Experimental report

02/10/2023

Proposal: 5-23-787 **Council:** 10/2022

Title: Determination of the superstructure of BF-BT-based lead-free piezoelectric ceramics

Research area: Materials

This proposal is a new proposal

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Samples: (0.75-x)BiFeO3-0.25BaTiO3-xBi0.5K0.5TiO3

Instrument	Requested days	Allocated days	From	To
D2B	2	2	19/06/2023	21/06/2023
D20	2	0		

Abstract:

The dual improvement of piezoelectric coefficient (d33) and ferroelectric-paraelectric phase transition temperature (TC) of BF-BT-based lead-free piezoelectric ceramics can be achieved by introducing BKT with tetragonal phase. The study of X-ray and Raman spectroscopy showed that the phase evolution from R3c to P4mm. In particular, the ceramic with x=0.010 located in the MPB region exhibits excellent performances with d33=149 pC/N, kp=38 %, Pr=28.2 ¿C/cm2 and TC=615 °C. Meanwhile, the temperature dependence of dielectric permittivity (¿r-T) showed that the introduction of BKT helps to improve TC and an additional frequency dispersion dielectric peak (T1) appeared at low temperature. In situ Raman spectroscopy and in situ d33 testing indicate the BiFeO3-BaTiO3-0.010Bi0.5K0.5TiO3 has excellent thermal stability (Td=570 °C). The knowledge of the structure is important for understanding the macroscopic properties and that this is the basis for further improving the materials. Therefore, it is necessary to use the neutron powder diffraction (NPD) to further clarify the formation of the T1, the origin of the TC improvement and mechanism of d33 variation with temperature.

EXPERIMENT N°: 5-23-787 **INSTRUMENTS:** D2B

DATES OF EXPERIMENT: 18/06/2023 - 21/06/2023

TITLE: Determination of the superstructure of BF-BT-based lead-free piezoelectric ceramics.

REPORT 28/09/2023

Piezoelectric materials are widely used in many fields, such as aerospace, deep-sea investigation or precision manufacturing, where maintaining their piezoelectric properties while working under harsh conditions in required. With the aim of replacing lead-containing materials, several piezoelectrics have been studied. $BiFeO_3$-$BaTiO_3$ systems outstand among them due to exhibiting moderate piezoelectric activity and a high Curie Temperature, making them promising for high temperature applications. Because of their electrical properties been able to be tuned by the co-substitution of different valence states in different sites, the understanding of the role played by the aforementioned substitution in the properties change is of great interest.

On this line, a set of (0.75-x)$BiFeO_3$-0.25$BaTiO_3$-x$Bi_{0.5}K_{0.5}TiO_3$ (x = 0, 0.01, 0.03) was measured for 72h at D2B diffractometer at ILL. The goal was to elucidate the effect of KBT doping on the structure, superstructure, oxygen vacancies and occupations of the sample.

Each sample was measured at three different temperatures: 25, 400 and 700°C (below, close to and above the depolarization temperature, T_d, respectively), with a working wavelength of 1.59Å. The experimental procedure was the following: the sample was inserted in the furnace at D2B, and a routine was configured to continuously increase the temperature of the sample until the desired temperature was reached. Subsequent to the temperature stabilization, neutron diffraction data was acquired. The obtained diffractograms are shown in Figure 1.

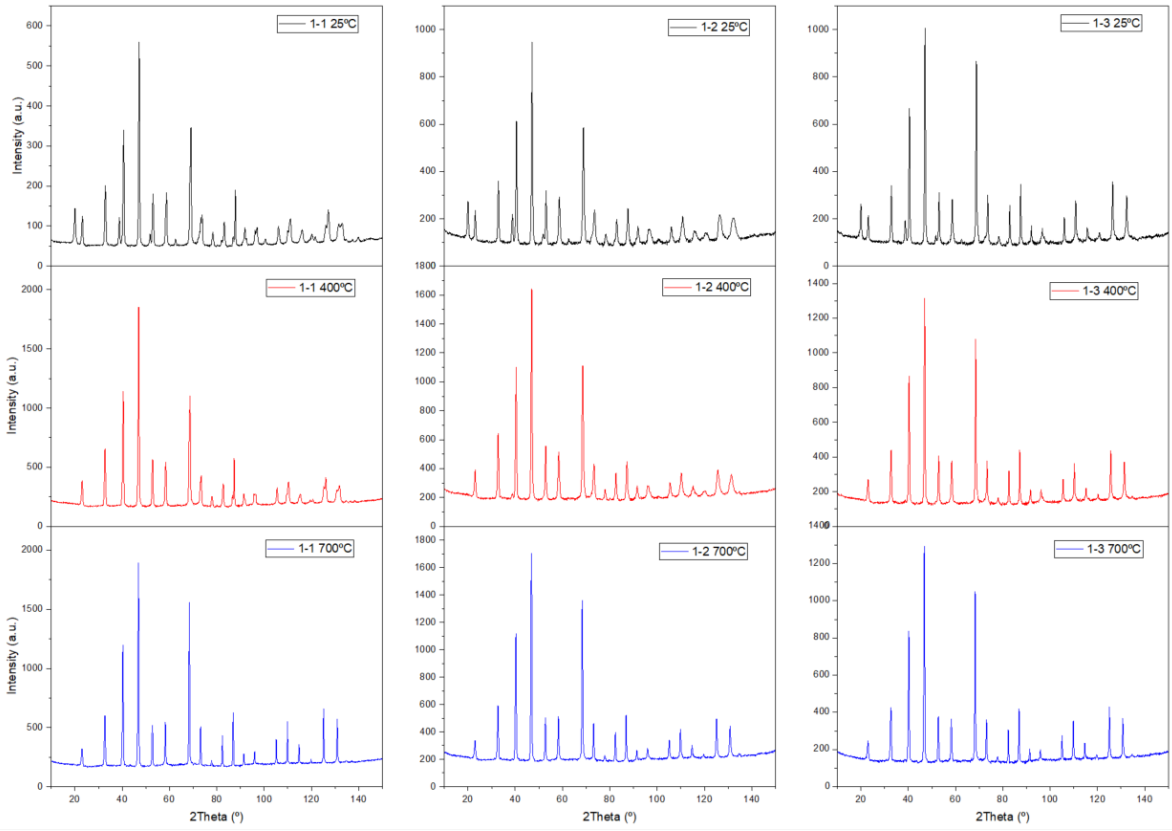

Figure 1. Powder Neutron Diffraction diffractograms obtained at 25^{o}C (black), 400^{o}C (red) and 700^{o}C (blue). First column corresponds to 1-1 sample (x=0); second column, to 1-2 sample (x=0.01) and third column, to 1-3 sample (x=0.03).

Preliminary Rietveld analysis performed in the x=0.01 sample at 300^{o}C shows a clear R3c structure for both BaTiO3 and $BiFeO_3$. The system appears to be a two-phases-system, rather than a solid solution. Further analysis will be performed to determine the lattice parameters of each unit cell, alongside with the atomic occupations and vacancies.