

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 spray-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 $4.3 \mu_B$ and saturation magnetization of $24 \text{ Am}^2\text{kg}^{-1}$ were observed for sample sintered at 1000°C .*

Keywords: *Yttrium Iron Garnet, Spray-Drying Synthesis, Magnetic Properties*

1. Introduction

Yttrium iron garnet ($\text{Y}_3\text{Fe}_5\text{O}_{12}$, 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 ($\geq 1600^\circ\text{C}$) and necessity to apply long annealing times. The successful 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 spray-drying synthesis from nitrates solutions in form of $1\mu\text{m}$ spheres. Then the prepared powders were calcined and sintered at different temperatures in interval of $800\text{--}1000^\circ\text{C}$ 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 $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ and $\text{Y}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ (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 ($\text{CuK}\alpha$ 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).

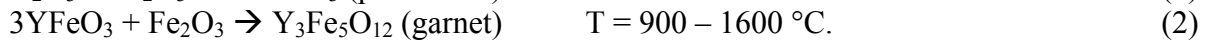
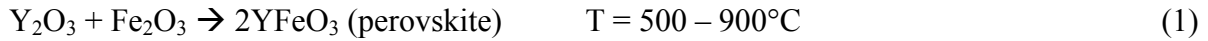
3. Results and discussion

The list of prepared samples with calculated content of $\text{Y}_3\text{Fe}_5\text{O}_{12}$ is in Tab.1 (col. 1-3). The prepared precursor powder (YIGP) after 650 °C calcination, was polycrystalline, with content of $\text{Fe}_4\text{Y}_4\text{O}_{12}$ and hematite $\alpha\text{-Fe}_2\text{O}_3$ 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 $\text{Y}_3\text{Fe}_5\text{O}_{12}$ 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 [$\text{Am}^2\text{kg}^{-1}$]	Remanence M_R [$\text{Am}^2\text{kg}^{-1}$]
YIG1	800	0	$1,87 \times 10^6$	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), $\text{Y}_3\text{Fe}_5\text{O}_{12}$ and $\alpha\text{-Fe}_2\text{O}_3$ crystalline phases. Also increasing of portion of $\text{Y}_3\text{Fe}_5\text{O}_{12}$ phase with increasing sintering temperature was observed. The samples sintered at lower temperatures (800, 900 °C) contain mainly $\alpha\text{-Fe}_2\text{O}_3$ 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 $\alpha\text{-Fe}_2\text{O}_3$ (0,8 wt. %). The presence of the residual phases is the result of incomplete YIG formation according to the reaction scheme:



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 $\text{Y}_3\text{Fe}_5\text{O}_{12}$ 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 ($< 1600 \text{ Am}^{-1}$).

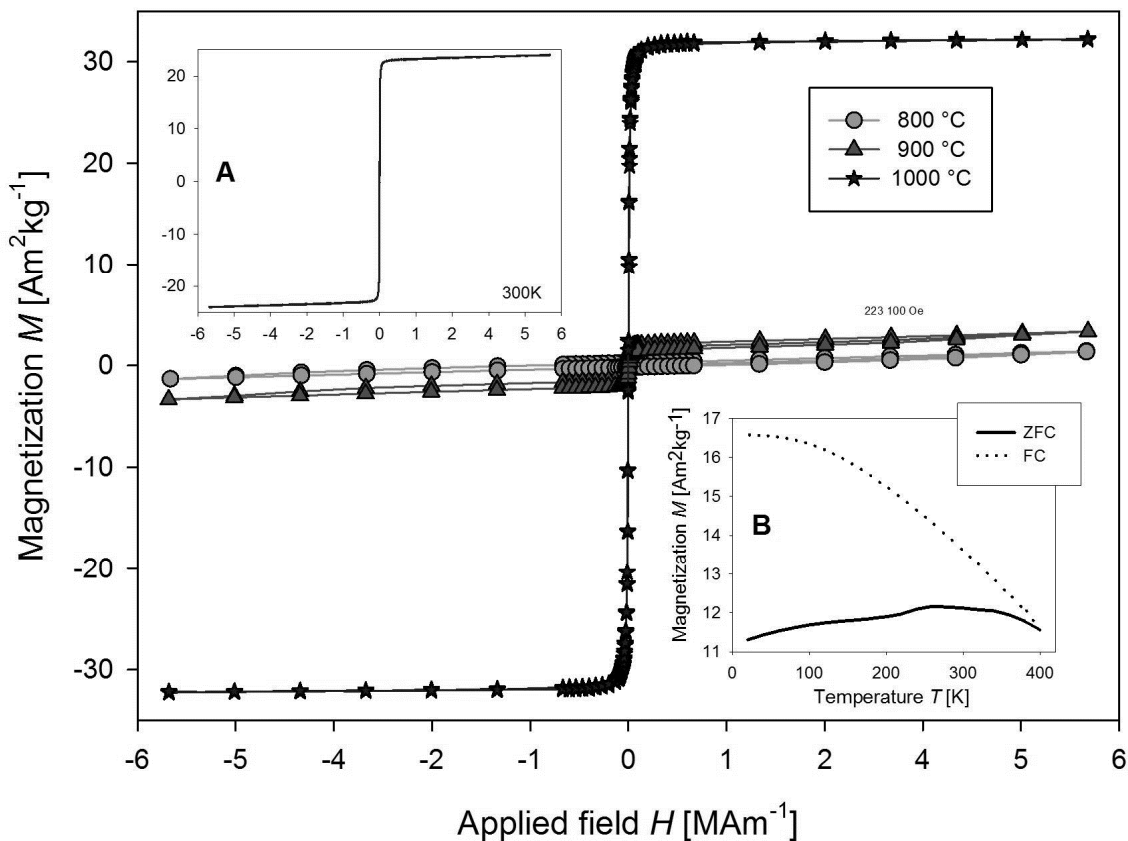


Fig. 1 Hysteresis loops recorded at the temperature of 5K for the samples sintered at three different temperatures. The inset A shows room temperature hysteresis loop for the sample YIG3 annealed at 1000 °C. The inset B shows ZFC and FC magnetization curves measured in a field of 8000 Am^{-1} for the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

The experimental value of the magnetic moment for this sample is $4,3 \mu_B$. The inset A of Fig. 1 shows hysteresis loop for the YIG3 sample measured at the room temperature, with the saturation magnetization of $24 \text{ Am}^2\text{kg}^{-1}$. These values are lower than expected for bulk YIG ($5 \mu_B$, $26,8 \text{ Am}^2\text{kg}^{-1}$ [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 $T_S \geq 400 \text{ K}$; (2) irreversibility is observed below T_S , with $M_{ZFC} < M_{FC}$; (3) the maximum value of the ZFC magnetization is observed at $T_M < T_S$ ($\sim 260 \text{ K}$).

4. Conclusions:

Three samples with YIG composition were prepared by spray-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 $4,3 \mu_B$ and saturation magnetization of $24 \text{ Am}^2\text{kg}^{-1}$ was measured for this sample. These results show the possibility of simple preparation of YIG materials with interesting magnetic properties.

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References

- [1] Nguyet DTT, Duong NP, Satoh T, Anh LN, Hien TD. Temperature-dependent magnetic properties of yttrium iron garnet nanoparticles prepared by citrate sol–gel. *Journal of Alloys and Compounds*, 541: 18-22, 2012.
- [2] Zhang W, Guo C, Ji R, Fang C, Zeng Y. Low-temperature synthesis and microstructure-property study of single-phase yttrium iron garnet (YIG) nanocrystals via a rapid chemical coprecipitation. *Materials Chemistry and Physics*, 125(3): 646-651, 2011.
- [3] Fu HP, Hong RY, Wu YJ, Di GQ, Xu B, Zheng Y, Wei DG. Preparation and Faraday rotation of Bi-YIG/PMMA nanocomposite. *Journal of Magnetism and Magnetic Materials*, 320(21): 2584-2590, 2008.
- [4] Grasset F, Mornet S, Demourgues A, Portier J, Bonnet J, Vekris A, Duguet E. Synthesis, magnetic properties, surface modification and cytotoxicity evaluation of $\text{Y}_3\text{Fe}_{5-x}\text{Al}_x\text{O}_{12}$ ($0 \leq x \leq 2$) garnet submicron particles for biomedical applications. *Journal of Magnetism and Magnetic Materials*, 234(3): 409-418, 2001.