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Microstructure and magnetic properties of yttrium-iron garnet prepared by a sol–gel method

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Abstract— **Yttrium iron garnet particles were synthesized by the sol-gel method of auto-combustion. Their crystalline structures and magnetic properties were investigated by using X-ray diffraction (XRD), vibrating sample magnetometer (VSM), and Mössbauer spectrum. The crystallite size and magnetic properties were observed and studied. The size of magnetic nanoparticles increased with increasing calcination temperature. The study of magnetic properties by VSM at room temperature showed that the samples are ferromagnetic, having the specific magnetizations of 20,6, 19 and 18.3 emu/g for $Y_3Fe_5O_{12}$ samples calcined at 900, 1000, and 1100°C, respectively. It is concluded that samples prepared via sol gel technique appear to be an attractive route due to the possible reduction of cost of production and the enhanced magnetic and structural properties of the sample.**

Index Terms— **Yttrium iron garnet, Mössbauer spectroscopy, XRD, Sol–gel method, Magnetic properties.**

I. INTRODUCTION

Ferromagnetic garnets are assigned to cubic structure (space group Ia3d), every cell contains eight $R^{3+}{}_3Fe^{3+}{}_5O_{12}$ molecules. R^{3+} ion cannot occupy the octahedral sites and tetrahedral sites because of its large ionic radius, it can only occupy dodecahedral sites which have large space. In the case of ferromagnetic garnet $R_3Fe_5O_{12}$, the ion distribution structure can be represented by writing the garnet formula as $\{R_3\}[Fe_2](Fe_3)O_{12}$, where { }, [ ], and ( ) are represented for 24c (dodecahedral), 16a (octahedral), and 24d (tetrahedral), respectively. The dodecahedron shares two edges with tetrahedra, four with octahedra, and four with other dodecahedra. Each octahedron shares six edges and each tetrahedron shares two edges with dodecahedra. The octahedra and tetrahedra share only corners. The dodecahedron has four different cubic-edge lengths, and the octahedron and tetrahedron two different ones. Consequently, the coupling of different polyhedra with different cubic-edge lengths is complicated. The fundamental magnetic properties of iron garnet originate from the magnetic ions and their relationship to the surrounding oxygen ions. The oxygen ions influence the electronic configuration of the enclosed iron ions and provide the superexchange between the iron ions in different sites [1].

Yttrium Iron Garnet (YIG – $Y_3Fe_5O_{12}$) is the most representative and well-known compound among the rare-earth garnets. $Y_3Fe_5O_{12}$ is a material used widely in electronic devices for the microwave region as well as in magnetic bubble domain-type digital memories. Recently, polycrystalline YIG also found its application in magneto-optical recording media devices [2-5]. However, to improve their present applications and develop new uses, a full understanding of their magnetic properties is required. Ferrimagnetic garnets are very well suited for magnetism studies, as these materials have a uniquely defined cation distribution and do not present any site inversion problems which can arise in other ferrites [6]. In recent years, scientists' interest was focused on the research of the properties of garnet structures depending on the size of particles. Some magnetic properties, such as saturation magnetization, remanence and coercitivity, depend critically on the structure and microstructure of the materials. Therefore, it is important to develop techniques to produce garnets with a strict control of the composition, homogeneity, size and particle shape [7]. In this regard, alternative methods of synthesis of garnet structures that ensure weak-agglomerated homogeneous in size nanoparticles actively began to be developed and implemented.

This paper studies the magnetic microstructure and magnetic properties of polycrystalline Yttrium Iron Garnet obtained by sol-gel method of auto-combustion.

II. EXPERIMENTAL

A. *Preparation of samples*

Studied material was synthesized by the sol-gel method of auto-combustion. As initial reagents there were used: $Y(NO_3)_3 \cdot 6H_2O$, $Fe(NO_3)_3 \cdot 9H_2O$ (nitrates crystalline hydrates of Y and Fe respectively), $C_6H_8O_7 \cdot H_2O$ (citric acid) and NH_4OH (ammonia). A solution of initial components was obtained with pH = 1. By increasing the pH value of solution, the single-phase garnet could be formed at a higher temperature. So, there is no advantage in terms of powder formation by increasing the pH precursor solution. Initial components for the formation of dispersed yttrium iron garnet were obtained by the method of deposition of colloidal solutions. The advantage of this method is that due to significant outgassing finely dispersed weak-agglomerated powder, in which metal oxides are mixed almost on molecular level, produce. Synthesized by us material for production of polycrystalline $Y_3Fe_5O_{12}$ was annealed at different

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temperatures.

B. Experimental techniques

The phase identification of the calcined powders was performed by X-ray diffraction (XRD) on a DRON-3 X-ray diffractometer using $Cu_{K\alpha}$ – radiation (λ = 1.5405 Å). The average crystallite sizes of the synthesized powders were determined by using the XRD patterns, via the well known Scherrer equation.

Mössbauer study conducted using a spectrometer MS-1104Em (Co^{57}:Cr), line width metallic α-Fe was 0.29 mm/s, calibration isomer shift occurred relative to α-Fe.

Magnetic measurements are carried out at room temperature using the vibrating sample magnetometer (7404 VSM, Lake Shore Cryotronics, Inc.).

III. Results and Discussion

Figure 1 shows the X-ray diffraction patterns for the $Y_3Fe_5O_{12}$ powders calcined at different temperatures. The XRD pattern of the sample with pH=1 indicates that the powder is a single-phase YIG. The direct formation of crystalline YIG during combustion and heat treating at 700°C is surely due the higher degree of compositional homogeneityand the greater heat generated from the exothermic reaction of nitrates and citric acid.

Using the Scherer's formula, the variations of full width at half maximum (FWHM – β) at different XRD peaks was calculated average crystallite size annealed at different temperature.

$$D=\frac{k\lambda}{\beta\cos\theta}, k=0.9, \lambda=0.15405\,\text{нм}$$

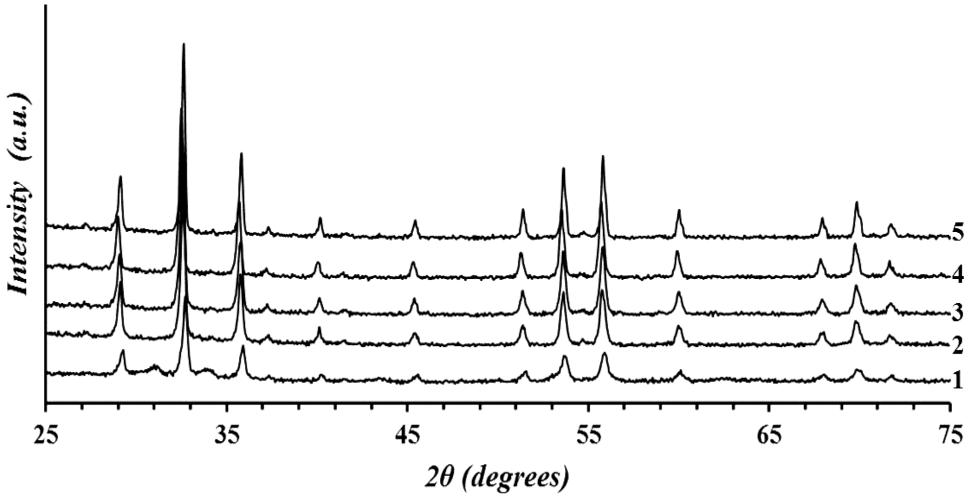

Fig. 1. XRD patterns of $Y_3Fe_5O_{12}$ samples calcined in air at (1) 700, (2) 800, (3) 900, (4) 1000 and (5) 1100 ºC for 2 h.

Calcination at high temperature leads to the reduction in the full width at half maximum due to the improvement in crystallinty and grain growth. The crystalline sizes increase as the treatment temperature (fig. 2).

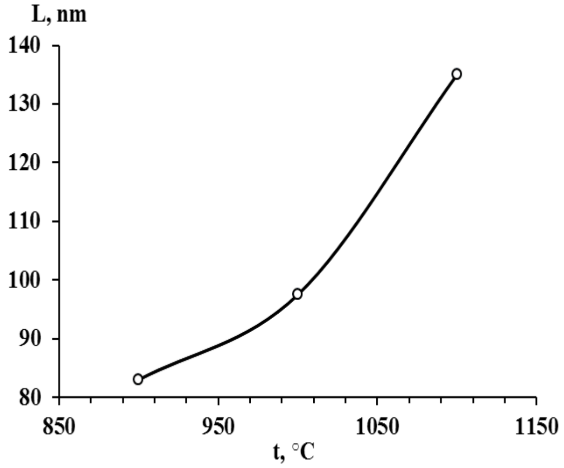

Fig. 2. The dependence of the size of coherent scattering blocks of annealing temperature.

The increase of temperature of isothermal annealing leads to decrease of the lattice constant of garnet. Structural changes caused by increasing the value of temperature of isothermal annealing of garnet forming systems lead to corresponding changes of the magnetic microstructure and the resulting changes of magnetic macroparameters of $Y_3Fe_5O_{12}$.

Mössbauer spectra at room temperature for the $Y_3Fe_5O_{12}$ powders annealed at different temperatures are shown in fig. 3. The ^{57}Fe Mössbauer spectrum is typical of single-phased $Y_3Fe_5O_{12}$ and can clearly identify octahedral [A] and tetrahedral [D] positions ions Fe^{3+}. Mössbauer parameters for the $Y_3Fe_5O_{12}$ powders annealed at different temperatures at room temperature are listed in Table. 1. The relative spectral areas show that the Fe^{3+} ions are distributed over the [A] and (D) sublattices in the usual 2:3 ratio.

$$\frac{S_{a_1}+S_{a_2}}{S_{d_1}+S_{d_2}+S_{d_3}}\approx\frac{2}{3}$$

Annealing at 900 ºC in air for two hours leads to the formation of the garnet single-phase system in which iron ions are in two non-equivalent a-positions with magnetic fields H_{a1}=474 kOe and H_{a2}=489 kOe, and in three non-equivalent d-positions with magnetic fields H_{d1}=392 kOe, H_{d2}=408 kOe and H_{d3}=371 kOe.

The presence of a significant number of non-equivalent positions of Fe^{3+} ions (both a- and d-positions) with different in magnitude magnetic fields and isomer shifts can be associated with the distortion of oxygen octahedrons and tetrahedrons, respectively.

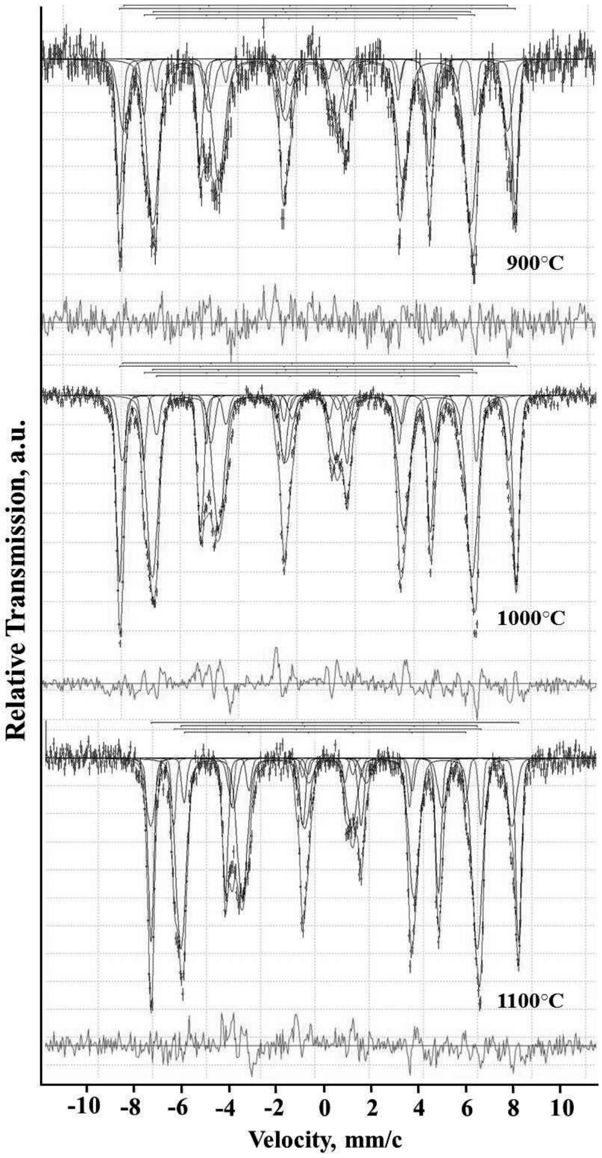

Fig. 3. ^{57}Fe Mössbauer spectrum recorded from the YIG particles prepared at 900-1100°C at room temperature.

Table 1

Mössbauer parameters at room temperature for the $Y_3Fe_5O_{12}$ powders annealed at different temperatures.

t, °C	δ, mm/s	ΔQ, mm/s	a, mm/s	H, kOe	S, %
900	0,43	-0,19	0,46	474	19
	0,35	0,12	0,28	489	21
	0,16	0,04	0,56	392	49
	-0,06	0,29	0,23	408	7
	0,10	-0,26	0,21	371	4
1000	0,45	-0,29	0,34	476	12
	0,35	0,10	0,30	488	27
	0,15	0,04	0,55	394	48
	-0,05	0,29	0,23	410	7
	0,12	-0,24	0,30	372	6
1100	0,41	-0,27	0,45	480	16
	0,36	0,12	0,27	488	24
	0,16	0,04	0,52	394	48
	-0,04	0,29	0,22	408	7
	0,12	-0,24	0,26	374	5

Further isothermal annealing of investigated system at 1000 °C and 1100 °C has not made any significant changes of the magnetic microstructure. Slight decrease of line width of some sextets of the Mössbauer spectrum is apparently caused by decrease of the number of oxygen vacancies as a result of increasing the annealing temperature, which is confirmed by the growth of the magnetic field at the iron nuclei in a-position from 474 kOe to 480 kOe.

The specific magnetization curves of calcined $Y_3Fe_5O_{12}$ sample obtained from room temperature are presented in fig.4. These curves are typical for a soft magnetic material.

Specific saturation magnetization (Ms) values of 20,6, 19 and 18.3 emu/g were observed for $Y_3Fe_5O_{12}$ samples calcined at 900, 1000 and 1100 °C, respectively. With increasing the annealing temperature the value of saturation magnetization decreases. The functional dependence of the coercitive force on the value of temperature of synthesis could not be set, but it can be definitely asserted that this value is less than 25 Oe in all cases.

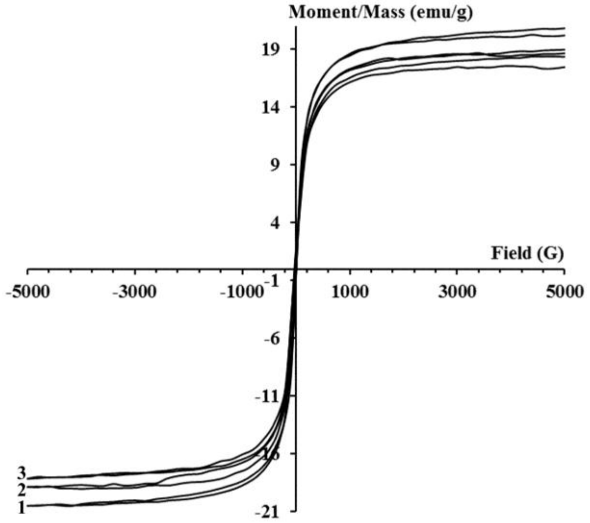

Fig. 4. Magnetisation curves (M–H loops) of YIG samples sintered at different temperatures: **1** - 900°C, **2** - 1000°C, **3** - 1100°C.

IV. CONCLUSION

We have synthesized $Y_3Fe_5O_{12}$ with a single garnet phase sample by a sol–gel method. This method provides the chemical homogeneity and activity of obtained powders, which enables to get finely dispersed polycrystalline porous material with a homogeneous size particles.

The sol-gel technique appears to be the favourable technique, because it provides the 40 and 50% reduction of sintering temperature and of sintering time. Formation of single phase garnet systems based on iron oxide and yttrium derived from acid medium, is provided at the temperature of 700 °C.

The size of magnetic nanoparticles increased with increasing calcination temperature. The calcined $Y_3Fe_5O_{12}$ nanopowder samples are ferromagnetic. The saturation magnetization is reduced with increasing the crystallite size. It takes the values of 20,6, 19 and 18.3 emu/g for Y3Fe5O12 samples calcined at 900, 1000 and 1100 °C, respectively.

REFERENCES

[1] Cheng Zhongjun, Cui Yuming, Yang Hua, Chen Yan "Effect of lanthanum ions on magnetic properties of $Y_3Fe_5O_{12}$ nanoparticles", J. Nanopart Res, 2009, 11, pp.1185–1192.

[2] X.Z. Guo, B.G. Ravi, P.S. Devi, J.C. Hanson, J. Margolies, R.J. Gambino, J.B. Praise, S. Sampath "Synthesis of yttrium iron garnet (YIG) by citrate–nitrate gel combustion and precursor plasma spray processes", J. Mag. Mater., 2005, 295, pp. 145–154.

[3] C.S. Kim, Y.R. Uhm, S.B. Kim, J.-G. Lee "Magnetic properties of $Y_{3-x}La_xFe5O12$ thin filmsgrown by a sol–gel method" J. Magn. and Magn. Mater., 2000, 215, pp.551–353.

[4] P.W. Peterman, M. Ye, P. Wigen "Controlling high frequency chaos in circular YIG films", J. Appl. Phys., 1994, 76, pp.6886–6888.

[5] S. Geller, G.P. Espinosa, P.B. Crandall "Thermal expansion of yttrium and gadolinium iron,gallium and aluminum garnets", J. Appl. Cryst., 1969, 2, pp.86–88.

[6] R.D. Sanchez, J. Rivas, P. Vaqueiro, M.A. Lopez-Quintela, D. Caeiro "Particle size effects on magnetic properties of yttrium iron garnets prepared by a sol–gel method", Journal of Magnetism and Magnetic Materials, 2002, 247, pp.92-98.

[7] Vaqueiro Paz, Arturo Lopez-Quintela M., Rivas Jose "Synthesis of yttrium iron garnet nanoparticles via coprecipitation in microemulsion", J. Mater. Chem., 1997, V.7, #3, pp.501–504.