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Characteristics of $BaNd_2Ti_5O_{14}$ powders directly prepared by high-temperature spray pyrolysis

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Abstract

$BaNd_2Ti_5O_{14}$ powders were directly prepared by high-temperature spray pyrolysis. The powders prepared at temperatures of 1300 and 1500 °C exhibited a pure $BaNd_2Ti_5O_{14}$ phase. The powders prepared at 1300 °C were spherical in shape. However, the powders prepared at 1500 °C showed non-spherical shapes. The $BaNd_2Ti_5O_{14}$ powders had a composition similar to that of the spray solution. The mean sizes of the $BaNd_2Ti_5O_{14}$ powders increased from 0.23 to 0.60 μm when the concentration of the spray solution was increased from 0.01 to 0.2 M. At a sintering temperature of 1100 °C, bridge-like structures were formed between the powders. Pellets sintered at 1300 °C exhibited a dense structure comprising rod-like crystals.

Keywords: A. Powders: gas phase reaction; A. Powders: chemical preparation

1. Introduction

Spray pyrolysis has several advantages in the preparation of multicomponent ceramic powders containing particles with a fine size and spherical shape [1–3]. For instance, spray pyrolysis enables a high degree of mixing of the constituents of the multicomponent ceramic, thereby decreasing the preparation temperature. However, the residence times of the powders inside the hot wall reactor are short, less than tens of seconds. Therefore, the precursor powders obtained by spray pyrolysis require post-treatment at high temperatures to improve crystallinity and phase purity. The post-treatment process causes deformation of the spherical shape of the particles and leads to particle aggregation. In particular, it is observed that fine powders comprised of particles having sizes in the range of several hundred nanometers possess low thermal stability.

High-temperature spray pyrolysis has been applied to the direct preparation of multicomponent ceramic powders without the post-treatment process [4–7]. The particles of ceramic powders directly prepared by high-temperature spray pyrolysis had a spherical shape, showed high crystallinity, and did not undergo particle aggregation. However, this pyrolysis method will cause composition deviation due to the evaporation of some components of the multicomponent ceramics.

$BaNd_2Ti_5O_{14}$ (BNT), an important compound of the Ba–Nd–Ti–O system, is known to be an important microwave dielectric material because of its high dielectric constant, low dielectric loss, and near-zero temperature coefficient of resonant frequency [8–12]. Fine powders are required to decrease the sintering temperature of BNT. BNT powders prepared by conventional solid-state reaction and liquid solution methods exhibited large particle sizes and irregular morphologies.

In this study, BNT powders were directly prepared by high-temperature spray pyrolysis. The effects of the preparation temperature on the morphologies and crystal structures of the BNT powders were investigated. The effect of the concentration of the spray solution on the mean particle size was also investigated.

2. Experimental

The spray pyrolysis equipment used consisted of six ultrasonic spray generators operating at 1.7 MHz, a tubular

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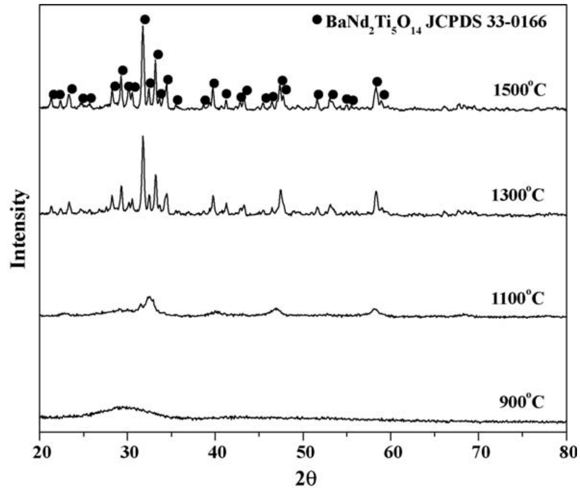

Fig. 1. XRD patterns of the BNT powders directly prepared by spray pyrolysis.

alumina reactor of 1000 mm length and 25 mm ID, and a bag filter. The general flow diagram of the spray pyrolysis process is given elsewhere [13]. The BNT powders were prepared by spray pyrolysis at temperatures between 900 and 1500 °C. The starting materials used in the synthesis of BNT powders were barium carbonate, neodymium nitrate, titanium tetra-*iso*-propoxide (TTIP). A small amount of nitric acid was used to peptize the hydrolyzed TTIP and form a clear solution. The concentrations of metal components were changed from 0.01 to 0.2 M. The flow rate of air used as the carrier gas was 5 L/min. The prepared BNT powders were pelletized under 500 MPa pressure into a 15 mm diameter and thickness of 1 mm. The pellets were then sintered at temperatures between 1000 and 1300 °C for 3 h and cooled naturally to room temperature.

The crystal structures of the BNT powders were investigated using X-ray diffraction (XRD) with Cu Kα radiation (λ 1.5418 × 10^{-1} nm). The morphological characteristics of the powders were investigated using scanning electron microscopy (SEM). The specific surface areas were measured by Brunauer–Emmet–Teller (BET) method using N_2 adsorption. The pore size distributions of the powders were analyzed by Barrett–Joyner–Halenda (BJH) method.

3. Results and discussion

The crystal structures of powders directly prepared by spray pyrolysis at various temperatures are shown in Fig. 1. The concentration of the spray solution was 0.2 M. The powders prepared at 900 °C had an amorphous phase. XRD patterns of the powders prepared at 1100 °C revealed broad crystalline peaks. BNT powders were not obtained when spray pyrolysis was carried out at temperatures below 1100 °C because of the

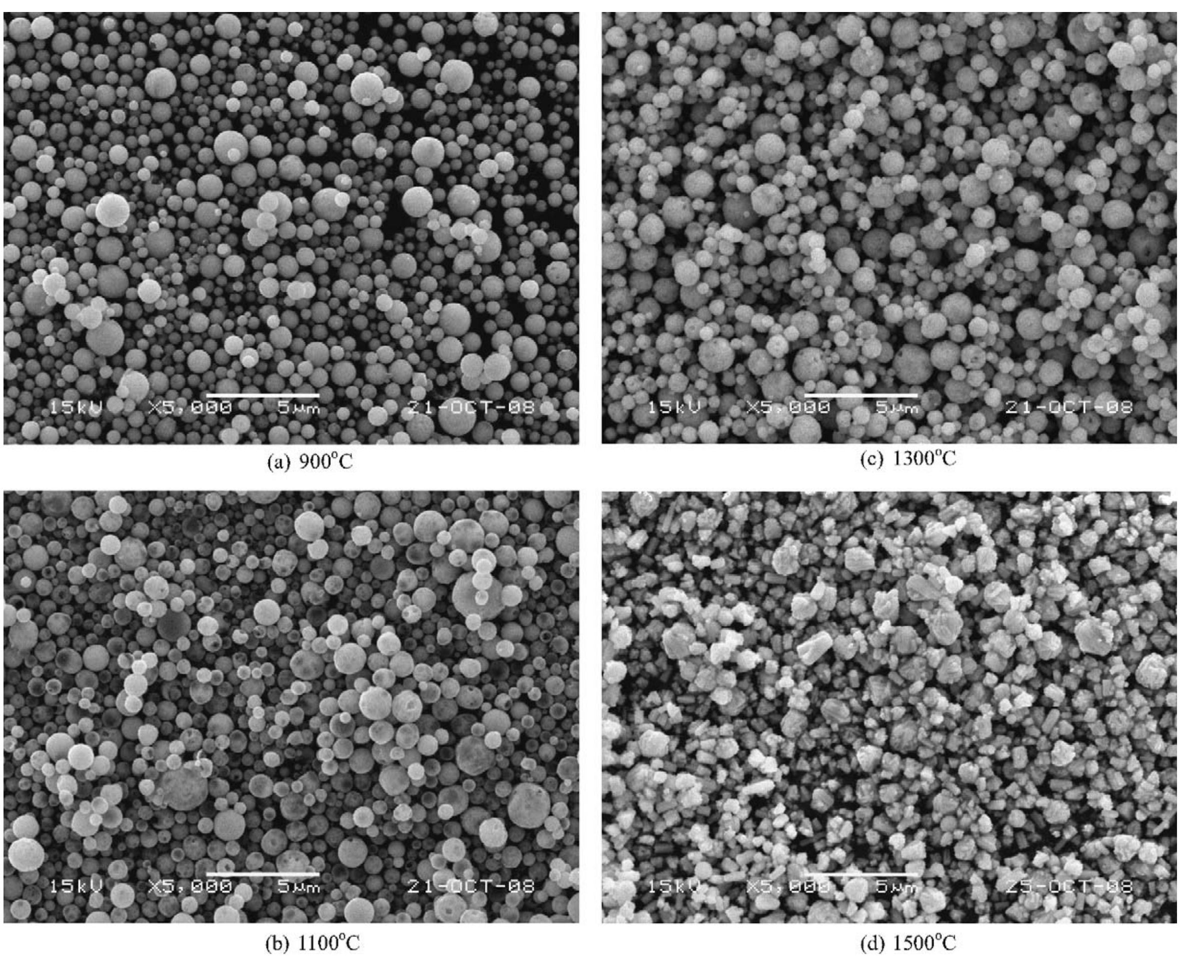

Fig. 2. SEM images of the BNT powders directly prepared by spray pyrolysis.