BILINGUAL PUBLISHING CO.
Pioneer of Global Academics Since 1984

Electrical Science & Engineering
https://ojs.bilpublishing.com/index.php/ese

ARTICLE

Ferroelectric Properties of $CuFe_2O_4$, $BaFe_2O_4$, $Ba_{0.2}La_{0.8}Fe_2O_4$ Nanoparticles

U.Naresh[1*] R. Jeevan Kumar[1] N.V.S. Gupta[2] D.Kothandan[3]

1. Sri Krishnadevaraya University, Ananthapuram, AP, 515055, India
2. G. Pulla Reddy engineering college, Kurnool, AP, 515055, India
3. Srinivasa Institute of Technology and Management Sciences, Chittore, A.P, 515055, India

ARTICLE INFO

Article history
Received: 20 March 2019
Accepted: 17 April 2019
Published Online: 30 April 2019

Keywords:
Hydrothermal technique
X-ray diffraction
Ferroelectric properties

ABSTRACT

In this article, we report ferroelectric properties of copper ferrite CuFe2O4 nanoparticles (CFN), Barium ferrite nanoparticles BaFe2O4 (BFN) and La substituted barium ferrite Ba0.2La0.8Fe2O4 (BLFN) nanoparticles synthesized via hydrothermal technique. The X-Ray diffraction for the synthesized particles reflects the cubic phase formation for CuFe2O4, orthorhombic phase structure for the BaFe2O4 and cubic formation of Ba0.2La0.8Fe2O4 (BLFN). The structural parameters such as crystallite size and micro-strain are computed from XRD and Williamson-Hall(W-H) analysis. The polarization- electric field (P-E) loop studies gave information about the ferroelectric nature of the synthesized samples. It was noticed that the CFN particle has a lossy dielectric nature whereas BFN, BLFN samples exhibit a multiferroic nature.

1. Introduction

Nano-materials get vital impact materials of nano-science and nanotechnology. Nanostructure science and technology may well be a broad and knowledge domain space of analysis and development activity that has been growing explosively worldwide inside the past few years [1-2]. It's the potential for revolutionizing the ways that within which materials and product are created and therefore vary and nature of functionalities that may be accessed. It's already having a major industrial impact, which is able to assuredly increase within thefuture. The ferrite nanoparticles are the class of the magnetic materials. The ferrites have enormous applications in the electronic and engineering field like memory devices, electromagnetic shielding application, etc., and biomedical application such as drug delivery systems, hyperthermia treatment, etc[3-5]. The soft ferrite has its characteristics like wide permeability in the gigahertz frequency make it into the electronic application [4]. The soft type ferrites having minimum coercively and maximum electrical resistive properties make it as a core material in the transformers and electronic communication application. In addition, superparamagnetism of ferrite nanoparticles extends their use in biomedical application like hyperthermia, drug delivery systems, and magnetic resonance imaging technology massively. In a similar way, the ferrite-based composites such as polyaniline, polypyrrole mixed with ferrites could be permitted for the fabrication of multi-efficient materials for electronic devices and sensors applications [11].

Basically, ferrites classified into 3 classes viz. spinel,

**Corresponding Author:*
U.Naresh,
Sri Krishnadevaraya University, Ananthapuram, AP, 515055, India;
Email: chandrababu954@gmail.com

DOI: https://doi.org/10.30564/ese.v1i1.929

garnet, and magneto-plumbite. However, the suitable elemental composition of ferrites exhibits its ownstructural, electrical and magnetic properties revealing technological applications. In general, ferrites are of AB_2O_4 cubical structure, wherever 'A' represents the power metal cations like $Fe^{2+,}$ Ni^{2+}, Mg^{2+}, Mn^{2+}, Co^{2+}, Zn^{2+} and Cu^{2+} and 'B' may be a powerful iron ion (Fe3+) [6-8]. The crystal structure primarily depends on chemical element anions and remaining cations organized in an exceedingly face-centered cubical closed packing system. Garnets have the final formula of $REFe_5O_{12}$, wherever 'RE' represents to lanthanide power cations like La^{3+}, Gd^{3+}, Y^{3+}, Dy^{3+}, Ce^{3+}, Eu^{3+}, $Sm^{3+,}$ etc [9]. These materials perform fascinating properties in microwave communication system [10]. The hexa ferrites (magneto-plumbite ferrites) square measure related to a general formula of $HFe_{12}O_{19}$ (H = Ba, Sr, Co, Ca, Pb, etc…) [9]. The incorporation of rare earth elements into the ferrite systems leads to the enhancement of physical properties. However, the replacement of rare earth elements in place of small radii could be made the probability of imperfections in the unit cell.

The previous reports show the importance of the Ba and La-doped barium ferrites interestingly. Qing Lin et.al prepared the nanoparticles as incorporating Ba^{+2} into the lanthanum magnesium ferrite nanoparticles by citrate sol-gel method. The structural analyses by using the XRD and FTIR, they found as decreases the Ba^{+2} concentration an additional $MgFe_2O_4$ phase was detected while the orthorhombic structure of LaFeO3 was not changed this was attributed by the calcination process was carried out at the time of synthesis [10]. Sagar M. Mane et.al [7,8] studied the composite (1-x)(Ba0.8Sr0.2TiO3)-x (Co0.9Ni0.1Fe2O4) properties which are synthesized through the co-precipitation method followed by a microwave calcination process. PE loop of the synthesized particles is shown ferroelectric behavior and ferromagnetic material nature. The magnetodielectric composites show the maximum value of the magneto-capacitance. The ferroelectric properties are observed in this case. The saturation magnetization (Ms) increases with increase in the ferrite concentration while the saturation polarization decreases (Ps) decreases revels the formation of multiferroic composites [11].

The synthesis of the nanoparticles is the significant step in the preparation of the nanoparticles. We have many synthesis techniques to prepare the nanomaterials such as sol-gel technique, auto combustion method, solvothermal and hydrothermal, polyol method among all of the techniques the hydrothermal technique is more accurate method in the preparation of the nanoparticle [12]. The consistency in the formation of single phase formation, economically feasible, low operating temperature, purity of the sample taken into account we opted the hydrothermal method in this work. In this work, we prepared $BaFe_2O_4$ (BFN), $CuFe_2O_4$ (CFN), and $Ba_{0.2}La_{0.8}Fe_2O_4$ (BLFN) particles vial hydrothermal technique and reported structural and ferroelectric properties of the synthesized sample.

2. Experimental Work

The Hydrothermal synthesis was chosen to prepare the Cu, Ba Ferrite nanoparticles. The chemicals of AR grade Cu $(NO_3)_26H_2O$, Ba $(NO_3)_26H_2O$, Fe $(NO_3)_3$ $9H_2O$, La (NO_3) $9H_2O$ & NaOH purchased from Sigma- Aldrich co ltd. $Cu(NO_3)_26H_2O$, Fe $(NO_3)_3$ $9H_2O$ & $Ba(NO_3)_26H_2O$, Fe $(NO_3)_3$ $9H_2O$ mixed in a laboratory glass beaker using magnetic stirrer in distilled water (1:4 Nitrates: water) separately as per the planned stoichiometric calculation. The stirring process continued by constant pH values through the addition of the NaOH agent slowly (pH = 11.8) to the mixer. After, the homogeneously mixed brown colored solution was transferred into 300 ml autoclave (steel lined Teflon jar). The locked autoclave was fetched into a hot air oven. The temperature of the oven kept as 150°C and time setup is 8 hrs for both CFN, BFN samples. Further, the reacted solution was washed with the acetone and distills water several times till it got purified, then this purified content dried at 100°C in the oven for two hours. Finally, fine powder type samples were collected. These samples are characterized by the study of structural and ferroelectric properties using X-ray diffraction and PE Loop tracer.

3. Result and Discussion

3.1. XRD Study

Figure 1a, 1b predicted hydrothermally synthesized $CuFe_2O_4$, $BaFe_2O_4$ nanoparticles X-ray diffraction patron. The patterns Figure 1a revealed the single phases of the prepared nanoparticle exhibited as cubic structures with the agreement of JCPDS card no. 25-0283 reflection planes for the CFN particles whereas the consistency of JCPDC cord no.46-0113 reflection planes with the figure 1b recognized as orthorhombic phase formation of BFN particles clearly. Figure 1c illustrates the patterns of X-ray diffraction for the synthesized material of $Ba_{0.2}La_{0.8}Fe_2O_4$. The XRD outline could be evidence that the synthesize nanoparticles showing the well developed cubic spinel phase formation. All the reflected planes (111), (220), (311), (222), (400), (511), (440), (533), (444) are in good quality catch with the standard JCPDS pattern containing the cubic spinel single phase. Moreover, the structural parameters such as average crystallite size (D) 25 nm for

 DOI: https://doi.org/10.30564/ese.v1i1.929

CFN and 19.26 nm for the BFN nanoparticles whereas for the BLFN particles we found 21nm and also, lattice constants for CFN, BFN, BLFN particles unit cell volume V, micro-strain (ε) was premeditated using principle ε = β/4tanθ of all the synthesized samples calculated using the followed formulas and the results presented in table 1.

The crystallite size (D) was calculated using the average crystallite D = 0.9λ/βcosθ, where λ= wavelength of X- rays and β is full width half maximum [13]. On the other hand, the lattice parameter a = d $(h^2 + k^2 + l^2)^{½}$ where the h, k, l are the miller indices. The radii of synthesized elements can be affected in the octahedral tetrahedral occupation Ba^{+2}:1.35 Å, La^{+3}: 1.032 Å, Fe^{+3}: 0.645 Å [13]. The difference between Ba^{+2} and La^{+3} radii makes strain in lattice it is due to the distribution of cations of different radii among the tetrahedral site and octahedral site it will be evaluated using the formula. The incorporation of La^{+3} atoms replaces Fe^{+2} atoms viewing to the ionic radii of La^{+3} greater than the Fe radii it leads to a larger value of lattice parameter it could become a reason for the larger volume which is found as 955.67 $Å^3$.

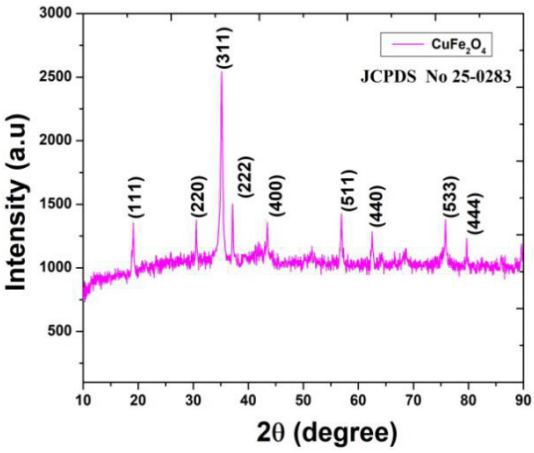


Figure 1a. XRD pattern of CFN particle

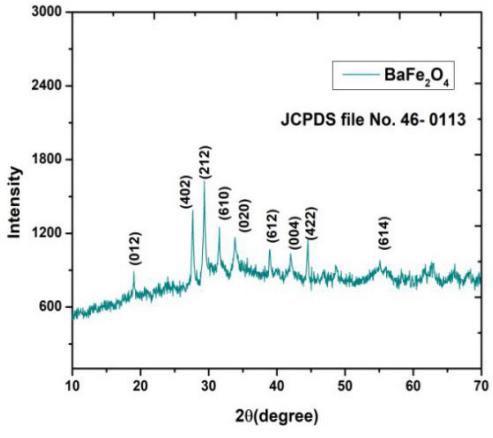


Figure 1b. XRD pattern of BFN particle

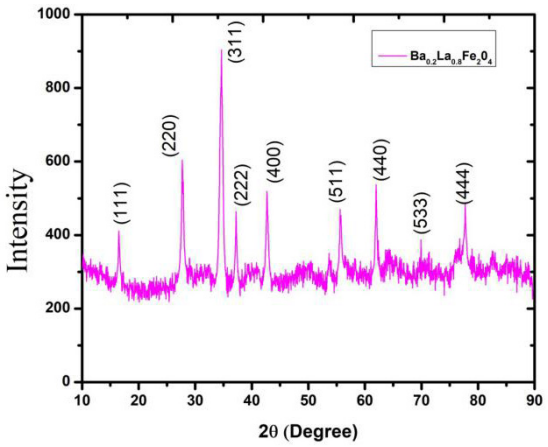
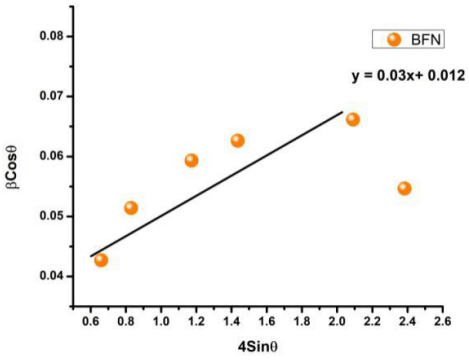


Figure 1c. X-ray diffraction spectra of the BLF nanoparticles

3.2 Williamson Hall methods

Williamson hall methods are used to find the strain induced by the crystal imperfections. Generally, the strain calculated from the X-ray diffraction using the formula ε = β/tanθ, where βis the full width half maximum and θ is the angle of diffraction. The above relation dependency on the tangent relation, to overcome this tangent dependency the w-h method shows the alternate method to determine the microstrain and the crystallite size. The WH method fallows as the plots drawn between βcosθ with respect to 4sinθ for the oriented reflection planes in the XRD patron. The slope (y = c +mx) of the plots related to the micro-strain and crystallite size as βCosθ = kλ/D' + ε' 4sinθ where ε' is referred as the microstrain and D' is crystallite size of WH method. The figure 2 shows the W-H plots of the synthesized samples such as CFN, BFN, BLFN particles respectively. The linear fitting of the plots gives the slop of the plots which relates the particles microstrain ε' and crystallite size D' the numerical values of ε', D' values are listed in table 1.



DOI: https://doi.org/10.30564/ese.v1i1.929

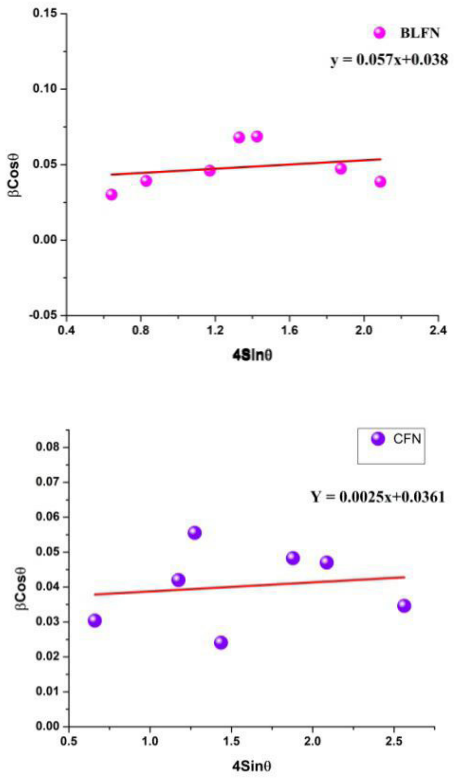


Figure 2. W-H plot analysis

Table 1. Structural factors of the CFN, BFN, and BLFN particles.

x	CFN	BFN	BFLN
a(Å)	8.434	19.36 b =5.03 c =8.9	9.85
D(nm)	25	19.26	21
Volume(nm)	599	584	955.67
ε	0.0031	0.0051	0.21
ε*	0.03	0.057	0.025
D’	30	23	34

3.3 Ferroelectric Properties

Ferroelectric properties of CFN, BFN, BLFN particles studied using with the help of P-E loop (polarization-electric field loops) as shown figure 3. The figure illuminates the BFN particles saturation polarization at 0.067 whereas in the CFN particles P-E loop illuminates no saturation polarization. The non saturation polarization indicates high dielectric loss of the CFN particles that is not ferroelectric nature. The ferroelectric parameters raveled very low coercive field 0.5kV/cm, remnant polarization are 0.04mC/cm^2. The unsaturated polarization of P-E loop given the information leakage current enhancement. This type of studies was reported by [15]. The substituent of iron cations in the ferroelectric element shows ferroelectric loop clearly it attributes the enhancement of magneto capacitance.

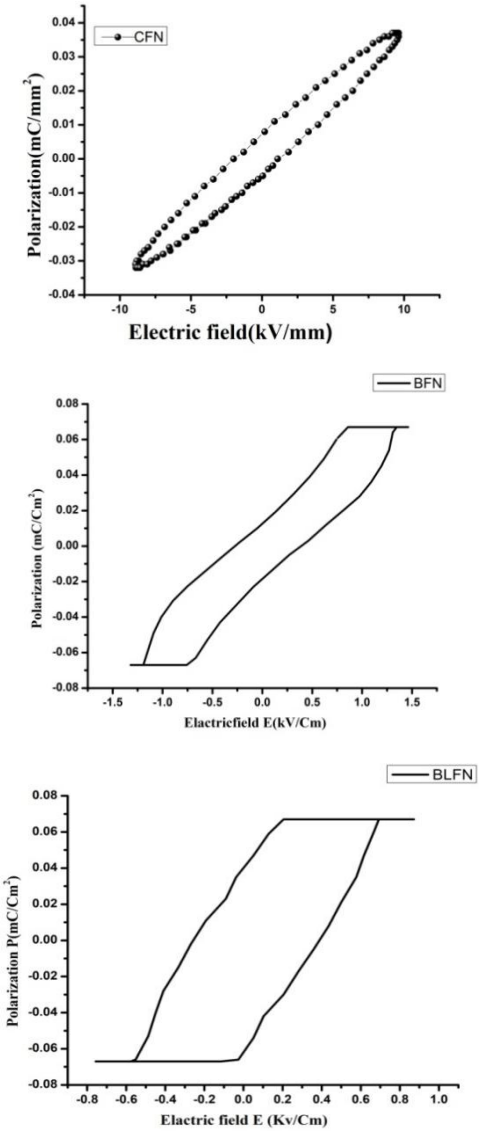


Figure 3. P-E loops of CFN, BFN, BLFN particles

On the other hand, P-E Loop of the lanthanum substituted barium ferrite nanoparticles shown fig3.6. Moreover, The ferroelectric parameters are such as remanent polarization (Pr), saturation polarization (Ps) is found from the loop as 0.02mC/Cm^2). Herein, the P-E loops of BLFN particles are saturated; this may be attributed to the low leakage current at the grain boundaries. The grain boundaries block the movement of charges developed due to applied alternating electric field resulting

 DOI: https://doi.org/10.30564/ese.v1i1.929

in low values of remanant and saturation polarization of these samples [16-18].

4. Conclusion

The $BaFe_2O_4$ (BFN), $CuFe_2O_4$ (CFN), $Ba_{0.2}La_{0.8}Fe_2O_4$(BLFN) particles are prepared via hydrothermal technique. The structural properties of synthesized particles studied with the help of X-ray diffraction analysis, Williamson hall plot linear fitting method and Ferroelectric properties were studied. The crystallite size was noticed as 25, 19, and 21 in nanometers. The microstrain of the particles is found small and whereas lanthanum substituted barium ferrite nanoparticles exhibit larger strain than the CFN, BFN particles this was attributed due to the lanthanum larger radii. In addition, the lattice parameters and unit cell volume are calculated. Also, the ferroelectric properties were studied with the help of the PE loop measurement. The obtained P-E hysteresis curve of CFN shows the material is kind of dielectric loss materials.

5. Acknowledgment

The authors thankful UGC New Delhi for providing financial support under SAP [NO.F 530/5/DRS-II/2016(SAP I)].

References

[1] K. Chandra Babu Naidu, W. Madhuri, Microwave processed bulk and nano NiMg ferrites: A comparative study on X-band electromagnetic interference shielding properties, Materials Chemistry and Physics, 2017, 187: 164-176.

[2] K. Chandra Babu Naidu and W. Madhuri,Ceramic nanoparticle synthesis at lower temperatures for LTCC and MMIC technology, IEEE Transactions on Magnetics, 2018, 54.
DOI: 10.1109/TMAG.2018.2855663

[3] Warren B. Cross, Louise Afeck, Maxim V. Kuznetsov,Ivan P. Parkin, Quentin A. Pankhurst Self-propagating high-temperature synthesis of ferrites MFe2O4 (M = Mg, Ba, Co, Ni, Cu, Zn); reactions in an external magnetic field, J. Mater. Chem., 1999, 9: 2545 -2552.

[4] S. Vadivelan, N. Victor Jaya, Investigation of magnetic and structural properties of copper substituted barium ferrite powder particles via co-precipitation method, Results in Physics, 2016, 6: 843–850.

[5] Muhammad Tahir Farid, Ishtiaq Ahmad, Muddassara Kanwal, Ghulam Murtaza, Irshad Ali, Muhammad Naeem Ashiq, Sajjad Ahmad Khan, Magnetic and electric behavior of praseodymium substituted Cu-PryFe2-yO4 ferrites, Journal of Magnetism and Magnetic Materials, 2017, 422: 337–343.

[6] D. Kothandan, R. J. Kumar & K. C. B. Naidu, Barium titanate microspheres by the low-temperature hydrothermal method: studies on structural, morphological, and optical properties, Journal of Asian Ceramic Societies, 2018, 6: 1-6.

[7] N. S. Kumar, R. P. Suvarna, K. C. B. Naidu, Grain, and grain boundary conduction mechanism in sol-gel synthesized and microwave heated PbLa0.8-yCoyTiO3 (y = 0.2-0.8) nanofibers, Materials Chemistry and Physics, 2019, 223: 241-248.

[8] S.F. Mansour, M.A. Abdo, F.L. Kzar, Effect of Cr dopant on the structural, magnetic and dielectric properties of Cu-Zn nano ferrites, Journal of Magnetism and Magnetic Materials, 2018.
https://doi.org/10.1016/j.jmmm.2018.05.104

[9] T. Ramaprasad, R. J. Kumar, U. Naresh, M. Prakash, D. Kothandan, K.C. B. Naidu, Effect of pH Value on Structural and Magnetic Properties of CuFe2O4 Nanoparticles Synthesized by Low-Temperature Hydrothermal Technique, Materials Research Express, 2018.
https://doi.org/10.1088/2053-1591/aad860

[10] Qing Lin, Jinpei Lin, Xingxing Yang, Yun He, Liping Wang, and Jianghui Dong, The effects of Mg2+ and Ba2+ dopants on the microstructure and magnetic properties of doubly-doped LaFeO3 perovskite catalytic nano crystals, Ceramics International.
https://doi.org/10.1016/j.ceramint.2018.10.246.

[11] S.M. Mane, P.M. Tirmali, B. Ranjit, M. Khan, N. Khan, A.N. Tarale, S.B. Kulkarni, Studies on magnetocapacitance, dielectric, ferroelectric, and magnetic properties of microwave sintered (1-x) (Ba0.8Sr0.2TiO3) - x (Co0.9Ni0.1Fe2O4) multiferroic composite, Solid State Sciences, 2018.
DOI: 10.1016/j.solidstatesciences.2018.05.004

[12] Anjana V, Sara John, Pooja Prakash, Amritha M Nair, Aravind R Nair, Sreedha Sambhudevan and Balakrishnan Shankar, Magnetic Properties of Copper Doped Nickel Ferrite Nanoparticles Synthesized by Co-Precipitation Method, IOP Conf. Series: Materials Science and Engineering, 2018, 310: 012024.
DOI: 10.1088/1757-899X/310/1/012024

[13] N. S. Kumar, R. P. Suvarna and K. C. B. Naidu, Sol-Gel Synthesized and Microwave Heated Pb0.8-yLayCo0.2TiO3 (y= 0.2–0.8) Nanoparticles: Structural, Morphological and Dielectric Properties, Ceramics International, 2018, 44:18189-18199.

[14] H. Ghayour & Majid Abdellahi & Mazyar Ghadiri Nejad & Amirsalar Khandan & Saeed Saber-Saman-

DOI: https://doi.org/10.30564/ese.v1i1.929

dari, Study of the effect of the Zn+2 content on the anisotropy and specific absorption rate of the cobalt ferrite: the application of Co1-xZnxFe2O4 ferrite for magnetic hyperthermia, J. Aust. Ceram. Soc. 2018, 54: 223-230.

[15] U. Naresh, R. J. Kumar, K. C. B. Naidu, Optical, Magnetic and Ferroelectric Properties of Ba0. 2Cu0.8-xLaxFe2O4 (x = 0.2 - 0.6) Nanoparticles, Ceramics International, 2019, 45: 7515-7523.

[16] S.F. Mansour, M.A. Abdo, F.L. Kzar, Effect of Cr dopant on the structural, magnetic and dielectric properties of Cu-Zn nano ferrites, Journal of Magnetism and Magnetic Materials, 2018. DOI: https://doi.org/10.1016/j.jmmm.2018.05.104

[17] Poonam Pahujaa, Chandra Prakash, R.P.Tandon "Comparative study of magnetoelectric composite system Ba0.95Sr0.05 TiO3 – Ni0.8Co0.2Fe2O4 with ferrite prepared by different methods", CeramicsInternational, 2013. http://dx.doi.org/10.1016/j.ceramint.2013.11.012

[18] K. C. B. Naidu, V. N. Reddy, T. S. Sarmash, D. Kothandan, T. Subbarao, N. S. Kumar, Structural, morphological, electrical, impedance and ferroelectric properties of BaO-ZnO-TiO2 ternary system, J Aust Ceram Soc., 2018. https://doi.org/10.1007/s41779-018-0225-0

 DOI: https://doi.org/10.30564/ese.v1i1.929