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Short communication

A coprecipitation technique to prepare $Mg_4Nb_2O_9$ powders

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Abstract

A simple coprecipitation technique had been successfully applied for the preparation of pure ultrafine single phase $Mg_4Nb_2O_9$ (MN). Sodium hydroxide was used to precipitate Mg^{2+} and Nb^{5+} cations as hydroxides simultaneously. This precursor on heating at 750 °C, produces MN powders. For comparison, MN powders were also prepared by the traditional solid state method. The phase contents and lattice parameters were studied by the powder X-ray diffraction (XRD). Particle size and morphology was studied by transmission electron spectroscopy (TEM).

Keywords: B. Electron microscopy; Ceramics; Electronic materials; X-ray diffraction

1. Introduction

Magnesium niobate (2:1) with corundum-like structure has attracted interest because of possible application as microwave dielectric material due to their low dielectric loss and high dielectric constant. [1–7]. The requirement of ceramic dielectric resonators used at microwave frequencies is high dielectric constant, a high Q value (reciprocal of dielectric loss) and a low temperature coefficient of resonant frequency. $Mg_4Nb_2O_9$ is also reported to be room temperature photo luminescent material. It also finds use as a suitable buffer layer material for fabrication of ferroelectric memory devices. Recently, Lu and yang [6] have shown that $Mg_4Nb_2O_9$ (MN) as a better precursor material for the successful preparation of single phase relaxor ferroelectric perovskite ($Pb(Mg_{2/3}Nb_{1/3})O_3$) which is becoming increasingly important for transducer, electrostrictor and actuator applications. Generally, MN powders are prepared by standard ceramic technique at 900 °C [2,4]. It was also reported [3,5] to be prepared using potassium niobate as a precursor. The properties of ceramics are greatly affected by the characteristics of the powder, such as particle size, morphology, purity and chemical composition. Using chemical methods, e.g., coprecipitation, sol–gel, hydrothermal and colloid emulsion technique have been confirmed to efficiently control the morphology and chemical composition of prepared powder. Among the reports of these wet chemical techniques sol–gel using alkoxides, hydrothermal and colloid emulsions are time consuming and involve highly unstable alkoxides and difficult to maintain reaction conditions. Coprecipitation is one of the more successful techniques for synthesizing ultrafine ceramic powders having narrow particle size distribution [8–10]. The purpose of this study was to prepare ultrafine $Mg_4Nb_2O_9$ powder using coprecipitation technique from simple water-soluble inorganic salts. This process can avoid complex steps, such as refluxing of alkoxides, resulting in less time consumption compared to other techniques. This method is not reported for the preparation of MN powders in the literature.

2. Experimental

For preparing $Mg_4Nb_2O_9$, niobium (V) oxide, magnesium nitrate and sodium hydroxide were used as starting materials, and all were of AR grade (LOBA cheme). A stoichiometric amount of $Mg(NO_3)_2{\cdot}6H_2O$ was dissolved in distilled water and Nb_2O_5 was dissolved in minimum

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amount of HF after heating at hot water bath for 20 h. Then, required quantity of NaOH solution was added with constant stirring to the above solution mixture until pH ∼12 to ensure complete precipitation. After filtration, the precipitate was washed several times and dried in an oven at 100 °C for 12 h. For comparison, MN samples are also prepared by ceramic method. The corresponding oxides or carbonates are taken in stoichiometric ratio and mixed, ground several times and heated at 900 °C for 12 h. Various techniques, such as XRD (Rigaku miniflex Diffractometer) and TEM were employed to characterize these powders. The powder X-ray pattern were recorded for all the samples sintered at various temperatures by using Philips PW-1710 model X-ray diffractometer using Cu Kα. For lattice parameter and interplanar distance (d) calculation, the samples were scanned in the 2θ range of 10–80° for a period of 5 s in the step scan mode. Silicon was used as an internal standard. Least squares method was employed to determine the lattice parameters. The TEM picture was recorded with JEOL model 1200 EX instrument at the accelerating voltage of 100 kV. The fine powders were dispersed in amyl acetate on a carbon coated TEM copper grid.

3. Result and discussion

The hydroxide precursor obtained is calcined at 750 °C for 6 h. Fig. 1 shows the XRD pattern of the calcined powder indicating formation of phase pure MN. The crystal structure of MN is hexagonal and all the d-lines pattern match with reported values [JCPDS 38-1459). The calculated lattice parameters by least square fit are $a = 5.12$ Å and $c = 14.047$ Å. At initial stages of heating and for lesser duration (<6 h) of heating, coulombite $MgNb_2O_6$ phase was found to be formed. This phase disappears on longer duration of heating as well at higher temperatures. Conventional solid-state method also forms MN phase at 900 °C [3] with comparatively larger particle size of ∼1 μm. The particle size and morphology of the calcined powders were examined by transmission electron microscopy. Particle morphology of calcined powder (750 °C for 6 h) prepared by the coprecipitation process was irregular in shape, with an average primary particle size around 150 nm (Fig. 2). The average particle size calculated from Scherrer's formula ($t = K\lambda/B\cos\theta_B$) where t is the average size of the particles, assuming particles are spherical, $K = 0.9$, λ is the wavelength of X-ray radiation, B is the full width at half maximum of the diffracted peak and θ_B is the angle of diffraction, is 200 nm. The average particle size of MN

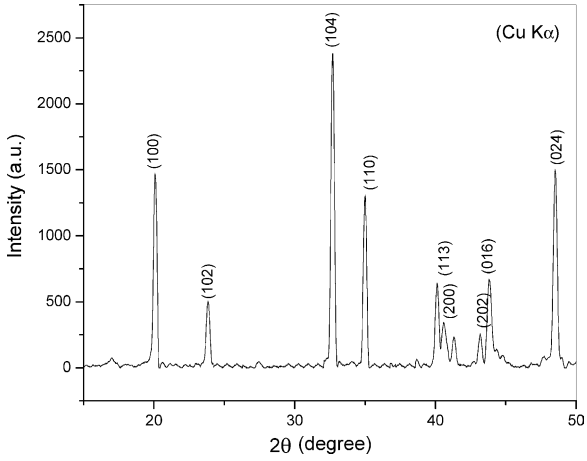

Fig. 1. XRD of MN powder calcined at 750 °C.

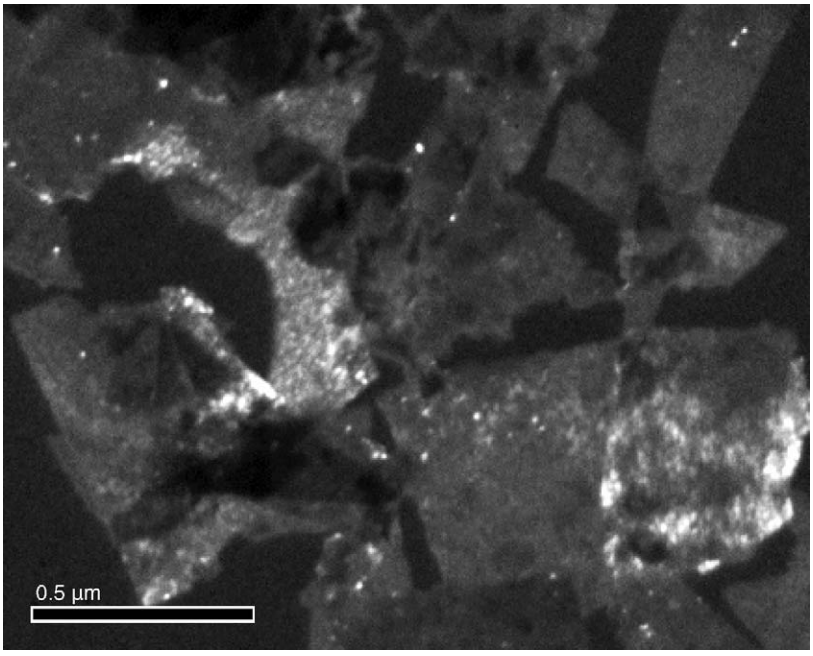

Fig. 2. TEM of MN powder calcined at 750 °C.

powders prepared by conventional ceramic method was in the range of 1–2 μm (not shown).

4. Conclusions

A simple coprecipitation method was used to prepare ultrafine particles of MN. The MN phase was found to be formed at 750 °C with average particle size of 150 nm.

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