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Short communication

Novel hydroxide precursors to prepare $NaNbO_3$ and $KNbO_3$

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Abstract

Firstly, fresh niobium hydroxide was precipitated from NbF_5 solution using an aqueous ammonium hydroxide under basic conditions. Then a simple procedure of mixing potassium (or sodium) and niobium hydroxides together and heating at a low temperature (<400 °C) produced $KNbO_3$ (KN) or $NaNbO_3$ (NN) powders. This is the lowest temperature so far reported for the formation of these phases. The phase content and lattice parameters are determined by X-ray diffraction (XRD). The average particle size and morphology were studied by scanning electron microscopy (SEM).

Keywords: A. Powders, chemical preparation; B. X-ray methods; D. Niobates; D. Perovskites

1. Introduction

Potassium niobate ($KNbO_3$) and sodium niobate ($NaNbO_3$) have attracted considerable interest among material scientists because of its high-temperature ferroelectricity, piezoelectricity, large electro-optic coefficient, excellent photorefractive properties and the moderate dielectric constant [1–10]. It has wide applications such as optical wave guiding, frequency doubling, holographic storage and surface acoustic wave devices. As it is lead free, it is a promising piezoelectric material, which can be an alternative to lead-based materials and they are currently used in majority of ultrasonic applications [11–20]. Since K_2O is a volatile component over 800 °C and its evaporation from the system is accelerated by the humidity in the sintering environment, $KNbO_3$-based ceramics are very difficult to synthesize by conventional solid-state reaction because the high-temperature sintering often leads to stoichiometric deviation in the composition of the final product.

Many methods were developed to synthesis $KNbO_3$, such as hydrothermal, homogeneous precipitation method, polymerized complex (PC) method, polymeric precursor method, and metal alkoxides route [1–20]. The purpose of this study was to investigate the possibility of lowering temperature of formation of KN and NN phases. The procedure consists of preparing corresponding hydroxides and mixing thoroughly and subsequent heating at 400 °C leads to formation of KN (or NN) 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 $KNbO_3$ and $NaNbO_3$, AR grade (Loba chemie) niobium(V) oxide, sodium and potassium hydroxides, hydrofluric acid (HF (40%)) and standard ammonia solution were used as starting materials. Required quantity of Nb_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 NbF_5 solution to precipitate niobium as hydroxide under basic conditions. The precipitate was washed free of anions and dried at 100 °C in an oven. A stoichiometric amount of NaOH (or KOH) was mixed with $Nb(OH)_5{\cdot}xH_2O$ 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*)

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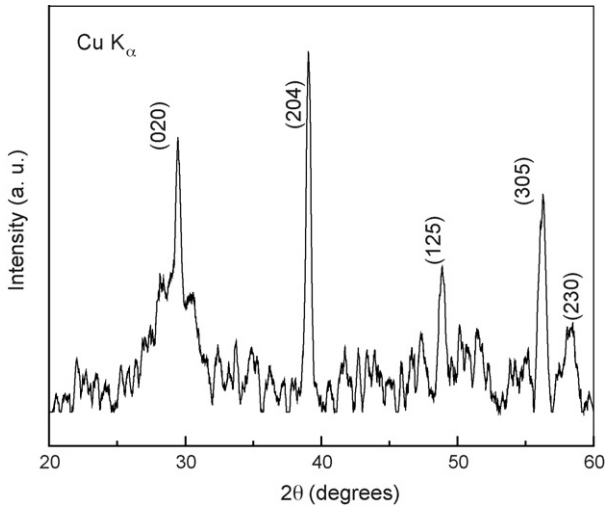

Fig. 1. XRD of NN precursor powder calcined at (a) 200 °C.

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. The SEM images were observed using a Leica Cambridge 440 microscope. All the powders were dispersed in amyl acetate for SEM studies.

3. Result and discussion

Fig. 1 shows the XRD pattern of the NN powder calcined at 200 °C. All the *d*-line peaks are similar to that reported in the literature. This is the lowest temperature reported so far for the formation of NN phase. The crystal structure of $NaNbO_3$ is orthorhombic (JCPDS: 14-603). The calculated lattice parameters by least squares fit are $a = 5.511$ Å, $b = 5.557$ Å and $c = 15.540$ Å. In the case of KN powders heated at 200 °C it shows many *d*-line peaks and authentically could not indexed. However, when calcination temperature was increased to 400 °C, it leads to formation of major $KNbO_3$ phase with minor impurities (Fig. 2). The crystal structure of $KNbO_3$ is

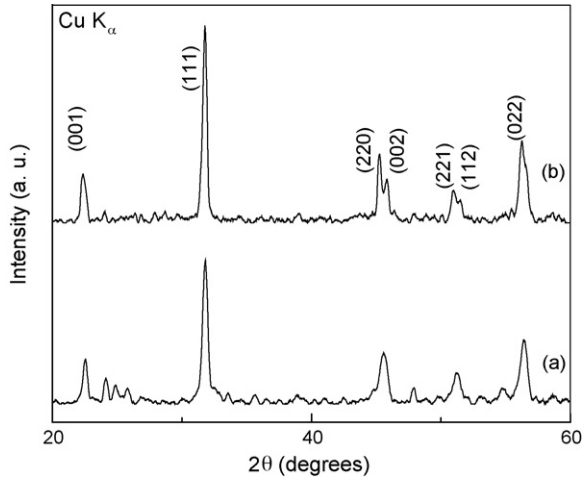

Fig. 2. XRD of KN precursor powder calcined at (a) 400 °C and (b) 700 °C.

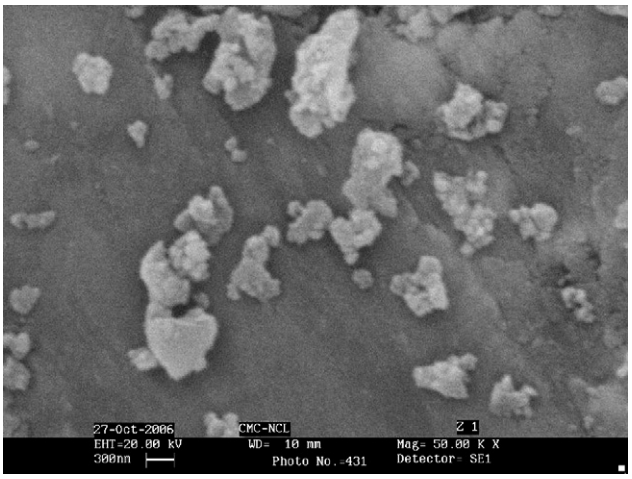

Fig. 3. SEM image of NN powder calcined at 400 °C.

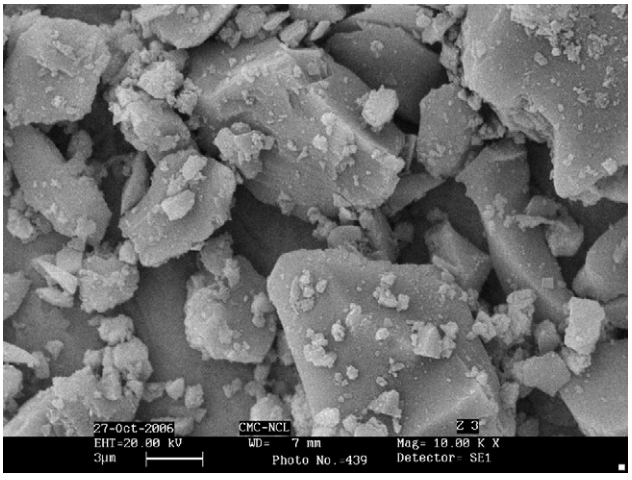

Fig. 4. SEM image of KN powder calcined at 400 °C.

orthorhombic and all the *d*-line patterns match with reported values (JCPDS: 32-0822). The calculated lattice parameters by least squares fit are $a = 5.69554$ Å, $b = 5.721$ Å and $c = 3.973$ Å. There was no change in the *d*-lines pattern when KN powders were heated at 700 °C. The low intense peaks corresponding to intermediate phases were also disappeared as observed from Fig. 2. The average particle size and morphology of these calcined powders (both NN and KN) were examined by scanning electron microscopy. Figs. 3 and 4 show the SEM images for NN and KN, respectively. In the case of NN, there was not much agglomeration with an average particle size of 70 nm (Fig. 3). The SEM picture of KN illustrated the presence of two types of particles with different morphologies and sizes (Fig. 4).

4. Conclusions

A simple procedure of using hydroxide precursors for the preparation of ultrafine particles of $KNbO_3$ and $NaNbO_3$ was elucidated. The formation of NN phase occurs at 200 °C whereas for KN, it is ~400 °C. The average particle size and morphology of these powders were investigated by SEM.

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