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Original Research Paper

Synthesis of <alpha>-cordierite nanoparticles from bentonite using thermal shock assisted solid-state reaction method

A. Cheraghi [a,b], H. Shoushtarizadeh [a], R. Malekfar [a,*], O. Alizadeh [c]

[a] Department of Physics, Tarbiat Modares University, P.O. Box 14115-175, Tehran, Islamic Republic of Iran
[b] Faculty of Basic Sciences, Shahid Sattari University, Tehran, Islamic Republic of Iran
[c] Department of Materials Engineering, Tarbiat Modares University, Tehran, Islamic Republic of Iran

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ABSTRACT

For the first time, we believe, nanosized <alpha>-cordierite glass-ceramics are produced using bentonite, talc, alumina, and kaolin as the raw materials and applying thermal shock to the precursor powders and sintered at 1100, 1200, and 1300 °C. A combination of a furnace at about 800 °C and liquid nitrogen was used for the applied thermal shock with a total temperature difference of about 1000 °C. The effects of thermal shock process and sintering temperature on <alpha>-cordierite formation and microstructure have been investigated. The results show that <alpha>-cordierite was formed above 1160 °C and its weight ratio increased continuously as sintered temperature increased to 1300 °C. By increasing the temperature, <alpha>-cordierite nanoparticles grain sizes and the intensity of FTIR peaks started to increase. Applying thermal shock to precursor powders reduced the grain size of each consisting mineral and resulted in nanosized <alpha>-cordierite powder.

1. Introduction

<alpha>-cordierite is the most popular glass-ceramic for its interesting properties including high mechanical strength, good wear resistance, low thermal expansion, high refractoriness, and relatively low dielectric coefficient [1–6]. <alpha>-cordierite glass-ceramics are widely used in many industrial branches such as heat exchangers, gas turbines, and high speed radomes [3,7–9]. Several approaches have been proposed to synthesize cordierite; mainly categorized in two groups; based on solid-state reaction of stoichiometric oxides and on wet chemical methods [6,10–12]. Expensive precursors, low yield, and sophisticated processing are main disadvantages of wet chemical methods. Replacement of high purity oxides with natural and more affordable raw materials have been widely investigated in recent years. Talc, kaolin, andalusite, rice husk, gold tailing, and aluminium rich anodizing sludge have been used as raw materials in solid-state reaction method [2,5,8,13–16].

Solid state reaction method based on thermal shock can be considered as a low cost and very simple technique in comparison to the sol-gel method. Radomes and cordierite family ceramic compounds synthesized by this method have excellent properties and can be used as suitable high temperature electrical insulators. They also have low thermal expansion coefficients resulting in their high thermal resistivity.

In this work, <alpha>-cordierite nanoparticles were prepared by thermal shock assisted solid-state reaction method using kaolin, talc, bentonite, and alumina as raw materials. A combination of a furnace at about 800 °C and liquid nitrogen (−196 °C) was used for the applied thermal shock with a total temperature difference of about 1000 °C. XRD, SEM, FTIR, Raman spectroscopy, and TGA/DTA are used to characterize phases, morphologies, and temperature of phase formation.

2. Experimental procedures

2.1. Materials

In order to synthesize cordierite glass-ceramic nanopowders by solid reaction method, naturally occurring minerals taken from different mines in Iran were used as raw precursor materials Bentonite, talc, kaolin, and alumina were obtained from Ferdows, Naeen, Yazd, and Isfahan mines (Iran), respectively.

2.2. Equipment

Differential thermal analysis (DTA) and thermo-gravimetric analysis (TGA) of the powders, and for a typical sample mass of 5 mg, were performed (using a DTA-TG Netzsch STA 409 PC/PG

* Corresponding author.
E-mail address: malekfar@modares.ac.ir (R. Malekfar).

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instrument) up to a temperature of 1200 °C with the heating rate of 10 °C·min^{-1} and Al_2O_3 as the standard sample. The type of crystalline phases of the heat-treated samples was identified from XRD patterns, obtained by an X-ray diffractometer (model X'pert 1480 manufactured by Philips Co.) using Co K_α radiation. Scanning Electron Microscope (SEM) with EDAX apparatus (model XL30, manufactured by Philips Co.) was used to characterize mineral composition, size and morphology of the nanoparticles. The FTIR absorption spectra of the grown samples were collected using an FTIR spectrometer (model Nexus 670 manufactured by Thermo Nicolet Co.) with 4 cm^{-1} resolution. Raman spectra were obtained using a back-scattering dispersive Raman spectrometer (model Almega Dispersive, manufactured by Thermo Nicolet Co.) in the spectral region of 350–1150 cm^{-1} with 4 cm^{-1} resolution.

2.3. Preparation of <alpha>-cordierite nanoparticle

Talk ($Mg_2Si_4O_{10}(OH)_2$), kaolinte ($Al_4Si_4O_{10}(OH)_8$), bentonite ($Na,Ca)_{0.22}(Al,Mg)_2Si_4O_{10}(OH)_2$, and alumina ($Al_2O_3$), with the composition concentrations shown in Table 1 have been used. The mixtures with near stochiometric ratios near to cordierite ($2MgO.2Al_2O_3.5SiO_2$) formula and according to Table 2 were used.

Results of the EDAX analysis of raw mineral materials are presented in Table 3. The EDAX results were used to adjust the wt.% ratio of each raw mineral in the starting powder according to the nominal stoichiometry of <alpha>-cordierite.

For selecting the precursor particles in a limited size range of 37–74 μm, starting powders were sieved using 200 and 400-mesh stainless steel sieves and dry-milled for 24 h at a rotation speed of 250 rpm using a planetary ball mill. To prepare nanosized <alpha>-cordierite, a 1000 °C thermal shock was applied to the precursor powders as follows: the precursor powder was heated to 800 °C at a rate of 10 °C·min^{-1} and with a soaking period of 2 h at 800 °C. Then, the heated powder was suddenly dropped into liquid nitrogen (−196 °C) with subsequent dry milling of the resulting powder for 10 h at a rotation speed of 250 rpm using a planetary ball mill. This process was repeated for each sample.

In order to synthesize <alpha>-cordierite nanoparticles, the above mentioned powders were mixed according to the required stoichiometric ratios and the resulting powder was heated up to 1100, 1200, and 1300 °C at a rate of 2 °C·min^{-1} and sintered at these temperatures for 4 h.

Table 1
The mixture composition used for the synthesis of cordierite ceramic from raw precursor minerals (at.%).

Alumina	Bentonite	Kaolinte	Talk	Compositions
0	11.92	46.55	60.64	SiO_2
98.6	43.48	39.5	0.45	Al_2O_3
0	12.82	0	0.1	Fe_2O_3
0	14.39	0	31.1	MgO
0	0	0	1.67	CaO
1.4	17.87	13.96	7.44	Other impurities

Table 2
The mixtures of raw mineral materials used with the near stochiometric ratios to cordierite ($2MgO{\cdot}2Al_2O_3{\cdot}5SiO_2$) formula.

32.26	0.73	1.64	64.17	Wt.%
Alumina	Bentonite	Kaolinte	Talk	Raw mineral material

Table 3
The EDAX results of bentonite, talc, kaolin, and alumina as the raw materials (wt.%).

Element	Bentonite	Talc	Kaolin	Alumina
Mg	0.72	9.69	0.31	0.4
Al	5.6	1.02	9.76	19
Si	33.32	32.2	19.94	3.93
K	0.35	0.18	0.28	5.63
Ca	1.49	1.37	0.2	0.84
Ti	0.18	0.2	0.27	0.53
Fe	0.82	0.25	0.4	2.03
O	56.92	54.7	68.12	66.94

3. Results and discussion

The TGA/DTA curves of the synthesized <alpha>-cordierite nanoparticles are depicted in Fig. 1. Below 220 °C, there is an endothermic DTA peak at 111.3 °C with about 3.6% mass loss, which occurs due to the removal of physically adsorbed water. The second stage of mass loss between 220 and 600 °C, accompanied with an endothermic DTA peak at 461.4 °C can be attributed to the removal of the water trapped in the structural pores of the raw mineral materials with about 7.6% mass loss. The third stage occurred at 600–1140 °C with two endothermic DTA peaks at 958.9 and 1072.5 °C is associated with a mass loss of 3% due to the desorption of coordination water from bentonite, talc, kaolin, and alumina.

An exothermic peak was observed at 1160.8 °C without mass loss in the DTA curve; indicating a phase transition. In other words, the minimum formation temperature of <alpha>-cordierite is 1160.8 °C.

Fig. 2a–c shows the XRD patterns of the powder mixtures after sintered at 1100–1300 °C. At 1100 °C, the main phases were enstatite (JCPDS No. 19-0605) and dolomite (JCPDS No. 79-1342), and spinel phase (JCPDS No. 75-1796) was as an impurity and small amount of <alpha>-cordierite (JCPDS No. 84-1219) crystallized. The main phase at 1200 °C was still enstatite but the intensities of this phase decreased and the intensities of spinel and <alpha>-cordierite increased. This is in agreement with TGA/DTA results. At 1300 °C, the main phase is <alpha>-cordierite (JCPDS No. 84-1219). Due to the grain growth, the FWHM of <alpha>-cordierite peaks decreased as the temperature increased from 1200 to 1300 °C.

In Tables 4–6 the diffraction angles (peak positions) of the XRD patterns due to the glass-ceramic synthesized at 1100, 1200 and 1300 °C and their other related characteristics are tabulated.

As it is clear from Tables 4–6 at temperature 1100 °C the dominant phases in the sample are quartz and spinel and the cordierite phase gradually starts to appear. At 1200 °C due to the increase in

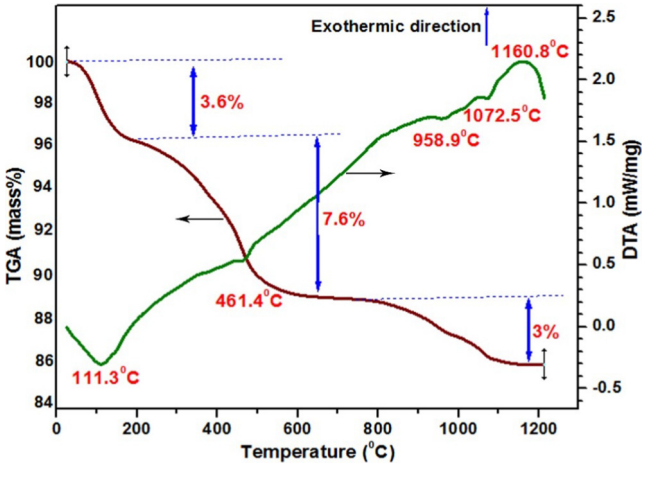

Fig. 1. TGA/DTA curves of the synthesized <alpha>-cordierite nanoparticles.

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