Materials Letters 175 (2016) 165–168

Contents lists available at ScienceDirect

Materials Letters

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

Rapid and efficient disposal of radioactive contaminated soil using microwave sintering method

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ARTICLE INFO

Article history:
Received 14 December 2015
Received in revised form
1 March 2016
Accepted 2 April 2016
Available online 4 April 2016

Keywords:
Radioactive contaminated soil
Microwave
Sintering
X-ray techniques
Glass waste forms

ABSTRACT

To effectively dispose the radioactive contaminated soil, a new technology to immobilize radionuclide is desired. In this study, microwave sintering technology was employed to vitrify the radioactive contaminated soil waste and immobilize the radionuclide. Radioactive contaminated soil has been sintered by microwave and conventional processes. The glass waste forms can be obtained after microwave sintering at low temperature (1300 °C) for short time (30 min). The microwave sintered samples were denser than the samples conventionally sintered at higher temperature (1500 °C) for the longer time (2 h). Moreover, it was found that the simulated radionuclide Nd can be successfully immobilized and uniformly distributed in glass waste forms.

1. Introduction

Radioactive contaminated soil is a kind of nuclear waste mainly comes from the nuclear test, nuclear weapons usage and nuclear accident etc [1–3]. Such nuclear waste pose a considerable biological hazard [4]. Thus, how to safe and efficient disposal of the radioactive contaminated soil is a pressing environmental problem faced by the nuclear industry today. Vitrification of these radioactive contaminated soils is an emerging option [5]. The most commonly used treatment method for radioactive contaminated soil is conventional sintering [6–8]. As a conventional sintering method to dispose the radioactive contaminated soil, the process is very sample, but high sintering temperature and long holding time are essential. Therefore, the development of a rapid and efficient technology for treatment of the radioactive contaminated soil has become increasingly important and essential. Microwave sintering, a promising synthesis method, has been widely employed to prepare various materials in recent times [9–11]. Although microwave heating was introduced over 50 years ago, its use in the treatment of radioactive contaminated soil is relatively new. There are some advantages, including high efficiency and no byproduct [12,13], if this method is introduced to dispose radioactive nuclear waste. Compared with conventional sintering treatment technologies, the microwave technique with its characteristics of polar oscillation and effect of dielectric losses offers the advantage of selective, uniform and rapid heating [14]. The application of microwave radiation heating is widely used in many fields, including sintering of ceramics [15–18], synthesis of composites [19,20]. Moreover, a growing interest in the immobilization of metal ions in soil through microwave radiation has also been reported [21]. Therefore, it is possible to use microwave sintering method to effectively dispose radioactive contaminated soil.

This project aims to investigate the disposal of radioactive contaminated soil by microwave sintering method. Three main goals were pursued: (1) determine the proper procedure for disposal of radioactive contaminated soil by microwave sintering, (2) discuss the effects of sintering temperature and holding time, (3) understand the benefit of microwave sintering.

2. Experimental

The composition of soil was listed in Table 1. The soil (200 mesh) and simulated radionuclide Nd^{3+}(Nd_2O_3, AR) were mixed with the weight ratio of 1:0.03, then sintered under microwave (2.45 ± 0.025 GHz) at 1100–1300 °C for 15–30 min in ambient air.

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http://dx.doi.org/10.1016/j.matlet.2016.04.018
0167-577X/

Table1
Composition of soil.

Oxide	SiO_2	Al_2O_3	Fe_2O_3	CaO	K_2O	MgO	Na_2O	TiO_2
Content (wt%)	66.32	16.57	5.87	4.67	2.86	1.66	0.81	0.74

In order to make it easier to understand the benefit of microwave sintering, some of the mixtures were sintered by the conventional sintering method at 1200–1500 °C for 0.5–2 h in ambient air.

The composition of the soil was measured by X-ray fluorescence analysis (XRF, PANalytical B.V. Axios). The crystal structure of soil samples before and after sintering processes were determined by X-ray diffraction (XRD) to determine the species variation. The microstructure of the sintered samples was observed by scanning electron microscope (SEM, Ultra 55. Germany). The density of each sample was measured at room temperature using the Archimedes method with water as an immersing liquid. Raman spectra were acquired using a Renishaw Invia Raman microscope at the room temperature.

3. Results and discussion

Samples have been analyzed by XRD to confirm their amorphous or crystalline nature. Fig. 1 shows the XRD results of simulated radioactive contaminated soil samples before and after sintered by microwave and conventional method. As seen from the figures, the patterns of samples sintered by microwave are typical of X-ray amorphous when sintering temperature reached 1300 °C. However, as can be seen from the patterns of samples sintered by conventional method, the amorphous phase occurs when sintering temperature reached as high as 1500 °C. The XRD analysis of samples indicates the amorphous character is expected. It was found that the simulated radioactive contaminated soil converted into glass after being radiated with microwave at low temperature (1300 °C) for short time (30 min). However, when the conventional sintering method was employed, the sample converted into glass after being sintered at higher temperature (1500 °C) for longer time (2 h). This may be due to the microwave technique with its characteristics of polar oscillation and effect of dielectric losses offers the advantage of rapid heating [14].

Fig. 2 shows the Raman spectra of samples sintered by microwave at different temperatures. Generally, the band located in the range of 1100–1300 cm^{-1} is associated with the asymmetric

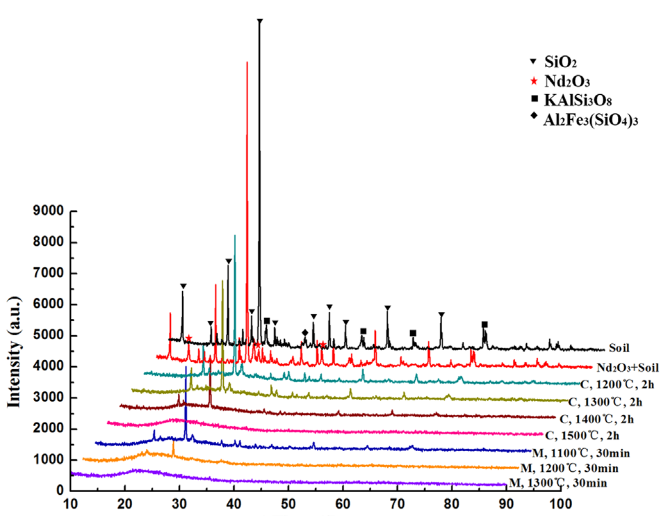

Fig. 1. XRD patterns of samples. (M: microwave sintering, C: convention sintering).

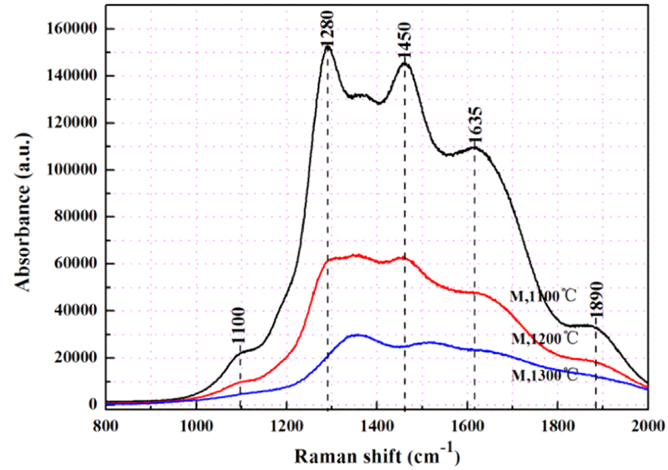

Fig. 2. Raman spectra of samples sintered by microwave at different temperatures.

stretching Si–O–Si mode, the peaks shift to longer wavelengths has been attributed to a gradual shift in the Si–O stretching frequency or to the Al neighbors connected to SiO_4 tetrahedron [22]. According to the electronic energy levels of neodymium [23], the weak bands were found around 1900 cm^{-1} in the Raman spectra of neodymium silicate [24]. It can be found that the full width at half maximum (FWHM) increased and the wave crest shifted to the high band region with increasing sintering temperature. This may be due to the high sintering temperature result in increasing irregularities of bond-lengths and bond-angles and a general breaking-up of the structure [25]. It indicates that the samples transformed into an amorphous structure. This is good agreement with the result of XRD. Thus, the glass waste forms can be obtained by microwave sintering at 1300 °C for 30 min.

The densities of the glass waste forms obtained from microwave and conventional sintering were given in Fig. 3. For microwave sintering, the density increased from 1.91 to 2.84 g/cm^3 as the temperature increased from 1100 to 1300 °C. However, the density of glass obtained from conventional sintering increased from 1.86 to 2.28 g/cm^3 as the temperature increased from 1200 to 1500 °C. The sintered density slightly increased as increasing temperature and time. This phenomenon was also found by Fang et al. [26]. This result indicates that the denser glass waste forms can be obtained by means of microwave sintering method at lower sintering temperature and shorter time. When conventional sintering was employed, the densification kinetics was sluggish in the temperature range up to 1500 °C because of the incompact and

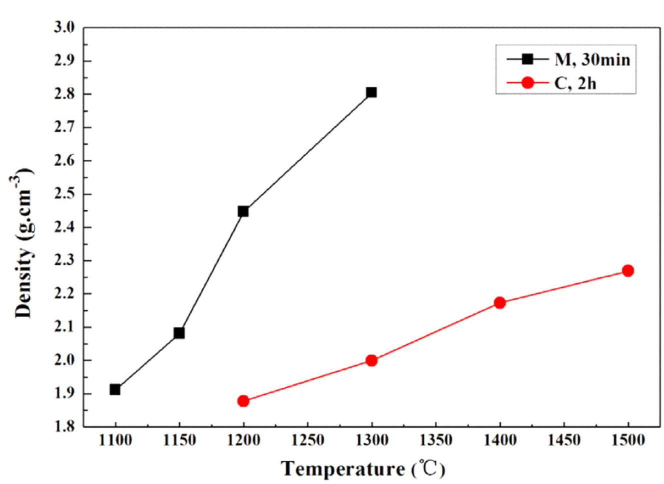

Fig. 3. The density of the obtained glass waste forms.