



Effect of rare earth Ce on the far infrared radiation property of iron ore tailings ceramics



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ABSTRACT

A kind of far infrared radiation ceramics was prepared by using iron ore tailings, CaCO₃ and SiO₂ as main raw materials, and Ce as additive. The result of Fourier transform infrared spectroscopy showed that the sample exhibits excellent radiation value of 0.914 when doping 7 wt.% Ce. Ce⁴⁺ dissolved into iron diopside and formed interstitial solid solution with it sintered at 1150 °C. The oxidation of Fe²⁺ to Fe³⁺ caused by Ce⁴⁺ led to a decrease of crystallite sizes and enhancement of Mg–O and Fe–O vibration in iron diopside, which consequently improved the far infrared radiation properties of iron ore tailings ceramics.

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1. Introduction

Far infrared ceramics, which can radiate infrared light waves with specific wavelengths, are widely used in fuel saving, air purification, health care, etc. For example, far infrared ceramics have the ability to facilitate atomization and full-combustion of fuel oil by way of reducing its surface tension [1]. Moreover, far infrared ceramic coatings have a catalytic function. It can generate OH⁻ under the sunlight, which could effectively remove indoor formaldehyde, benzene, sulfide, ammonia, or odor material. Therefore, far infrared ceramics are considered to be good functional ceramics for nature environment.

Tailings are a kind of solid waste derived from mining, attracting more and more attention as a secondary resource recycling [2–6]. It can be applied to materials preparation in many fields, such as building brick, concrete, mesoporous materials, and microcrystalline glass [7–13]. Using iron ore tailings (IOT) to prepare far infrared ceramics are based on the following reasons: first, similar chemical compositions. IOT are mainly composed by SiO₂, Al₂O₃, Fe₂O₃, CaO, and MgO, corresponding to the minerals of quartz, hematite, calcite, feldspar, etc. Coincidentally, far infrared

ceramics with high emissivity are just composed by these materials mentioned above. Second, the lower production cost. Far infrared materials derived from purified oxides or expensive rare minerals increase the production cost, while IOT with low price are relatively easy to obtain. Furthermore, this approach has played an important role in turning waste into treasure.

Rare earths have the reputation of “industrial vitamin”, which are used in the field of ceramics in order to improve their performance. Liang et al. [14–17] have reported the performances and applications of some kinds of far infrared ceramics prepared by rare earth and minerals. It was found that appropriate content of rare earth could enhance the property of far infrared ceramics. Two reasons are summarized here: (1) in the view of quantum mechanics, the decrease of multi-phonon relaxation rate can promote property of far infrared radiation ceramics [18–20]. (2) Lattice distortion and grain refinement are the main affecting factors [21,22]. Rare earth can either cause internal defect through replacing cation in ceramic matrix [23,24], or form independent phase at grain boundaries [25–27]. For example, rare earth La is able to enhance Fe–O vibration via replacing Fe in the perovskite [28,29]. In addition, CeO₂ and Eu₂O₃ can limit the crystal growth and reduce the average grain size of ceramics through moving to the grain boundaries, forming an isolating ultra-thin layer around the grains [28].

In this paper, IOT was used to replace natural minerals as the main raw materials, and Ce as additive to prepare far infrared ceramics. The effect of Ce on the far infrared radiation property was

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systematically studied. It is expected that this study can be a significant step in promoting the functional applications of IOT.

2. Experimental

2.1. Synthesis of samples

The raw materials were IOT, CaCO_3 , SiO_2 , and $\text{Ce}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ (analytical grade, 99.0%). IOT came from Hebei province of China, which is the remaining materials derived from iron ore by crushing, sieving, grinding, and flotation. Its chemical compositions were given in the mass ratio as follows: SiO_2 , 30.09%; Fe_2O_3 , 28.02%; Al_2O_3 , 11.73%; TiO_2 , 10.97%; CaO , 5.64%; MgO , 4.33%; Na_2O , 1.78%; loss on ignition, 5.67%; others, 1.77%. The weight percentages of Ce in the experimental group are 3, 5, 7, 9, and 11 wt.%, noted as C3, C5, C7, C9, and C11, respectively; while in the control group was noted as C0.

Given below are the experimental details. The raw materials were first dissolved in deionized water to obtain a mixed solution with an appropriate proportion (IOT: CaCO_3 : SiO_2 = 6:1:1, the ratio was obtained according to results of orthogonal experiments).

The solution was dried at 120 °C for 6 h and then pressed into ceramic body at 70 MPa. Then the ceramic bodies were put into a furnace for finally sintering. The prepared ceramics were crushed into fine powder with the help of an agate mortar. The sintering process is as follows: Stage I, room temperature to 900 °C with heating rate of 2.4 °C/min; Stage II, 900–1150 °C with heating rate of 2.8 °C/min; Stage III, 1150 °C was kept for 2 h; Stage IV, slowly cooled to room temperature.

2.2. Characterization

The far infrared emission and absorption properties of samples were tested by Fourier transform infrared spectroscopy (FTIR, Bruker-80V, Germany). The crystalline phases were identified by X-ray diffraction (XRD, Philips-X'Pert MPD, Netherlands) with $\text{CuK}\alpha$ radiation ($\lambda = 0.15406$ nm). The X-ray photoelectron spectra (XPS, Thermo Fisher Scientific K-Aepna, USA) were carried out using a spectrometer with AlK α X-ray source ($h\nu = 1486$ eV), and an electron energy analyzer with a constant pass energy of 50 eV. Scanning electron microscope (SEM, FEI NanoSEM 430, USA) and transmission electron microscopy (TEM, Philips Tecnai G²F20, Netherlands) were used to characterize the microstructure of samples.

3. Results and discussion

3.1. Far infrared radiation property

The histograms of far infrared emissivity are plotted as Fig. 1. It is important to note that with increasing doping Ce the value reaches its maximum at $x=7$, and gradually decreases with a further increasing in Ce content. In our experiment, the value sequence for the far infrared emissivity is $\text{C7} > \text{C5} > \text{C9} > \text{C11} > \text{C3} > \text{C0}$. The optimal doping content of Ce for the sample is 7 wt.%. The upper inset in Fig. 1 presents the far infrared emission spectrum of C7.

3.2. Phase compositions and lattice parameter

XRD patterns of the as-prepared samples are shown in Fig. 2. As can be seen, the main crystalline phases are anorthite [$(\text{Na}_{0.45}\text{Ca}_{0.55})(\text{Al}_{1.55}\text{Si}_{2.45}\text{O}_8)$, JCPDS No. 71-0748] and iron diopside [$\text{Ca}_{0.991}(\text{Mg}_{0.641}\text{Fe}_{0.342})(\text{Si}_{1.6}\text{Fe}_{0.417})\text{O}_6$, JCPDS No. 89-0837], while the second crystalline phases are ceria [CeO_2 , JCPDS No. 81-0792] and pseudobrookite [Fe_2TiO_5 , JCPDS No. 70-2728]. The sharp

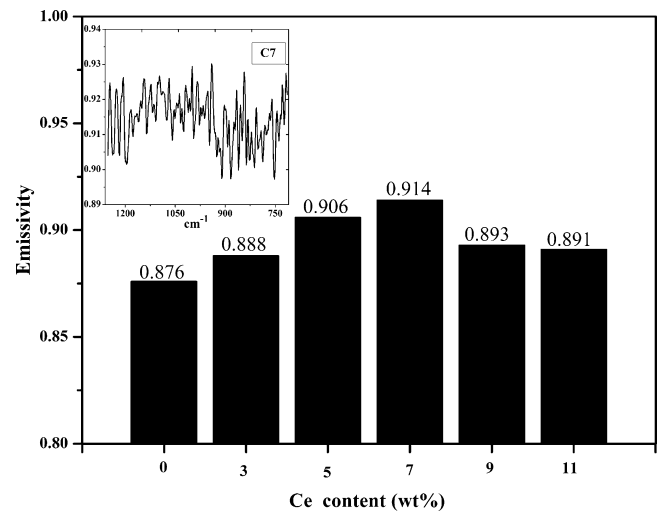


Fig. 1. Far infrared emissivity of the ceramic powder samples.

diffraction peaks of CeO_2 indicate the good crystallinity with increasing Ce content.

Further analysis shows that iron diopside's crystallite size changed to a certain extent when calculated by Debye Scherrer's formula [30,31]:

$$D = \frac{k\lambda}{\beta \cos \theta_{hkl}} \quad (1)$$

where D is the crystallite size (nm), k is the shape factor ($k = 0.89$), λ is the X-rays wavelength ($\lambda = 0.154056$ nm), θ_{hkl} is Bragg diffraction angle (°), and $\beta = \sqrt{\beta_{\text{measured}}^2 - \beta_{\text{instrumental}}^2}$ is the full width at half maximum (FWHM) of an estimated peak (rad). The result of crystallite sizes corresponding to various planes and their averages (\bar{D}) are shown in Table 1.

It can be seen that \bar{D} decrease first and then increase, reaching its minimum with doping 7 wt.% Ce. This result has a certain correlation with the far infrared emissivity of samples. The most likely explanation for this is that the transformation from Fe^{2+} to Fe^{3+} oxidized by Ce^{4+} because of the radius of Fe^{2+} (0.078 nm) is larger than that of Fe^{3+} (0.0645 nm) [32,33].

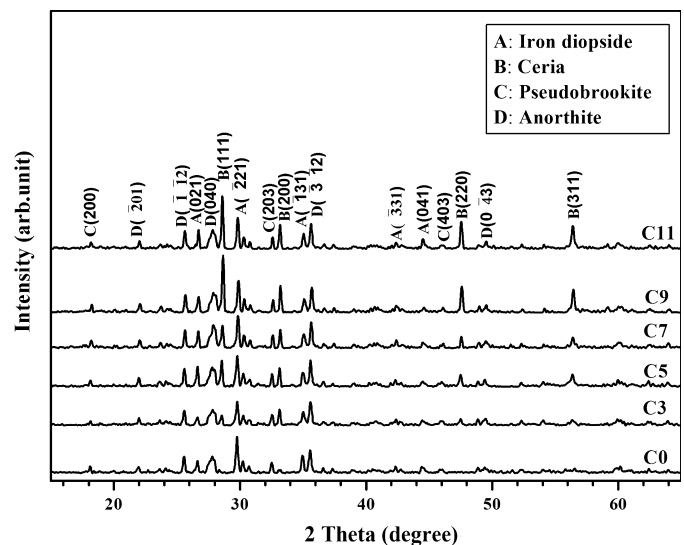


Fig. 2. XRD patterns of samples doped with different Ce content.

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