Effect of Preheating Temperature on Synthesis of Pure BiFeO$_3$ via Sol–Gel Method

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Abstract. BiFeO$_3$ (BFO) nanopowder was synthesized in a pure form via a sol-gel method based on glycol gel reaction. Effect of drying and preheating temperature on preventing other phases was studied. Many parameters were studied as calcination temperature and time & stirring temperature as well. The prepared powder was characterized by X-Ray Diffraction of powder (XRD) and Transmission Electron Microscope (TEM). High pure BiFeO$_3$ was obtained by preheated process at 400 °C for 0.5 h and calcination at 600 °C for 0.5 h without any impurities compared to dry at 110 °C.

Keywords: BiFeO$_3$, sol–gel, preheating temperature, drying temperature

Introduction

So far, there is a great interest to discover substances that have properties stable in room temperature, which has widespread applications in the manufacturing of devices. Of these, multiferroic materials which defined as substance coupling between two or more of the following property: ferroelectricity, ferromagnetic, ferroelasticity [1]. There are many types of multiferroic materials were studied, but the common issue with them is the ferroic properties are low at room temperature. Consequently, there is difficult to use in many applications. Considerable attention has been paid to BiFeO$_3$ because it considers the only material has stable multiferroic properties at room temperature [2–4]. Its ferroelectricity cure temperature is 830 °C and the ferromagnetic Neel’s temperature is 370 °C [5–7]. BiFeO$_3$ has been utilized in many applications such as industry of radio, television, microwave and satellite communications, audio-video, digital recording and, as permanent magnets, information storage [8], transducer and magnetoelectric coupling [9, 10]. However, technological application of BiFeO$_3$ is limited because of forming secondary phases during synthesis, weak magnetic characteristics, lower magnetoelectric coupling coefficients, a large difference in ferroic transition temperatures and high leakage current [11].
Much research on the synthesis of BiFeO$_3$ using different methods have been done. Of these, solid-state [12], mechano-chemical [13, 14], solution chemistry methods such as Pechini method [15], precipitation/coprecipitation [16, 17], sol–gel [18–20], alkali metal ions-assisted controllable synthesis hydrothermal method [21–23], sonochemical [24], solution-combustion method [25] and soft chemical method [26]. Most of the mentioned procedures need high-temperature treatments (>800 °C) and the appearance of second phases still the major issue of them[27]. To avoid bismuth volatilization and minor phase formation, the development of low-temperature synthesis methods is essential.

The sol–gel method is a very effective method for fabricating a uniform ultrafine porous powder and can also be moved up to accommodate industrial-scale production [28]. There are several techniques for sol–gel method as glycol–gel reaction [29–31], Metal complex [32–35], modified Pechini [36], Polymer complex solution [37–39]. Among them, glycol–gel reaction considers the best because of low chemical used in this reaction and several roles of ethylene glycol [40].

Many researchers used sol–gel method for preparing BFO by studying different parameters for obtaining a pure form. Of these, calcination temperature and time [41–43], precursor type [44], excess amount of Bi$^{3+}$ [45–47], few studies have done on the impact of reaction temperature (stirring temperature) [48] but no one studies the difference of drying and preheats temperature of the resulted gel and its relation to obtaining pure BFO.

The paper provides a comparison study of the impact of different drying temperature on forming a pure form of BiFeO$_3$ via a sol–gel method based on glycol gel reaction was reported.

**Experimental**

*Materials*

Precursors of Bi(NO$_3$)$_3$.5H$_2$O, Fe(NO$_3$)$_3$.9H$_2$O, nitric acid and ethylene glycol were of Sigma-Aldrich highly pure grade and used without further purifications.

*Methodology*

The process of synthesis BiFeO$_3$ is presented in *Fig 1*. A mixed solution of equimolar (0.2M) of Bi(NO$_3$)$_3$.5H$_2$O and Fe(NO$_3$)$_3$.9H$_2$O in 50 ml ethylene glycol was prepared. The solution mixture was stirred until completely dissolved and a clear red solution obtained. After dissolving, the mixture was stirred at 80 °C until the brown gel was formed. The obtained gel was divided into two parts:

- **Part I:** was dried at 110 °C for 48h. The resulted powder was then calcinated from 400– 600 °C for different intervals (0.5 h – 3 h).
- **Part II:** was preheated at 400 °C for 30 min, then calcinated 500–600 °C for 0.5 h.

Nitric acid (10%) was used (when the gel color is started to transfer into green, the acid was added to prevent forming this color) to prevent reduction of Fe$^{3+}$ to Fe$^{2+}$. Otherwise, electric neutrality required, which lead to a high number of oxygen vacancies [49, 50].

*Effect of stirring temperature*

Stirring temperature was varied from 20–80 °C after that the gel was dried at 110 °C and was calcined at 600 °C for 0.5 and 2 h.
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**Characterization**

The structure and phase composition of the synthesized bismuth ferrite powders were investigated using X-ray diffractometer (XRD) BrukerAXSD8 Germany Cu K$_\alpha$ radiation 0.154 and Transmission Electron Microscope (TEM) JEOL (JEM-HR-2100 ELECTRON MICROSCOPE).

**Results and discussion**

**Effect of calcination temperature**

There is no doubt that, temperature affects both the formation and stability of BiFeO$_3$ nanopowder. This is may because that the elevated temperature leads to the following: i) instability of BiFeO$_3$ at high temperatures where, BiFeO$_3$, undergoing a phase transition at about 850 °C [51], ii) it can decompose into Fe$_2$O$_3$ and Bi$_2$O$_3$, as shown in Eq. (1):

$$2\text{BiFeO}_3 \xrightarrow{\Delta} \text{Fe}_2\text{O}_3 + \text{Bi}_2\text{O}_3.$$  (1)

Various heating temperatures were reported by many researchers ranged from 60–140 °C for drying and evaporation of the solvent followed by calcination at varying temperature for forming BiFeO$_3$ [11, 41, 44, 53–57]. Furthermore, according to previous studies [42, 53], 400 °C was considered as the crystallization temperature of BiFeO$_3$, oppositely, it decomposed above 600 °C and second phases observed [58, 59]. As a result of that, direct heating
at 400 °C was applied in this work and the maximum calcination temperature was chosen to be 600 °C.

As can be seen, on drying the powder of BFO at 110 °C (Fig. 2 and Table 1), the highest weight percentage of BFO was obtained at calcination temperature 500 °C with 76.9% and crystal size of 71.3 nm and Bi$_2$O$_3$ & Bi$_2$Fe$_4$O$_9$, as other phases were formed.

On the other hand, on heating immediately at 400 °C, the XRD (Fig. 3) indicates that sharp and intense peaks of high crystallinity of BiFeO$_3$ were observed at calcination temperature 600 °C without any second phase formation. The crystal size of BFO determined from XRD at 2$\theta$ = 32 was found to be 62.1 nm (Table 1).

In comparison between the two parts I, II, Table 1, the overall trend of the increasing temperature on forming BFO the same. As calcination temperature increases the impurity decrease and BFO crystallization raise. Heating gel directly at 400 °C without further drying led to a higher weight percentage of BFO than drying process.

Data obtained in previous studies indicated that, second phases was regarded using sol–gel method for synthesis of BiFeO$_3$ as follows: Bi$_2$Fe$_4$O$_9$, Bi$_{36}$Fe$_{24}$O$_{57}$ and Bi$_2$O$_3$ [60]; Bi$_2$Fe$_4$O$_9$, Bi$_{22}$FeO$_{29}$ and Bi$_2$O$_3$ [20]; $\gamma$-Fe$_2$O$_3$ [61], Bi$_2$Fe$_4$O$_9$, Bi$_{25}$FeO$_{40}$ [47, 48]; Bi$_2$Fe$_4$O$_9$ [62–64] and Bi$_{12}$FeO$_{40}$ [10, 17]. Likewise, Sharma et al. heat at 400 °C for 1 h and calcinated at 700 °C for 2 h, found Bi$_2$Fe$_4$O$_9$ and Bi$_{25}$FeO$_{40}$ as a second phase formed with BFO [65]. Zhang et al. use three different temperatures for obtaining a pure form of BFO as follows: 100 °C for solvent evaporation and gel formation, 350 °C for organic removal for 1h, finally calcinated at 550 °C for 2 h [66].

![Fig. 2. Effect of calcination temperature on drying sol–gel method](image-url)

Table 1. Comparison between the sol–gel method according to calcination temperature after either drying or preheating

| Temperature (°C) | Drying on 110 °C for 48 h | Preheating on 400 °C for 0.5 h |
|------------------|----------------------------|-------------------------------|
|                  | Wt. (%)  Crystallite Size (nm) | Wt. (%)   Crystallite Size (nm) |
| 400              | 10.4    47.6              | 70.9       28.6              |
| 500              | 76.9    71.3              | 9.2        23.5              |
| 600              | 70.4    71.4              | 100        62.1              |
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Effect of calcination time

Effect of calcination time was studied on part I (the dried powder of BFO) to improve the purity of BiFeO$_3$ obtained by this route of sol–gel. This impact was studied within the range of 0.5 h – 3 h. From XRD pattern, Fig. 4 and Table 2, it was clear that 0.5, 1 and 3 h nearly analogous to each other, where the weight percent of BFO were 76.9, 78.4 and 78.4%, respectively. The purity of BFO was found to be time independent. Other studies were found that for sol–gel method, 600 °C calcination temperature with time of 2 h is enough for forming crystals of BiFeO$_3$ [67–70].

![Fig. 3. Effect of calcination temperature, 600 °C, on preheating method of sol–gel](image)

**Fig. 3. Effect of calcination temperature, 600 °C, on preheating method of sol–gel**

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**Fig. 4. Effect of calcination time on drying method of sol–gel**

**Table 2. Effect of calcination temperature and time on drying sol–gel method**

| Calcination temperature (°C) | Weight percent (%) at different times (h) |
|------------------------------|-------------------------------------------|
|                              | 0.5 | 1   | 2   | 3   |
| 400                          | 10.4| 72  | 9.8 | 33.5|
| 500                          | 76.9| 78.4| 44.5| 78.4|
| 600                          | 70.4| 15.4| 23.6| 31.8|
Effect of stirring temperature

The reaction temperature is considered as one of the crucial factors affecting sol and gel formations [71]. The study of the effect of stirring temperature was applied to part I. In comparison, of the results obtained from the effect of calcination temperature for preheating method (600 °C for 0.5 h) and with other researchers like Fukumura et al. [68] (600 °C for

Table 3. Comparison between stirring temperature for samples calcinated for 0.5 and 2 h

| Stirring temperature (°C) | BiFeO₃ 600 °C for 0.5 h | BiFeO₃ 600 °C for 2 h |
|---------------------------|-------------------------|-----------------------|
|                           | Crystallite size (nm)   | Wt. (%)               | Crystallite size (nm) | Wt. (%)               |
| 20                        | 78.3                    | 76.7                  | 90.1                   | 83.2                  |
| 40                        | 110.8                   | 38.7                  | 105.8                  | 96.7                  |
| 60                        | 97.6                    | 84.3                  | 98.9                   | 82.6                  |
| 80                        | 109.8                   | 70.4                  | 29.5                   | 23.6                  |

Fig. 5. Effect of stirring temperature on synthesis of BiFeO₃ using drying sol–gel method for 2 h

Fig. 6. Effect of stirring temperature on synthesis of BiFeO₃ using drying sol–gel method for 0.5 h
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2 h), the effect of stirring temperature was conducted within the range of 20–80 °C at calcination temperature of 600 °C and calcination time of 0.5 h and 2 h in a comparative study.

It was regarded that, there was an improvement in weight percentage of BiFeO$_3$ on change stirring temperature. From Table 3 and Figs 5, 6, the sharpest, most intense peaks and purity of the multiferroic nano powder were fabricated at stirring temperature of 40 °C for 2 h calcination time with 96.7% weight percent. As well, from Table 4, at stirring temperature 40 °C, the time required for forming sol and gel was very small compared with other temperatures. Whereas, Surech and Srinath have studied reaction temperature on the range of 80–400 °C for synthesis BFO through a metal complex reaction sol–gel method and found that, 250 °C reaction temperature give more pure BFO than other studied temperatures [48].

### TEM micrographs

Transmission Electron Microscopy (TEM) micrographs of BiFeO$_3$ Fig. 7 illustrated that, BFO prepared by drying sol–gel method consists of spherical and rectangular shapes with a particle size of 13.5–24.6 nm. Instead, Fig. 7c,d, HR-TEM gave the inter-planer of 0.47 nm of the crystal. For BFO synthesized by preheat sol–gel method, TEM micrographs, Fig. 8a,b, exhibit spherical crystals and irregular atomic clusters with particle size ranging from 87.6–179.84 nm. The appearance of agglomerated clusters, as a result of high temperature the sample exposed [72].

![TEM micrographs](image)

**Fig. 7.** TEM morphology of (a) BFO prepared with drying sol-gel method for 1 h calcination at 500 °C (c) and (d) HR-TEM image
Conclusion

BFO synthesized using a sol–gel method with two different drying temperatures resulted in- 
dicated that variation on drying temperature (110 or 400 °C) led to a variation on purity, 
morphology and crystal size of the final product. In comparison, between the two tem- 
p eratures, drying on 110 °C, 96% BFO was formed with crystal size 105.8 nm at calcination 
temperature 600 °C for 2 h with 40 °C stirring temperature. On the other hand, extra phase 
was observed with BFO nanopowder. In contrast, for preheating on 400 °C, pure BFO 
nanopowder without any secondary phases resulted with crystal size 62.1 nm and some 
irregular morphology.

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