

## On the synthesis and characterization of iron-containing garnets ( $\text{Y}_3\text{Fe}_5\text{O}_{12}$ , YIG and $\text{Fe}_3\text{Al}_5\text{O}_{12}$ , IAG)

E. Garskaite <sup>a</sup>, K. Gibson <sup>b</sup>, A. Leleckaite <sup>a</sup>, J. Glaser <sup>b</sup>, D. Niznansky <sup>c</sup>,  
A. Kareiva <sup>a,\*</sup>, H.-J. Meyer <sup>b,\*</sup>

<sup>a</sup> Department of General and Inorganic Chemistry, Vilnius University, Naugarduko 24, LT-03225 Vilnius, Lithuania

<sup>b</sup> Institut für Anorganische Chemie, Eberhard-Karls-Universität Tübingen, Auf der Morgenstelle 18, D-72076 Tübingen, Germany

<sup>c</sup> Department of Inorganic Chemistry, Charles University of Prague, Albertov 2030, CZ-12840, Prague 2, Czech Republic

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### Abstract

A sol–gel method based on in situ generation of mixed-metal chelates by complexing metal ions with 1,2-ethanediol or tartaric acid in an aqueous media has been elaborated to prepare iron containing garnets,  $\text{Y}_3\text{Fe}_5\text{O}_{12}$  (YIG) and  $\text{Fe}_3\text{Al}_5\text{O}_{12}$  (IAG). The X-ray diffraction patterns (XRD) of the powders sintered at 1000 °C showed the formation of monophasic YIG, however the formation of iron aluminium garnet at the same synthesis conditions does not proceed. The phase composition of the samples was additionally characterized by infrared (IR) spectroscopy. Micro-structural features of the polycrystalline samples were studied by scanning electron microscopy (SEM). The magnetic properties of all these sol–gel derived materials were investigated by Mössbauer spectroscopy and magnetic susceptibility measurements.

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### 1. Introduction

Since their discovery the iron-containing oxide phases with  $\text{A}_3\text{B}_5\text{O}_{12}$  cubic garnet structure have been the subject of extensive investigations. These oxides possess unique magnetic, magneto-optical, thermal, electrical and mechanical properties such as ferrimagnetism, excellent creep and radiation damage resistance, high thermal conductivity, high electrical resistivity, controllable saturation magnetization, moderate thermal expansion coefficients, energy-transfer efficiency, narrow linewidth in ferromagnetic resonance and others. These properties make iron-containing garnets suitable for numerous device applications, including magnetic materials (circulators, oscillators, phase shifters for microwave region), sensors, lasers,

phosphorescent sources, microwave and electrochemical devices [1–4]. Many recent publications showed that the behaviour of yttrium iron garnet largely depends on the valence-uncompensated doping [5] or substitution of iron in tetrahedral or octahedral sites [6,7], or substitution of yttrium in dodecahedral sites [3,8–11] by different other metallic cations.

For instance, Zhao et al. [11] have demonstrated that the Bi-substitution can drop relative dielectric constant and loss tangent of YIG very much. The improved magneto-optical properties of higher Faraday rotation angle and lower temperature dependence showed the YbBi:YIG crystal ( $\text{Y}_{2.289}\text{Yb}_{0.246}\text{Bi}_{0.465}\text{Fe}_5\text{O}_{12}$ ) [9]. This was a feasible way to improve magneto-optical properties of YIG by combining two opposite temperature coefficient sign rare-earth iron garnets.

Recently, Grasset et al. [6] have illustrated that the substitution of  $\text{Al}^{3+}$  in YIG increases the specific surface area and decreases the Curie temperature as expected.

\* Corresponding authors. Tel.: +370 5 2336214; fax: +370 5 2330987.  
E-mail addresses: [aivaras.kareiva@chf.vu.lt](mailto:aivaras.kareiva@chf.vu.lt) (A. Kareiva), [juergen.meyer@uni-tuebingen.de](mailto:juergen.meyer@uni-tuebingen.de) (H.-J. Meyer).

The variation of the magnetic moment of the compound  $Y_3Fe_{5-x}Al_xO_{12}$  with  $x < 0.7$  indicated that aluminium cations replace iron cations in tetrahedral sites. Since these particles have also been shown to be non-cytotoxic, they would be suitable for biotechnological applications. Structural and magnetic characterization of  $(Y,Bi)_3(Fe,Ga,Al)_5O_{12}$  layers obtained on different substrates proved the suitability of such layers for applications as magneto-optic sensors [7].

It is interesting to note that the substitution of Bi for Y can even decrease the garnet phase formation temperature and the sintering temperature of YIG [11]. This allows to prepare micron-thick layers of bismuth doped magnetic garnet phase by lower thermal treatments [8]. However, YIG garnet is still hardly used in multilayer microwave components such as isolators, circulators and phase shifters because of high sintering temperature ( $>1350^\circ C$ ) of YIG. Moreover, all above mentioned properties of YIG are highly sensitive not only to the changes in dopant composition or host stoichiometry, but also to the processing conditions, which are very much responsible for the crystallinity, crystal shape, crystal size, crystal size distribution and phase purity of the resulting powders [12].

Previously, for the preparation of nanocrystalline yttrium aluminium garnet ( $Y_3Al_5O_{12}$ , YAG), new mixed-metal  $Y_3Sc_xAl_{5-x-y}Ga_yO_{12}$  ( $0 \leq x, y \leq 5$ ) (YSAGG) garnets and yttrium gallium garnet ( $Y_3Ga_5O_{12}$ , YGG) we have developed very simple aqueous sol-gel process using mixtures of inorganic salts or oxides of the respective elements [13–16]. It has been demonstrated that the sol-gel process offers considerable advantages such as better mixing of the starting materials and excellent chemical homogeneity in the final product. Moreover, the molecular level mixing and the tendency of partially hydrolyzed species to form extended networks facilitate the structure evolution thereby lowering the crystallization temperature.

In the present work, the sinterability and formation of yttrium iron garnet  $Y_3Fe_5O_{12}$  (YIG) and iron aluminium garnet  $Fe_3Al_5O_{12}$  (IAG) powders by aqueous sol-gel processes are investigated. The microstructural evolution and magnetic features of synthesized compounds are also discussed.

## 2. Experimental

Two YIG samples having the same nominal composition,  $Y_3Fe_5O_{12}$  were synthesized by aqueous sol-gel method using different complexing agents in the gelation process. The gels were prepared using stoichiometric amounts of analytical-grade  $Y_2O_3$  and  $Fe(NO_3)_3 \cdot 9H_2O$  as starting materials. In the sol-gel process, yttrium oxide (0.001875 mol; 0.4238 g) was first dissolved in 100 mL of 0.2 M  $CH_3COOH$ . A clear solution of yttrium acetate was obtained after stirring for 10 h at  $65^\circ C$  in a beaker covered with a watch glass. To this solution, iron nitrate (0.00625 mol; 2.5250 g) dissolved in 75 mL of distilled water was added and the resulting mixture was stirred for

1 h at the same temperature. In a following step, 1,2-ethanediol (2 mL) (YIG – route 1) or tartaric acid (0.45 g) dissolved in 20 mL of distilled water (YIG – route 2) as complexing agents were added to the above solution.

The precursor gel for  $Fe_3Al_5O_{12}$  were prepared using stoichiometric amounts of analytical-grade  $Fe(NO_3)_3 \cdot 9H_2O$  and  $Al(NO_3)_3 \cdot 9H_2O$  as starting materials. In the sol-gel process, iron nitrate (0.00375 mol; 1.5150 g) was first dissolved in 100 mL of 0.2 M  $CH_3COOH$ . To this solution, after stirring for 1 h at  $65^\circ C$  in a beaker covered with a watch glass, aluminium nitrate (0.00625 mol; 2.3438 g) dissolved in 75 mL of distilled water was added and the resulting mixture was stirred for 1 h at the same temperature. In a following step, 1,2-ethanediol (2 mL) as complexing agent was added to the above solution.

After concentrating the solutions by slow evaporation at  $65^\circ C$  under stirring the Y-Fe-O and Fe-Al-O sols turned into brown transparent gels. The oven dried ( $110^\circ C$ ) gel powders were ground in an agate mortar and preheated for 2 h at  $800^\circ C$  in air. Since the gels are very combustible slow heating ( $\sim 3-4^\circ C \text{ min}^{-1}$ ) especially between 100 and  $400^\circ C$  was found to be essential. After an intermediate grinding in an agate mortar the powders were additionally sintered for 10 h at  $1000^\circ C$  in air. The aqueous sol-gel syntheses of YIG and IAG were repeated several times (about 8–10). It is significant to note that the proposed sol-gel preparation gave very reproducible results. The investigated properties of each YIG and IAG ceramic specimens were found to be almost identical.

The infrared (IR) spectra were recorded as KBr pellets on a Perkin-Elmer FTIR Spectrum BX II spectrometer. The X-ray powder diffraction (XRD) studies were performed on a Philips Xpert diffractometer operating with  $Cu K\alpha_1$  radiation. Scanning electron microscope (SEM) JEOL 820 was used to study the morphology and microstructure of the ceramic samples. The Mössbauer spectra were recorded at room temperature on a Wissel spectrometer equipped with  $^{57}Co/Rh$  source. The magnetic susceptibility behaviour of garnets was studied with a SQUID magnetometer (Quantum Design, MPMS) in the temperature region between 20 and 300 K using a static field.

## 3. Results and discussion

To obtain crystalline  $Y_3Fe_5O_{12}$  and  $Fe_3Al_5O_{12}$  phases the appropriate precursor gels were calcined and annealed at  $1000^\circ C$  in air. Fig. 1 shows the IR spectra of iron-containing ceramics obtained at  $1000^\circ C$ . IR spectra of the  $Y_3Fe_5O_{12}$  samples (see Fig. 1(a) and (b)) contain several intense peaks in the range of  $900-500 \text{ cm}^{-1}$  (889, 870, 845, 778, 755, 710, 591,  $537 \text{ cm}^{-1}$ ) which are due to the metal-oxygen vibrations in the ceramic samples. Moreover, according to Vaqueiro and Lopez-Quintela [1,17] these bands are characteristic for YIG and YAG structures and could be attributable to the stretching mode of the tetrahedral units present in the garnet structure. Thus, according to the stretching frequencies observed and the

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