. The two Mg ions in the unit formula form octahedral units with O tying the rings together. In addition, the other three tetrahedral aluminum groups are interspaced between the magnesia octahedra separating the predominantly silicate rings. The rings are distributed in reflection planes parallel to the base at heights 0, $c/2$, and c [7]. However, there were some limitations to the use of cordierite glass ceramics: they have a high melting point (~1500 C); high viscosity even at high temperatures; high corrosive melt. For shaping cordierite glass; it is required to overheat the glass above its melting point because of its high viscosity [8]. Cordierite glasses were usually melted in the electric resistance furnaces using alumina and Pt crucibles [9]. There are some downsides to using these crucibles: 1) Pt crucible is very costly, 2) although an alumina crucible is a good choice for the melting of cordierite glass, the issue of corrosion by the melt still

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remains and 3) the maximum temperature of the melt, which is important for shaping the glass, is 1650 °C in these crucibles. The high corrosivity of cordierite glass melts and the need to overheat the melt in order to be able to better shape the glass induces one to consider CCIM for melting of cordierite glass. Production of glass using CCIM has been reported in some papers [10–14]. Recently, Crum et al. [15] reported the result of the investigation on the capability of the CCIM process for making the glass for immobilization of nuclear waste. But no report has yet been published about the melting of cordierite glasses using CCIM. Unfortunately, in most papers about the production of glass using CCIM, the physical properties of the glasses were not reported and were not compared with the melted glasses using conventional methods. In this research, non-stoichiometric cordierite glass was melted using CCIM at a temperature of approximately 2200 °C. For comparison, the glass was also melted in an alumina crucible. Physical properties of cordierite glass and glass-ceramic were reported.

2. Experiment

2.1. Materials

The chemical composition of the cordierite glass was chosen based on a previous report [3]. The composition was $56SiO_2$–$20Al_2O_3$–$15MgO$–$9TiO_2$ by weight percent. The raw materials were SiO_2, $MgCO_3$, Al_2O_3 and TiO_2 powder with reagent grade. The raw materials were well mixed using a wet ball mill and dried in an oven. After drying, they were used for the melting experiments using both the CCIM and alumina crucible methods.

2.2. Electrical resistivity measurement

In the melting with the high frequency induction melting method, it is necessary to know the electrical resistivity (ER) of the molten glass. Frequency of the induction generator depends on the ER of the material [15]. Two-wire method was used for measuring the ER. The two-wire method is a low accuracy method for making quick laboratory measurements or monitoring an industrial process stream [16,17]. The ER of cordierite glasses was measured in the range of 1100 to 1550 °C. Schematic of the used setup was shown in Fig. 1. The crucible and the wires consisted of platinum. HCl (4 wt.% concentration) solution, with known conductivity, was used for the determination of the cell constant. The wires were placed into an alumina tube. Next, the tube was placed in the furnace from the top and the wires were immersed in the melt. The depth of immersion was controlled carefully to be 5 mm. By using a power supply, the loaded voltage was changed and the induced current was measured by an ampere meter. After that, using Eq. (1), the ER of the melt was calculated.

$$R = \rho C \quad (1)$$

In this equation, R is resistivity, ρ is the specific electrical resistance of the glass melt and C is the cell constant. For more accuracy, the test was performed three times on the each sample. According to the results of the tests error bars were derived.

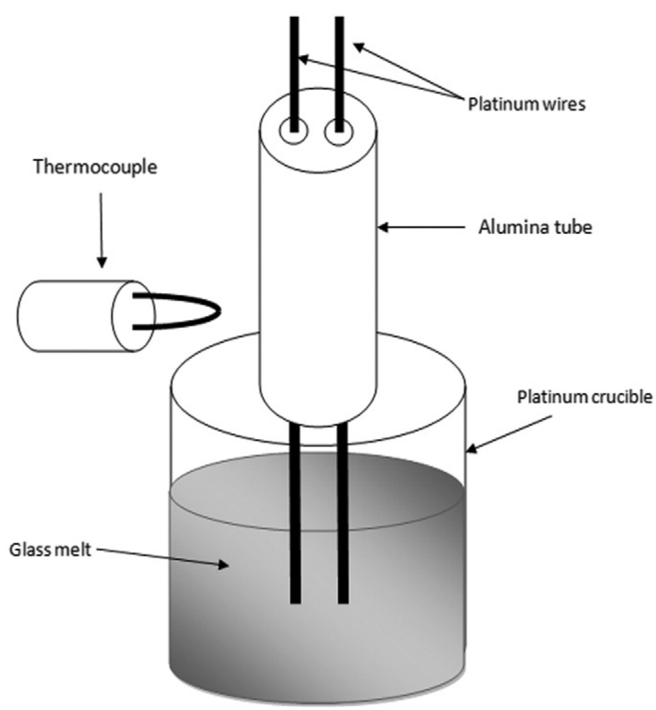

Fig. 1. The schematic of the setup of electrical resistivity of the glass melt.

2.3. Melting setup

An induction generator with the frequency of 2 MHz and power of 60 kW was used. The diameter and width of the one turn induction coil were 15 and 5 cm, respectively. A segmented cold crucible was designed and fabricated from a copper tube (Fig. 2). Water was circulated in the segments which cooled them so that when the melt reached to the segments, it cools down and makes a solid thin layer which was named Skull [18]. The temperature of the melt was measured using a pyrometer on the top of the crucible. The starting melt was made by using the graphite rings as the susceptor for induction current. Graphite was used as the starter of the melting process because it can absorb the induction current and warm up to 2700 °C [19]. After reaching the critical volume of the starting melt under the effect of the high-frequency field, the entire charge was gradually melted. The layer near the cold walls of the crucible solidified just after the melting and formed a shell, acting as the self crucible for the melt. Ten grams of the graphite rings (which is equal to 0.3% of total batch) were placed in the middle of the cold crucible which was filled with 3000 g of the glass batch. The graphite rings were covered by the raw materials. After applying the induction current, the starting melt was formed in a few seconds and the melted glass absorbed the induction current by itself. The entire batch was melted in 2 h. For comparison, 100 g of the batch was melted in an alumina crucible in an electric furnace (Carbolite 1600). The melting time was 5 h. After melting, the resulting glass was poured into a preheated steel mold.

2.4. Thermal analysis

DTA was performed (Shimadzu-DTG60AH) in order to investigate the thermal behavior of glass samples. The reference material in these experiments was α-Al_2O_3 and the heating rate was 10 $°C \cdot min^{-1}$. The glass transition temperature (T_g) and dilatometric softening point (T_d) were also determined by utilizing the onset temperature and minimum of the endothermic peak in the DTA trace. T_g was determined from the change in the slope of the elongation vs. temperature plot; and the dilatation softening temperature, T_d was obtained from the maximum of the expansion trace corresponding to the onset of viscous deformation. These temperatures were used to demarcate the glass-in-glass phase separation and nucleation temperature [20]. According to the Ray procedure [21] the narrowest peak of the DTA traces was chosen for determining the best nucleation time.

2.5. XRD analysis

In order to identify the crystallization phases, the parent glasses and the heat treated samples were subjected to X-ray diffraction (XRD). Cu-Kα radiation (Siemens-D500) at 20 kV and in the 2Θ ranges from 10°–60° was used for the examination of heat-treated specimens.