

Preparation and Synthesis of Columbite $M\text{Nb}_2\text{O}_6$ Ceramics by Chemical Reaction (Where $M = \text{Zn, Ni, Co}$)

Praveen Kumar* Bharat Mishra* Mukul Pastor**

*Department of Physical Science, MGCGV, Chitrakoot, Satna (M.P.)

**Innovation centre, Bundelkhand University, Jhansi(U.P.)

ABSTRACT

Columbite $M\text{Nb}_2\text{O}_6$ ($M = \text{Zn, Ni, Co}$) ceramics produce by the chemical reaction process (sol gel) were investigated. Ethylene glycol was used to precipitate M (Zn, Ni, Co) and Nb^{+5} cation as hydro were also prepared by the traditional solid state method. The phase evaluation were studied by the powder X-ray diffraction (XRD).

Keywords : - Ceramics, Materials, Sol-gel, XRD.

1. INTRODUCTION

Columbite material have been widely investigated for the use of resonators and filters in the satellite and mobile communication system. [1, 2] The binary niobate ceramic, with a general formula $M\text{Nb}_2\text{O}_6$ where M is a divalent alkalineearth or transition metal cation with ionic-radius less than 1\AA , usually crystallize in the columbite structure. Raw material Ta_2O_5 is expansive as compared with Nb_2O_5 , $M\text{Nb}_2\text{O}_6$ (where $M = \text{Mg, Zn, Ni, Ca, Cu, Mn}$ and Co) columbite niobate compound of low sintering temperature and low cost have received much attention [3-6]. $\text{Zn Nb}_2\text{O}_6$ (ZN) ceramics were reported to exhibit excellent dielectric properties. Columbite niobate compounds are investigated for the application in electromagnetic properties.

The powder of reactants are homogenized and activated in high energy vibromill in this mechanical technique. The sol gel process is a wet chemical method used for the fabrication of both glassy and ceramic materials. In this process the sol (solution) involves gradually towards the formation of a gel like network containing both a liquid phase and a solid phase.

2. EXPERIMENTAL PROCEDURE :

All samples in this study were prepared from sol-gel method. Appropriate amounts of raw material for stoichiometric $M\text{Nb}_2\text{O}_6$ were weighted milled in acetone with zirconia balls for 12 h.

2.1. Prepare the ZnNb_2O_6 by Sol-gel method :

The pure and doped ZnO nanoparticles were synthesized in sol-gel methods, with the help of precursor solution of ethylene glycol and zinc acetate with in 1 : 1 ratio a precursor solution was prepared in deionized water.

We prepare the columbite material ($ZnNb_2O_6$) by solid state reaction. The solid state reaction route is the most widely used method for the preparation of polycrystalline solids from a mixture of solid starting materials. We take the raw material ZnO and Nb_2O_5 same ratio 2:2. We grinded the both material in pestle. Then finally a powder was calcined at $1000^{\circ}C$ for 4 hours.

2.2. Prepare $NiNb_2O_6$ by sol-gel method

The pure and doped NiO nanoparticles were synthesized in sol-gel method, with help of precursor solution of ethylene glycol and Nickel nitrate [$Ni(NO_3)_2$] with in 1:1 ratio a precursor solution was prepared in a deionized (DI) water. Formerly Nickel nitrate hexahydrate [$Ni(NO_3)_2 \cdot 6H_2O$]. A continuous stirring solution with 3-4 hours at $50^{\circ}C$, a green solution was acquired. The homogeneous mixture was maintained under reflux at $130-150^{\circ}C$ for 8 hours. A wet gel was attained after the evaporation of excess solvents. Then finally a powder was calcined at $800^{\circ}C$ for 4 hours and then grinded.

Prepared the columbite material ($NiNb_2O_6$) by solid state reaction. The traditional solid state reaction route for the synthesis of meltferroic consist of mixing by milling the appropriate metal oxides or carbonates and then calcinating at high temperature to allow interdiffusion of the cation the chemical reaction occurs by solid state diffusion of the ions, which is characterized by a slow kinetic rate.

We collect the raw material NiO and Nb_2O_5 same ratio 2 : 2. We grinded the both material in pestle. Finally a powder was calcined at $1000^{\circ}C$ for 4 hours.

2.3. Prepare the $CoNb_2O_6$ by sol-gel method

The pure and doped CoO nanoparticles were synthesized in sol-gel methods, with help of precursor solution of ethylene glycol and citric acid [$C_6H_8O_7$] with in 1:1 ratio a precursor solution was prepared in a deionized (DI) water. Formerly copper nitrate trihydrate [$Co(NO_3)_2 \cdot 3H_2O$]. A continuous stirring of solution with 3-4 hours at $50^{\circ}C$, a green solution was acquired. The homogeneous mixture was maintained under reflux at $130-150^{\circ}C$ for 8 hours. A wet gel was attained after the evaporation of excess solvents. Then finally a powder was calcined at $800^{\circ}C$ for 4 hours and then grinded.

We prepare the columbine material ($CoNb_2O_6$) by solid state reaction. Solid state reaction are performed in the absence of solvents by either grinding or melting the starting materials together or simply applying heat to a mixture of starting materials.

We take raw material CoO and Nb_2O_5 same ratio 2 : 2. We grinded the both material in pestle. Then finally a powder was calcined at $1000^{\circ}C$ for 4 hours.

3. RESULT AND DISCUSSION :

3.1. $ZnNb_2O_6$

Final calcinations powder X-ray diffraction was conducted on a Phillips analytical XPERT diffractometer using a Cu K α radiation ($\lambda=1.54056 \text{ \AA}$) in range 2θ and 10^0 - 80^0 .

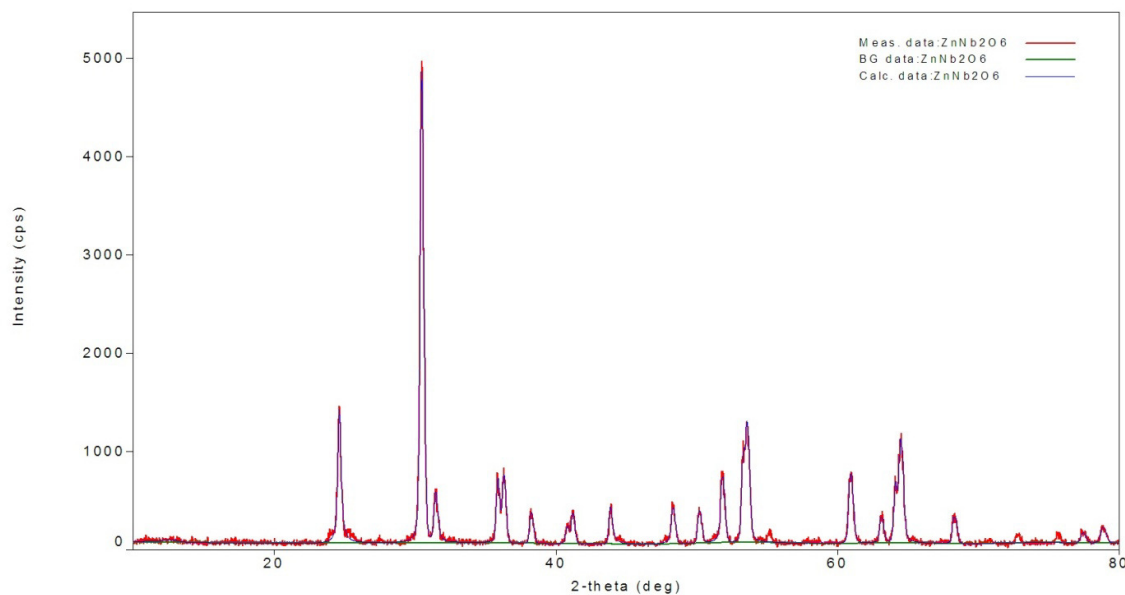


Figure No. 1 : XRD of prepared sample oxide at 1000^0C calcinations temperature.

3.2. NiNb_2O_6

Finally calcinations powder X-ray diffraction was conducted on a Phillips analytical XPERT diffractometer using a Cu K α radiation ($\lambda=1.54056\text{\AA}$) in range 2θ and 10^0 - 80^0C .

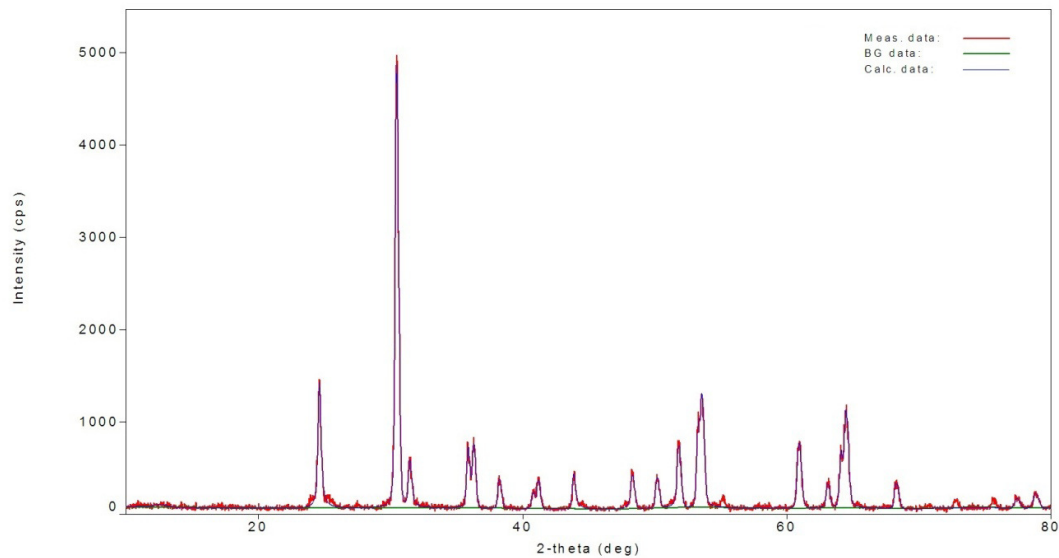


Figure No. 2 : XRD of prepared sample oxide at 1000⁰C calcinations temperature.

3.3. CoNb_2O_6

Final calcinations powder X-ray diffraction was conducted on a Phillips analytic XPERT diffractometer using a Cu K α radiation ($\lambda=1.54056\text{\AA}$) in range 2θ and 10° - 80°C .

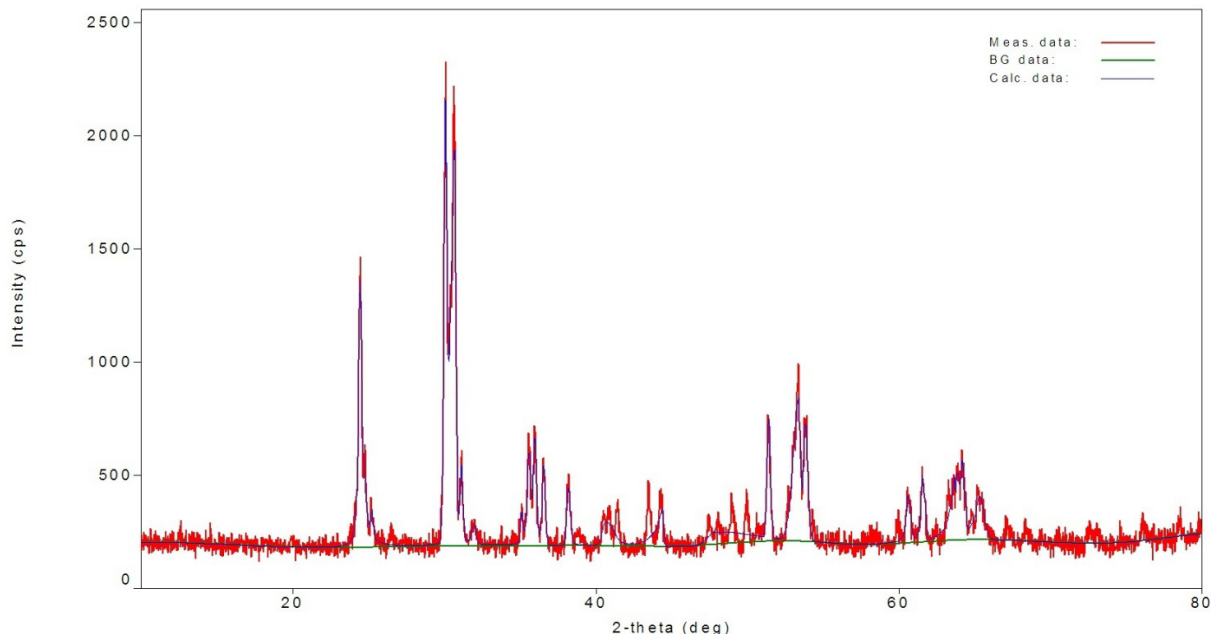


Figure No. 3 : XRD of prepaid sample oxide at 1000°C calcinations temperature.

The calcinations powder were analyzed by X-ray diffraction (XRD) to check the formed phases.

4. CONCLUSION

Pure clumbite MNb_2O_6 ($M = \text{Zn}, \text{Co}, \text{Ni}$) phase was obtained successfully by chemical reaction process. After 1000°C sintering for 2 hours, a density 4.37 g/cm^3 were obtained.

REFERENCES

1. S. Kawashima, Nishida, L Ueda, H. Ouchi. $\text{Ba}(\text{Zn}_{1/3}\text{Ta}_{2/3})\text{O}_3$ ceramics with low dielectric loss at microwave frequencies, J. Am. Ceram. Soc. 66 (6) (1983) 233-241.
2. S. Nomura, K. Toyama. K. Kaneta, $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{2/3})\text{O}_3$ ceramics with temperature stable high dielectric constant and low microwave loss, Jpn. J. Appl. Phys. 21 (1982) L624—L626.
3. M. Meada, T. Yamamura, T. Ikeda, Dielectric characteristics of several complex oxide ceramics at microwave frequencies, Jpn. J. Appl. Phys. Supp. 26-2 (1987) 76-79.
4. H.J. Lee, I.T. Kim_ K.S. Hong, Dielectric properties of AB_2O_6 compounds at microwave frequencies ($A = \text{Ca}, \text{Mg}, \text{Mn}, \text{Co}, \text{Ni}, \text{Zn}$ and $B = \text{Nb}, \text{Ta}$). Jpn. J. Appl. Phys. 36, part 2 (10A) (1997) L318—L320.

5. H.J. Lee, K.J. Hong, S.J. Kim, I.T. Kim, Dielectric properties of $M\text{Nb}_2\text{O}_6$ compounds (where $M = \text{Ca}, \text{Mn}, \text{Co}, \text{Ni}$ or Zn), *Mater. Res. Bull.* 32 (7) (1997) 847-855. .461
6. C.S. Hsu, C.L. Huang, J.F. Tseng, C.Y. Huang, Improved high-Q micro-wave dielectric resonator using CuO-doped MgNb_2O_6 ceramics, *Mater. Res. Bull.* 38 (2003) 1091-1099.
7. Y.C. Lion, Tseng, Stoichiometric $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ perovskite ceramics produced by reaction-sintering process, *Mater. Res. Bull.* 38 (8) (2003) 1351-1357.
8. Y.C. Lion, Shih, C.H. Yu, Stoichiometric $\text{Pb}(\text{Fe}_{1/2}\text{Nb}_{1/2})\text{O}_3$ perovskite ceramics produced by reaction-sintering process, *Mater. Lett.* 57 (2003) 1977-1981.
9. Y.C. Lion, C.T. Wu, K.H. Tseng, T.C. Chung, Synthesis of BaTi_4O_9 ceramics by reaction-sintering process, *Mater. Res. Bull.* 40 (9) (2005) 1483-1489.
10. Y.C. Lion, LH_ Chen, H.W. Wang, C.Y. Liu, Synthesis of $(\text{Ba}_x \text{Sr}_{1-x})(\text{Zn}_{1/3} \text{Nb}_{2/3})\text{O}_3$ Ceramics by Reaction-Sintering Process and Microstructure. *Mater. Res. Bull.* 41 (3) (2006) 455-460.
11. Y.C. Lion, W.H. Shine, C.Y. Shill. Microwave ceramics $\text{Ba}_5\text{Nb}_4\text{O}_{15}$ and $\text{Sr}_5\text{Nb}_4\text{O}_{15}$ prepared by a reaction-sintering process, *Mater. Sci. Eng. B* 131 (2006) 142-146.
12. L. Srisombat, S. Ananta, S. Phanichphant, Chemical synthesis of magnesium niobate powders, *Mater. Lett.* 58 (2004) 853-858.
13. D. Saha, A. Sen, H.S. Maiti, Solid-state synthesis of precursor MgNb_2O_6 for the preparation of $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$, *J. Mater. Sci. Lett.* 13 (1994) 723-724.
14. Y.S. Hong, H.B. Park, S.J. Kim, Preparation of $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ powder using a citrate-gel derived columbite MgNb_2O_6 precursor and its dielectric properties, *J. Eur. Ceram. Soc.* 18 (1998) 613-619.
15. S. Ananta, R. Brydson, N.W. Thomas, Synthesis, formation and characterization of MgNb_2O_6 powder in a columbite-like phase, *J. Eur. Ceram. Soc.* 19 (1999) 355-362.
16. Y. Yu, C. Feng, C. Li, Y. Yang, W. Yao, H. Yan, Formation of columbite-type precursors in the mixture of $\text{MgO-Fe}_2\text{O}_3\text{-Nb}_2\text{O}_5$ and the effects on fabrication of perovskites, *Mater. Lett.* 51 (2001) 490-499.
17. Y.C. Lion, C.Y. Shine, Preparation of NiNb_2O_6 columbite ceramics by a reaction-sintering process, *Mater. Res. Soc. Symp. Proc.* 848 (2005) 115-119.
18. M. Thinunal, A.K. Ganguli, Synthesis and dielectric properties of magnesium niobate-magnesium tantalite solid solutions, *Mater. Res. Bull.* 36 (2001) 2421-2427.
19. R.C. Pullar, K. Okeneme, N.M. Alford, Temperature compensated niobate microwave ceramics with the columbite structure, $\text{M}^{2+}\text{Nb}_2\text{O}_6$, *J. Eur. Ceram. Soc.* 23 (2003) 2479-2481
20. Y.C. Zhang, Z.X. Yue, Z.L. Gui, L.T. Li, Effect of CaF₂ addition on the microstructure and microwave dielectric properties of ZnNb_2O_6 ceramics, *Ceram. Int.* 29 (2003) 555-559.
21. L.B. Kong, J. Ma H Huang, R.F. Zhang, T.S. Zhang, Zinc niobate derived from mechanochemically activated oxides, *J. Alloys Compd.* 347 (2002) 308-313.
22. Y.C. Zhang, L.T. Li, Z.X. Yue, Z.L. Gui, Effects of additives on micro-structures and microwave dielectric properties of ZnNb_2O_6 ceramics, *Mater. Sci. Eng. B* 99 (2003) 282-285.
23. Y.C. Zhang, J. Wang, Z.X. Yue, Z.L. Gui, L.T. Li, Effects of Mg^{2+} substitution on microstructure and microwave dielectric properties of $(\text{Zn}_{1-x}\text{Mg}_x)\text{Nb}_2\text{O}_6$ ceramics, *Ceram. Int.* 30 (2004) 87-91.