



# Solution Combustion Approach For The Synthesis Of $\text{BiFe}_{1-x}\text{Pb}_x\text{O}_3$ ( $x = 0, 0.05, 0.15, 0.25, 0.35$ ) Multiferroic Ceramic Samples Using Nitrates And Urea Fuel As Primary Precursors: Studies Related With Dielectric Characteristics

Yogesh A. Chaudhari<sup>+</sup>

Assistant Professor and Head, Department of Physics,

Shri Pancham Khemraj Mahavidyalaya (Autonomous), Sawantwadi, Sindhudurg, Maharashtra, India

## ABSTRACT:

This paper presents the synthesis of samples such as,  $\text{BiFeO}_3$ ,  $\text{BiFe}_{0.95}\text{Pb}_{0.05}\text{O}_3$ ,  $\text{BiFe}_{0.85}\text{Pb}_{0.15}\text{O}_3$ ,  $\text{BiFe}_{0.75}\text{Pb}_{0.25}\text{O}_3$  and  $\text{BiFe}_{0.65}\text{Pb}_{0.35}\text{O}_3$  multiferroic ceramics via solution combustion method (SCM). These  $\text{BiFeO}_3$ ,  $\text{BiFe}_{0.95}\text{Pb}_{0.05}\text{O}_3$ ,  $\text{BiFe}_{0.85}\text{Pb}_{0.15}\text{O}_3$ ,  $\text{BiFe}_{0.75}\text{Pb}_{0.25}\text{O}_3$  and  $\text{BiFe}_{0.65}\text{Pb}_{0.35}\text{O}_3$  ceramic materials were formulated by using metal nitrates and urea as an initial starting precursors. The temperature dependence of dielectric constant shows a dielectric anomalies at various temperatures in  $\text{BiFe}_{0.95}\text{Pb}_{0.05}\text{O}_3$ ,  $\text{BiFe}_{0.85}\text{Pb}_{0.15}\text{O}_3$ ,  $\text{BiFe}_{0.75}\text{Pb}_{0.25}\text{O}_3$  and  $\text{BiFe}_{0.65}\text{Pb}_{0.35}\text{O}_3$  ceramics at 1 kHz, 3 kHz and 5 kHz frequencies. Each of these powdered samples were ground in an acetone medium, then calcined and sintered at elevated temperatures before being pelletized.

**Keywords:** Multiferroics, Pure  $\text{BiFeO}_3$ , Pb doped  $\text{BiFeO}_3$ , SCM, Dielectric, applications.

## I. INTRODUCTION:

Electrical and magnetic order occurring simultaneously in multiferroic materials [1].  $\text{BiFeO}_3$  has an antiferromagnetic Neel temperature ( $T_N$ ) of 640 K and a ferroelectric Curie temperature ( $T_C$ ) of 1100 K [2]. The multiferroic  $\text{BiFeO}_3$  ceramics have number of potential applications in different sectors such as magnetocapacitive transducers [3], ferroelectric memory storage [4], microelectronic devices [5], photovoltaic devices [6], spin field effect transistors, nanoelectronics [7].

The pure and doped  $\text{BiFeO}_3$  multiferroic ceramics have been synthesized using different formulation routes such as solid state reaction [8], sol- gel method [9], Pechini method [10], Auto-combustion Technique [11], combustion method [12] and hydrothermal method [13].

## II. EXPERIMENTAL PROCEDURE:

The preparation of pure  $\text{BiFeO}_3$  and Pb-doped samples such as  $\text{BiFe}_{0.95}\text{Pb}_{0.05}\text{O}_3$ ,  $\text{BiFe}_{0.85}\text{Pb}_{0.15}\text{O}_3$ ,  $\text{BiFe}_{0.75}\text{Pb}_{0.25}\text{O}_3$  and  $\text{BiFe}_{0.65}\text{Pb}_{0.35}\text{O}_3$  ceramics were formulated using solution combustion method (SCM).

## MATERIALS:

The principal components are urea, ferric nitrate, lead nitrate, and bismuth nitrate.

## SYNTHESIS PROCESS:

The preparation of  $\text{BiFeO}_3$ ,  $\text{BiFe}_{0.95}\text{Pb}_{0.05}\text{O}_3$ ,  $\text{BiFe}_{0.85}\text{Pb}_{0.15}\text{O}_3$ ,  $\text{BiFe}_{0.75}\text{Pb}_{0.25}\text{O}_3$  and  $\text{BiFe}_{0.65}\text{Pb}_{0.35}\text{O}_3$  ceramic samples were carried out using the precursors such as bismuth nitrate, lead nitrate, ferric nitrate as oxidizers while urea was used as a fuel. The oxidizer (O) to fuel (F) ratio was precisely taken into account when producing the sample combination using the oxidizing and reducing valences of the metal nitrates and fuel [14]. Distilled water was used to dissolve stoichiometric amounts of bismuth nitrate, lead nitrate, ferric nitrate, and urea in various beakers. These solutions were then mixed and boiled on a gas burner in a Pyrex dish. Subsequently the constant heating, the water gets evaporated and lastly a combustion takes place with formation of  $\text{BiFeO}_3$ ,  $\text{BiFe}_{0.95}\text{Pb}_{0.05}\text{O}_3$ ,  $\text{BiFe}_{0.85}\text{Pb}_{0.15}\text{O}_3$ ,  $\text{BiFe}_{0.75}\text{Pb}_{0.25}\text{O}_3$  and  $\text{BiFe}_{0.65}\text{Pb}_{0.35}\text{O}_3$  ceramic samples. The experimental procedure was provided by Chaudhari et.al. [15], these powders were grinded in an acetone medium, finally calcined and sintered at  $425^\circ\text{C}$ ,  $450^\circ\text{C}$ ,  $475^\circ\text{C}$ ,  $500^\circ\text{C}$ ,  $525^\circ\text{C}$  for 3 hours in a furnace and lastly carried out for pelletization. The process of producing  $\text{BiFeO}_3$  ceramic samples is shown in Fig.1. Fig.2 presents the experimental procedure used for developing  $\text{BiFeO}_3$  ceramics. Figure 3 displays the  $\text{BiFeO}_3$  samples in powdered form, and Figure 4 displays the  $\text{BiFeO}_3$  pellet that was formed. The  $\text{BiFe}_{0.95}\text{Pb}_{0.05}\text{O}_3$ ,  $\text{BiFe}_{0.85}\text{Pb}_{0.15}\text{O}_3$ ,  $\text{BiFe}_{0.75}\text{Pb}_{0.25}\text{O}_3$ , and  $\text{BiFe}_{0.65}\text{Pb}_{0.35}\text{O}_3$  ceramic samples are synthesized using the flowchart in Fig.5, and the experimental approach to produce doped  $\text{BiFe}_{0.95}\text{Pb}_{0.05}\text{O}_3$ ,  $\text{BiFe}_{0.85}\text{Pb}_{0.15}\text{O}_3$ ,  $\text{BiFe}_{0.75}\text{Pb}_{0.25}\text{O}_3$ , and  $\text{BiFe}_{0.65}\text{Pb}_{0.35}\text{O}_3$  ceramic samples is shown in Fig. 6. Fig.7 (a), (b), (c), (d) shows the synthesized powder samples of the  $\text{BiFe}_{0.95}\text{Pb}_{0.05}\text{O}_3$ ,  $\text{BiFe}_{0.85}\text{Pb}_{0.15}\text{O}_3$ ,  $\text{BiFe}_{0.75}\text{Pb}_{0.25}\text{O}_3$  and  $\text{BiFe}_{0.65}\text{Pb}_{0.35}\text{O}_3$  ceramic samples and the developed pellets of the doped  $\text{BiFe}_{0.95}\text{Pb}_{0.05}\text{O}_3$ ,  $\text{BiFe}_{0.85}\text{Pb}_{0.15}\text{O}_3$ ,  $\text{BiFe}_{0.75}\text{Pb}_{0.25}\text{O}_3$  and  $\text{BiFe}_{0.65}\text{Pb}_{0.35}\text{O}_3$  ceramic samples are displayed in Fig. 8 (a), (b), (c), (d). Fig. 9 (a), (b), (c), (d) and (e) presents the temperature dependence of dielectric constant for the  $\text{BiFe}_{0.95}\text{Pb}_{0.05}\text{O}_3$ ,  $\text{BiFe}_{0.85}\text{Pb}_{0.15}\text{O}_3$ ,  $\text{BiFe}_{0.75}\text{Pb}_{0.25}\text{O}_3$  and  $\text{BiFe}_{0.65}\text{Pb}_{0.35}\text{O}_3$  samples at 1kHz, 3kHz and 5kHz frequencies. The dielectric studies exhibits a dielectric anomalies in at different temperatures in  $\text{BiFe}_{0.95}\text{Pb}_{0.05}\text{O}_3$ ,  $\text{BiFe}_{0.85}\text{Pb}_{0.15}\text{O}_3$ ,  $\text{BiFe}_{0.75}\text{Pb}_{0.25}\text{O}_3$  and  $\text{BiFe}_{0.65}\text{Pb}_{0.35}\text{O}_3$  ceramics at different frequencies like 1 kHz, 3 kHz and 5 kHz are given in the table 1.

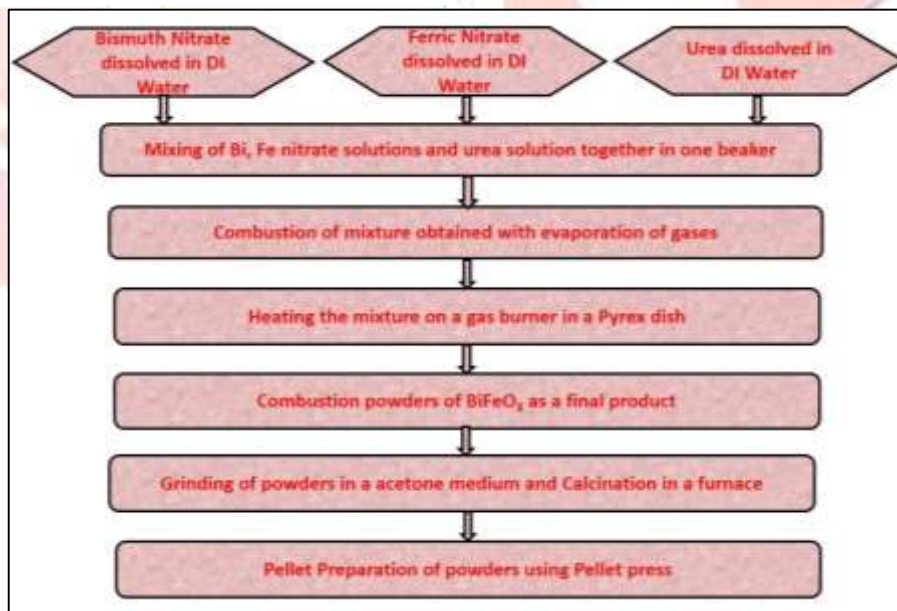


Fig.1. Flowchart of synthesis of  $\text{BiFeO}_3$  samples by SCM.

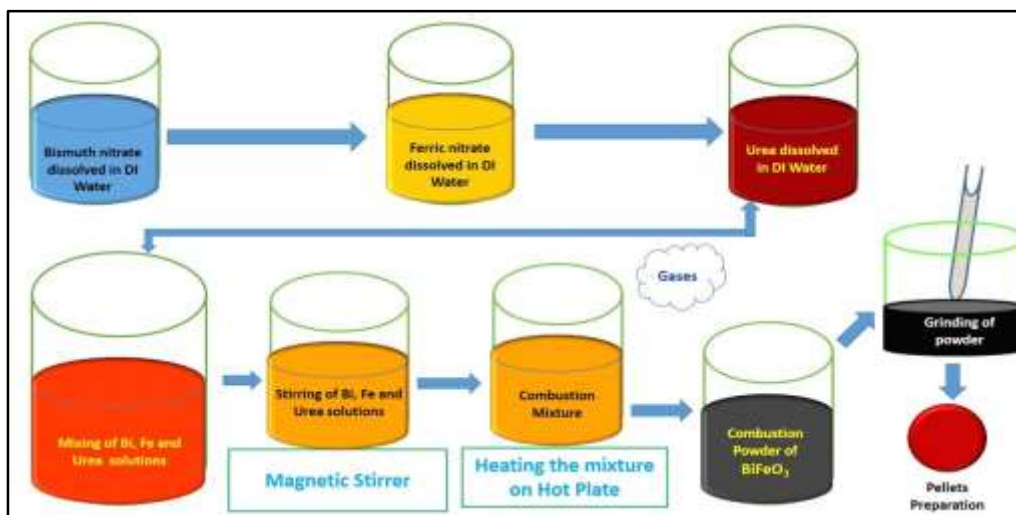


Fig. 2. Experimental procedure of synthesis of BiFeO<sub>3</sub> nanoceramic samples by SCM.

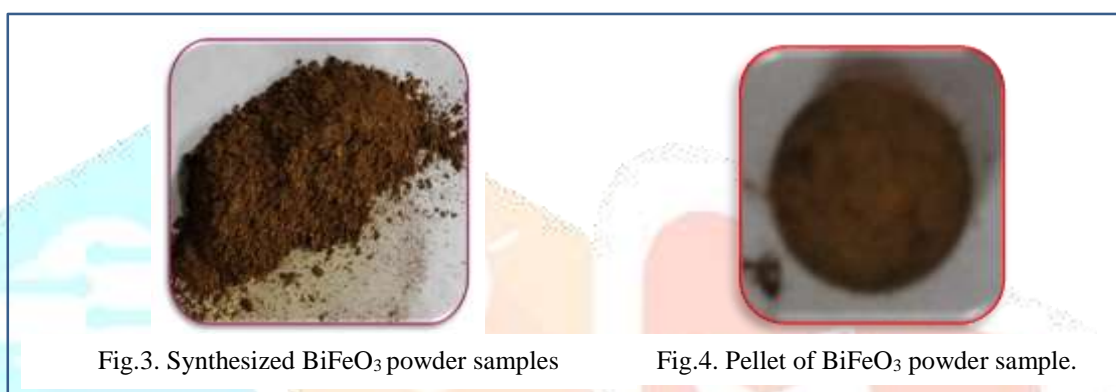


Fig.3. Synthesized BiFeO<sub>3</sub> powder samples

Fig.4. Pellet of BiFeO<sub>3</sub> powder sample.

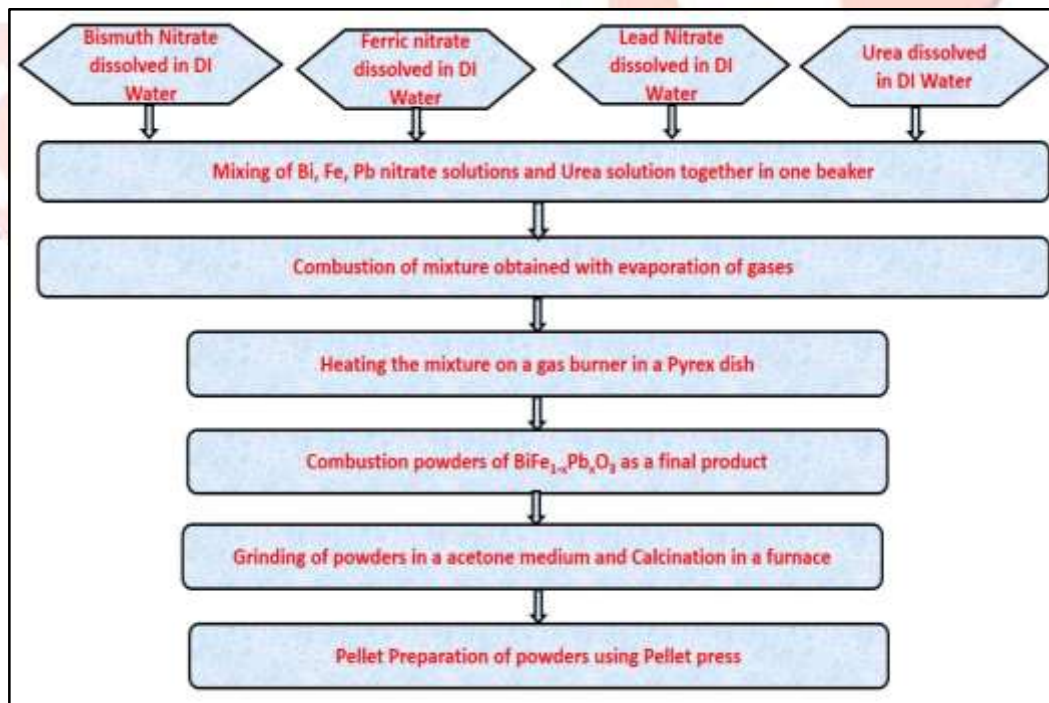


Fig.5. Flowchart of synthesis of BiFe<sub>0.95</sub>Pb<sub>0.05</sub>O<sub>3</sub>, BiFe<sub>0.85</sub>Pb<sub>0.15</sub>O<sub>3</sub>, BiFe<sub>0.75</sub>Pb<sub>0.25</sub>O<sub>3</sub> and BiFe<sub>0.65</sub>Pb<sub>0.35</sub>O<sub>3</sub> ceramic samples by SCM.



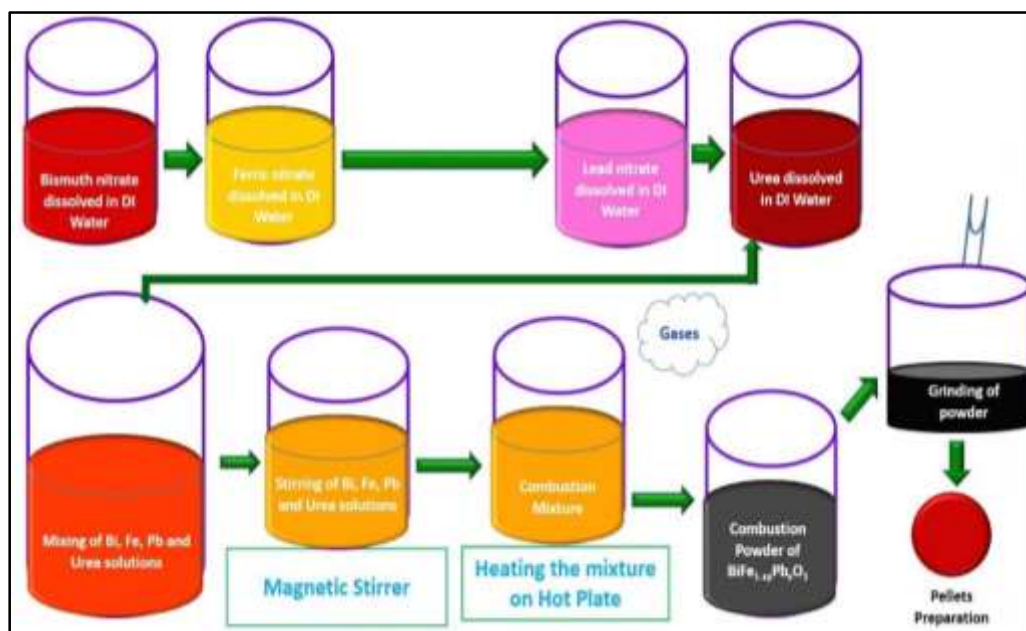


Fig.6. Experimental process of synthesis of BiFe<sub>0.95</sub>Pb<sub>0.05</sub>O<sub>3</sub>, BiFe<sub>0.85</sub>Pb<sub>0.15</sub>O<sub>3</sub>, BiFe<sub>0.75</sub>Pb<sub>0.25</sub>O<sub>3</sub> and BiFe<sub>0.65</sub>Pb<sub>0.35</sub>O<sub>3</sub> ceramic samples by SCM.

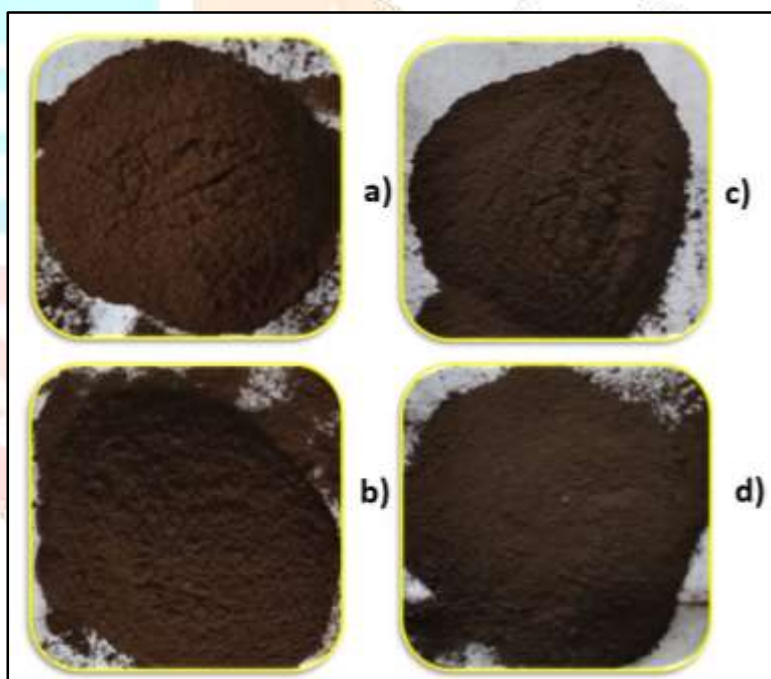


Fig.7. Synthesized powder samples of a) BiFe<sub>0.95</sub>Pb<sub>0.05</sub>O<sub>3</sub>, b) BiFe<sub>0.85</sub>Pb<sub>0.15</sub>O<sub>3</sub>, c) BiFe<sub>0.75</sub>Pb<sub>0.25</sub>O<sub>3</sub> and d) BiFe<sub>0.65</sub>Pb<sub>0.35</sub>O<sub>3</sub> ceramic samples.

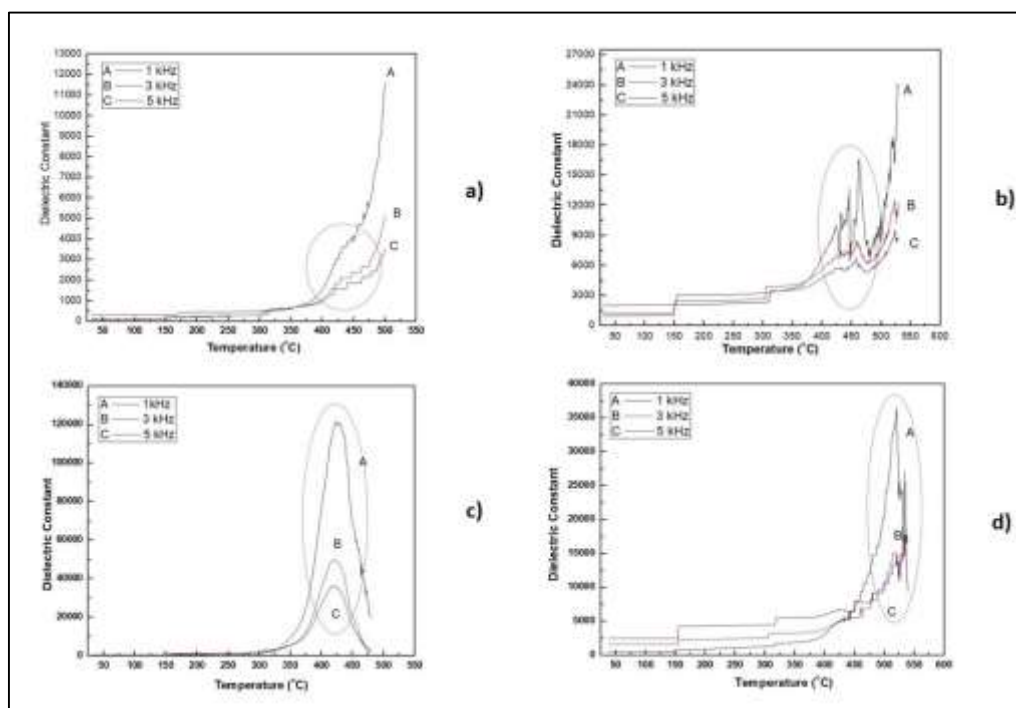


Fig. 9 shows the dielectric constant as a function of temperature a)  $\text{BiFe}_{0.95}\text{Pb}_{0.05}\text{O}_3$ , b)  $\text{BiFe}_{0.85}\text{Pb}_{0.15}\text{O}_3$ , c)  $\text{BiFe}_{0.75}\text{Pb}_{0.25}\text{O}_3$  and d)  $\text{BiFe}_{0.65}\text{Pb}_{0.35}\text{O}_3$  ceramic samples.

Sr. No.	Samples	1 kHz	3 kHz	5 kHz
1.	$\text{BiFe}_{0.95}\text{Pb}_{0.05}\text{O}_3$	450 °C	425 °C	415 °C
2.	$\text{BiFe}_{0.85}\text{Pb}_{0.15}\text{O}_3$	440 °C	450 °C	465 °C
3.	$\text{BiFe}_{0.75}\text{Pb}_{0.25}\text{O}_3$	430 °C	425 °C	410 °C
4.	$\text{BiFe}_{0.65}\text{Pb}_{0.35}\text{O}_3$	520 °C	530 °C	540 °C

Table 1. Temperature dependence of dielectric constant of a)  $\text{BiFe}_{0.95}\text{Pb}_{0.05}\text{O}_3$ , b)  $\text{BiFe}_{0.85}\text{Pb}_{0.15}\text{O}_3$ , c)  $\text{BiFe}_{0.75}\text{Pb}_{0.25}\text{O}_3$  and d)  $\text{BiFe}_{0.65}\text{Pb}_{0.35}\text{O}_3$  samples.

## RESULTS AND DISCUSSION:

Fig. 1 displays the flowchart used to create the  $\text{BiFeO}_3$  ceramic sample. The experimental procedure to make  $\text{BiFeO}_3$  ceramics is described in Fig.2. The  $\text{BiFeO}_3$  sample's synthesized powder is shown in Fig.3, and the produced  $\text{BiFeO}_3$  pellet is shown in Fig.4. In Fig.5, the synthesis mechanism for the ceramic samples  $\text{BiFe}_{0.95}\text{Pb}_{0.05}\text{O}_3$ ,  $\text{BiFe}_{0.85}\text{Pb}_{0.15}\text{O}_3$ ,  $\text{BiFe}_{0.75}\text{Pb}_{0.25}\text{O}_3$ , and  $\text{BiFe}_{0.65}\text{Pb}_{0.35}\text{O}_3$  is displayed. Fig. 6 shows the experimental procedure for creating doped  $\text{BiFe}_{0.95}\text{Pb}_{0.05}\text{O}_3$ ,  $\text{BiFe}_{0.85}\text{Pb}_{0.15}\text{O}_3$ ,  $\text{BiFe}_{0.75}\text{Pb}_{0.25}\text{O}_3$  and  $\text{BiFe}_{0.65}\text{Pb}_{0.35}\text{O}_3$  ceramic samples. The Fig.7 (a), (b), (c), (d) shows the synthesized powder samples of the  $\text{BiFe}_{0.95}\text{Pb}_{0.05}\text{O}_3$ ,  $\text{BiFe}_{0.85}\text{Pb}_{0.15}\text{O}_3$ ,  $\text{BiFe}_{0.75}\text{Pb}_{0.25}\text{O}_3$  and  $\text{BiFe}_{0.65}\text{Pb}_{0.35}\text{O}_3$  ceramic samples and Fig. 8 (a), (b), (c), and (d) display the fabricated pellets of the ceramic samples doped with  $\text{BiFe}_{0.95}\text{Pb}_{0.05}\text{O}_3$ ,  $\text{BiFe}_{0.85}\text{Pb}_{0.15}\text{O}_3$ ,  $\text{BiFe}_{0.75}\text{Pb}_{0.25}\text{O}_3$  and  $\text{BiFe}_{0.65}\text{Pb}_{0.35}\text{O}_3$ . Fig. 9 (a), (b), (c), (d) and (e) presents the temperature dependence of dielectric constant for the  $\text{BiFe}_{0.95}\text{Pb}_{0.05}\text{O}_3$ ,  $\text{BiFe}_{0.85}\text{Pb}_{0.15}\text{O}_3$ ,  $\text{BiFe}_{0.75}\text{Pb}_{0.25}\text{O}_3$  and  $\text{BiFe}_{0.65}\text{Pb}_{0.35}\text{O}_3$  samples at 1kHz, 3kHz and 5kHz frequencies. The dielectric studies exhibits a dielectric anomalies in  $\text{BiFe}_{0.95}\text{Pb}_{0.05}\text{O}_3$ ,  $\text{BiFe}_{0.85}\text{Pb}_{0.15}\text{O}_3$ ,  $\text{BiFe}_{0.75}\text{Pb}_{0.25}\text{O}_3$  and  $\text{BiFe}_{0.65}\text{Pb}_{0.35}\text{O}_3$  samples at different temperatures at 1 kHz, 3 kHz and 5 kHz frequencies are given in the table 1.

## CONCLUSIONS:

We have successfully prepared the pure  $\text{BiFeO}_3$  as well as the Pb substituted  $\text{BiFeO}_3$  samples like  $\text{BiFe}_{0.95}\text{Pb}_{0.05}\text{O}_3$ ,  $\text{BiFe}_{0.85}\text{Pb}_{0.15}\text{O}_3$ ,  $\text{BiFe}_{0.75}\text{Pb}_{0.25}\text{O}_3$  and  $\text{BiFe}_{0.65}\text{Pb}_{0.35}\text{O}_3$  ceramics were formulated through solution combustion method (SCM).

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