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DIELECTRICPROPERTIES OF Ba6-3xNd8+2xTi18O54CERAMICS

LEE CHIAN HENG

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Abstract of thesis presented to the Senate of Universiti Putra Malaysiainfulfillment of the requirement for the degree of Master of Science

DIELECTRICPROPERTIES OF $Ba_{6-3x}Nd_{8+2x}Ti_{18}O_{54}$CERAMICS

By

LEE CHIAN HENG

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Chairman: Jumiah Hassan, PhD

Faculty: Science

In this research, high dielectric constant material especially in microwave frequency region was investigated. Pure barium titanate showed low dielectric constant value at high frequency region. In order to improve the properties, rare earth dopant was introduced to this material. The doping mechanism of neodymium ion on barium titanate could be promising a new material for applications in miniature microwave technology and mobile communication systems. The processing of the material always influences materials properties. Therefore, microstructural and complex permittivity of $Ba_{6-3x}Nd_{8+2x}Ti_{18}O_{54}$, with x=0.15 ceramics at different sintering temperatures were investigated. The phenomenon of polarization produced by the applied electric field was studied. The samples were prepared by the wet solid state method and sintered at a temperature range from 600°C to 1300°C. Sintering effects on the crystallite structure and surface morphology were studied and characterized by XRD and FESEM. The transformation of majority of the phases in the system from barium titanate to barium neodymium titanate was confirmed by XRD pattern due to change in sintering temperature. The shrinkage of each sintered ceramic was determined compared with the original dimension. The change in sample densities was determined using Archimedes' method. BNT ceramic with highest sintering temperature obtained full densification. This is confirmed by the micrograph from FESEM, which showed grains are in rectangular shape. Two activation energies of 0.0698 eV for low sintered ceramics and 0.3348 eV for high sintered ceramics were observed by using estimated diffusion process.The dielectric properties with respect to the frequency from 1 MHz to 1.5 GHz were measured using the Impedance Analyzer, and the results were compared and analyzed. The highest dielectric constant and lowest loss tangent were defined among the samples.The BNT ceramic sintered at 1300°C obtained the highest dielectric constant of 30 at 1 GHz and 0.09 for loss tangent. The dielectric constant of this ceramic also showed almost independent of frequency. The complex dielectric modulus was used to differentiate the contribution of the grain and grain boundary. The results showed the semicircular arc changing when sintering temperature increased due to the influence of the secondary phases. For further investigation, BNT ceramic with x=0.25 was fabricated in order to compare with BNT x=0.15 at low frequency region and temperature effect. Higher Nd dopant definitely decreased the dielectric valuefrom

26.5 for BNT x=0.15 to 25.9 for BNT x=0.25 at 1 MHz at room temperature, but both ceramics showed independence of temperature and frequency. The desirable characteristics of $Ba_{6-3x}Nd_{8+2x}Ti_{18}O_{54}$for x=0.15 sintered at 1300°Cinclude high dielectric constant of 30, low loss tangent of 0.09, and high quality factor developed a new field for electronic applications.

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SIFAT DIELEKTRIKBAGI$Ba_{6-3x}Nd_{8+2x}Ti_{18}O_{54}$SERAMIK

Oleh

LEE CHIAN HENG

Oktober 2014

Pengerusi: Jumiah Hassan, PhD

Fakulti: Sains

Barium titanat yang tulen menunjukkan nilai pemalar dielektrik yang rendah pada rantau frekuensi tinggi. Dalam usaha untuk memperbaiki sifat-sifat tersebut, pendopan nadir bumi telah diperkenalkan kepada bahan ini. Mekanisme pendopan neodimium ion pada barium titanat boleh menjanjikan bahan baru untuk aplikasi dalam teknologimikrogelombang dan sistem komunikasi mudah alih. Pemprosesan bahan sentiasa mempengaruhi sifat bahan. Oleh itu, ketelusan mikrostruktur dan kompleks $Ba_{6-3x}Nd_{8+2x}Ti_{18}O_{54}$, dengan x = 0.15 seramik pada suhu pensinteran yang berbeza telah dikaji. Fenomena polarisasi yang dihasilkan oleh medan elektrik yang digunakan telah dikaji. Sampel telah disediakan dengan kaedah keadaan pepejal basah dan disinter pada pelbagai suhu dari 600°C hingga 1300°C. Kesan pensinteran ke atas struktur crystallite dan morfologi permukaan telah dikaji melalui kajian XRD dan FESEM. Transformasi fasa majoriti dalam sistem daripada barium titanat kepada barium neodimium titanat telah disahkan oleh corak XRD disebabkan perubahan suhu pensinteran. Pengecutan setiap seramik tersinter ditentukan berbanding dengan dimensi asal. Perubahan dalam ketumpatan sampel ditentukan dengan menggunakan kaedah Archimedes. BNT seramik dengan suhu pensinteran tertinggi yang diperolehi sepenuhnya pemadatan juga disahkan oleh keputusan mikrostruktur melalui FESEM, yang menunjukkan butiran dalam bentuk segi empat tepat. Dua tenaga pengaktifan 0.0698eV untukseramiktersinterrendah dan0.3348eV untukseramiktersintertinggi diperhatikan dengan menggunakan anggaran proses penyebaran. Sifat-sifat dielektrik berkenaan dengan frekuensi daripada 1 MHz hingga 1.5 GHz telah diukur dengan menggunakan Impedance Analyzer, dan keputusan dibandingkan dan dianalisis. Pemalar dielektrik yang paling tinggi dan tenaga kehilangan yangpaling rendah telah ditakrifkan antara sampel. BNT seramik tersinter pada suhu 1300°Cperolehi pemalar dielektrik tertinggi 30 dan0.09untuk kehilangantangen. pada 1 GHz, dan ia menunjukkan hampir tidak bersandar pada frekuensi. Modulus dielektrik kompleks digunakan untuk membezakan sumbangan butiran dan sempadan butiran. Keputusan menunjukkan arka separuh bulatan berubah apabila pensinteran suhu meningkat kerana dipengaruhi oleh fasa-fasa yang lain.Untuk kajian lanjut, BNT seramik

dengan x = 0.25 telah dihasilkan untuk dibandingkan dengan BNT x = 0.15 di rantau frekuensi rendah dan kesan suhu. Pendopan Nd yang tinggi telah menurunkan nilai pemalar dielektrik, iaitudaripada26.5untukBNTx =0.15 kepada 25.9untukBNTx =0.25pada 1MHz untuksuhu bilik,tetapi kedua-dua seramik menunjukkan tidak bersandar pada suhu dan frekuensi. Ciri-ciri wajar $Ba_{6-3x}Nd_{8+2x}Ti_{18}O_{54}$untuk x=0.15disinter pada1300°Ctermasukpemalardielektrik tinggidengan nilai 30, kehilangan tenaga rendahdengan nilai 0.09, dan faktor kualiti tinggi telah membangunkan satu bidang baru bagi aplikasi elektronik .

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Approval Sheet 1

I certify that an Examination Committee has met on **13 October 2014**toconduct the final examination of **Lee Chian Heng**on his **master**thesis entitled "**DIELECTRICPROPERTIES OF $Ba_{6-3x}Nd_{8+2x}Ti_{18}O_{54}$CERAMICS**" in accordance with UniversitiPertanianMalaysia (Higher Degree) Act 1980 and UniversitiPertanian Malaysia(Higher Degree) Regulations 1981. The Committee recommends that thestudent be awarded the (Name of relevant degree).Members of the Examination Committee were as follows:

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NORITAH OMAR, PhD
Professor and Deputy Dean
School of Graduate Studies
Universiti Putra Malaysia
Date:

This thesis was submitted to the Senate of Universiti Putra Malaysia andhas been accepted as fulfillment of the requirement for the degree of Master of Science. The members of the Supervisory Committee were as follows:

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Faculty of Science
Universiti Putra Malaysia
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Institute of Advanced Technology
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at%	Atomic percentage
wt%	Weight percentage
°C	Degrees Celsius
τ_f	Resonant frequency
ε_{eff}	Effective dielectric constant
kHz	kilo Hertz
MHz	Mega Hertz
GHz	Giga Hertz
XRD	X-ray diffraction
eV	Electron volt
λ	Wavelength
d	Interplanar spacing
FESEM	Field Emission Scanning Electron Microscope
ρ	Density
µm	Micrometer
nm	Nanometer
ε^*	Complex dielectric constant
ε’	Dielectric constant

ε”	Dielectric loss factor
M*	Complex dielectric modulus
M’	Real part of dielectric modulus
M”	Imaginary part of dielectric modulus
$\tan\delta$	Dielectric loss tangent
σ_{ac}	AC conductivity
h	Planck constant
c	Speed of light
|Z|	Magnitude of impedance
θ	Phase
C	Capacitance
V	Voltage
ω	Angular frequency
i	Current
Q	Charges
E	Electric field
ε_0	Permittivity of vacuum
μ	Dipole moment
P	Polarization

CHAPTER 1

INTRODUCTION

1.0 Introduction

In this chapter, scope and objective of the research were discussed. The purpose of this research was to investigate the dielectric materials with excellent dielectric properties. However, the properties of the materials are usually related to the microstructure and macrostructure inside the materials, especially for the composite materials. The motivation of this research is stated in problem statement. The objectives of the research were also defined in order to resolve the problems. Besides, the properties of the related elements and compounds were also introduced in this chapter. The information such as descriptions of the elements and compounds were presented. The change of the properties when the reaction occurred was briefly explained here.

1.1 Introduction of properties of elements and compounds

1.1.1 Barium, Neodymium, Titanium, and Oxygen

Barium is a soft silvery metallic alkaline earth metal which is listed in Group 2 of the periodic table. Barium was first discovered by an English chemist, Sir Humphry Davy at 1808. The thin films of barium are used as rotors of anodes in vacuum X-ray tubes. Neodymium was discovered by an Australian chemist, Carl Auer von Welsbach in 1885. The main use of neodymium is as a colourant for glasses, which can produce colours ranging from pure violet to wine red through to warm grey. When neodymium is mixed with iron and boron, it produced powerful permanent magnets, and these magnets are used in computers, cell phones, motors, wind turbines, and audio systems. Titanium was first discovered by a British clergyman and mineralogist, William Gregor, but named by a German chemist, Martin Heinrich Klaproth in 1791. Titanium is a silvery white transition metal found in mineral rutile or ore ilmenite. Titanium is used in many applications such as aerospace, industrial, medical, and other recreational fields. It has been used in various applications due to its light weight, high temperature performance, high level of corrosion resistance, and good biocompatibility and durability for human implants. Oxygen was discovered by Carl Wilhelm Scheele in 1773, but the related work was published by Joseph Priestley in 1774. Oxygen was named by Antoine Lavoisier in 1777 when he disproved the phlogiston theory of combustion and corrosion. Oxygen are widely used in steel production that remove the impurity carbon element by producing carbon dioxide gases, as an oxidant for rocket fuel, and respiration of plants and animals. The important properties of barium, neodymium, titanium, and oxygen elements were shown in Table 1.1.

Table 1.1: Physical and chemical properties of barium, neodymium, titanium, and oxygen (David 2010)

Properties	Barium	Neodymium	Titanium	Oxygen
Symbol	Ba	Nd	Ti	O
Atomic number	56	60	22	8
Atomic radius (Å)	1.34	1.81	1.47	0.68
Atomic weight (amu)	137.327	144.242	47.87	15.9994
Density at 20°C (g cm^{-3})	3.62	7.01	4.506	0.001429
Molar Volume at 20°C (cm^3/mol)	38.16	20.59	10.64	17.36
Melting (°C)	727	1016	1668	-218.79
Boiling (°C)	1897	3074	3287	-182.953
Crystal structure	BCC	Hexagonal	Hexagonal	Cubic
Electrical resistivity at 20°C (nΩ.m)	332	643	390	-
Dielectric constant at infinite frequency (calculated)	1.00000000033353	1.00000000515081	1.00000002004916	1

1.1.2 Barium Carbonate, Neodymium Oxide, and Titanium dioxide

Barium carbonate or witheritewas discovered from barite by William Withering in 1784. It is a heavy, odorless, white-to-cream colored chemical with the chemical formula $BaCO_3$. Unlike Group I carbonates,barium carbonate which is listed in Group II carbonates is insoluble in water.Carbonates are the compounds that contain CO_3^{2-} ions. It has been noticed that barium carbonate can be thermally decomposed to produce barium oxide and carbon dioxide gas at about temperature 1360°C with larger scale size. However, barium carbonate is the most difficult to decompose compare to other carbonates due to its high thermal stability. Barium carbonate is widely used in chemical applications to remove the sulfates in phosphoric acid production and chlorine alkali electrolysis. It is also used as a flux in ceramic, and as an ingredient in glass production. The thermal decomposition of barium carbonate is shown below:

$$\mathrm{BaC}O_3 \xrightarrow[heating]{} \mathrm{BaO} + \mathrm{C}O_2 \qquad (1.1)$$

Neodymium (III) oxide is one of the rare earth oxides that are insoluble in water. It is formed as a sesquioxide because neodymium element has oxidation state +3 that oxidize completely to Nd_2O_3 when heating the neodymium element in air about 150°C. The applications of neodymium (III) oxide are similar to the neodymium element, but it is also used as polymerization catalyst. The chemical reaction of Nd_2O_3 formed as follows:

$$\mathbf{4Nd} + \boldsymbol{3O_2} \xrightarrow[\boldsymbol{heating}]{} \boldsymbol{2Nd_2O_3} \qquad (\mathbf{1.2})$$

Titanium dioxide is one of organic compounds with chemical formula TiO_2. It is a white solid in color and insoluble in water. Titanium has +4 oxidation state, and it formed titanium dioxide when heated in air. The general chemical reaction for TiO_2 is showed in equation 3. Titanium dioxide was widely used as pigments, sunscreen, and photocatalyst. The important properties of barium carbonate, neodymium oxide, and titanium dioxide were shown in Table 1.2.

$$\mathbf{Ti} + \boldsymbol{O_2} \xrightarrow[\boldsymbol{heating}]{} \boldsymbol{TiO_2} \qquad (\mathbf{1.3})$$

Table 1.2: Physical and chemical properties of barium carbonate, neodymium oxide, and titanium dioxide in nano size (David, 2010)

Properties	Barium Carbonate	Neodymium Oxide	Titanium Oxide
Chemical formula	$BaCO_3$	Nd_2O_3	TiO_2
Chemical Abstracts Service Registry Number	513-77-9	1313-97-9	13464-67-7
Molecular weight (amu)	197.336	336.482	79.866
Density at 20°C (g cm-3)	4.308	7.24	4.17
Melting (°C)	811	2233	1843
Boiling (°C)	1360	3760	3000
Crystal structure	orthorhombic	hexagonal	tetragonal

1.2 Tungsten Bronze structure

Tungsten bronzes are the nonstoichiometric compounds that have the general formula $A_xMO_{3-y+zx/2}$, where A is electropositive metal, M is transition metal, and O is oxide of the compounds. For an ideal tungsten bronze, y in the composition is equal to zx/2, therefore the formula become A_xMO_3. The composition x of the compound not only affects the chemical properties, but also physical properties. It has been noticed that if the composition x is smaller than 0.3, the compounds are semiconducting materials. However, if the composition x is greater than 0.3, the compound becomes a conductor due to the influence of the Fermi level. The stability of the tungsten bronze depends on the crystallite size of A in the chemical formula. The stability of tungsten bronze increased with increase in the ionic radius of A. In this research, barium and neodymium were selected as electropositive metal, and titanium was chosen as transition metal. As mentioned above, Ba^{2+} and Nd^{3+} are divalent and trivalent cationic form of barium and neodymium. In order to achieve the electrostatic stability, three Ba^{2+}ions can be replaced by two Nd^{3+} ions and a vacancy. The chemical equation is showed below.

$$3Ba^{2+} \xleftrightarrow[electrostatic \quad stability]{} 2Nd^{3+} + vacancy \qquad (1.4)$$

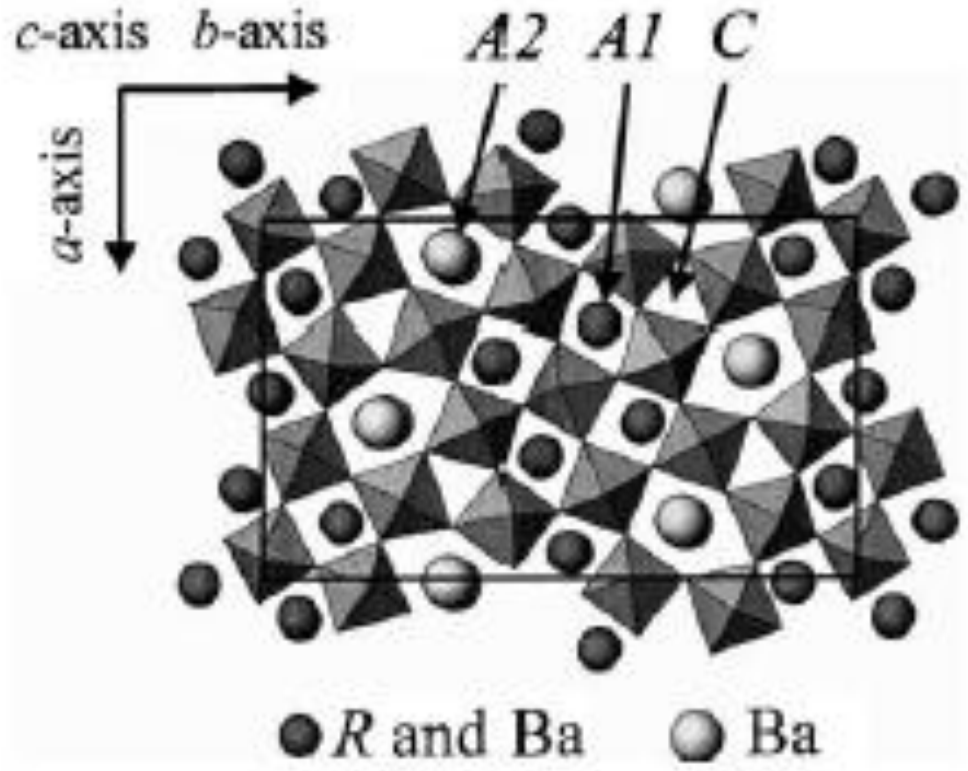

Figure 1.1 Projection of tungsten bronze type structure of $Ba_{6-3x}R_{8+2x}Ti_{18}O_{54}$ solid solutions in c-plane (Ohsato and Imaeda, 2003)

Figure 1.1 shows crystal structure of $Ba_{6-3x}R_{8+2x}Ti_{18}O_{54}$ tungsten bronze type solid solutions, where R=Nd. This structure constructed by corner sharing of TiO_6 octahedral formed three cation sites. The largest cation site which is A2 in the structure has pentagonal shape, and always filled with Ba ions. The medium site of the structure that has rhombic spacing is A1, mainly partially occupied by Nd ions and Ba ions. The smallest trigonal spacing in C site was empty according to the compositional formula. It actually can be filled by small ionic radius ions such as Li or Mg ions, but not in this case. The tungsten bronze types like $Ba_{6-3x}R_{8+2x}Ti_{18}O_{54}$solid solutions were found by Varfolomeev (2003). These types of materials are currently widely used in the development of microwave telecommunication technology due to their high dielectric constant. The cations in the solid solutions are important to the dielectric properties of the compounds. The chemical formula $Ba_{6-3x}R_{8+2x}Ti_{18}O_{54}$enable to clearly show the effect on the replacement of Ba with Nd elements. In this case, the atomic ratio of titanium to oxygen is 1:3 which is similar to perovskite structure ABX_3. This means that the solid solutions $Ba_{6-3x}R_{8+2x}Ti_{18}O_{54}$can also be used as application of perovskite structure such as sensors, memory devices, and spintronics.

1.3 Problem statement

Nowadays, miniaturization of electronic components has created new challenge for materials research. To maintain high performance in decreasing the size, a deep research on the microstructure and the properties of the materials is needed.

Microstructure, especially the grain size strongly influences the dielectric properties of the materials. The more crystalline material will increase the dielectric constant. The fast growth of the nanotechnology is opening a new field of science in order to understand the material properties at nanoscale. The processing of the materials is one of the factors that affect the quality of the materials.$BaTiO_3$ is the well-known material that has high dielectric constant. However, the properties of doping ion into $BaTiO_3$ are still not yet clear especially during the sintering process. Evolution of the microstructure and dielectric properties will be studied.

1.4 Objective of the research

General Objective:

The aim of this project is to investigatephases, microstructural, and dielectric properties of $Ba_{6-3x}R_{8+2x}Ti_{18}O_{54}$, with R = Nd (rare-earth oxide)at x=0.15 and 0.25prepared by wet solid-state reaction method and sintered at different sintering temperatures.

Specific Objectives:

1. To prepare $Ba_{6-3x}Nd_{8+2x}Ti_{18}O_{54}$and study its crystalline structure and physical characteristics such as density, porosity, and grain size of sintered ceramics.
2. To determine the dielectric properties in microwave region, and relate it to the microstructure.
3. To analyze the samples using complex dielectric impedance.

1.5Significance of Research

Recently microwave dielectric materials are extensively investigated because of their growing potential for the applications to mobile communication systems. The desirable characteristics of the microwave materials include low dielectric loss, high quality factor, high dielectric constant and small temperature coefficient of resonant frequency. For microwave frequency applications, high dielectric constant materials based on $BaO-R_2O_3-TiO_2$(R = rare earth) phase systems are widely used in the electronics industry. The compounds that possess excellent dielectric properties and temperature stability are an interesting topic in microwave devices. The study of replacement of barium by rare earth material in this research is to find out which could be a promising material for application in microwave technology.

1.6 Naono science and its trends

In view of science and engineering, nano science is to understand the object in terms of atoms, molecules, and nano size particles. Nowadays, nanotechnology is developed due to its unclear applications. The same materials with different dimensions will give impact to their applications. For example, combination of the nano size particles is more easily compared to the micro size particles during the formation period. Not only that, the space inside the shaped-sample can be well minimized by smaller size particles. One nano meter is equal to 10^{-9} meter length. Thanks to miniaturization of electronic components, materials researchers started to produce material with particle size below 100 nm. The applications of nano science are widely used in many fields. One of the interesting applications is spintronic device, which is utilized in the spinning behavior of electrons. For this high frequency application, it needs smaller dimension to enhance the properties of the devices. For dielectric materials, most of the researchers and industries have the interest to find the high dielectric constant materials in order to block current or store charges.

REFERENCES

Agilent 4291B RF Impedance/Material Analyzer datasheet

Alfa Mirage Electronic Densimeter MD-300S Instruction Manual

Bhowmik, R. N., and Panneer Muthuselvam, I. (2013). Dielectric properties of magnetic grains in $CoFe_{1.95}Ho_{0.05}O_4$ spinel ferrite. *Journal of Magnetism and Magnetic Materials, 335*(0), 64-74.

Callister, W.D., and Rethwisch, D.G. (2010). Materials science and engineering: an introduction 8th edition. (pp. 108-113)

Clugston, M.; and Flemming, R. (2000). Advanced Chemistry. Oxford University Press. pp. 290-291.

David R. Lide, *CRC Handbook of Chemistry and Physics, 90th Edition (CD-ROM Version 2010),* CRC Press/Taylor and Francis, Boca Raton, FL

Fang, L. Xiang, F., Liao, W., Liu, L.J., Zhang, H., and Kuang, X.J. "Dielectric Properties and High-Temperature Dielectric Relaxation of $Ba_3Ti_4Nb_4O_{21}$ Ceramic." *Materials Chemistry and Physics* 143.2 (2014): 552-6.

Gedam, R. S., and Ramteke. D. D. "Electrical, Dielectric and Optical Properties of La_2O_3 Doped Lithium Borate Glasses." *Journal of Physics and Chemistry of Solids* 74.7 (2013): 1039-44. Print.

Hirose, N., Skakle, J. M. S., and West, A. R. (1999). Doping mechanism and permittivity correlations in Nd-doped $BaTiO_3$. *Journal of Electroceramics, 3*(3), 233-238.

Kaur, D., and Narang, S. B. (2012) Electronic properties of complex barium-neodymium titanates in microwave regime.*International Conference on Emerging Trends in Computer and Electronics Engineering*

Kaur, D., Narang, S. B., and Thind, K. S. (2006). Processing, dielectric behavior and conductivity of some complex tungsten-bronze dielectric ceramics. *Journal of Ceramic Processing & Research*, *7*(1), 31-36.

Kim, D. H., Park, H., Kim, S., and Lee, K. (2005). Characteristics of Ni 8 wt%-doped titanium dioxide photocatalyst synthesized by mechanical alloying. *Catalysis Letters*, *100*(1-2), 49-52

Lassner, E., and Schubert, W.Tungsten. (1999). Tungsten: properties, chemistry, technology of the element, alloys, and chemical compounds. Springer pp. 160-164

Lee, S. M., Jo, Y. H., Kim, H. E., Mohanty, B. C., and Cho, Y. S. (2012). Barium neodymium titanium borate glass-based high k dielectrics. *Journal of the American Ceramic Society, 95*(4), 1356-1359.

Li, Y.X., Yao, X., and Zhang, L.Y. (2004). High permittivity neodymium-doped barium titanate sintered in pure nitrogen.*Creamics International,* 30, 1325-1328

Narang, S. B., and Kaur, D. (2009). Temperature dependent dielectric behavior of neodymium substituted barium titanate ceramics. *Ferroelectrics, 393*(1), 106-113.

Ohsato, H. (2001). Science of tungstenbronze-type like $Ba_{6-3x}R_{8+2x}Ti_{18}O_{54}$ (R=rare earth) microwave dielectric solid solutions. *Journal of the European Ceramic Society, 21*(15), 2703-2711

Ohsato, H., and Imaeda, M. (2003). The quality factor of the microwave dielectric materials based on the crystal structure—as an example: The $Ba_{6-3x}R_{8+2x}Ti_{18}O_{54}$ (R = rare earth) solid solutions. *Materials Chemistry and Physics*, *79*(2–3), 208-212

Ohsato, H., and Imaeda, M. (2003). The quality factor of the microwave dielectric materials based on the crystal structure—as an example: The $Ba_{6-3x}R_{8+2x}Ti_{18}O_{54}$ (R = rare earth) solid solutions. *Materials Chemistry and Physics, 79*(2–3), 208-212.

P. Debye, (1929) Polar Molecules, Dover Publications, New York.

Pornprasertsuk, R., Yuwapattanawong, C., Permkittikul, S., and Tungtidtham, T. (2012). Preparation of doped $BaZrO_3$ and $BaCeO_3$ from nanopowders. *International Journal of Precision Engineering and Manufacturing, 13*(10), 1813-1819.

Raju, G. G. (2003). Dielectric loss and relaxation - I. *Dielectrics in Electric Fields*. CRC Press.

Raju, G. G. (2003). Dielectric loss and relaxation - II. *Dielectrics in Electric Fields*. CRC Press.

Raju, G. G. (2003). Polarization and static dielectric constant. *Dielectrics in Electric Fields*. CRC Press.

Rejab, N., Sreekantan, S., Abd Razak, K., and Ahmad, Z. (2011). Structural characteristics and dielectric properties of neodymium doped barium titanate. *Journal of Materials Science: Materials in Electronics, 22*(2), 167-173.

Samkaria, R.,and Sharma, V. (2013) "Effect of Rare Earth Yttrium Substitution on the Structural, Dielectric and Electrical Properties of Nanosized Nickel Aluminate." *Materials Science and Engineering: B* 178.20: 1410-5.

Singh, P., Agarwal, A.,Sanghi, S., Navneet Singh, N.,and Khasa, S. "Dielectric Characterization of Bismuth Layered $(Bi_2O_3)(Na_xFe_{1-x}O_3)$ Ceramics." *Physica B: Condensed Matter* 436 (2014): 64-73.

Snashall, A. L., Norén, L., Liu, Y., Yamashita, T., Brink, F., and Withers, R. L. (2012). Phase analysis and microwave dielectric properties of $BaO–Nd_2O_3–5TiO_2$ composite ceramics using variable size TiO_2 reagents. *Ceramics International, 38, Supplement 1*(0), S153-S157.

Wagner, K.W. (1973). The distribution of relaxation times in typical dielectrics, *Annals of Physics*, 40 817–819.

Waseda, Y., Matsubara, E., and Shinoda, K. (2011). Fundamental properties of X-rays. (pp. 1-20). Springer.

Waseda, Y., Matsubara, E., and Shinoda, K. (2011). Scattering and diffraction. (pp. 67-106). Springer.

Wu, M., Hsieh, M., Yen, C., Huang, Y., Huang, W., and Su, W. (2007). Low sintering $BaNd_2Ti_4O_{12}$ microwave ceramics prepared by CuO thin layer coated powder. *Journal of the European Ceramic Society, 27*(8–9), 2835-2839.

Yao, X., Lin, H., Zhao, X., Chen, W., and Luo, L. (2012). Effects of Al_2O_3 addition on the microstructure and microwave dielectric properties of $Ba_4Nd_{9.33}Ti_{18}O_{54}$ ceramics. *Ceramics International, 38*(8), 6723-6728.

Yao, Z., Liu, H., Liu, Y., Wu, Z., Shen, Z., Liu, Y., and Cao, M. (2008). Structure and dielectric behavior of Nd-doped $BaTiO_3$ perovskites. *Materials Chemistry and Physics*, 109(2–3), 475-481.

Zhang, L., Chen, X. M., Qin, N., and Liu, X. Q. (2007). Upper limit of x in $Ba_{6-3x}Nd_{8+2x}Ti_{18}O_{54}$ new tungsten bronze solid solution. *Journal of the European Ceramic Society, 27*(8–9), 3011-301

Zhang, W., Cao, L., Su, G., and Liu, W. (2013). Influence of microstructure on dielectric properties of Nd-doped barium titanate synthesized by hydrothermal method. *Journal of Materials Science: Materials in Electronics, 24*(6), 1801-1806.

Zheng, X. H., Chen, X. M., and Wang, T. (2006). Aging effects on dielectric properties of barium neodymium titanium tantalate ceramics. *Journal of Materials Science: Materials in Electronics, 17*(7), 543-547.

Zhou, L., Zhou, H., Shao, H., and Zhi, H. (2012). Microstructure and microwave dielectric properties of $Ba_{6-3x}Sm_{8+2x}Ti_{18}O_{54}$ ceramics with various $Ba_xSr_{1-x}TiO_3$ additions. *Journal of Rare Earths, 30*(2), 142-145.