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Utilizing maleic acid as a novel fuel for synthesis of $PbFe_{12}O_{19}$ nanoceramics via sol–gel auto-combustion route

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ABSTRACT

$PbFe_{12}O_{19}$ nanostructures were prepared in an aqueous solution by the sol–gel auto-combustion method using $Pb(NO_3)_2$ and $Fe(NO_3)_3$ as starting materials and various carboxylic acids, including oxalic acid, malonic acid, succinic acid and maleic acid as fuel and reducing and capping agents. The as-synthesized products were characterized by X- ray diffraction, scanning electron microscopy, and X-ray energy dispersive spectroscopy. The effect of carboxylic acid type, Pb^{+2} to carboxylic acid molar ratio, and calcination temperature was investigated on the morphology of the products and several experiments were carried out to obtain the optimal reaction conditions. It was found that the phase and the morphology of the products are influenced by the investigated parameters. Furthermore, vibrating sample magnetometer (VSM) was used to study the magnetic properties of $PbFe_{12}O_{19}$ samples.

1. Introduction

Nanomaterials have been broadly investigated for the fundamental scientific and technological interests in accessing new classes of functional materials with unique properties and applications [1–4].

Among magnetic materials, ferrite ceramics are currently the focus of considerable interest. Hexagonal ferrite ceramics are generally applied as permanent magnets, high-density magnetic recording media, and microwave devices because of their unique properties including large magnetocrystalline anisotropy, high saturation magnetization, coercivity, and corrosion resistivity [5,6]. $PbFe_{12}O_{19}$, an important member of this family, has been less investigated due to its low saturation magnetization, anisotropy field, and undesirable environmental effects which are originated from the chemical stability of this material (i. e. decomposition to lead oxide at 950 °C) compared with the other hexagonal ferrites ($MFe_{12}O_{19}$, M: Ba, Sr) [7]. However, recently PbM has attracted great attentions owning to its crystallization temperature which is lower than barium and strontium hexaferrite. This feature has changed PbM into attractive materials and researchers have focused on the preparation and characterization of these compounds. Investigating the effect of Pb substitution in BaM is one of the appealing reports in recent years [8–10].

Among various methods developed for the synthesis of $PbFe_{12}O_{19}$ including hydrothermal [11], sol–gel [12], glass crystallization [13], and co-precipitation [14], sol–gel route has been mostly used because it is considered as a versatile technique for preparing chemical compounds and inorganic materials. This method providing various advantages, including chemical homogeneity, easy component adjustment, low calcination temperature, and low cost is one of the important routes for the synthesis of nanomaterials [15,16].

In this work, various carboxylic acids were employed as capping agents and fuels for autocombustion process and the results showed that maleic acid could be considered as an efficient capping agent and fuel. Also, for obtaining a pure product, the various molar ratios of Pb^{2+} to maleic acid as well as different calcination temperatures were applied. The products were characterized by X-ray diffraction (XRD), scanning electron microscopy (SEM), and X-ray energy dispersive spectroscopy analysis (EDS). Vibrating sample magnetometer (VSM) was utilized to study the magnetic properties of $PbFe_{12}O_{19}$ samples.

2. Experimental

The $PbFe_{12}O_{19}$ samples were prepared as follows: first, an aqueous solution containing 0.6 mmol of $Pb(NO_3)_2$ was prepared, then 7 mmol of $Fe(NO_3)_3$ was added and the solution was kept vigorously stirred at 50 °C for 30 min. Second, an aqueous solution (0.14 M) containing 7 mmol of desirable carboxylic acid (oxalic acid, malonic acid, succinic acid or maleic acid) was dropwise added to the above solution. The above solution was heated at 100 °C and stirred for 1–2 h. Evaporation of the solution caused the formation of a highly viscous gel which was dried at 100 °C. The formed gel was calcined at 900 °C for 2 h. The products were characterized by XRD, SEM and TEM.

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Table 1
Preparation conditions for samples 1–7.

Sample no.	Pb^{+2}/fuel	Kind of fuel	Calcination temperature (°C)
1	13	Oxalic acid	900
2	13	Malonic acid	900
3	13	Succinic acid	900
4	13	Maleic acid	900
5	6.5	Maleic acid	900
6	26	Maleic acid	900
7	13	Maleic acid	800

3. Results and discussion

To investigate the effect of different parameters on the morphology, purity, and particle size of the products, various experiments were carried out. All of the preparation conditions are listed in Table 1.

Various pH ranges were tried in this work and it was observed that the aimed gel is only formed within a certain pH range (pH = 4.5–5.5). In this pH range, a dark gel was created and precipitation occurred in the condensed phase. It is noteworthy to mention that transparency of the formed gel is an indicator of homogeneity of the composition. In addition, maleic acid that acts as a reducing agent is oxidized by nitrate ions during the combustion reaction and provides the required energy for reaction progress.

3.1. SEM images

SEM images reveal that the morphology of product changes using different conditions. SEM images of synthesized products using various carboxylic acids, including oxalic acid (sample no. 1), malonic acid (sample no. 2), succinic acid (sample no. 3), and maleic acid (sample no. 4) are illustrated in Fig. 1a–d, respectively. By considering images in Fig. 1, it can be observed that with increasing chain length of the

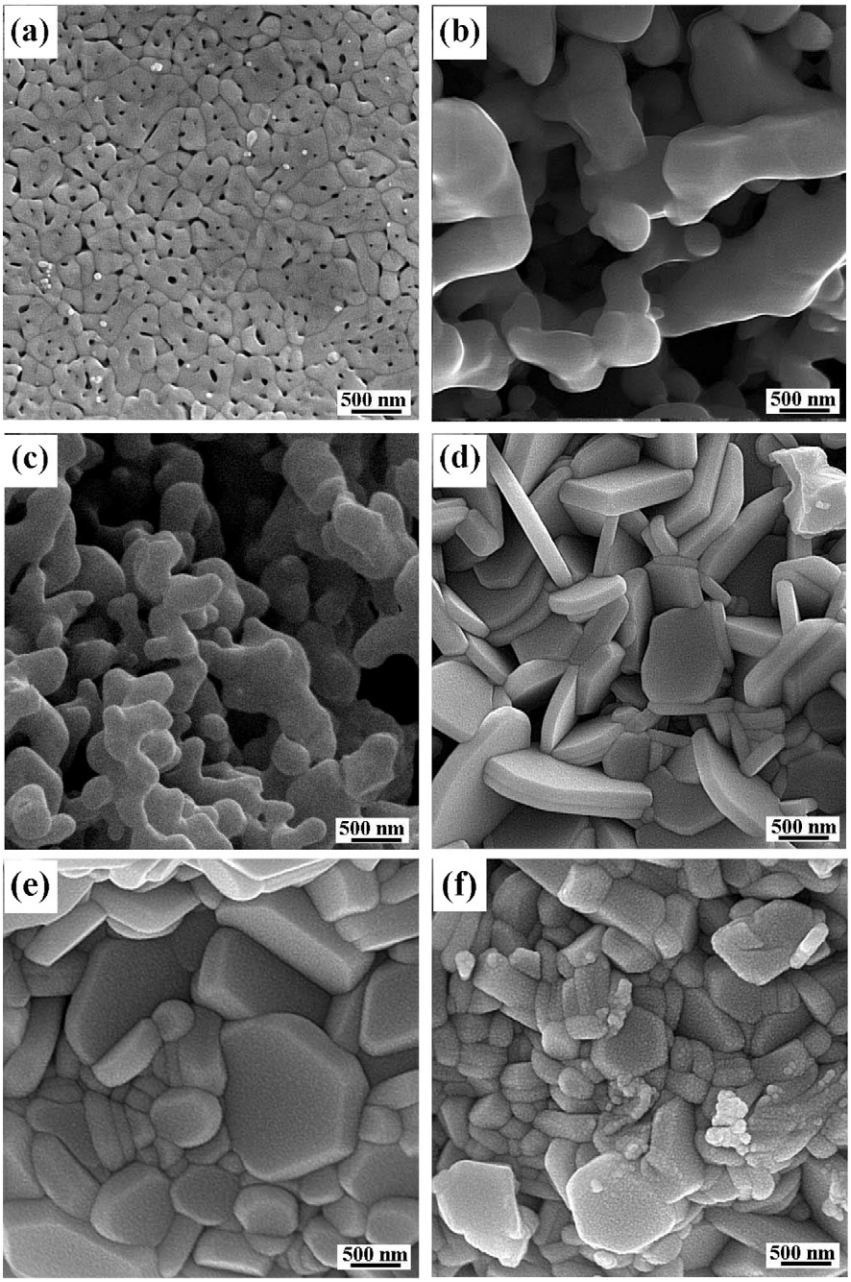

Fig. 1. SEM images of $PbFe_{12}O_{19}$ of (a) sample no. 1, (b) sample no. 2, (c) sample no. 3, (d) sample no. 4, (e) sample no. 5 and (f) sample no. 6.