Journal of Non-Crystalline Solids 355 (2009) 780–784

Contents lists available at ScienceDirect

Journal of Non-Crystalline Solids

journal homepage: www.elsevier.com/locate/jnoncrysol

The study on the phosphate glass melted by microwave irradiation

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ARTICLE INFO

Article history:
Received 17 November 2007
Available online 18 April 2009

PACS:
81.05.Kf
81.20.Ev
81.70.Pg
83.80.Ab

Keywords:
Microwave
Glass transition
Phosphates

ABSTRACT

In this study, $NH_4H_2PO_4$, Li_2CO_3, and $Ca(OH)_2$ raw materials were mixed with and without adding water or pressing pressure. The three types of mixture (i.e., raw mixtures, waterish mixtures, and dense mixtures) were then subjected to microwave irradiation. The samples were characterized by various methods of analysis method. With adding water, the particles changed from irregularity into aggregation. Simultaneously, the existence of H_3PO_4 and increase in hydroxyl group of waterish mixtures is due to the decomposition of $NH_4H_2PO_4$, which could promote microwave absorption. In addition, dense mixtures could change into glass after microwave irradiation with increasing pressing pressure. The connection between mixtures with and without adding water or pressing pressure and degree of microwave absorption, and how they are influenced on glass formation is discussed.

1. Introduction

Nowadays, microwave processing of materials has opened a new area of research in industrial and medical domains [1]. The research areas include the basic theory of the interaction between microwave and materials, microwave sintering [2], refining and reheating for forming and thermal toughening [3], joining, drying, and synthesis, microwave remediation of nuclear waste [4], glass sealing [5], optical glass [6], bio-glass [7], and so on. Microwave technology uses electromagnetic waves that pass through material and cause its molecules to oscillate and then generate heat. As compared with conventional heating, microwave heating generates heat within the material and heats the entire volume at about the same rate, which results in volumetric heat generation.

Phosphate glasses with low dispersion and relatively high refractive indices were developed for achromatic optical elements for several years ago [10]. By adding different elements, phosphate glasses have been developed for a variety of specialty applications. For example, alkali aluminophosphate compositions are used for specialty hermetic seals [11], zinc phosphate compositions can be co-formed with high temperature polymers to produce unusual organic/inorganic composites [12], iron phosphate glasses have led to their development as nuclear waste hosts [13], Biocompatible phosphate glasses have medical applications [14], and amorphous lithium phosphate [15] and phosphorus oxynitride glasses [16] are useful as solid state electrolytes. Traditionally, phosphate glasses were frequently made by conventional heating at high temperature in furnace following mixing of raw materials, which consumes time and energy. By using microwave heating, some of the main advantages include very short processing times obtainable due to rapid, selective and volumetric heating. Thus, the main interest in microwave processing lies in discovering new ways to utilize the microwave energy as an alternative in materials processing to substantially save time and energy consumption. Consequently, in this study, the main objectives are the evaluation and understanding of microwave irradiating on forming of phosphate glass.

2. Experimental procedure

Ammonium dihydrogen phosphate ($NH_4H_2PO_4$) (0.72 mol), 0.07 mol of calcium hydroxide ($Ca(OH)_2$), and 0.21 mol of lithium carbonate (Li_2CO_3) were used as raw materials. In order to compare the effect of different types for mixtures on microwave heating, three different types of samples are used in this study: raw mixtures, waterish mixtures, and dense mixtures. Appropriate amounts of each salt were dry mixed together in a polypropylene bottle for 6 h to ensure through mixing by using a Turbula powder mixer, that is, raw mixtures. The waterish mixtures are produced by adding suitable amount of water (25 wt%) into raw mixtures and well mixed. In addition, the raw mixtures were putted into a mold and then prepared using a hydraulic press operated at the

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doi:10.1016/j.jnoncrysol.2009.04.002

maximum pressure of 10 MPa to obtain dense mixtures. Afterward, all types of mixtures were placed into clay crucibles and then transferred to a microwave oven (SANYO EM-23PDS). The MW irradiation was performed with a power setting of 900 W/h and irradiation time of 1–15 min, respectively.

The skin temperature of mixtures in microwave oven was measured by Infrared Ray (IR) pyrometer. The dielectric measurement (tangent loss) was carried out at frequency 1 MHz using LCR meter (Agilent Technologies, 4284A) with a voltage of 5 V. The test pellets (12.9 mm in diameter and 0.5 mm in thickness) of mixtures are pressed under pressure of 250 kgf/cm^2. In order to detect and identify the crystalline phases formed during the irradiation, the products formed in a crucible was ground in an agate mortar and then subjected to X-ray diffraction [(XRD), SHIMADZU XRD-6000]. The surface morphology of the samples was characterized by scanning electron microscopy [(SEM), JEOL JSM-5160]. Fourier transform infrared absorption spectroscopy [(FTIR), BIO-RAD FTS-3000MX] was employed to characterize the bonding configurations of these samples after microwave irradiation. The samples for FTIR analyses were mixed with KBr powder (The weight ratio of the sample to KBr is 1:200.) and pressed into pellets. Portions of each sample were crushed to powder and the glass transition temperature (Tg) determined using Thermogravimetric Analyzer [(TGA), TA Q50], performed under nitrogen with a heating rate of 5 °C/min.

3. Results and discussion

Fig. 1 presents SEM morphology of raw mixtures, waterish mixtures, and dense mixtures, respectively. The particle morphology of raw mixtures prepared by different powders after well mixing for 6 h are not of uniform size and irregularity, as shown in Fig. 1(a). Simultaneously, separation of the majority of particles is also observed in the raw mixtures. From Fig 1(b), with adding water into raw mixtures, the morphology changes into aggregation. On the other hand, separation particle of raw mixtures turn into aggregated and dense after pressing, as shown in Fig. 1(c). The influence of raw mixtures with and without adding water or pressing pressure on microwave irradiation was examines as follows.

During microwave irradiation, the material dielectric loss (or loss tangent) is a function of microwave frequency, which is responsible to convert electric energy into heat. Thus, dielectric loss tangent is a measure of the dipole's ability to absorb microwave energy [8,9]. In this study, the loss tangent of $NH_4H_2PO_4$, Li_2CO_3, and $Ca(OH)_2$ are 0.015, 0.01, and 0.157, respectively. For this reason, the temperature of mixtures of $NH_4H_2PO_4$, Li_2CO_3, and $Ca(OH)_2$ increase after irradiating microwave (described later). Fig. 2 gives the ratio of weight loss of raw mixtures and waterish mixtures as a function of irradiation time. The ratio of weight loss is defined as ratio of weight difference of mixtures before and after microwave irradiation to weight of mixtures before microwave irradiation, which represents the degree of species vaporized by microwave irradiation. From Fig. 2, the ratios of weight loss of waterish mixtures are larger than that of raw mixtures all the irradiation time, which implies waterish mixtures are subjected to high temperature. In addition, the ratios of both mixtures increase abruptly with increasing irradiation time after irradiating for 0–3 min, which indicates lots of species vaporized from both mixtures. Simultaneously, the ratios increase gradually while the irradiation time varied from 3 to 6 min. Furthermore, one sees that the ratios are nearly the same when irradiation time exceeded 6 min, resulting in the species no longer vaporized. The reason for higher temperature of waterish mixture than that of raw mixtures after microwave irradiation will be discussed as follows.

Fig. 3 shows the FTIR absorption spectra of raw mixtures and waterish mixtures before microwave irradiation. The main characteristic modes of pure Li_2CO_3 and $Ca(OH)_2$ powders before and after adding water are almost the same except for increase in the intensity of hydroxyl peak (not shown), which deserve to be mentioned. In the spectra presented in Fig. 3, the characteristic mode of raw mixtures at ~3500 cm^{-1} is due to the stretching vibration of OH group connected with the presence of $Ca(OH)_2$ [17]; simultaneously, the intensity of polar hydroxyl peak increases after adding water into raw mixture. Cheshkova and Stoilova reported that OH groups (3570–3500 cm^{-1}) perturbed through their oxygen atoms due to formation of ammonia [18]. Also, NH_4^+ in the ammonia should be responsible for the broad and strong absorption band

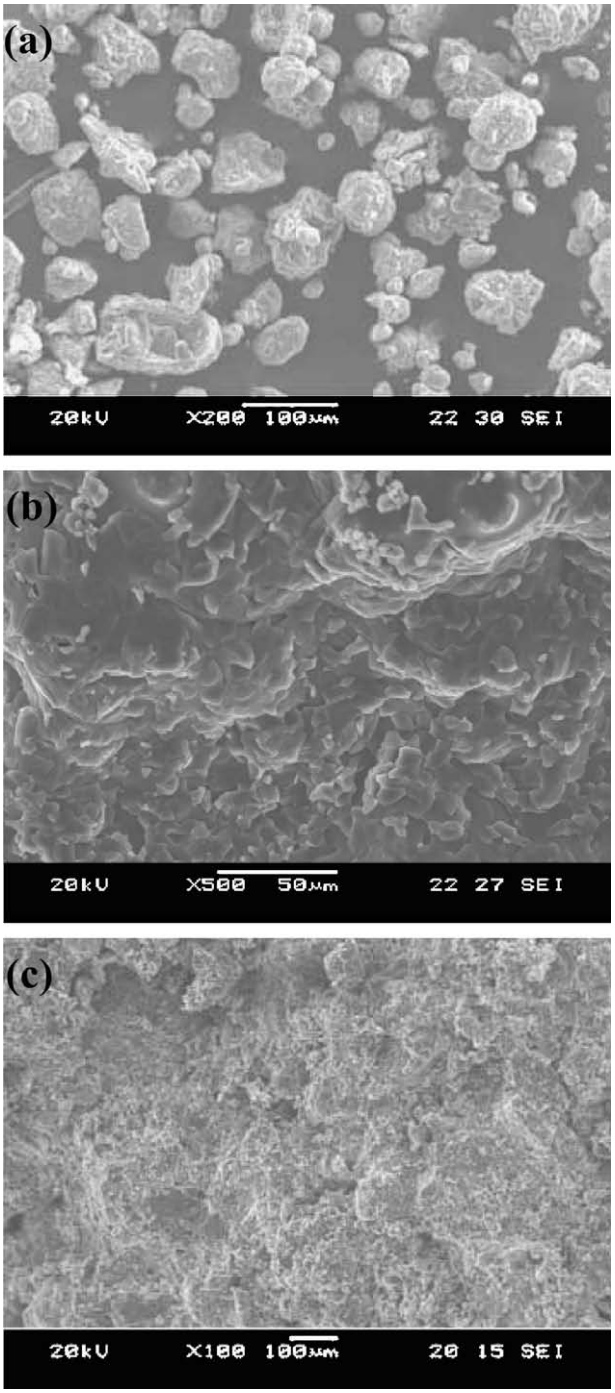

Fig. 1. SEM micrographs of (a) raw mixtures, (b) waterish mixtures, and (c) dense mixtures.