Magnetic Properties of Yttrium Iron Garnet Polycrystalline Material Prepared by Spray-Drying Synthesis

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Abstract. *The yttrium iron garnet polycrystalline powder was prepared by spry-drying synthesis from nitrates solution. The calcined powder was pressed into pellets and sintered at various temperatures for 2 hours. Prepared samples were characterized by XRD analysis and magnetic properties were measured. The magnetic moment of 4.3 μ_B and saturation magnetization of 24 Am^2kg^{-1} were observed for sample sintered at 1000 °C.*

Keywords: Yttrium Iron Garnet, Spry-Drying Synthesis, Magnetic Properties

1. Introduction

Yttrium iron garnet ($Y_3Fe_5O_{12}$, YIG) is one of the most important ferromagnetic materials and has been widely applied for tunable microwave devices, circulators, isolators, phase shifters, nonlinear devices, magnetic bubble domain-type digital memories [1], etc. due to its excellent electromagnetic properties, including low dielectric loss, narrow ferromagnetic resonance linewidth in microwave region, adjustable saturation magnetization, good temperature and chemical resistance [2]. In recent years, research attention has been committed to the investigation of YIG and doped YIG nanocrystals since these highly divided materials may be used in Faraday magneto-optical devices for telecommunications [2,3] or for localized hyperthermia by induction heating as biomedical application [4].

Preparation of the YIG materials in polycrystalline or monocrystalline form is associated with technological complications related with high temperatures (≥1600°C) and necessity to apply long annealing times. The successfully preparation of uniform polycrystalline powders with very high homogeneity and particle size distribution by spray drying synthesis and subsequent controlled crystallization is a reasonable economic solution.

In this work, YIG materials were prepared by spry–drying synthesis from nitrates solutions in form of 1μm spheres. Then the prepared powders were calcined and sintered at different temperatures in interval of 800-1000 °C for 2 hours. The sintered samples were characterized by XRD analysis, magnetic properties were measured, quantities of individual crystalline phases were calculated, and the results are reported.

2. Experimental

As starting materials for solution, nitrates $Fe(NO_3)_3 \cdot 9H_2O$ and $Y(NO_3)_3 \cdot 6H_2O$ (Sigma-Aldrich Chemie Gmbh, USA) in a stoichiometric ratio were used. The required amounts of nitrates were weighed and then dissolved in distilled water to a final volume of 250 ml. This way prepared solution was fed into spray-dry apparatus (Büchi Mini-spray dryer B-290). The prepared powder was calcined at 650 °C for 4 h, pressed in the form of pellets with diameter of 8 mm and then sintered at different temperatures (800, 900, 1000 °C) for 2 hours. The X-ray diffraction analyses were performed on a Panalytical Empyrean device (CuKα radiation, 2Θ range 10-80°), to estimate the degree of crystallinity and for qualitative evaluation of individual crystalline phases in samples. The obtained diffraction data have been evaluated by the High Score Plus software (v.3.0.4, PAN Analytical, Netherlands) equipped with the Open Crystallographic Database (OCD, v. 2013). The thermal behaviour of raw material in the temperature range of 35-1200 °C with heating rate of 10 °C/min was studied by DSC analysis (Netzsch STA 449 F1 Jupiter). For this measurement, O_2 atmosphere and platinum crucibles with sample weight of approx. 15 mg were used. After this examination, 5 weight % of Si was added to sintered samples and content of individual crystalline phases was calculated from X-ray diffraction patterns.

DC magnetization measurements were performed on a Quantum Design MPMS XL-7 SQUID magnetometer. Magnetization as a function of temperature was measured in the temperature range of 1,8–400 K. The samples were cooled from the room temperature to the lowest temperature in zero applied field and the magnetization was measured after the application of the field (8000 Am^{-1}), while warming (ZFC, zero-field-cooled measurement). For field-cooled (FC) measurements, the samples were cooled in the presence of field and magnetization was measured as a function of temperature while cooling. Measurement of the magnetization as a function of field was made at two different temperatures (5, 300 K).

3. Results and discussion

The list of prepared samples with calculated content of $Y_3Fe_5O_{12}$ is in Tab.1 (col. 1-3). The prepared precursor powder (YIGP) after 650 °C calcination, was polycrystalline, with content of $Fe_4Y_4O_{12}$ and hematite α-Fe_2O_3 as a major phases. In DTA record of YIGP, the small exothermic effect with maximum at temperature 998 °C was observed, which can be assigned to $Y_3Fe_5O_{12}$ phase formation.

Table 1. Magnetic properties of samples prepared at three different temperatures.

Sample name	Sintering temp. [°C]	XRD YIG quantity[wt %]	Coercitivity H_C [Am^{-1}]	Saturation M_S [Am^2kg^{-1}]	Remanence M_R [Am^2kg^{-1}]
YIG1	800	0	$1{,}87\times10^6$	1,5	5,9
YIG2	900	7	9390	2,7	7,4
YIG3	1000	88	1519	24,0	4,2

From comparison of XRD records of samples was concluded that all prepared samples were polycrystalline with content of $YFeO_3$ (yttrium iron perovskite), $Y_3Fe_5O_{12}$ and α-Fe_2O_3 crystalline phases. Also increasing of portion of $Y_3Fe_5O_{12}$ phase with increasing sintering temperature was observed. The samples sintered at lower temperatures (800, 900 °C) contain mainly α-Fe_2O_3 and $YFeO_3$ and small portion of YIG crystalline phase. The YIG3 sample (Tab.1) contain high portion of YIG phase (88 wt. %), smaller content of $YFeO_3$ (10,8 wt. %) and α-Fe_2O_3 (0,8 wt. %). The presence of the residual phases is the result of incomplete YIG formation according to the reaction scheme:

$$Y_2O_3 + Fe_2O_3 \rightarrow 2YFeO_3 \text{ (perovskite)} \qquad T = 500 - 900°C \qquad (1)$$
$$3YFeO_3 + Fe_2O_3 \rightarrow Y_3Fe_5O_{12} \text{ (garnet)} \qquad T = 900 - 1600\ °C. \qquad (2)$$

Hysteresis loops (Fig. 1) recorded at the temperature of 5 K indicate that with increasing of the sintering temperature, the saturation magnetization rises with increasing amount of the $Y_3Fe_5O_{12}$ phase. The profile of the magnetization for the YIG3 sample suggest obtaining of a soft ferromagnetic material, because sample is very sensitive to an external magnetic field, reaches its saturation at relatively small field and the coercivity is small (< 1600 Am^{-1}).

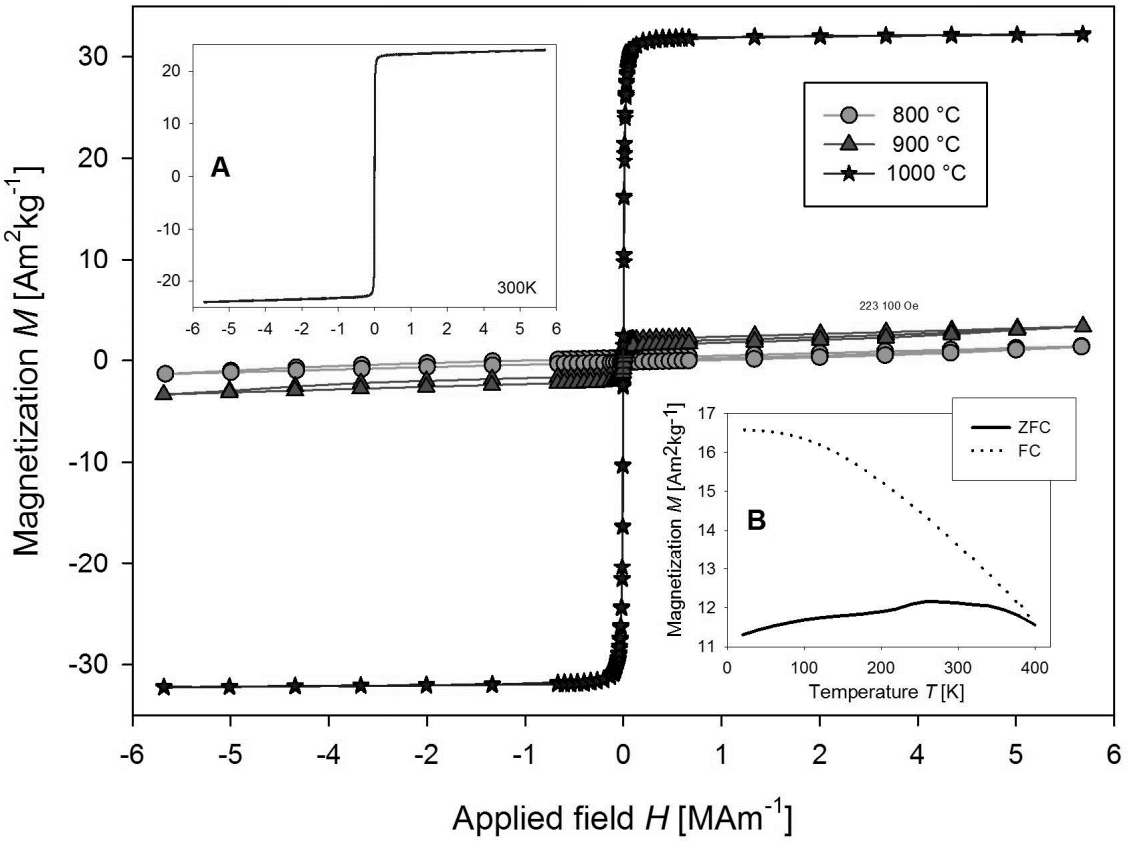

Fig. 1 Hysteresis loops recorded at the temperature of 5K for the samples sintered at three different temperatures. The inset A shows room temperature hysteresis loop for the sample YIG3 annealed at 1000 °C. The inset B shows ZFC and FC magnetization curves measured in a field of 8000 Am^{-1} for the YIG3 sample.

The experimental value of the magnetic moment for this sample is 4,3 μ_B. The inset A of Fig. 1 shows hysteresis loop for the YIG3 sample measured at the room temperature, with the saturation magnetization of 24 Am^2kg^{-1}. These values are lower than expected for bulk YIG (5 μ_B, 26,8 Am^2kg^{-1} [1,2,3]) and it could be attributed to the presence of residual phases. Magnetic properties (for 300 K measurement) of prepared samples are summarized in Tab. 1 (col. 4-6). The inset B of Fig. 1 shows the temperature dependence of the ZFC/FC magnetization measured at applied field of 8000 Am^{-1} for the YIG3 sample in the temperature range of 20–400 K. Although, the Curie temperature T_C for YIG is higher than 400 K [1], the following features can be observed: (1) if we could increase the temperature, both curves will probably collapse above $T_S \geq 400$ K; (2) irreversibility is observed below T_S, with $M_{ZFC} < M_{FC}$; (3) the maximum value of the ZFC magnetization is observed at $T_M < T_S$ (~ 260 K).

4. Conclusions:

Three samples with YIG composition were prepared by spry-drying synthesis and sintered at 800, 900 and 1000 °C for 2 hours. All prepared samples were studied by XRD analysis and magnetic properties were measured**.** The samples sintered at lower temperatures contained mainly perovskite and hematite phases. In contrast, sample sintered at 1000 °C contained 88 % of YIG phase. The magnetic moment of 4,3 μ_B and saturation magnetization of 24 Am^2kg^{-1} was measured for this sample. These results show the possibility of simple preparation of YIG materials with interesting magnetic properties.

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