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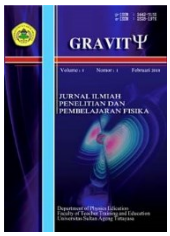

Evaluation of thermal testing and X-ray diffraction of $Ka_{0,5}Na_{0,5}NbO_3$

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ABSTRACT

The structure of perovskite-based material contained in the niobate and titanate. The fabric was a perovskite crystal structure having the formula ABO_3. Ferroelectric and piezoelectric materials had a perovskite structure. This material could store an electric charge, which was good because of the polarization resulting in a material that was a dielectric. Unleaded piezoelectric material, $K_{0,5}Na_{0,5}NbO_3$ (KNN), was synthesized using reliable state methods. Synthesis was done by first setting up K_2CO_3, Na_2CO_3, and Nb_2O_5 as a base KNN system. Studies cover X-ray diffraction, thermal analysis TGA-DTA and lattice parameter analysis. From the TGA-DTA analysis obtained for KNN calcination temperature at 700^0C for 2 hours can produce a single-phase ABO_3 where A = (K, Na) and B = (Nb). Orthorhombic perovskite structure KNN material owned by P4mm space group with lattice parameters a = 3,572 Å; b = 3,570 Å; and c = 3,565 Å.

Keywords: piezoelectric, perovskite, solid state, $K_{0,5}Na_{0,5}NbO_3$

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INTRODUCTION

Known perovskite structure-based material contained in the niobate and titanate (Desmelinda, 2015). It is known that niobate-based material has an advantage and a similar nature of PZT, but more environmentally friendly (Chen, et al., 2013). Perovskite crystal structure is Sebastian having a general formula ABO_3. This structure is based on cubic each cube face having an oxygen atom. A atoms occupy each corner of the cube while the atom B occupies the centre of the hub. The material has a perovskite structure that can have a lattice cube, tetragonal, orthorhombic, and others.

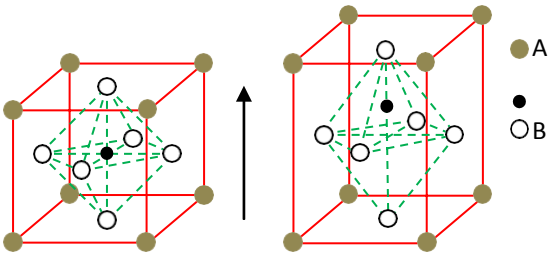

Figure 1. Perovskite structure in the cubic phase (left) and tetragonal phase (right) (Chiang, 1997).

The ferroelectric material is one of several types of materials that have a role in the world of science and industry. Examples of material utilization ferroelectric on electronic circuits such as: variation on the circuit microwave, modular electro-optical, dielectric materials that are good for the capacitor, FeRAM (ferroelectric RAM), ferroelectric tunnel junction, multiferroic materials, piezoelectric transducers, detectors pyroelectric, PTC

(positive temperature coefficient) and infrared sensors (Suasmoro et al., 2000 and Fu, 2009).

Piezoelectric and ferroelectric materials have a structure that is perovskite. Therefore, the content can store electrical charge properly — the electrical charge storage due to the polarization of the material. Polarization is the result of a cubic phase transition into a tetragonal phase at a specific temperature, which is also called a curie temperature. So that the electric field is relatively small given cation would shift the central grid due to Coulomb interaction. This polarization resulted in distortion of crystal that forms a dipole and the macroscopic scale separation of positive and negative charges or a dielectric material that is called. Ferroelectric has a high dielectric constant when applied to the field of relatively low frequency, for example, at room temperature, εr to barium titanate of 5000. Therefore, the capacitor is made of a dielectric material with ferroelectric properties (Callister & Rethwisch, 2012).

Piezoelectricity is a symptom when no force is applied to a material segment that causes the electric charge on the surface of the material sections. A piezoelectric material can alter the mechanical stress into an electrical charge given to him and change the electric field given to him becoming a mechanical stress. The piezoelectric material used as the material to produce electricity cantilever low-power mini electric motors, microphone and medical devices on ultrasonography (Ahda, 2012). Another application is as a base for the manufacture of transducers. The piezoelectric charge coefficient values are in the range of 1-100 PC/N (Saito, 2004). Known piezoelectric materials today are $PbZrTiO_3$ (known as PZT), which are known to have excellent piezoelectric properties and widely applied. But the lead oxide is a toxic material that is high and will increase the danger at high temperatures, especially in the calcining and sintering processes (Mardiyanto, 2010). So that research must be done to make these unleaded piezoelectric material to make it more environmentally friendly.

In the general situation, the unleaded piezoelectric material is a material with a perovskite crystal structure such as $BaTiO_3$ (BT), $KNbO_3$, $NaTaO_3$, and non-perovskite include bismuth layer structured ferroelectric materials (BLSF) and tungsten-bronze ferroelectric materials. According to research conducted by some experts, materials $BiNaTiO_3$ (BNT) is a good candidate to replace the PZT material. The amount of remnant polarization, namely BNT (P_r) = 38 μC/cm^2 and E_c = 73 kV/cm (Ni et al, 2011). But apparently, this BNT material still has shortcomings that made a dopant to improve the performance of the piezoelectric material.

$Ka_{0,5}Na_{0,5}NbO_3$ or commonly abbreviated as KNN is an excellent candidate to make unleaded piezoelectric material for piezoelectric properties and a strong ferroelectric. According to Saito (2004), a composition is having a high d_{31} values obtained in the phase between orthorhombic and tetragonal. KNN widely studied is a compound made of the carbonate such as K_2CO_3, Na_2CO_3, and Nb_2O_5. KNN that have been hot press having d_{33} ~ 160 pC/N (Desmelinda, 2015). Analysis of the structure and morphology of the piezoelectric material is performed using X-ray diffraction (XRD).

X-ray diffraction is a test to determine the crystal system in materials. This test also can explain the existence of the lattice parameters, the type of structure, the difference in the arrangement of atoms in the crystal, the crystal is not perfect, and the amorphous form in the material. X-rays produce electrons spread if the particle strikes a metal at high speed in a state of vacuum tubes. X-ray beam using a crystal to diffraction because of the order of X -ray wavelengths have the distances between atoms is nearly equal or smaller in a glass (Zulianingsih, 2012).

XRD testing utilizing the diffraction of X-rays. High voltage generator to function as power generating X-ray source in the x-ray tube. Samples that have been compressed powder form placed over a container that can be positioned. Then the X-ray beam strikes an example and is diffracted by the sample, into the tool counter. X-ray diffraction intensity captured by the detector and translated in the form of curves.

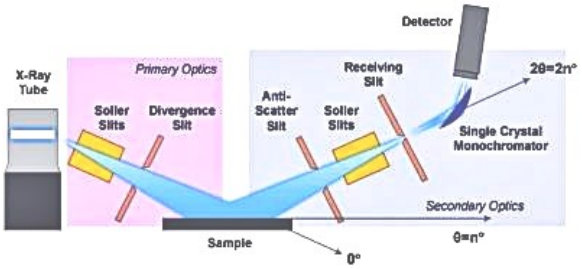

Figure 2. XRD Work Scheme (Ywcmatsci.yale.edu/xrd, 2020)

In addition to analysis using X-ray diffraction, testing the other is by thermal analysis. Thermal analysis is the measurement of the physical properties and chemical as a function of temperature. Thermal analysis has two analytical techniques that thermogravimetric analysis (TGA) and differential thermal (DTA). Thermogravimetric analysis (TGA) can measure the change in weight of the sample. In contrast, the thermal differential analysis (DTA) can measure the temperature difference between the sample with inert reference material as a function of temperature. DTA is used for the study of the structure of the glass, the determination of the phase diagram, the phase transition polymeric, kinetic energy, heat capacity, enthalpy, and decomposition.

The robust reaction method is a method for making ceramics made in the solid-state. It occurs at temperatures above 1000°C. Method with less solid-state synthesis is used to create a unique composition and morphology needed to produce desirable properties in crystals, piezoelectric, and other advanced materials. In general, the solid-state reaction is stoichiometric reaction powders at high temperatures. Keuntunggan of this method is simple, in the sense that everyone can do it, but it can be tricky, for instance, at a temperature and atmosphere used.

RESEARCH METHODS

Material Synthesis $Ka_{0,5}Na_{0,5}NbO_3$

Equipment and materials used during material synthesis $Ka_{0,5}Na_{0,5}NbO_3$ is O'haus PA214 digital scales, Laboratory planetary mill "Pulverisette 5" rotary evaporator "VV Micro," Furnace "Thermolyne" type F1500, and plate alumina crucible, mold pellet-shaped with a diameter of 13 mm, a spatula, powder K_2CO_3, Na_2CO_3, Nb_2O_5 (Merck) and 99% alcohol.

The first step in the synthesis of (K, Na) NbO_3 is a powder, each weighing 1,002 grams of K_2CO_3, 0.768 grams of Na_2CO_3, and 3,854 grams of Nb_2. Then the three powder is mixed with alcohol (99%) in a planetary milling, which included a zirconia balls and incorporated for 1 hour at a rotation speed of 150 rpm. The next process is to enter the material that has been mixed into the rotary evaporator "VV Micro" (Heidolph) so that the alcohol is gone and keep equipment so obtained homogeneous powders KNN. And lastly, calcining the KNN powder at a temperature of 700^0C for 2 hours.

The analysis used in this study are as follows: (1) *Thermal analysis*. Thermal Analysis performed with TGA-DSC test to determine the calcination temperature of the ingredients have been mixed. From this test chart obtained in the form of mass change of heat so it can know at what temperature the material has a constant mass. (2) Analysis Phase and Precursor Lattice parameters or materials that have been completed until calcination phase then testing X-Ray Diffraction. This test is used to look at any phase contained in the content that has been made. Data obtained in the form of the diffraction pattern when the material shot by the X-ray. The XRD pattern is the result of a comparison between the diffraction angle with high intensity. From the results of these XRD characteristics are known and analyzed with match software where the software already has a reference database and is matched with the test results. This XRD test can also see the material lattice parameters by smoothing calculated and measured patterns using Rietica software on the basis of the Rietveld method.

RESULTS AND DISCUSSION

Results of KNN Powder Thermal Analysis

Synthesis KNN ($K_{0,5}Na_{0,5}NbO_3$) was performed using a reliable reaction method (regular mixing). Mixing is done using the Planetary Milling equipped with zirconia balls

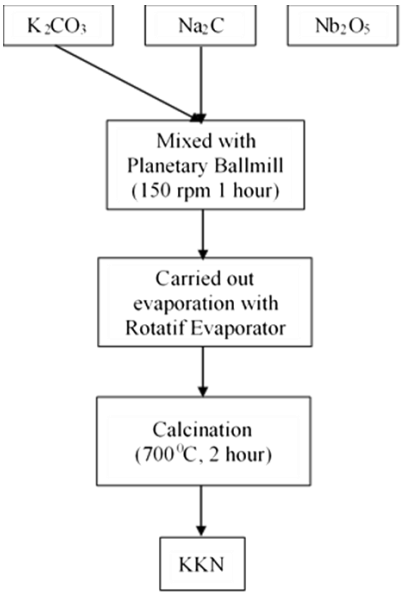

Figure 3. Flowchart synthesis $Ka_{0,5}Na_{0,5}NbO_3$ in 99% alcohol for 1 hour. Then dried using a rotatory evaporator to obtain the KNN powder while maintaining the homogeneity of the mixture. Once it is done, thermal analysis (TGA-DTA) to determine the reactions indicated by the presence of mass reduction and both endothermic and exothermic phenomena that accompany the specific range of temperatures that can be known calcination temperature.

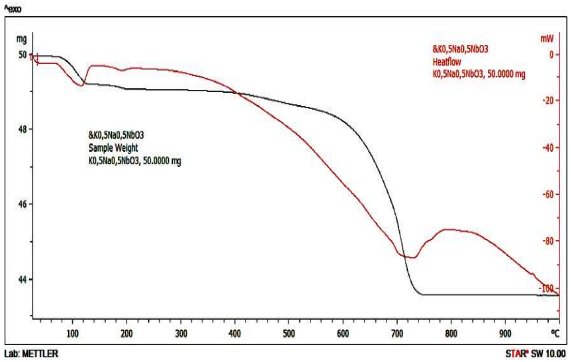

Figure 4. The test results of TGA-DSC 50 mg $Ka_{0,5}Na_{0,5}NbO_3$

From Figure 4 the results of TGA-DTA is known that a mixture of powdered K2CO3, Nb2O5, NaCO3 and decreased mass at a temperature of around 100°C and 650 to 750° C. Loss of mass that occurs at a temperature of about 100°C is a process of evaporation of water, where the water is still in powder have been mixed.

This is because the drying process is not perfect, whereas when the temperature is

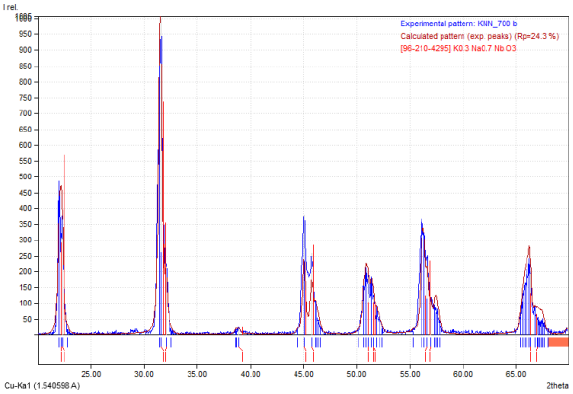

Figure 5. Results KNN powder diffraction pattern at a calcination temperature of 700°C.

650^{o}C to 750°C is possible because the CO_2 released from carbonate of approximately 11.17%.

After the temperature over 750°C, there was found no decrease in mass. From the results of the thermal analysis test it is believed that at temperatures around 650°C to 750°C the reaction occurs in the formation of KNN material.

Test Results X-ray Diffraction

After the thermal analysis is carried out, the powder mixture of K_2CO_3, $NaCO_3$ and Nb_2O_5 is calcined at a temperature that is expected to form the KNN reaction at 700°C with a holding of 2 hours. Then powder XRD results calcination testing to determine the phases present in the KNN powder. Figure 5 is an XRD pattern of results from the software origin calcined at a temperature of 700°C for 2 hours. The results of the XRD pattern shows that when a temperature of 700°C, KNN powder has been formed due to the phase generated only a single step and no secondary phase.

To clarify the evidence that there is only a single phase of this material can be analyzed using software match. Figure 6 is a picture of mountains owned by KNN calcined material 700^{0}C for 2 hours. The Figure shows that the single-phase already obtained. Determining the type of TiO2 phases are generated based on the location of the summit accord angle (2θ) with an individual specific datasheet.

ICSD data show that KNN orthorhombic crystal structure at room temperature. To find

out the truth, XRD results need further analysis using the software Rietica smoothing method.

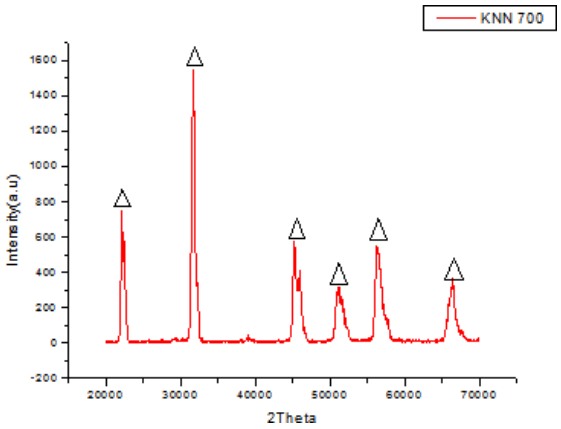

Figure 6. Results KNN powder diffraction pattern at a calcination temperature of 700°C

Rietveld analysis is a method for matching the measured diffraction pattern of research and obtained from the model. This method is carried out for the comminution process lattice parameters of the crystal structure of the material.

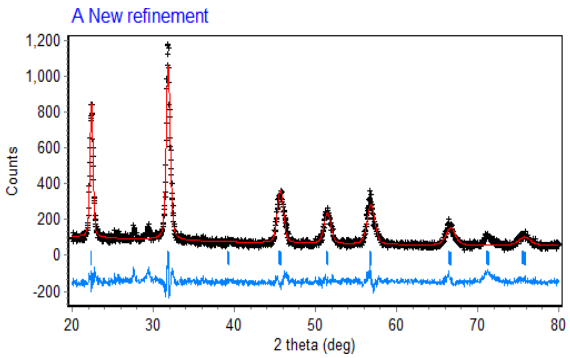

Figure 7. Results refinement KNN material

Figure 7 is the result of the refinement of the KNN material, which has a structure orthorhombic with P4mm space group with lattice parameters a = 3.572 Å; b = 3.570 Å; and c = 3.565 Å. In this figure, the data showed that between the ICSD with the results of the study have a lattice parameter value of less than 4 Å, which means that research can be said to have been by the data ICSD and has a single-phase so that the reliable reaction method with a composition that has been measured can be used for further research analysis.

CONCLUSION

Perovskite structure without timbale material synthesized by reliable reaction method. Their calcination at a temperature of 700°C for 2 hours is able to produce a single-phase ABO_3 where A = (K, Na) and B = (Nb). KNN material has a perovskite structure orthorhombic with P4mm space group and lattice parameters a = 3.572 Å; b = 3.570 Å; and c = 3.565 Å.

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