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Dielectric properties of nano-sized $Ba_{0.7}Sr_{0.3}TiO_3$ powders prepared by spray pyrolysis

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Abstract

Nano-sized $Ba_{0.7}Sr_{0.3}TiO_3$ powders are prepared by post-treatment of the precursor powders with hollow and thin wall structure at temperatures between 900 and 1100 °C. Ethylenediaminetetraacetic acid and citric acid improve the hollowness of the precursor powders prepared by spray pyrolysis. The mean sizes of the powders post-treated at temperatures of 900, 1000 and 1100 °C are 42, 51 and 66 nm, respectively. The densities of the $Ba_{0.7}Sr_{0.3}TiO_3$ pellets obtained from the powders post-treated at 900, 1000 and 1100 °C are each 5.36, 5.55 and 5.38 g cm^{-3} at a sintering temperature of 1300 °C. The pellet obtained from the powders post-treated at 1000 °C has higher maximum dielectric constant than those obtained from the powders post-treated at 900 and 1100 °C.

Keywords: C. Dielectric properties; Barium strontium titanate; Spray pyrolysis; Nano powders

1. Introduction

Barium strontium titanate (BST) [$Ba_{1-x}Sr_xTiO_3$, $0 < x < 1$] are widely used in the thick film forms in various fields because of their high dielectric constant and composition-dependent Curie temperature [1–7]. Pure BST thick films has to be sintered at high temperatures above 1300 °C, and therefore only platinum or refractory metals can be used as conductors [8,9]. In recent, nickel, copper and silver conductors with low sintering temperatures are mainly applied to reduce the production cost. Therefore, the sintering characteristics of BST thick films should be improved at low temperatures. One method of decreasing the sintering temperature of BST is to decrease the mean size of the powders. The performance of BST thick films is also highly dependent on the characteristics of powders, such as stoichiometry, phase homogeneity, powders size and its distribution [10].

Fine size BST powders with various compositions were widely prepared by liquid solution methods such as sol–gel, co-precipitation and hydrothermal [11–18]. However, the characteristics of BST powders prepared by gas phase reaction methods are scarcely studied. Jung et al. investigated the mean sizes, morphologies, and crystal structures of the $Ba_{0.5}Sr_{0.5}TiO_3$ powders prepared by flame spray pyrolysis using "CA-assisted" spray solution [19]. The mean size of the $Ba_{0.5}Sr_{0.5}TiO_3$ powders was 32 nm at a post-treatment temperature of 1000 °C. Branković et al. prepared the submicron-sized $Ba_{0.8}Sr_{0.2}TiO_3$ powders by ultrasonic spray pyrolysis from the polymeric precursors [20], in which one particle was formed from the one droplet. The sintering characteristics and dielectric properties of the nano-sized BST powders are strongly affected by the composition as well as the preparation process.

In this study, nano-sized $Ba_{0.7}Sr_{0.3}TiO_3$ powders were prepared by spray pyrolysis from the spray solution with ethylenediaminetetraacetic acid (EDTA) and citric acid. The sintering characteristics and dielectric properties of the $Ba_{0.7}Sr_{0.3}TiO_3$ powders prepared by spray pyrolysis were investigated.

2. Experimental

The schematic diagram of the equipment and the formation mechanism of the nano-sized $Ba_{0.7}Sr_{0.3}TiO_3$ powders in the spray pyrolysis are shown in Fig. 1. The precursor powders with hollow and thin wall structure were prepared by spray pyrolysis

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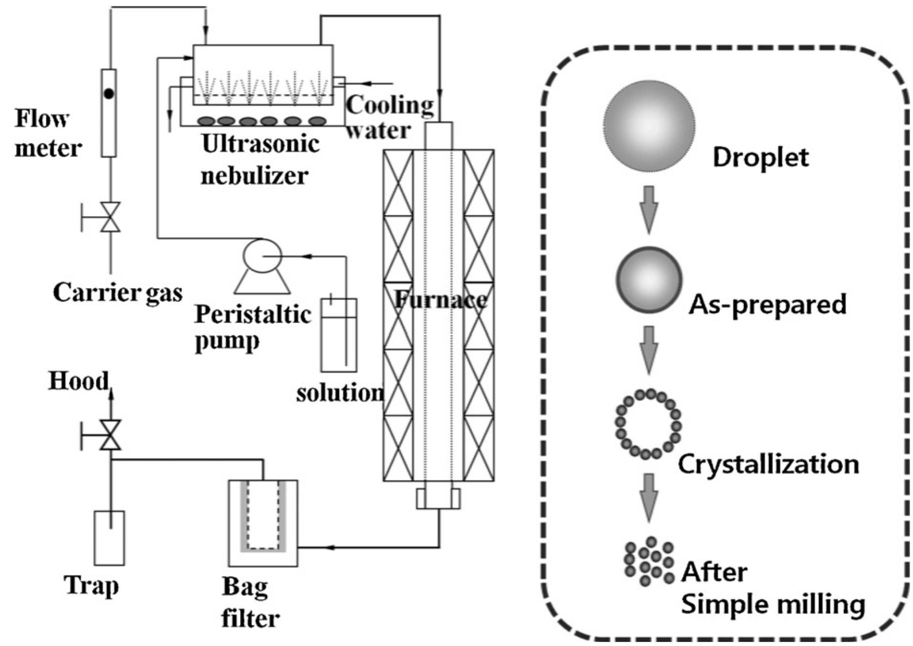

Fig. 1. Schematic diagram of spray pyrolysis process and formation mechanism of nano-sized ceramic powders.

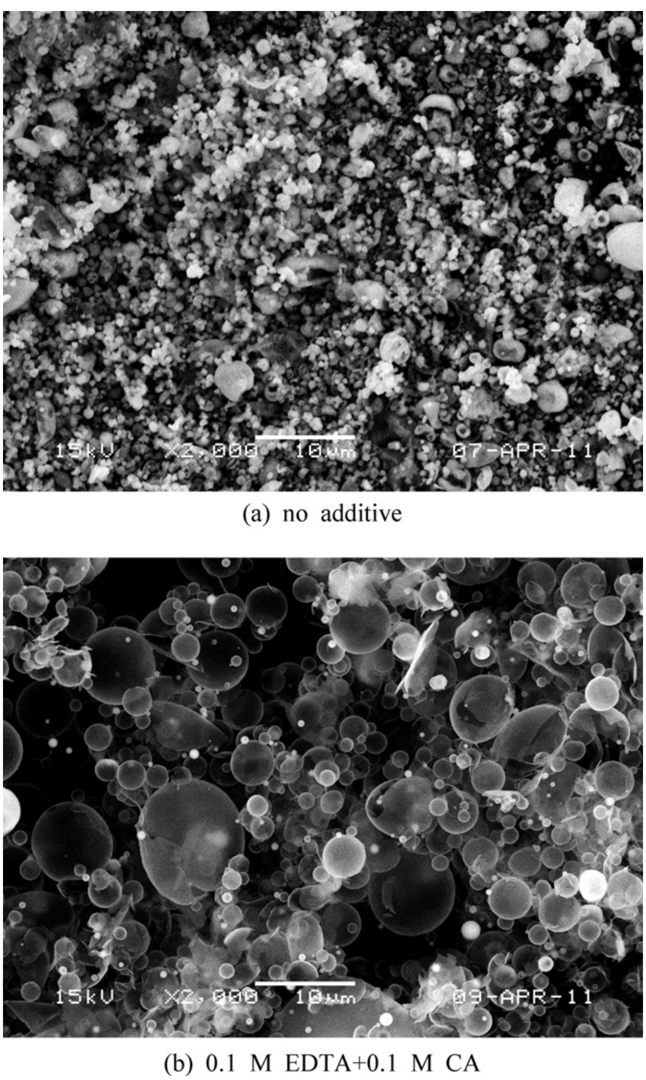

Fig. 2. SEM images of the precursor powders prepared by spray pyrolysis.

to obtain the nano-sized $Ba_{0.7}Sr_{0.3}TiO_3$ powders. The equipment consisted of six ultrasonic spray generators operating at 1.7 MHz, a tubular quartz reactor (length: 1200 mm; ID: 50 mm), and a bag filter. The starting materials for the synthesis were barium nitrate, strontium nitrate and 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 fixed at 0.1 M. Citric acid and ethylenediaminetetraacetic acid (EDTA) were used as chelating agents to improve the hollowness of the precursor particles. The concentrations of citric acid and EDTA were each 0.1 M. The spray pyrolysis temperature of the precursor powders was fixed at 900 °C. The flow rate the air carrier gas was fixed at 40 L min^{-1}. The precursor powders prepared by spray pyrolysis were post-treated at temperatures of 900, 1000 and

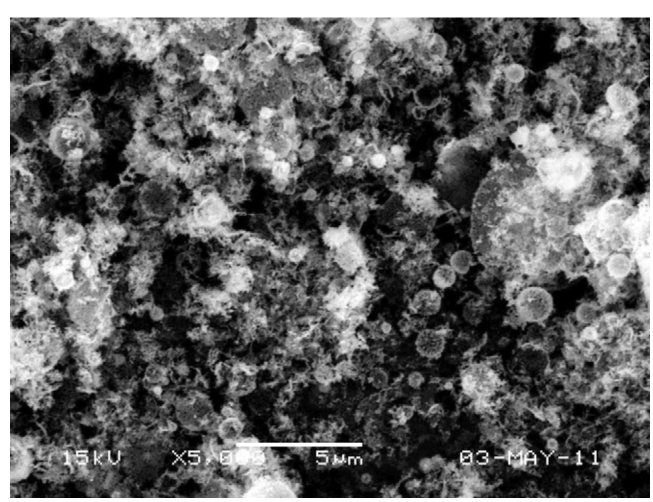

Fig. 3. SEM image of the BST powders post-treated at 1000 °C prepared from the spray solution with organic additive.