Potential of Chambersite Tailing as Raw Material for the Preparation of Glass Ceramic: Investigation on Crystallization Behavior and Crystal Structure

Qingguo Tang\textsuperscript{1,2} · Xiaozhan Mu\textsuperscript{3} · Xinhui Duan\textsuperscript{1,2} · C. Srinivasakannan\textsuperscript{4} · Jingsheng Liang\textsuperscript{1,2} · Debao Ding\textsuperscript{1,2} · Zhigang Zhao\textsuperscript{1,2}

Received: 15 March 2017 / Accepted: 3 December 2017 / Published online: 16 January 2018
© King Fahd University of Petroleum & Minerals 2018

Abstract

The feasibility of preparing glass ceramic from chambersite gravity tailings was explored. The crystallization parameters and state of glass ceramic were analyzed by Johnson–Mehl–Avrami and Augis–Bennett equations. The effects of sintering temperature and crystallization duration on bending strength and water absorption of glass ceramic were investigated, while the phase composition, crystal structure, lattice constants and surface topography were characterized. The results indicate the crystallization behavior of chambersite tailings glass ceramic is mainly body crystallization with clinopyroxene as a primary crystalline phase, the activation energy for crystallization ($E$) is 315.27 kJ/mol, the kinetics parameter for crystallization ($k$) is $7.98 \times 10^{14}$, and the main phase lattice constants of clinopyroxene were $a = 0.97392$ nm, $b = 0.88521$ nm, $c = 0.52748$ nm, $\beta = 107.062^\circ$. At sintering temperature of 890 $^\circ$C, pyroxene crystal with tetragonal column structure is formed, the diameter of the crystal is about 0.05–0.5 $\mu$m, while the crystallinity is more than 75%; the bending strength of the glass ceramic increases with the decrease in material particle size; at the D$_{90}$ of 5.71 $\mu$m and crystallization duration of 150 min, the bending strength of glass ceramic reaches the maximum of 128.3 MPa and the water absorption is between 0.06 and 0.02%.

Keywords Chambersite · Gravity tailings · Glass ceramic · Crystallization behavior · Crystal structure

1 Introduction

Glass ceramic is a kind of polycrystalline material with excellent properties produced in 1960s. It has outstanding mechanical properties and corrosion resistance [1], and the composition of the raw materials and the heat treatment has direct influence on the type, number, grain size and crystallinity of the main crystal phase in the formation of glass ceramics, further affecting the performance of the glass ceramics. Traditional glass ceramics mostly use pure industrial mineral as raw material for production, the cost is relatively high, with the growing environmental problems of China caused by industrial tailings, to produce glass ceramics using tailings and slag has become a new effective way to deal with these solid wastes. For example, Xu et al. [2] used potash feldspar tailings to prepare glass ceramic consisting of matrix glass and $\beta$-wollastonite crystal with grain size around 2 $\mu$m with Vickers hardness of 9.77 GPa. Chen et al. [3] used gold tailings as raw material to prepare CaO–Al$_2$O$_3$–SiO$_2$ glass ceramics having bending strength of 122 MPa with density of 2.836 g/cm$^3$. Du et al. [4] analyzed the crystallization characteristic and properties of Baiyunebo (China) tailing glass ceramics and found that an increase in FeO (wt%) content could greatly improve the crystallization characteristics of glass ceramics. Cetin et al. [5] prepared lightweight glass ceramic by the sintering of mining tailings at sintering temperature of 1050 $^\circ$C having density of sample in the range of 2.0–2.2 g/cm$^3$. 

---

Xinhui Duan
dxh1911984@aliyun.com

1 Key Laboratory of Special Functional Materials for Ecological Environment and Information (Hebei University of Technology), Ministry of Education, Tianjin 300130, China
2 Institute of Power Source and Ecomaterials Science, Hebei University of Technology, Tianjin 300130, China
3 CAS Key Laboratory of Green Process and Engineering, Institute of Process Engineering, Chinese Academy of Sciences, Beijing 100190, China
4 Chemical Engineering Program, The Petroleum Institute, P.O. Box 2533, Abu Dhabi, United Arab Emirates
Chambersite (Mn$_3$B$_7$O$_{13}$Cl) is a rare manganese borate mineral first discovered at Barber’s Hill salt dome, Chambers County, Texas, USA. The mineral is also known from the Dongshuichang deposits in Jixan, Tianjin, China, which is the only chambersite mineral deposit with known reserve of 242,000 tons \cite{6}. Chambersite has excellent abrasion resistance and electromagnetic properties and can produce special piezoelectric, ferroelectric, pyroelectric effect, which could be used as a wear-resistant materials and microwave dielectric substrate material \cite{7–9}. In order to alleviate the burden bought by the tailings to the environment, in the present paper, we attempt to investigate the potential of chambersite tailings as raw material for the preparation of glass ceramic for the purpose of enhancing resource reutilization. The crystallization behavior, activation energy for crystallization, phase composition and crystal lattice were investigated in the present work. In addition, the influence of sintering temperature, crystallization duration and material particle size on bending strength and water absorption of glass ceramics was also investigated. It is hoped to provide a theoretical basis for rational utilization of chambersite tailings.

2 Experimental Procedure

2.1 Experimental Materials

Jixian chambersite tailings were provided by Zhengzhou Institute of Multipurpose Utilization of Mineral Resources, Chinese Academy of Geological Sciences. Particle size of chambersite tailings milled by planetary high-energy ball mill (WL-IA, Tianjin City Jun Sheng Electric Equipment Co. Ltd.) was less than 0.076 mm. Table 1 shows the chemical composition of chambersite tailings tested by X-ray fluorescence spectrum (XRF) and atomic absorption spectrophotometer.

As listed in Table 1, the main chemical composition of chambersite tailings was SiO$_2$, MgO, MnO, B$_2$O$_3$, CaO, Fe$_2$O$_3$, accounting for 78.4% of the total composition. Based on the composition of chambersite tailings, the composition of glass ceramic was evaluated based on MgO–CaO–Al$_2$O$_3$–SiO$_2$ phase diagram. Additionally, talcum powder (powder size < 0.076 mm), quartz and aluminum oxide were added to improve the mechanical strength and thermostability of glass ceramic. The material composition of chambersite tailing glass ceramics utilized in this work was: chambersite tailings (70 wt%), talcum powder (9 wt%), quartz (13 wt%), aluminum oxide (8 wt%). In addition, the presence of B$_2$O$_3$ in the chambersite could function as the melting agent to reduce raw material melting temperature \cite{10}, while the Fe$_2$O$_3$, MnO$_2$ could act as nucleating agent \cite{11,12} in glass ceramics. Hence, addition of fluxing agent and nucleating agent was avoided.

2.2 Sample Preparation

Different ratios of materials were added into graphite crucible accurately and were mixed thoroughly. The homogenized mixture of these materials was melted in graphite crucible at 1200 °C in a rapid heating furnace (SX-G18123, Tianjin Zhonghuan Experiment Electric Furnace CO. LTD, China) for 120 min with heating rate of 7 °C/min. The molten glasses were then poured into cold water to form homogenous glass slag. Glass powder (size less than 0.075 mm) was prepared by drying and grinding. The chemical composition of the glass powder is shown in Table 2.

The dry pressing method was used for production of initial glass ceramic body by placing known quantity of glass powder into a specific mold (length of 40 mm, width of 8 mm). The glass powder was pressed at a pressure of 50 kN for 60s with the loading speed of 0.5–2 kN/s using a hydraulic press (TYE-300B, Wuxi Jianyi Instrument & Machinery CO., LTD, China). Then, initial glass ceramic bodies were rapidly heated from room temperature to glass transition temperature of 650 °C at the heating rate 7 °C/min and held for 60 min of duration, after that, the bodies were further heated at the rate of 5 °C/min to a crystallization temperature of 890 °C and held for a duration of 120 min. After sintering, glass

| Table 1 Chemical composition of chambersite tailings (wt%) |
|-----------------|---|---|---|---|---|---|
| Mineral         | SiO$_2$ | Al$_2$O$_3$ | CaO | MgO | MnO | B$_2$O$_3$ |
| Chambersite tailings | 30.14 | 2.18 | 5.23 | 18.42 | 12.52 | 7.82 |

| Table 2 Chemical composition of basic glass (wt%) |
|-----------------|---|---|---|---|---|---|---|
| SiO$_2$ | Al$_2$O$_3$ | CaO | MgO | MnO | Fe$_2$O$_3$ | B$_2$O$_3$ | Cl$^-$ | K$_2$O | Na$_2$O | TiO$_2$ |
| 40.84 | 10 | 6.22 | 17.68 | 10.12 | 3.41 | 6.27 | 1.57 | 1.16 | 0.09 | 0.09 |