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Short communication

A novel technique to prepare $LiTaO_3$ at low temperature

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Abstract

Firstly, fresh tantalum hydroxide was precipitated from TaF_5 solution using an aqueous ammonium hydroxide under basic conditions. Then a simple procedure of mixing lithium and tantalum hydroxides together and heating at a low temperature (450 °C) produced pure ultrafine single phase $LiTaO_3$ (LT). This is the lowest temperature so far reported for the formation of LT in the literature. The phase content and lattice parameters are determined by X-ray diffraction (XRD). The average particle size and morphology were studied by transmission electron microscopy (TEM).

Keywords: B. Electron microscopy; Chemical synthesis X-ray diffraction; Electronic materials; $LiTaO_3$

1. Introduction

Ceramics based on $LiNb_{1-x}Ta_xO_3$ have excellent electro-optical and photorefractive properties and find extensive applications such as optical wave guides, optical modulators and surface acoustic wave devices [1–6]. Although single crystals of $LiTaO_3$ (LT) find applications, there still are restrictions because of high cost and difficult fabrication. In contrast, polycrystalline LT ceramics can be made with a larger size and more complex shape. Since, microstructure affects critically the optical properties, the synthesis of LN ceramic powders with good sinterability and compositional homogeneity is necessary. Traditionally LT is prepared by solid state reaction which leads to inhomogeneity in composition and coarse particles. The wet chemical methods have their own advantages when compared with the traditional solid state method. The advantages include nanocrystalline size, homogeneity in composition, high reactivity and surface area, narrow particle size distribution and low sintering temperature. The wet chemical methods include coprecipitation, hydrothermal, combustion, alkoxide and citrate gel methods. Recently microwave hydrothermal technique was also used for the preparation of complex oxides. The purpose of this study was to investigate the possibility of lowering temperature of formation of $LiTaO_3$ phase. It consists of preparing corresponding hydroxides and mixing thoroughly and subsequent heating at 450 °C leads to formation of LT powders. This procedure is quite novel and can be used for large scale production of these powders. This method is not reported in the literature.

2. Experimental

For preparing $LiTaO_3$, AR grade (Loba chemie) tantalum (V) oxide, lithium hydroxide and standard ammonia solution were used as starting materials. Required quantity of Ta_2O_5 was dissolved in HF (40%) after heating in a hot water bath for 10 h. Then an aqueous ammonium hydroxide was added dropwise to the TaF_5 solution to precipitate tantalum as hydroxide under basic conditions. The precipitate was washed free of anions and dried at 100 °C in an oven. A stoichiometric amount of LiOH was mixed with $Ta(OH)_5{\cdot}xH_2O$ and ground well for ~1 h using acetone in a agate mortar. These powders were calcined at different temperatures from 200 to 500 °C for 6 h. The powder X-ray pattern was recorded for all the samples calcined at different temperatures by using a Philips PW-1710 model X-ray diffractometer and Cu Kα radiation. 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 used to determine the lattice parameters.

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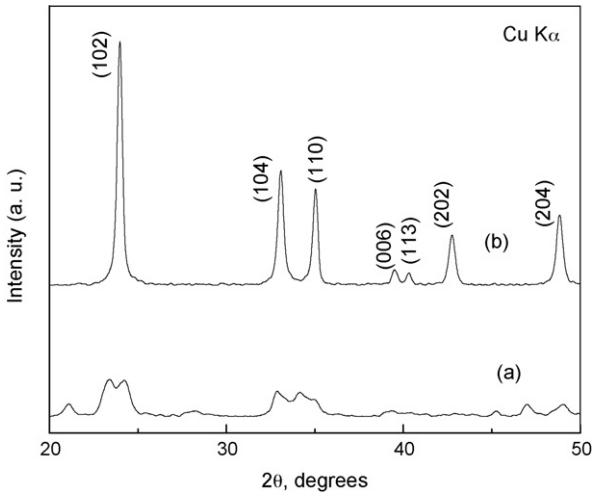

Fig. 1. XRD of LT precursor powder calcined at (a) 200 °C and (c) 450 °C for 6 h.

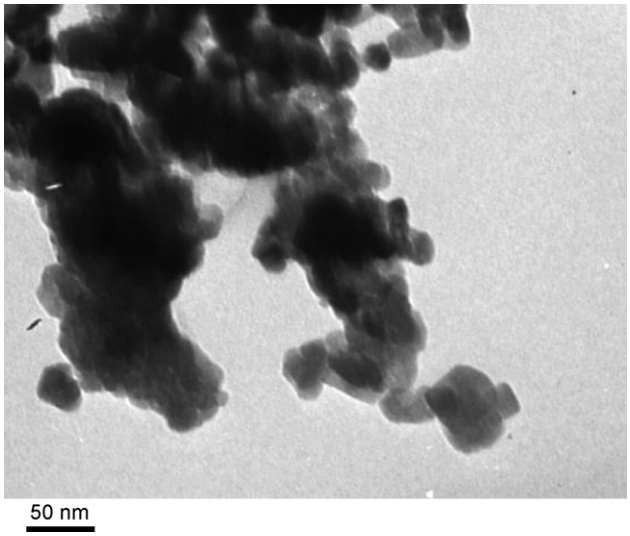

Fig. 2. TEM image of LT powder calcined at 450 °C for 6 h.

Transmission electron microscopy (TEM) observations were made with a JEOL model 1200 EX instrument at the accelerating voltage of 100 kV. All the powders were dispersed in amyl acetate on a carbon coated TEM copper grid.

3. Result and discussion

Fig. 1 shows the X-ray diffraction (XRD) patterns of the powder calcined at different temperatures. The powders heated at 200 °C consists of many intermediates of varying composition and authentically could not indexed. However, when calcination temperature was 450 °C and duration of ∼6 h, it leads to formation of major $LiTaO_3$ phase. This is the lowest temperature reported so far for the formation of LT phase. All the d-line peaks are similar to that reported in the literature. The crystal structure of LT is hexagonal and all the d-line patterns match with reported values (JCPDS-29–836). The calculated lattice parameters by least squares fit are $a = 5.154$ Å and $c = 13.756$ Å. The conventional solid state method gives LT phase at 900 °C after prolonged firing. The particle size and morphology of the calcined powders were examined by transmission electron microscopy. The particle morphology of calcined powder (450 °C for 6 h) was irregular in shape, with an average primary particle size around 50 nm (Fig. 2). The average particle size of 45 nm was calculated from Scherrer's formula

$$t = \frac{K\lambda}{B\cos\theta_B}$$

where t is the average size of the particles assuming particles to be spherical, $K = 0.9$, λ the wavelength of X-ray radiation, B the full width at half maximum of the diffracted peak and θ_B is the angle of diffraction.

4. Conclusions

A simple procedure of using hydroxide precursors for the preparation of ultrafine particles of $LiTaO_3$ was elucidated. The LT phase crystallized at 450 °C with average particle size of 50 nm.

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