

Effect of Average Ionic Radius of A-site, B-site in Ba_{0.85}X_{0.15}TiO₃, BaY_{0.15}Ti_{0.85}O₃ Systems Respectively (X = Pb, Ca, Co), (Y = Zr, Mn, Si) as a Perovskite Structure Ceramics (ABO₃) on Crystal Structure, Curie Temperature

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Research Article

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**Effect of Average Ionic Radius of A-site, B-site in $\text{Ba}_{0.85}\text{X}_{0.15}\text{TiO}_3$,
 $\text{BaY}_{0.15}\text{Ti}_{0.85}\text{O}_3$ Systems Respectively (X = Pb, Ca, Co), (Y = Zr,
Mn, Si) as a Perovskite Structure Ceramics (ABO_3) on Crystal
Structure, Curie Temperature**

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Abstract

Barium titanate BaTiO_3 is one of the most ferroelectrics. Its commonly used in capacitors due to its high dielectric constant. In this study, our goal is to determine the extent to which the change in the average ionic radius of A, B sites affects the structural, dielectric properties of this ceramic. In this study, barium titanate ($\text{Ba}_{0.85}\text{X}_{0.15}\text{TiO}_3$, $\text{BaY}_{0.15}\text{Ti}_{0.85}\text{O}_3$) (X = Pb, Ca, Co, Y= Zr, Si, Mn) was prepared by solid state reaction method in which the calcination temperature was 1200 °C for 2 hours. Structural characterization and dielectric properties were determined and calculated by using x-ray diffractometer, RCL-Meter (PM6303) at frequency 20 Hz using different temperature respectively. The results of the work showed that the average ionic radius at the A, B sites plays a large and clear role in determining the values of the dielectric constant and Curie temperature in perovskites in addition to the structural parameters.

1. Introduction

Barium titanate BaTiO_3 is one of the most common ferroelectrics. Ferroelectricity is the spontaneous polarization in the absence of an electric field. The spontaneous polarization is a consequence of the positioning of the Ba^{+2} , Ti^{+4} and O^{-2} within the unit cell as in the fig (1).

Under the ferroelectric curie temperature ($120\text{-}130\text{ }^{\circ}\text{C}$) the unit cell of BaTiO_3 is tetragonal in which spontaneous polarization occurs in which the titanium ions displaced along the c-axis, but when BaTiO_3 is heated above this temperature, the unit cell became cubic (nonpolar phase) [3,4,5]. BaTiO_3 has a perovskite structure ($\text{A}^{+2}\text{B}^{+4}\text{O}_3$) in which ($\text{A} = \text{Ba}^{+2}$, $\text{B} = \text{Ti}^{+4}$)

In this structure Ba^{+2} coordinated by 12 oxygen ions , Ti^{+4} coordinated by 6 oxygen ions which form an octahedron as structure in figure [6], this structure shows a cubic unit cell with large cation A on the corner, a smaller cation B in the body centers of the faces. BaTiO_3 was candidate for piezoelectric transducer applications and the ferroelectric multilayer ceramic capacitors(MLCCs) extremely important and widely used in many applications include capacitor, phonograph pickups, band pass filters, medical ultrasound ,RAMs, biocompatible,nanogenerators, etc [7,8,9] There are many studies that dealt with the preparation and study of BaTiO_3 . Sareecha et al.were studied the temperature dependence of dielectric constant for pure and Pb-doped BaTiO_3 ceramics [10]. Shuvo et al were found out that doped of BaTiO_3 with PbO in 20 % led to developing in dielectric properties [11]. Zhang et al. were elaborated the enternal mechanism for high piezoelectric response of $\text{Ba}(\text{Ti}_{0.8}\text{Zr}_{0.2})\text{O}_3 - (\text{Ba}_{0.7}\text{Ca}_{0.3})\text{TiO}_3$ [12]. Kim and Song were found that addition of Mgo (0.2 – 1 wt %) led to increasing of dielectric permittivity while decreasing with addition of Y_2O_3 (0.3-1.5 wt%) [13]. Butee et al. were prepared the $\text{Ba}_{1-x}\text{Pb}_x\text{TiO}_3$ and found that the curie temperature increasing from 120 C^0 to 180 C^0 for ($x=0.15$) [14]. Kadira et al. were studied the effect of Ca^{+2} ion ($x= 0.01 - 0.1$) on ferroelectric properties of $\text{Ba}_{1-x}\text{Ca}_x\text{TiO}_3$ and they found that the maximum Curie temperature at ($x=0.05$)[15]. Islam et al. were studied the effect of Mn^{+4} ion

on the dielectric, microstructural properties of Ba Mn_x Ti_{1-x} O₃ (x = 0.0-0.04)[16] the aim of this work is to indicate and verify that whether the radius of the ion added to the base ion in the (A-site = Ba⁺²) and (B-site = Ti⁺⁴) or in other words the average ionic radius has an effect on ferroelectric Curie temperature, note that there are articles that have shown that the ionic radius < r_{A-site}> of A-site ions effect strongly on the Curie temperature [17] .

2. Experimental Part

1- The compounds used :

2- Sample preparation :

After performing the operations of weighing the used compounds according to the proportions mentioned in table (2) in which the mole ratio for both BaCO₃ ,TiO₂ used in this work was [1 :1] according to [18 , 19]. the wet mixing process was performed by using magnetic stirrer with heating at 70 C⁰ after adding distilled water to the beakers that containing prepared powders for a better reaction process , the mixing process lasted for 1 hr.After that, these beakers were placed in the oven at 150 C⁰ for 2 hr to drying the samples. Then , these samples were ground by using a gate mortar for more homogenous , and to check the prepared phase , samples were examined by XRD technique using (Philips: PW 1316/90, single pen recorder/ Cu K_α Target). After that , the samples were calcined in a muffle furnace type () at 1200 C⁰ for 2 hr at heating rate 8 C⁰ / min . The calcined samples were strongly grounded by using ball milled process to get the smallest grain size possible in which the samples placed in a cylindrical container with two stainless steel balls in which the crushing process holder is performed after placing the container on a vibrating stand for 2 hr.

Then, the crushed samples were pressed under pressure ()to discs with diameter 2 cm and thickness (3 – 5 mm) as shows in figure () by using hydraulic press, and for a coherent , regular shape the PVA binder was used .The discs were sintering at 1200 C⁰ for 2 hr at heating

rate $6\text{ C}^0/\text{min}$ after remaining at 500 C^0 for 30 min to expel the binder and to prevent the cracks that led to damage the discs .

3- Grain size calculation : The grain size of samples were calculated depending on the XRD pattern by using Scherrer equation

$$D = \frac{K \lambda}{\beta \cos \theta} \quad (1)$$

where D is mean crystallite size, K is Scherrer's constant, λ is diffraction wavelength, β is FWHM

4- Dielectric constant vs temperature calculations:

To calculate the dielectric constant vs temperature, determination of Curie temperature, the equation (2-2) was used after measurement of capacity of samples by using RCL-Meter (PM6303) at frequency 20 Hz and electrical oven as a source of temperature.

$$\varepsilon_r = \frac{C d}{\varepsilon A} \quad (2)$$

In which C , d , A , ε represent the capacitance, thickness of samples, surface area of capacitors, space permittivity ($8.85 \times 10^{-12}\text{F/m}$) respectively

3. Result and Discussion

Figures (3,4,5,6,7,8,9) shows the XRD pattern of BaTiO_3 , $\text{Ba}_{0.85}\text{Pb}_{0.15}\text{TiO}_3$, $\text{Ba}_{0.85}\text{Ca}_{0.15}\text{TiO}_3$, $\text{Ba}_{0.85}\text{Co}_{0.15}\text{TiO}_3$, $\text{BaZr}_{0.15}\text{Ti}_{0.85}\text{O}_3$, $\text{BaSi}_{0.15}\text{Ti}_{0.85}\text{O}_3$, $\text{BaMn}_{0.15}\text{Ti}_{0.85}\text{O}_3$ respectively . Fig (3) shows the good growth of the BaTiO_3 phase with the appropriate intensity of the main peaks characteristic of this phase namely [100] , [101] , [111] , [002] , [200] , [201] , [211] , [202] , [220] , [301] , [310] at $2\theta = 22.317^\circ, 31.705^\circ, 39.06^\circ, 45.15^\circ, 45.51^\circ, 50.82^\circ, 56.356^\circ, 65.96^\circ, 66.242^\circ, 74.62^\circ, 75.23^\circ$. In addition to that , we don't find any impurities in this phase and that the calcination at 1200 C^0 to 2 hr is appropriate to prepare BaTiO_3 phase in BaO-TiO_2 by using solid state reaction method .

The figure(4,5) also show good growth of the $\text{Ba}_{0.85}\text{Pb}_{0.15}\text{TiO}_3$, $\text{Ba}_{0.85}\text{Co}_{0.15}\text{TiO}_3$ phases with no observed characteristic impurities except of the small intensity peak at ($2\theta = 29.17^\circ$) that belong to PbO compound in figure (2). In fig (6) we can observe two peaks with distinguish intensity belong to CaO compound , and This indicates the need to raise the calcination temperature to more than 1200 C^0 , and the evidence is that the phase growth has not been healed as required.

Figures (7,8,9) shows also good growth of $\text{BaZr}_{0.15}\text{Ti}_{0.85}\text{O}_3$, $\text{BaMn}_{0.15}\text{Ti}_{0.85}\text{O}_3$ $\text{BaSi}_{0.15}\text{Ti}_{0.85}\text{O}_3$, respectively with no observed characteristic impurities except in fig (5) at ($2\theta= 28.83^\circ$) belong to ZrO_2 (JCPDS, 42-1164) , fig (6) at ($2\theta= 26.35^\circ$) belong to MnO_2 · fig (7) at ($2\theta= 29.02^\circ$) belong to SiO_2 (JCPDS, 40-1498).

Tables 3,4 shows the crystal dimensions ($a,c,c/a$) and average crystallite size of prepared groups respectively . By looking at the tables 2, 3, we find that the ratio c/a increases with a decrease in the average ionic radius of the site A in the structure of perovskites ABO_3 except for the group (4) where it remains relatively constant . As for the B site, we note that there is no fixed relationship between the average ionic radius and the value of (a) , While we find that the (c/a)ratio is greatly affected by the average ionic radius, as it increases with its increase and decreases with its decrease

Figure (8) shows that the dielectric constant increases with increasing temperatures and at different rates from room temperature to temperatures close to Curie temperature, and this behavior is similar for all the prepared groups except for the group $\text{Ba}_{0.85}\text{Co}_{0.15}\text{TiO}_3$, where we clearly see that it is more stable in its dielectric constant within the thermal range ($60-90 \text{ C}^0$). We also note that upon reviewing the radii of the ions that were compensated at the two sites A, B in the composition of perovskites ABO_3 (table .2), we find that the decreasing in the average ionic radius leads to a clear and large decrease in the dielectric constant at Curie temperature, while increasing the average of the ionic radius at the site B leads to an increase

in the value of the dielectric constant at Curie temperature, as in the case when the relative substitution of Zr⁺⁴ ion instead of Ti⁺⁴ in the BaZr_{0.15}Ti_{0.85}O₃ group , Although it could not be generalized in absolute terms only after many appropriate experiments . The effect of the difference in the ionic radius for example between Zr⁺⁴ and Ti⁺⁴ ions in BaZr_{0.15}Ti_{0.85}O₃ group lead to octahedral oxygen distortion [],this can be generalized to the rest of the prepared groups and we believe that this may have the largest role in changing the values of the dielectric constant and Curie point.

4. Conclusions

In concluding remarks, the temperature of 120 °C for 2 hours is suitable for preparing BaTiO₃. The effect of the average ionic radius on the displacement rate of the main peaks of the prepared compound can be observed. The occurrence of a creep and a shift in the direction of the decrease in the 2θ value of the main peaks, especially when the value of the average ionic radius changes in the B-site where we find the large relative displacement of the (110) peak of the groups (BaZr_{0.15}Ti_{0.85}O₃, BaSi_{0.15}Ti_{0.85}O₃, BaMn_{0.15}Ti_{0.85}O₃). To develop a comprehensive general relationship linking the average ionic radius at the A,B sites in the composition of perovskites and between the crystal structure, its dimensions and the granular size, we must discuss and take into account other factors related to the physical and structural properties of the ion or the added oxide, the most important of which is the melting point. The average ionic radius at the A,B sites plays a large and clear role in determining the values of the dielectric constant and Curie temperature in perovskites.

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Table (1) The compounds that used in this work

No.	Compound	Origin	Purity (%)	Impurities (%)
1	BaCO ₃	Riedel-De Haen (Germany)	99	Ca (0.01) Fe (0.001) Cl (0.005) NO ₃ (0.005)
2	TiO ₂	BDH chemicals Ltd(poole England)	98	Fe (0.5) L.I (0.5)
3	ZrO ₂	BDH chemicals Ltd(poole England)	98	HCl(0.005) Fe (0.002)
4	PbO (analar)	BDH chemicals Ltd(poole England)	99	
5	CoO (black)	BDH chemicals Ltd(poole England)		
6	MnO ₂	Riedel-De Haen (Germany)	95	
7	CaCO ₃	Panreac Quimica SA (Bercelona – Espania)		
8	SiO ₂	Riedel-De Haen (Germany)	99.51	

Table (2) Prepared groups

No	Compositions	< r _{A-site} >	< r _{B-site} >
1	BaTiO ₃	1.61	0.605
2	Ba _{0.85} Pb _{0.15} TiO ₃	1.592	
3	Ba _{0.85} Ca _{0.15} TiO ₃	1.569	
4	Ba _{0.85} Co _{0.15} TiO ₃	1.480	
5	BaZr _{0.15} Ti _{0.85} O ₃		0.622
6	BaSi _{0.15} Ti _{0.85} O ₃		0.574
7	BaMn _{0.15} Ti _{0.85} O ₃		0.593

Table (3) The crystal dimensions of groups

No	Material	a (Å)	c (Å)	c/a
1	BaTiO ₃	3.982	4.012	1.0075
2	Ba _{0.85} Pb _{0.15} TiO ₃	3.980	4.030	1.0125
3	Ba _{0.85} Ca _{0.15} TiO ₃	3.971	4.023	1.0131
4	Ba _{0.85} Co _{0.15} TiO ₃	3.980	4.009	1.0073
5	BaZr _{0.15} Ti _{0.85} O ₃	3.996	4.028	1.0080
6	BaSi _{0.15} Ti _{0.85} O ₃	3.996	4.006	1.0025
7	BaMn _{0.15} Ti _{0.85} O ₃	3.995	4.013	1.0045

Table (4) Crystallite size of prepared groups

No.	Material	Crystallite size (nm)	Average crystallite size (nm)
1	BaTiO_3	252-335	293.5
2	$\text{Ba}_{0.85}\text{Pb}_{0.15}\text{TiO}_3$	388- 407	397.5
3	$\text{Ba}_{0.85}\text{Ca}_{0.15}\text{TiO}_3$	234.91- 266.5	250.70
4	$\text{Ba}_{0.85}\text{Co}_{0.15}\text{TiO}_3$	330 – 396	363
5	$\text{BaZr}_{0.15}\text{Ti}_{0.85}\text{O}_3$	356 – 374	365
6	$\text{BaSi}_{0.15}\text{Ti}_{0.85}\text{O}_3$	330 – 388.04	359.02
7	$\text{BaMn}_{0.15}\text{Ti}_{0.85}\text{O}_3$	213.23- 374.6	293.915

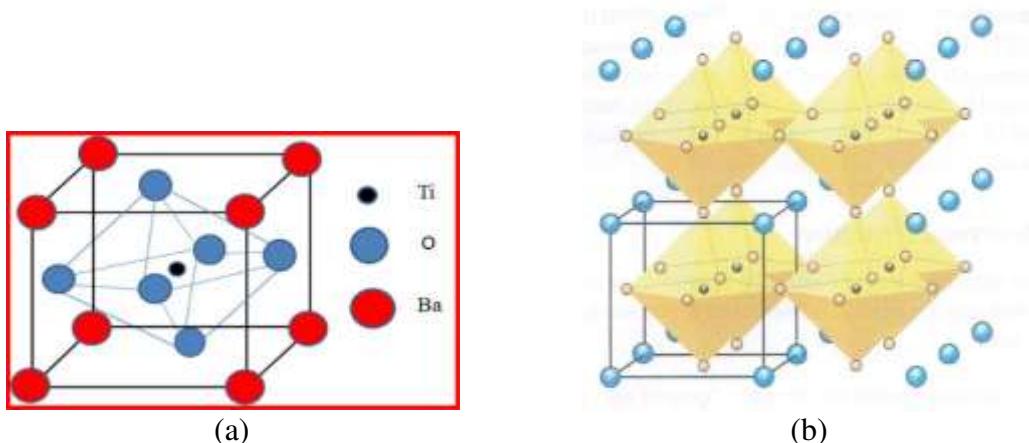


Fig. (1) (a) Structure of perovskite ABO_3 unit cell structure of BaTiO_3 , (b) Perovskite structure as connected network of octahedra

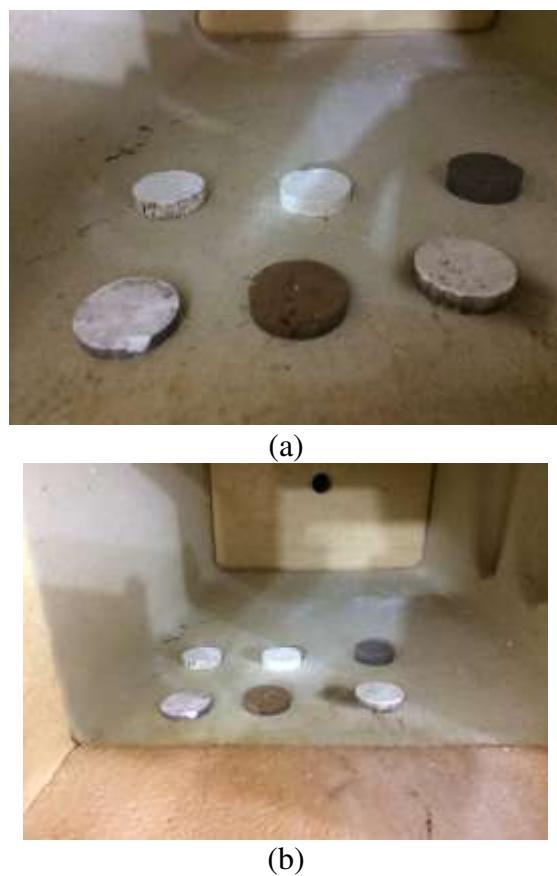


Fig. (2) Discs of prepared samples in (3-5 mm) thickness and in 20 mm diameter (in muffle furnace)

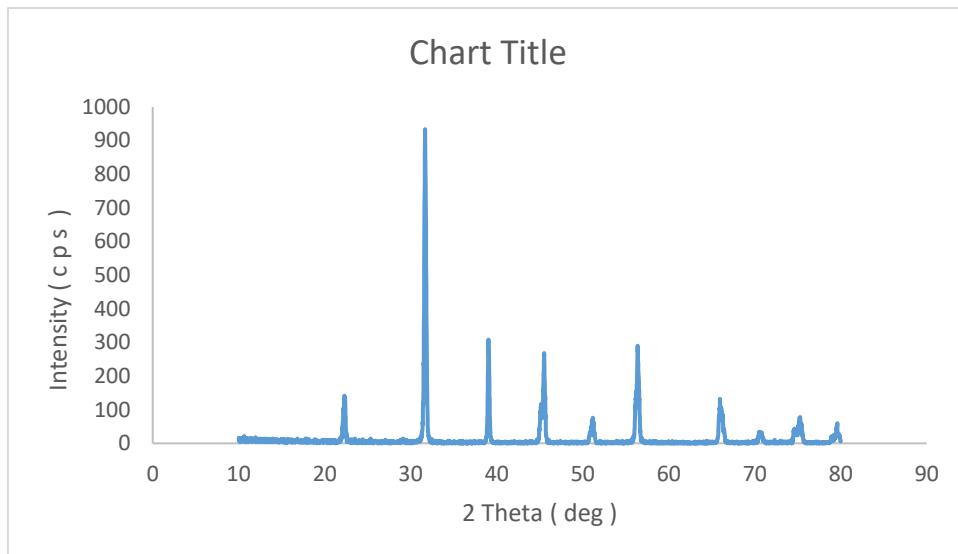


Fig (3) XRD pattern of BaTiO_3 ceramic calcined at 1200 C^0 for 2 hr

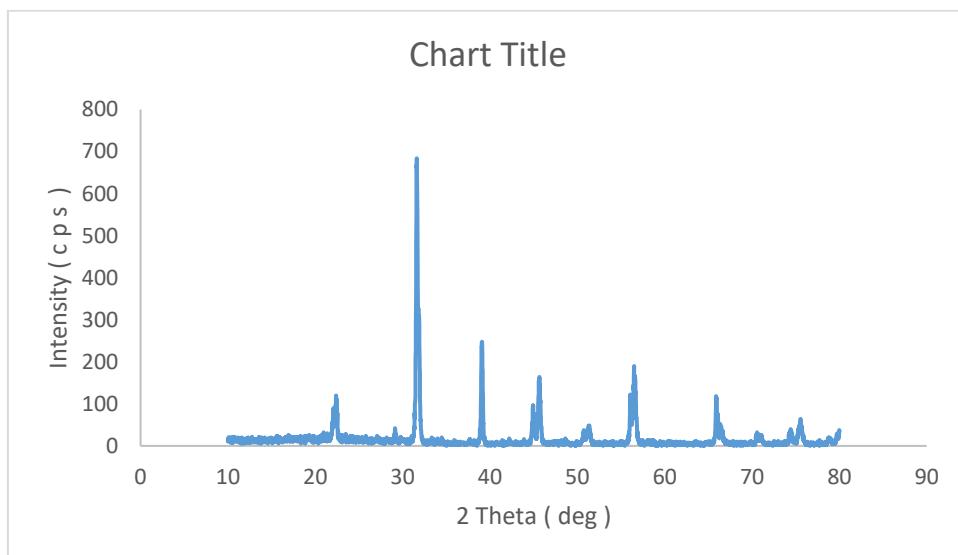


Fig. (4) XRD pattern of $\text{Ba}_{0.85}\text{Pb}_{0.15}\text{TiO}_3$ ceramic calcined at 1200 C^0 for 2 hr

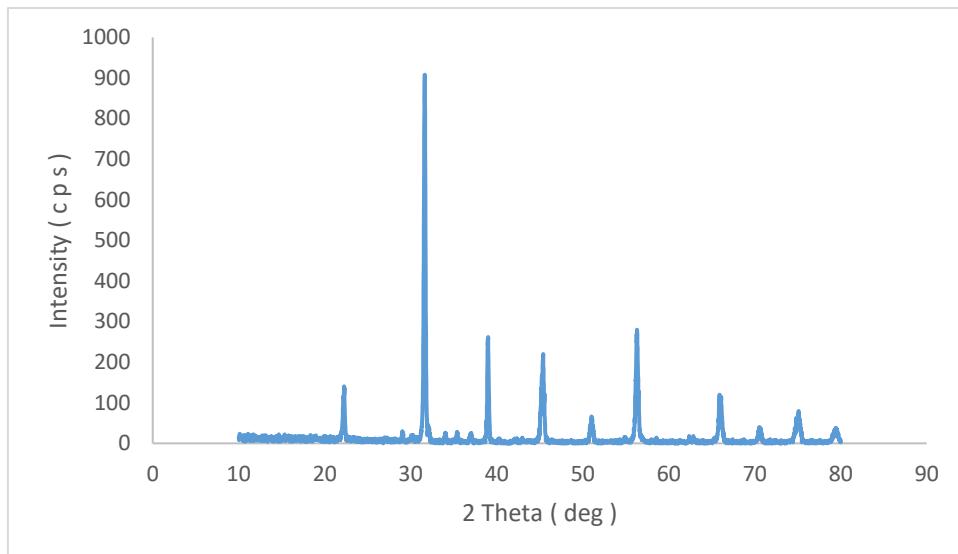


Fig. (5) XRD pattern of Ba_{0.85}Co_{0.15}TiO₃ ceramic calcined at 1200 C° for 2 hr

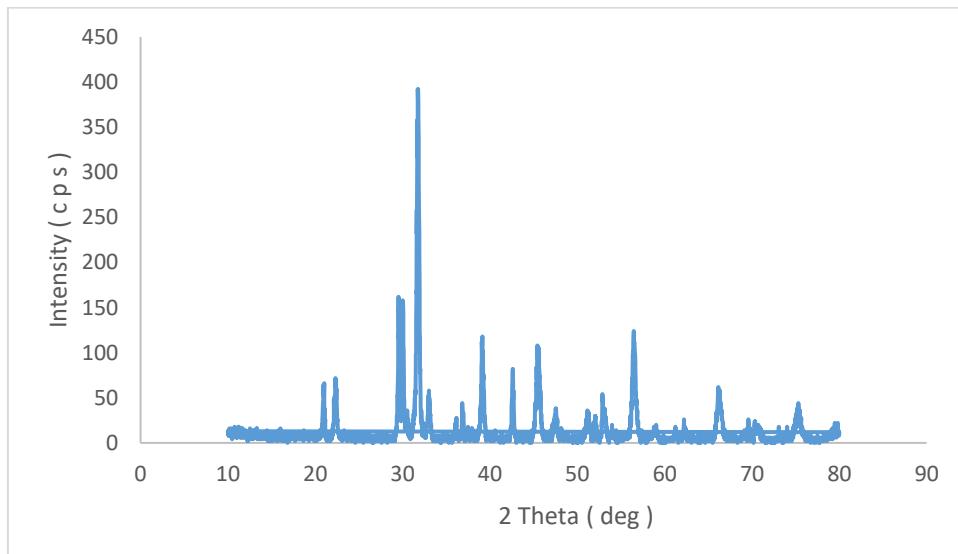


Fig. (6) XRD pattern of Ba_{0.85}Ca_{0.15}TiO₃ ceramic calcined at 1200 C° for 2 hr

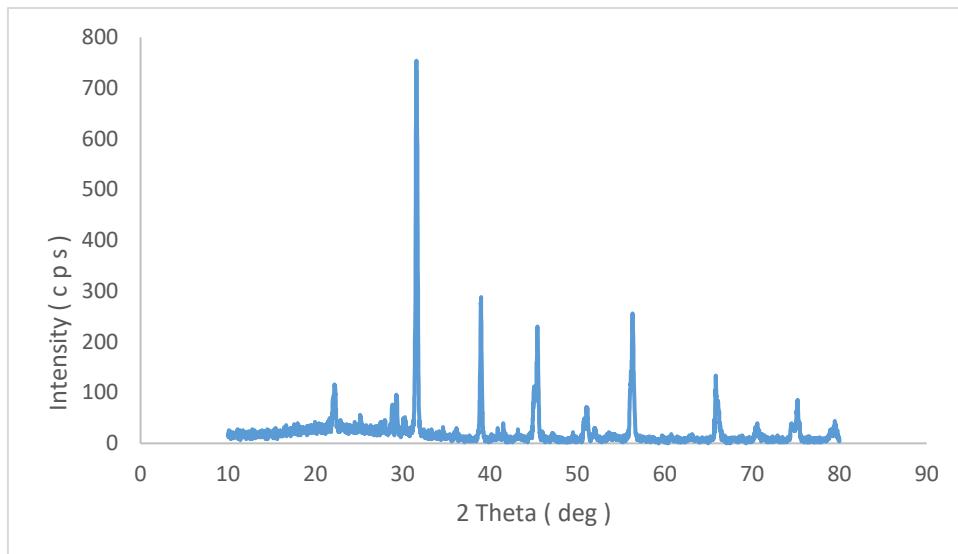


Fig. (7) XRD pattern of Ba Zr_{0.15}Ti_{0.85}O₃ ceramic calcined at 1200 C° for 2 hr

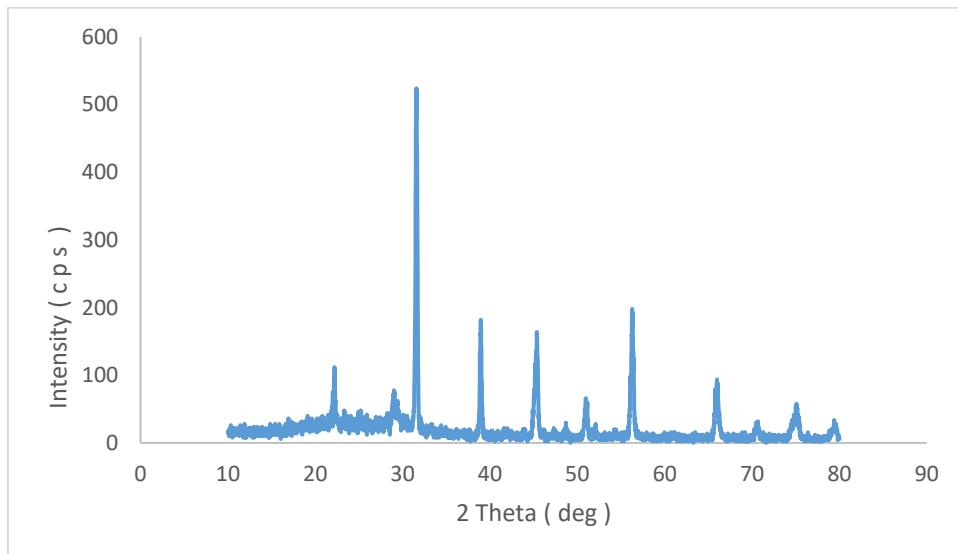


Fig. (8) XRD pattern of Ba Si_{0.15}Ti_{0.85}O₃ ceramic calcined at 1200 C° for 2 hr

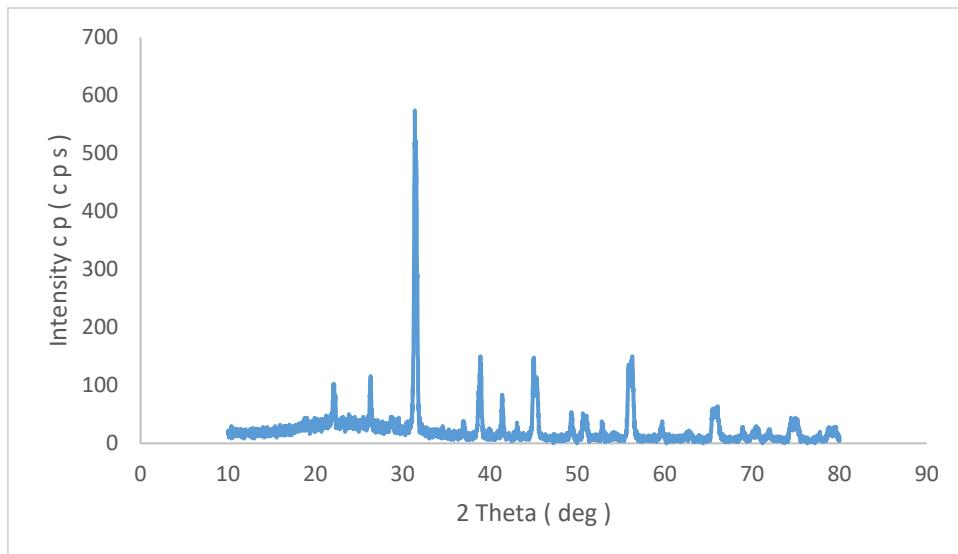


Fig. (9) XRD pattern of $\text{Ba Mn}_{0.15}\text{Ti}_{0.85}\text{O}_3$ ceramic calcined at 1200 C° for 2 hr

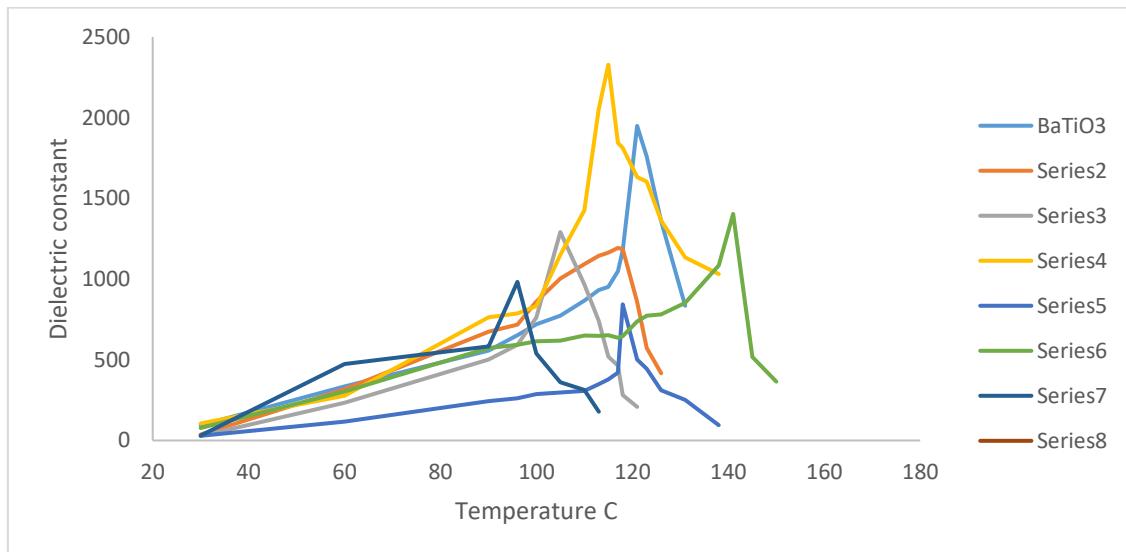


Fig. (10) Dielectric constant VS temperature at 20 Hz of prepared groups

Series 2: $\text{Ba Mn}_{0.15}\text{Ti}_{0.85}\text{O}_3$.., series 3: $\text{Ba Si}_{0.15}\text{Ti}_{0.85}\text{O}_3$..,
 Series 4 : $\text{Ba Zr}_{0.15}\text{Ti}_{0.85}\text{O}_3$.., series 5 : $\text{Ba}_{0.85}\text{Ca}_{0.15}\text{TiO}_3$
 Series 6 : $\text{Ba}_{0.85}\text{Pb}_{0.15}\text{TiO}_3$.., series 7: $\text{Ba}_{0.85}\text{Co}_{0.15}\text{TiO}_3$

Figures

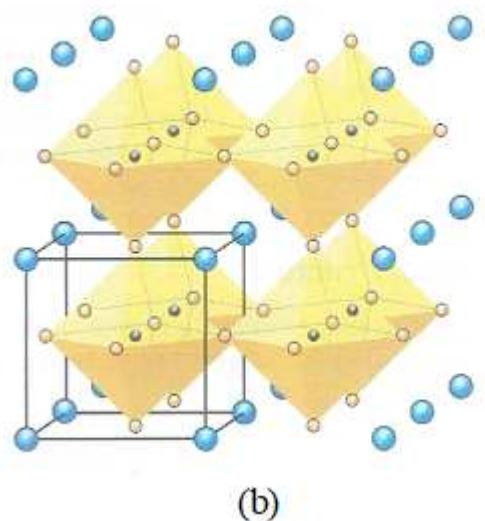
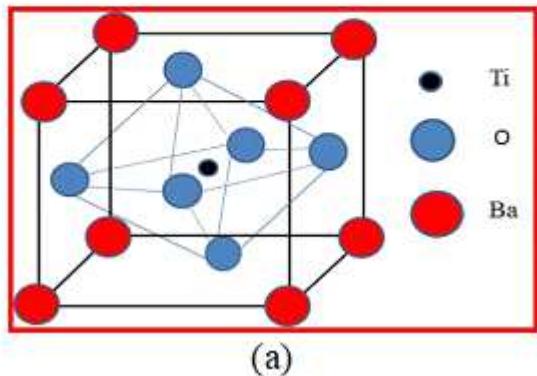


Figure 1

(a) Structure of perovskite ABO_3 unit cell structure of BaTiO_3 , (b) Perovskite structure as connected network of octahedra

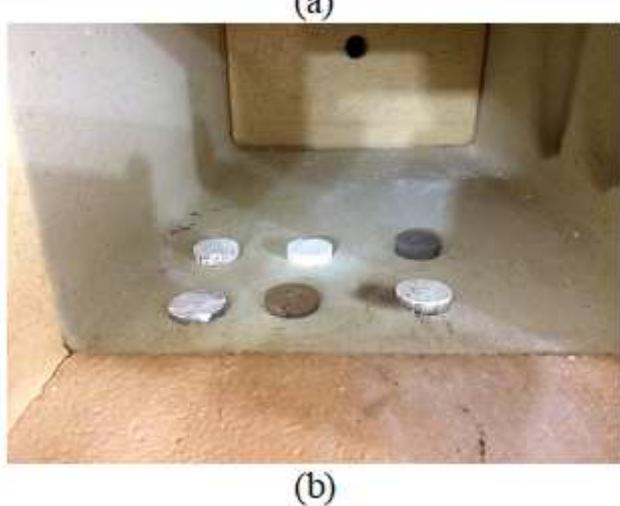


Figure 2

Discs of prepared samples in (3-5 mm) thickness and in 20 mm diameter (in muffle furnace)

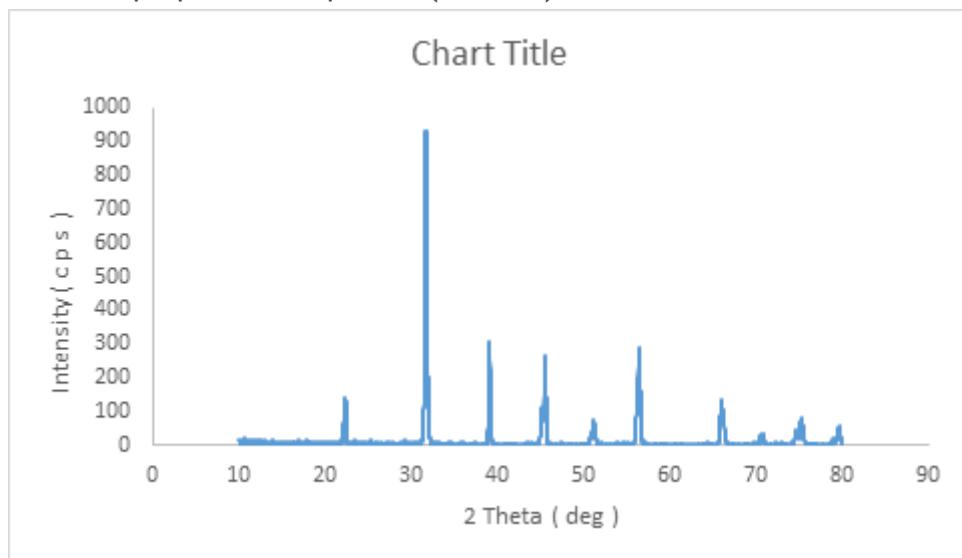


Figure 3

XRD pattern of BaTiO₃ ceramic calcined at 1200 C0 for 2 hr

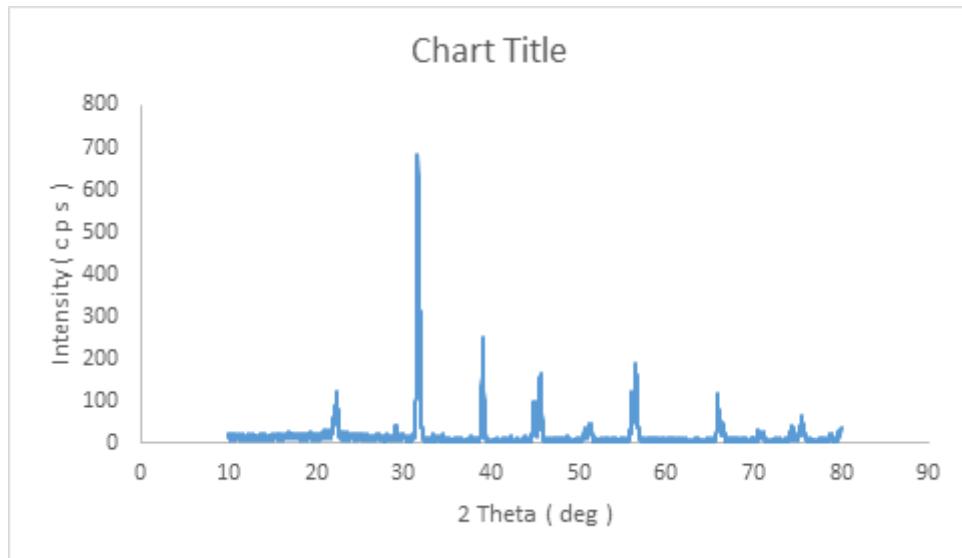


Figure 4

XRD pattern of Ba 0.85 Pb 0.15 TiO₃ ceramic calcined at 1200 C0 for 2 hr

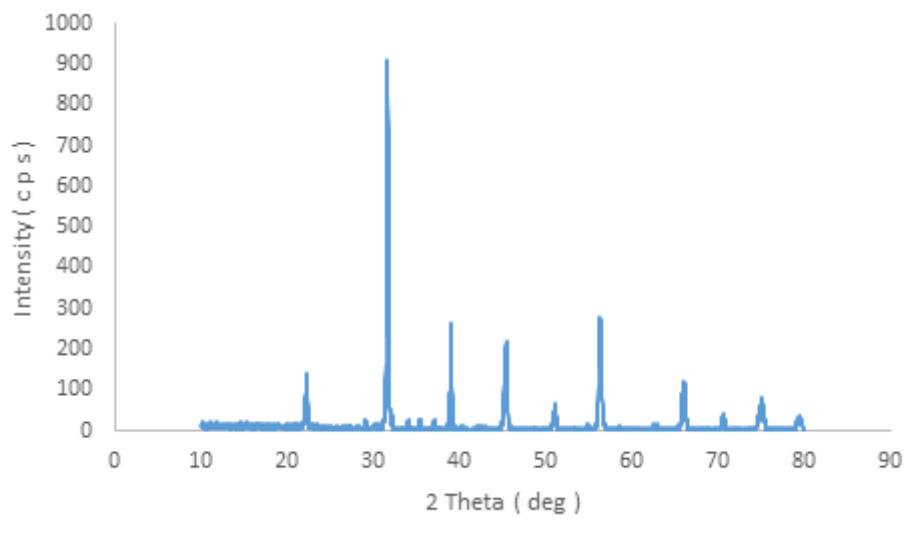


Figure 5

XRD pattern of Ba 0.85 Co 0.15 TiO₃ ceramic calcined at 1200 C0 for 2 hr

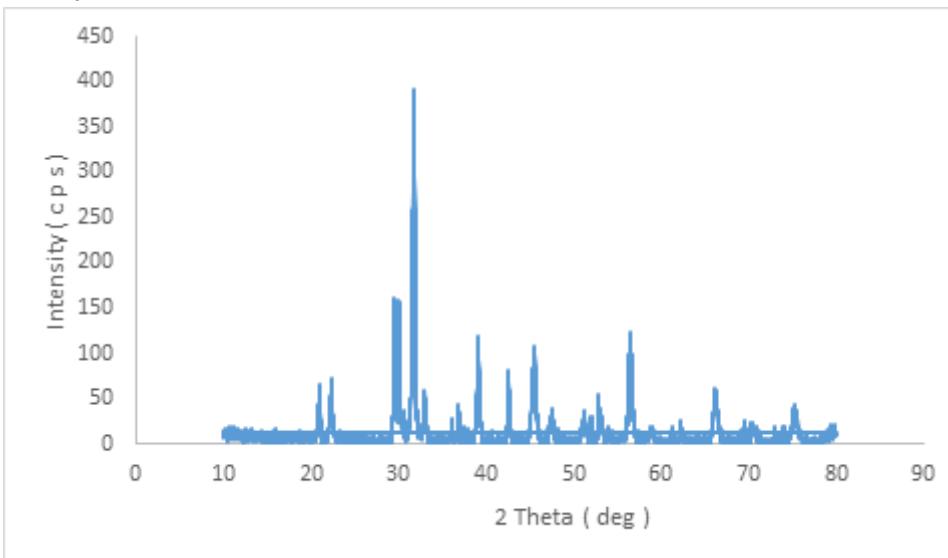


Figure 6

XRD pattern of Ba 0.85 Ca 0.15 TiO₃ ceramic calcined at 1200 C0 for 2 hr

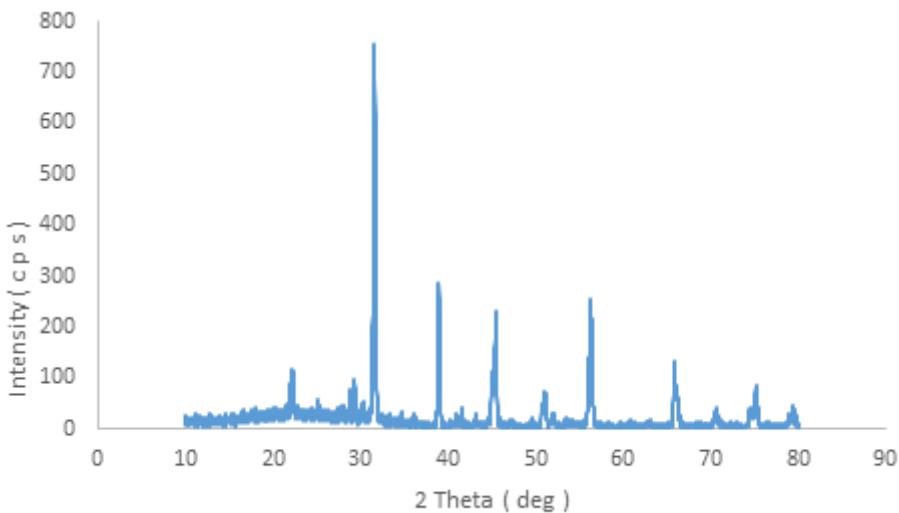


Figure 7

XRD pattern of Ba Zr 0.15 Ti 0.85 O₃ ceramic calcined at 1200 C0 for 2 hr

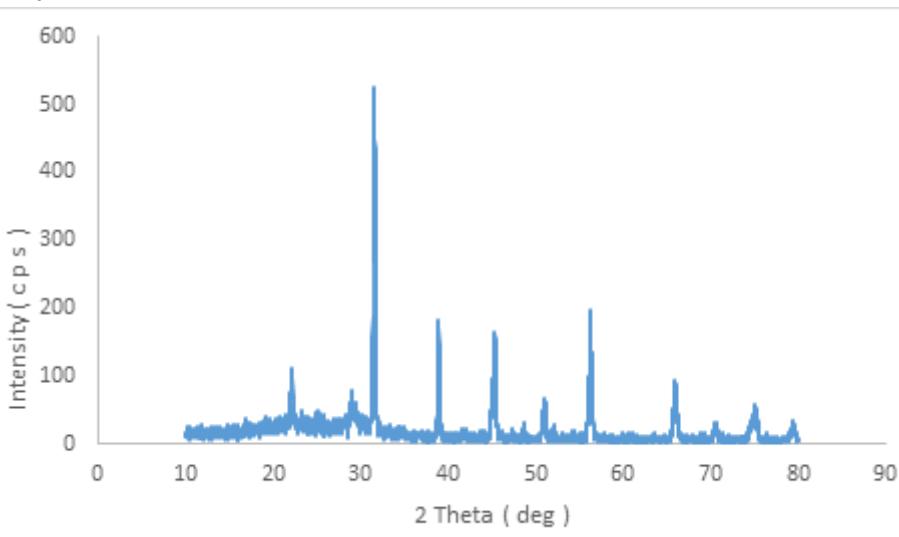


Figure 8

XRD pattern of Ba Si 0.15 Ti 0.85 O₃ ceramic calcined at 1200 C0 for 2 hr

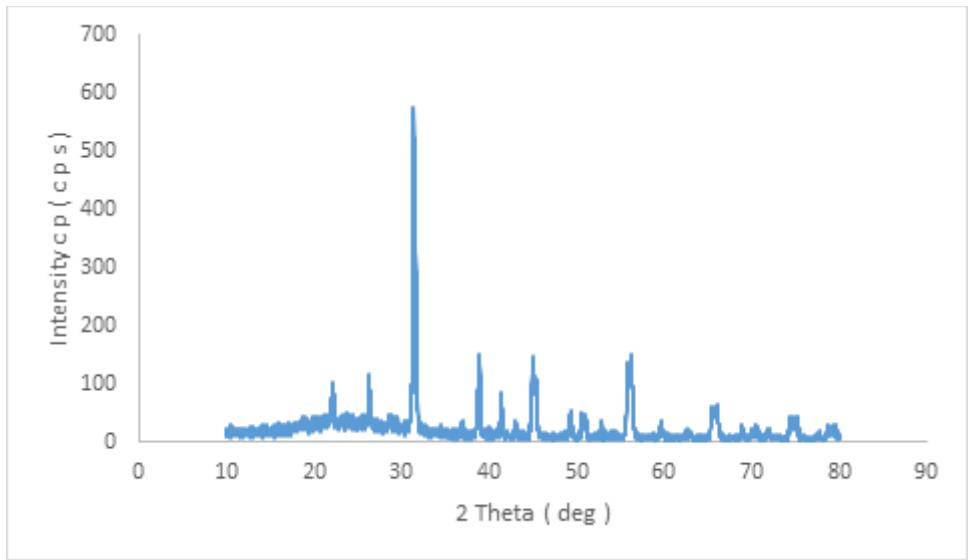


Figure 9

XRD pattern of Ba Mn 0.15 Ti 0.85 O₃ ceramic calcined at 1200 C0 for 2 hr

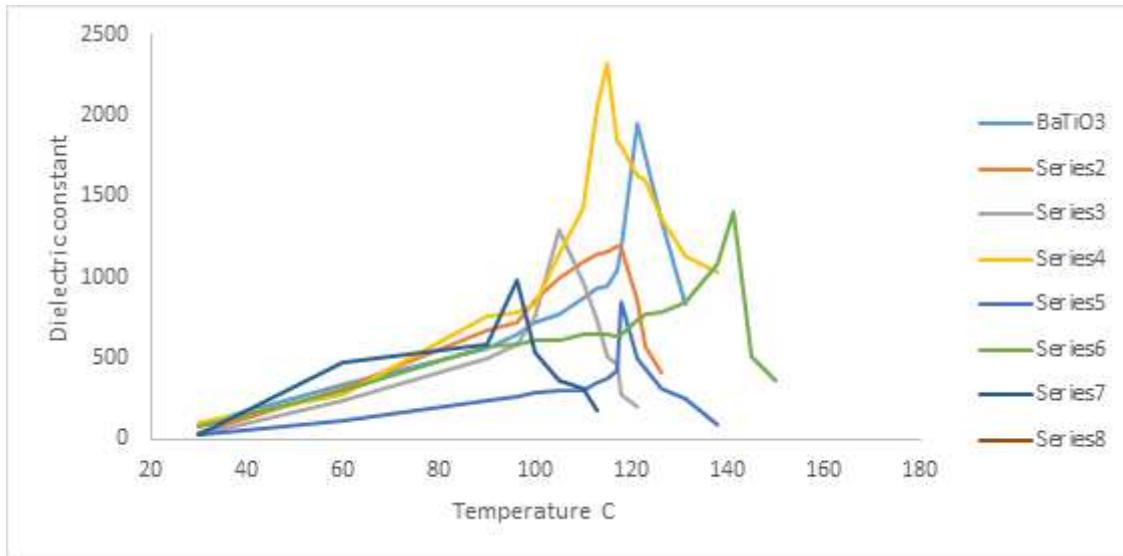


Figure 10

Dielectric constant VS temperature at 20 Hz of prepared groups Series 2: Ba Mn 0.15 Ti 0.85 O₃ , series 3: Ba Si 0.15 Ti 0.85 O₃ , Series 4 : Ba Zr 0.15 Ti 0.85 O₃ , series 5 : Ba 0.85 Ca 0.15 TiO₃ Series 6 : Ba 0.85 Pb 0.15 TiO₃ , series 7: Ba 0.85 Co 0.15 TiO₃