

Gd₃Fe₅O₁₂ garnet ceramic with mesoscale structuring processed by slip casting. Effect of magnetic field

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ABSTRACT

This study reports the fabrication of Gd₃Fe₅O₁₂ ferrimagnetic ceramic by magnetic slip casting under applied magnetic fields of 1T and 12T, followed by solid-state sintering. Structural, microstructural, and magnetic properties were investigated using X-ray diffraction, scanning and transmission electron microscopy, Mössbauer spectroscopy, and magnetometry. XRD and SAED confirmed the presence of single-phase cubic garnet in the dense bulk ceramic with no detectable crystallographic texture. Rietveld analysis revealed relatively large crystallite sizes in the range of 165–350 nm. SEM observations showed that magnetic fields during magnetic slip casting, especially of high value (12T), promote the organization of local mesoscale grain assemblies with a preferential spatial arrangement, forming two-dimensional magnetically correlated regions. Magnetic measurements indicate that this mesoscale structure significantly affects the low-field magnetic behavior, as evidenced by pronounced ZFC-FC irreversibility and anisotropic magnetic response. Despite distinct magnetic relaxation patterns caused by microstructural differences, all magnetic slip-casted samples exhibit the same elevated compensation temperature ($\theta_C = 336$ K), which is primarily attributed to the large crystallite sizes achieved through the processing route. Comparison with the 0T slip-casted reference sample indicates that the enhanced compensation behaviour is primarily associated with the processing-induced local structural/microstructural state rather than directly with the applied magnetic field. These findings demonstrate that magnetic slip casting is a practical method for engineering magnetic properties in rare-earth iron garnet ceramics by designing their microstructure at the mesoscale.

1. Introduction

In recent years, rare-earth iron garnets (RE₃Fe₅O₁₂, where RE = rare earth), have attracted a significant interest in their exceptional magneto-optical properties, which make them suitable for various applications including spintronics, microwave devices, magnetic sensors, and magneto-optical recording media [1–5]. The cubic crystal structure of garnet (space group Ia-3d) is advantageous because its magnetic configuration, composed of three sublattices, enables tunable magnetic properties such as net magnetization and compensation temperature through the interplay between the sublattice moments. Two Fe³⁺ ions occupy the octahedral crystal site (16a), three Fe³⁺ ions occupy tetrahedral crystal site (24d) and the rare earth ions occupy the dodecahedral

crystal site (24c) with each rare earth ion being surrounded by eight oxygen ions [6]. Hence, the total magnetization of the RE₃Fe₅O₁₂ arises from the contribution of these three sublattices as $M_t = |M_a + M_c - M_d|$, where a, c, d describe the octahedral, dodecahedral and tetrahedral sites, respectively.

One critical feature of iron garnet systems is the magnetic compensation at critical temperature (θ_C), the point at which the net magnetization approaches zero due to compensation between the magnetic moments of different sublattices. This effect is useful in novel magnetic switching devices, currently attracting much attention [7,8]. Bulk Gd₃Fe₅O₁₂ exhibits the highest θ_C among all rare-earth iron garnets, which is close to room temperature [9]. This temperature is highly sensitive not only to composition, but also to local structural disorder,

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defect chemistry, crystallite size, and processing conditions, which can modify the Fe–O–Gd exchange interactions. There are significant challenges related to properties control and optimization depending on processing techniques and conditions. For instance, a higher value of θ_C (~ 377 K) was observed in $\text{Gd}_3\text{Fe}_5\text{O}_{12}$ thin films on Si (001) substrate grown by Pulsed Laser Deposition [4], but lower values of θ_C (220–230 K) were observed by $\text{Gd}_3\text{Fe}_5\text{O}_{12}$ thin films with perpendicular magnetic anisotropy fabricated by magnetron sputtering [10]. On nanoparticles of $\text{Gd}_3\text{Fe}_5\text{O}_{12}$ synthesized by the sol-gel method [11], a cancellation of magnetization was observed at a $\theta_C \sim 286.5$ K and a Curie Temperature (T_C) of 560 K in agreement with the bulk behavior. Different studies have also focused on the influence of doping with rare-earth [12] or transition metal ions [13] on the structural, magnetic, and magnetocaloric properties of the $\text{Gd}_3\text{Fe}_5\text{O}_{12}$ garnet. Furthermore, special properties were expected in high entropy garnets. For example, high-entropy rare-earth iron garnet ceramic synthesized by a solid-state method with composition $(\text{Gd}_{0.2}\text{Dy}_{0.2}\text{Er}_{0.2}\text{Y}_{0.2}\text{RE}_{0.2})\text{Fe}_5\text{O}_{12}$ (RE = Sm, Eu, Tm, Yb) have shown saturation magnetization of 12.29–13.27 emu/g and coercivity of 18.46–19.87 Oe, suggesting the possibility of tunable magnetism through compositional/structural/microstructural design [14].

Controlling the structural orientation of particles/crystallites in various materials, usually the anisotropic ones, is a well-known procedure for enhancing their functional properties. Magneto-processing is recognized as a powerful technique and it was successfully applied e.g. for texturing of alumina platelets coated with iron oxide under ultra-low magnetic fields [15], orientation of SmCo-based alloys during solidification [16], for stacking of barium ferrite particles in magnetic fields during slip casting [17] or for fabrication of oriented Ti_3SiC_2 ceramic with increased fracture toughness and thermal conductivity [18]. Magneto-processing has shown its versatility not only for inducing orientation but also provided new possibilities to obtain new materials with peculiar architecture, structural and microstructural details and performance. Magnetic slip casting is a popular magneto-processing technique. It emerged as a promising technology for fabricating textured ceramic materials with controlled microstructure, thereby significantly enhancing magnetic, mechanical, thermal, and other properties [19–23]. In slip casting, a ‘suspension’ such as a colloidal solution or a slurry is casted and dried under an external magnetic field. Next, the as-prepared textured green body is sintered. To ensure uniform dispersion of the powder in the liquid and its stability during casting, suitable binders and dispersing agents are added to the suspension [24–26]. One dispersant, polyethylenimine (PEI), is often used to stabilize various suspensions of SiC [27], $\text{Y}_2\text{Si}_2\text{O}_7$ [28] or Y_2O_3 [29]. The applied magnetic field during slip casting provides the torque $\vec{T} = \vec{M} \times \mu_0 \vec{H}_0$ rotating the particles, if magnetization vector $\vec{M}$ of the particles is not aligned with an external magnetic field H_0 . Torque from rotation aligns magnetic domains or particles in a preferred direction. For efficient orientation, particles should fulfill different conditions related to magnetic properties, size and shape, while the liquid environment should allow free particle rotation and movement [30,31]. Although magnetic slip casting was applied to different materials, we note that there are limited reports on ferrimagnetic magnetic materials [21], most of the work being done on paramagnetic and diamagnetic ceramic materials such as oxides, carbides, and borides, as mentioned before.

This work explores the novel fabrication of ferrimagnetic bulk $\text{Gd}_3\text{Fe}_5\text{O}_{12}$ garnet via slip casting and solid-state sintering. The impact of magnetic slip casting on magnetic properties in sintered ceramics is assessed, considering new opportunities for additional control over magnetic properties. A better understanding of the complex relationships among specific structural and microstructural aspects promises new developments and advances in the functionality of $\text{Gd}_3\text{Fe}_5\text{O}_{12}$, as well as of other rare-earth garnets for spintronic and other applications. It is noteworthy that, although the applied magnetic field during slip casting is considered the key technological parameter investigated in

this work, the resulting magnetic properties may also depend on other parameters or on their combination taking into account specifics of slip casting with and without magnetic field, pressing or not of the green bodies and sintering. Consequently, the interpretation of magnetic behavior requires consideration of the processing-specific influence on structural and microstructural evolution which in turn will impact magnetic functionality of the samples.

2. Materials and methods

2.1. Samples preparation

Powders of Gd_2O_3 (Fluka, 99.9%) and Fe_2O_3 (Fluka, 97%) were mixed for 30 min in an agate mortar and pestle system. The mixture was designed to obtain the stoichiometry of the garnet. In the next step, the powder mixture was uniaxially pressed and a compact disk with 10 mm in diameter was obtained. Solid state reaction method [32] was applied: the disk was heated in air at 1400 °C. The dwell time was 10 h. Heating and cooling rates were 5 °C/min [33]. The $\text{Gd}_3\text{Fe}_5\text{O}_{12}$ sintered bulk was crushed with a vibrational mill that includes a tungsten carbide ball and vial, for a processing time of 1 h. As-milled powder was passed through a 200-sieve mesh. This powder, composed of single phase $\text{Gd}_3\text{Fe}_5\text{O}_{12}$ as determined by X-ray diffraction (XRD), was dispersed in ethyl alcohol (purity 99.5%), adding an optimal amount of polyethylenimine (PEI, Mw = 10000, Wako Pure Chemical Industries, Ltd. Tokyo, Japan) dispersing agent (2% wt. of the $\text{Gd}_3\text{Fe}_5\text{O}_{12}$ powder weight). The resulting suspension, composed of 30% volume of $\text{Gd}_3\text{Fe}_5\text{O}_{12}$ was ultrasonicated and stirred for 6 h for deagglomeration. Then, solution was poured into a plastic cylinder (10 mm diameter) glued with vacuum grease on an absorbing porous alumina plate. The cylinder containing the solution with the garnet powder was processed at room temperature without an applied magnetic field (0T green) or it was placed inside a superconducting magnet (Japan Superconductor Technology, Inc. JMTD-12T100) charged with an applied magnetic field (H_0) of 1 or 12 T. The applied magnetic fields of 1 T and 12 T were selected to represent two distinct regimes of magnetic-field-assisted processing covering the entire range of the available fields for processing. This choice allows a comparative evaluation of moderate-field versus high-field processing conditions rather than a systematic field-dependent study. The direction of the applied magnetic field was parallel to the length of the cylinder with solution and to casting direction (CD) (Fig. 1(a)). We assume that for a relatively small cylinder diameter (10 mm in our case) magnetic field in the sample's volume was uniform. The resulting green body presented a columnar-like structure when applied field was 12T (columns along CD direction are indicated with blue lines in Fig. 1(a)). After 48 h of drying, sample was removed from the plastic cylinder. A uniaxial pressure (P_0) of 80 MPa was applied on the sample, parallel (Fig. 1(b)) or perpendicular (Fig. 1(c)) to CD direction. Samples were labeled: 12T_{||} (see Fig. 1(b)) and 12T_⊥ (see Fig. 1(c)). The columnar-like structure was not observed in the sample slip-cast under an applied magnetic field of 1T. This green sample was not subject to pressing step, and it was labeled 1T. A reference sintered sample processed without the uniaxial magnetic field application during slip casting and without the uniaxial pressing step was labeled 0T. Green compacts after slip casting and drying denoted 0T green and 12T green, obtained without uniaxial pressing, were also used to compare with the corresponding sintered ceramics to extract information regarding the influence of the uniaxial pressing step. In the last step of the processing, all four green compacts (0T, 12T_{||}, 12T_⊥, and 1T) were sintered in air at 1400 °C for 10 h.

2.2. Samples characterization

X-ray diffraction (XRD) patterns were collected at room temperature using an Anton Parr XRDynamic 500 diffractometer (Cu K α 1 radiation, $\lambda = 1.5406$ Å). The precise alignment of the diffractometer was confirmed against the Standard Reference Material (SRM) NIST1976c.

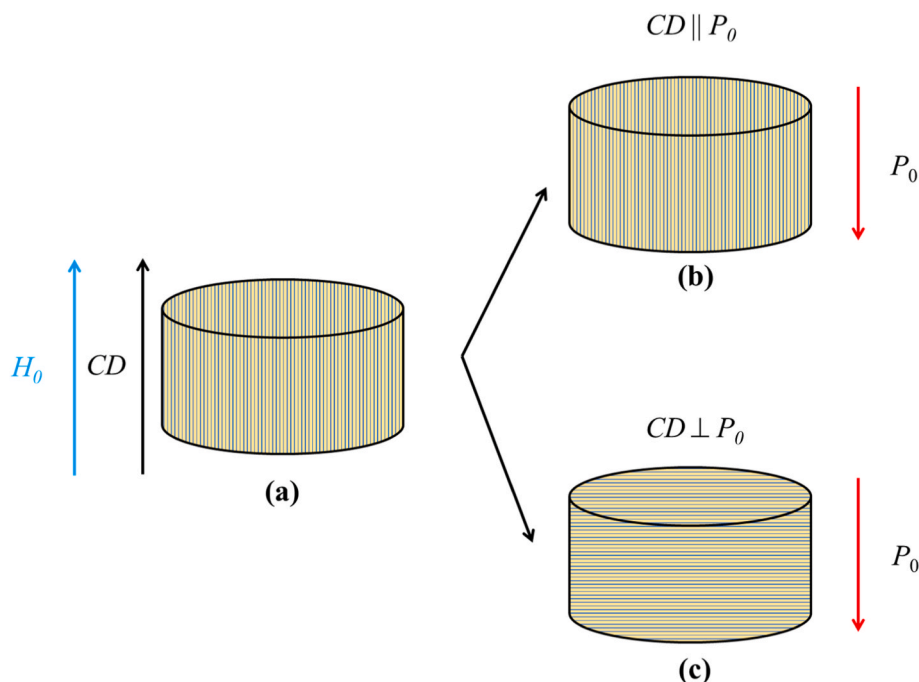


Fig. 1. Samples preparation: (a) - magnetic slip casting and drying; (b) and (c) - uniaxial pressing of green compacts. In (a), the applied magnetic field H_0 is perpendicular to the sample's surface, and it is also parallel to the casting direction (CD); In (b) and (c), uniaxial pressure P_0 is applied parallel and perpendicular to the slip casting direction (CD), respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

For phase identification, the ICDD Powder Diffraction database was used [34]. Rietveld analysis was performed with MAUD (Materials Analysis Using Diffraction) software version 2.98 to extract the structural parameters using an instrumental function obtained from the XRD data recorded on a sample of LaB_6 .

Scanning Electron Microscopy (SEM) observations and Energy-Dispersive X-ray spectroscopy (EDS) elemental analyses were performed with a TESCAN Lyra3 XMU Dual Beam Microscope equipped with a Bruker detector.

Transmission Electron Microscopy (TEM) investigations were carried out with a JEOL 2100 instrument (Japan). Samples for TEM observations were prepared by crushing the bulks into thin pieces. VESTA software was used for representing the structural models viewed from their respective local orientation determined with selected area electron diffraction in TEM (SAED).

Local magnetic configurations were investigated by temperature-dependent Mössbauer spectroscopy using a conventional constant-acceleration spectrometer (SEECO, USA) with a $^{57}\text{Co}(\text{Rh})$ radioactive source. Measurements were performed at different temperatures (7 K and 300 K) by inserting the sample into a closed-cycle cryostat (Janis, USA). Fitting of Mössbauer spectra was performed with NORMOS software [35]. Isomer shift values are reported relative to $\alpha\text{-Fe}$ at room temperature.

Hysteresis loops at different temperatures and field-cooling (FC), zero-field cooling (ZFC) curves were measured with a DynaCool PPMS-9T magnetometer (Quantum Design, USA). The measured samples were cubic in shape of $1 \times 1 \times 1 \text{ mm}^3$, being cut from the center of the sintered bulks.

3. Results and discussions

3.1. Structure and microstructure

XRD patterns confirmed that all sintered bulk samples crystallized in a single phase with a cubic garnet structure ($\text{Gd}_3\text{Fe}_5\text{O}_{12}$, space group $Ia\bar{3}d$), with no detectable secondary phases (Fig. 2, (a)- sample 12T $_{||}$, (b)-

sample 12T $_{\perp}$, (c)- sample 1T, and (d)- sample 0T). The XRD patterns (Fig. 2(a–c)) exhibit sharp and intense peaks with a low background, indicating a high level of samples crystallinity.

Rietveld refinements were carried out using the standard garnet structural model, with Gd^{3+} on the 24c dodecahedral sites, Fe^{3+} distributed over the 16a (octahedral) and 24d (tetrahedral) sites, and oxygen on the 96h positions. They revealed low-weighted-profile residuals ($R_{\text{wp}} = 6.4\text{--}7.3$, Table 1), demonstrating excellent agreement between the calculated and experimental patterns. The refined lattice parameters show a very narrow range around the expected value for stoichiometric $\text{Gd}_3\text{Fe}_5\text{O}_{12}$ ($a \approx 12.45 \text{ \AA}$, Table 1), and the differences between samples obtained by conventional solid state reaction route reported in literature [32,36] and those processed by magnetic slip casting from this work are within the refinement uncertainties. Nevertheless, there are some differences between samples fabricated by the two routes. In Ref. [32], the crystallite size was around 119 nm, and the microstrain was 1×10^{-4} . For our samples, a large crystallite size of 165–353 nm was estimated (Table 1), and the microstrain of $2.1\text{--}2.3 \times 10^{-4}$ remains in the same order of magnitude. In addition to crystalline coarsening, results indicate that slip casting does not alter the composition or structure of the garnet phase. Importantly, no XRD evidence of preferred structural orientation (texture) was observed in samples processed by slip casting under a magnetic field.

Selected-area electron diffraction (SAED) in TEM further confirms the high crystal quality of the magnetic slip-cast ceramics (Fig. 3). TEM images show grains with sharp corners and often straight edges. Grain boundaries in the upper agglomerate from Fig. 3(a) are well defined, and particle size is around 200–500 nm. Some objects with the appearance of large grains are present, and they have a high degree of structural coherence without amorphous phases (Fig. 3 a1–3, b4, c5). Presented aspects, reinforce XRD and Mössbauer (see Section 3.2) observations, namely that the magnetic slip casting route followed by sintering promotes grain growth without compositional and structural modifications. SAED patterns correspond to distinct orientations such as $[320]$, $[751]$, and $[331]$ of the cubic $\text{Gd}_3\text{Fe}_5\text{O}_{12}$ garnet structure (space group $Ia\bar{3}d$). For the observed orientations, simulated crystal structures are presented

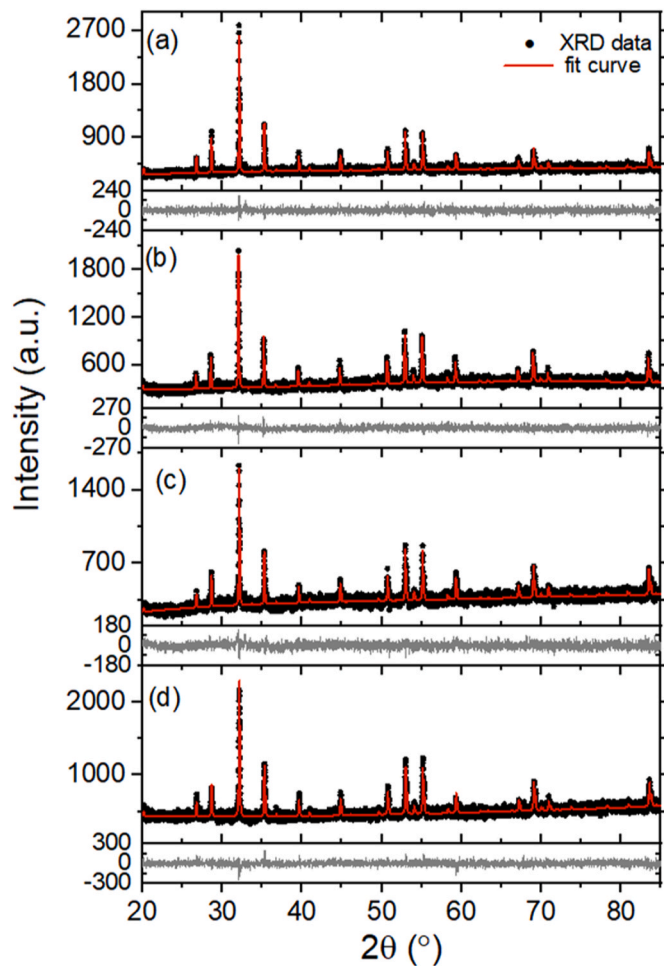


Fig. 2. Rietveld refinements of XRD patterns for $\text{Gd}_3\text{Fe}_5\text{O}_{12}$: (a) $12\text{T}_{||}$, (b) $12\text{T}_{\perp}$, (c) 1T , (d) 0T .

Table 1

Structural parameters obtained from Rietveld refinement are presented in Fig. 2: lattice parameter a , crystallite size (D_{eff}), lattice micro strain $\langle \epsilon^2 \rangle^{1/2}$ and the calculated R-factor, R_{wp} .

Sample	Structural parameters
$12\text{T}_{ }$	$a = 12.458 \pm 1 \cdot 10^{-3} \text{ \AA}$ $D_{\text{eff}} = 353 \pm 9 \text{ nm}$ $\langle \epsilon^2 \rangle^{1/2} = 2.4 \cdot 10^{-4} \pm 1 \cdot 10^{-5}$ $R_{\text{wp}} (\%) = 7.05$
$12\text{T}_{\perp}$	$a = 12.458 \pm 1.2 \cdot 10^{-3} \text{ \AA}$ $D_{\text{eff}} = 233 \pm 4 \text{ nm}$ $\langle \epsilon^2 \rangle^{1/2} = 2.1 \cdot 10^{-4} \pm < 1 \cdot 10^{-5}$ $R_{\text{wp}} (\%) = 7.26$
1T	$a = 12.457 \pm 1.5 \cdot 10^{-3} \text{ \AA}$ $D_{\text{eff}} = 251 \pm 6 \text{ nm}$ $\langle \epsilon^2 \rangle^{1/2} = 2.3 \cdot 10^{-4} \pm < 1 \cdot 10^{-5}$ $R_{\text{wp}} (\%) = 7.3$
0T	$a = 12.450 \pm 1.4 \cdot 10^{-3} \text{ \AA}$ $D_{\text{eff}} = 165 \pm 3 \text{ nm}$ $\langle \epsilon^2 \rangle^{1/2} = 2.3 \cdot 10^{-4} \pm < 1 \cdot 10^{-5}$ $R_{\text{wp}} (\%) = 6.4$

in Fig. 4. To arrive from one orientation to another, rotation in the ac-plane around b -axis with relatively small angles of 6.9 and 11.4° is required. Thus, a local misalignment of the crystalline domains in the large objects is likely. Some alignment, only between large grains, is also possible, and this idea will be considered and explored by SEM observation in the next paragraph. Other than these details, no defects were

revealed, and we shall note that XRD could not detect texture, and the samples are structurally randomly oriented.

SEM images taken on the surface (Fig. 5(a)) of sample 1T processed without the pressing step show mesoscale agglomerates, some of which consist of elongated grains stacked approximately parallel to one another. Images of the cross-section (Fig. 5(b)) reveal grains with flat surfaces, irregular shapes, and clean, distinct grain boundaries. Smaller grains with irregular morphology are also present. All magnetic slip-casted samples display microstructures characterized by well-developed, micrometer-sized grains with clear shapes, flat surfaces, and grain boundaries (Fig. 5(c–f)). In samples $12\text{T}_{||}$ and $12\text{T}_{\perp}$, there are subtle differences in how local mesoscale grain clusters are arranged. This means that uniaxial pressure P_0 (Fig. 1) can either maintain or disrupt the mesoscale grain assemblies, depending on its orientation relative to the CD. It is noteworthy that these mesoscale agglomerates with elongated morphology, which repeatedly appear across the samples, are apparently related to local particle arrangements established during magnetic slip casting. This SEM observation is convergent with results inferred from TEM. However, their characteristics, such as size and packing, depend on the magnitude of the applied field during slip casting and on the pressing direction relative to the CD. In contrast, the 0T reference sample exhibits a substantially more homogeneous randomly organized microstructure and does not show the elongated mesoscale grain assemblies observed in the magnetic-field-processed ceramics (Fig. 5g and h). Instead, the microstructure is dominated by grains without evident preferential mesoscale organization. This comparison suggests that the elongated mesoscale objects observed in the 1T and 12T samples originate during magnetic-field-assisted slip casting and are subsequently retained, at least partially, after sintering. Although the subsequent sintering treatment at 1400°C promotes grain growth, no crystallographic texture is detected by XRD (Fig. 2). Magnetic slip-casted samples ($12\text{T}_{||}$, $12\text{T}_{\perp}$, 1T) can be viewed as being composed of mesoscale objects (agglomerates) with a possible degree of orientation (Figs. 3 and 4) in a randomly oriented matrix. During sintering, the two components may behave differently, but some specific features of the microstructure from the green bodies are partially preserved in the sintered ceramic.

In summary, although there is no macroscopic texture, samples processed by magnetic slip casting exhibit a mesoscopic structure, which is expected to influence their magnetic behavior.

3.2. Mössbauer spectroscopy and magnetic properties

To obtain a highly localized view of the Fe^{3+} atomic environment, temperature dependent Mössbauer spectra were collected at 7 and 300 K for all three $\text{Gd}_3\text{Fe}_5\text{O}_{12}$ ceramic samples (Fig. 6). The hyperfine parameters (B_{hf} - hyperfine magnetic field, IS - isomer shift, ϵ - quadrupole correction) as well as the intensity ratio A_{23} (between the second and third absorption lines) and the relative area of spectral components as extracted from the fits are summarized in Table 2. Spectra present only sextet patterns, thus confirming that garnet ceramic samples are entirely magnetically ordered not only at 7 K , but also at 300 K . We note here the extensive debate on fitting the Mössbauer spectra with respect to the most convenient decomposition in the corresponding sextets. A complex fitting algorithm was provided by G. J. Long and F. Grandjean [36,37] assuming implicitly a reduction from the cubic to a rhombohedral symmetry. Accordingly, 3 sextet components arise from the octahedral sites, and 4 sextet components arise from the tetrahedral sites (all consistent with substantial quadrupole splitting values and specific orientations of the hyperfine magnetic field versus the crystalline axis). However, if the cubic symmetry is strictly considered, the usual procedure is for a decomposition into 2 sextet components at room temperature (RT) and 2 or 3 sextet components at low temperatures where magnetic relaxation phenomena are disregarded [32,38–40]. If 3 sextets are considered in the Mössbauer spectra of low temperatures, which are the most conclusive for the investigation of the local configurations, two

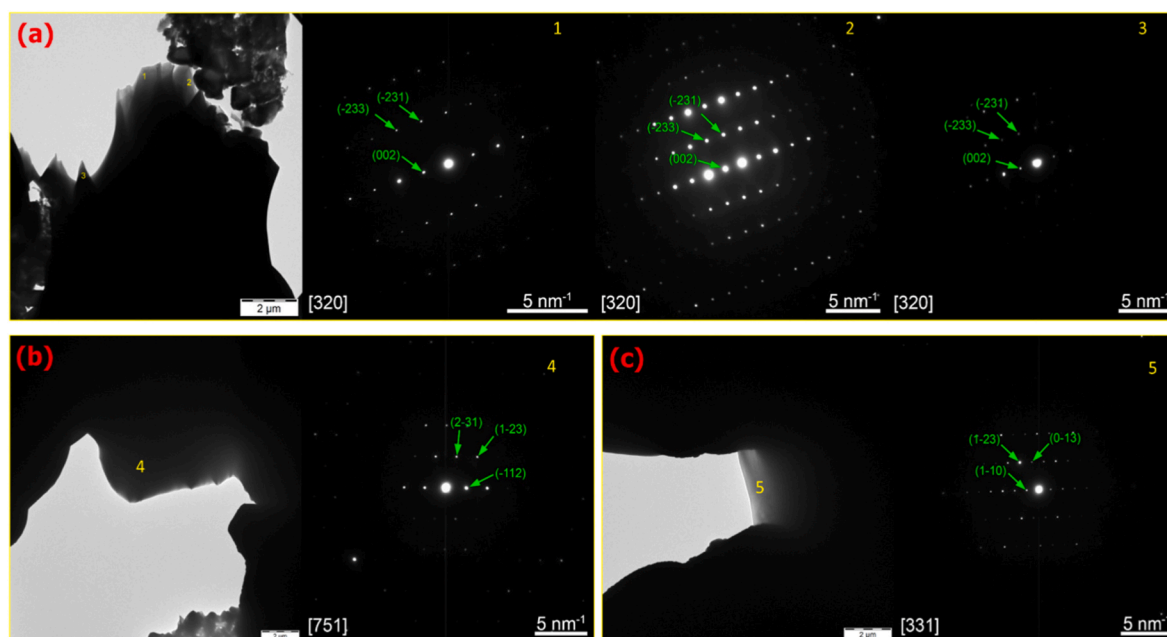


Fig. 3. TEM images of 12T1 sample on different grains (a, b, c) and their associated SAED patterns (1-5).

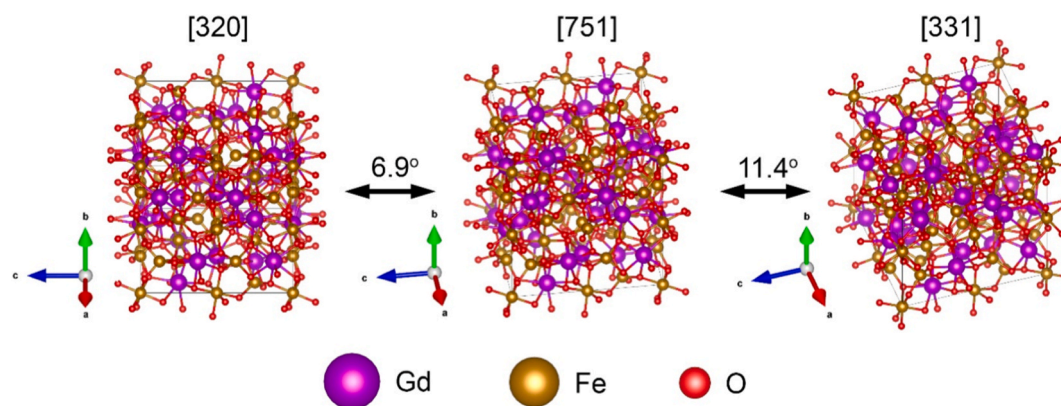


Fig. 4. Simulated crystal structure of $\text{Gd}_3\text{Fe}_5\text{O}_{12}$, and its orientation based on SAED patterns of 12T1 illustrated in Fig. 3 (a1-3: left, b4: middle, c5: right).

of them with a major contribution of the relative area and with hyperfine parameters specific to Fe^{3+} ions are safely assigned to the octahedral (16a) and tetrahedral (24d) positions, respectively. On the other side, the third, substantially less intense sextet component also assigned to Fe^{3+} ions, might have different origins, depending on sample composition and preparation conditions. This idea is supported by both the corresponding relatively spread values of hyperfine parameters and some related properties, as for example the compensation temperature for a same type of compound. However, due to its preponderant collapsing at room temperature (RT) over the sextet belonging to the octahedral Fe^{3+} ion, it might be attributed to either an additional octahedral Fe^{3+} configuration or to Fe^{3+} ions in the dodecahedral position specific to the RE ions. Although the last attribution of Fe^{3+} ions on the dodecahedral sites is less usual, it was supported by both theoretical [41] and experimental [32,42] arguments. For example, in Ref. [32], the hypothesis of the dodecahedral site occupancy by Fe^{3+} ions, as supported by the specific Mössbauer parameters of the third sextet at 7 K, was fully consistent with both XRD investigations and magnetic measurements. Moreover, it provides a reasonable explanation for the unexpected increased compensation temperature through straightening the Fe-Gd exchange interactions. Therefore, for the present compounds characterized by significantly higher compensation

temperatures as compared to the compounds with ideal site occupation (as it will be shown in the following), the treatment of the low temperature Mössbauer spectra will be based on the above-mentioned attribution for the third less intense Mössbauer sextet.

Firstly, we shall address the 7 K Mössbauer spectrum of sample OT. This was fitted by 3 sextet components. More specifically, according to the previous literature, the most external sextet with the largest hyperfine magnetic field (B_{hf}) of 55.13 T and with an isomer shift (IS) of 0.47 mm/s (27.7% of the relative spectral area) was clearly assigned to the octahedral positions whereas the most internal sextet with the lowest hyperfine magnetic field of 48.32 T and IS of 0.26 mm/s (61.8% of the relative spectral area) was assigned to the tetrahedral positions of the garnet structure. The less intense sextet (10.5% relative area) with an intermediate B_{hf} of 53.3 T and IS values of 0.57 mm/s (i.e. slightly higher than for the octahedral coordination) was assigned based on the arguments presented in Refs. [32,40] to Fe^{3+} on the dodecahedral positions. However, the relatively high absolute value of the quadrupole correction of 0.15 mm/s (as compared to the almost null value on octahedral and tetrahedral sites) suggests the presence of rather defect structures with 7 neighboring O ions surrounding the central Fe^{3+} ion on the dodecahedral position.

At low temperatures (7 K, Fig. 6-left), all Mössbauer spectra collected

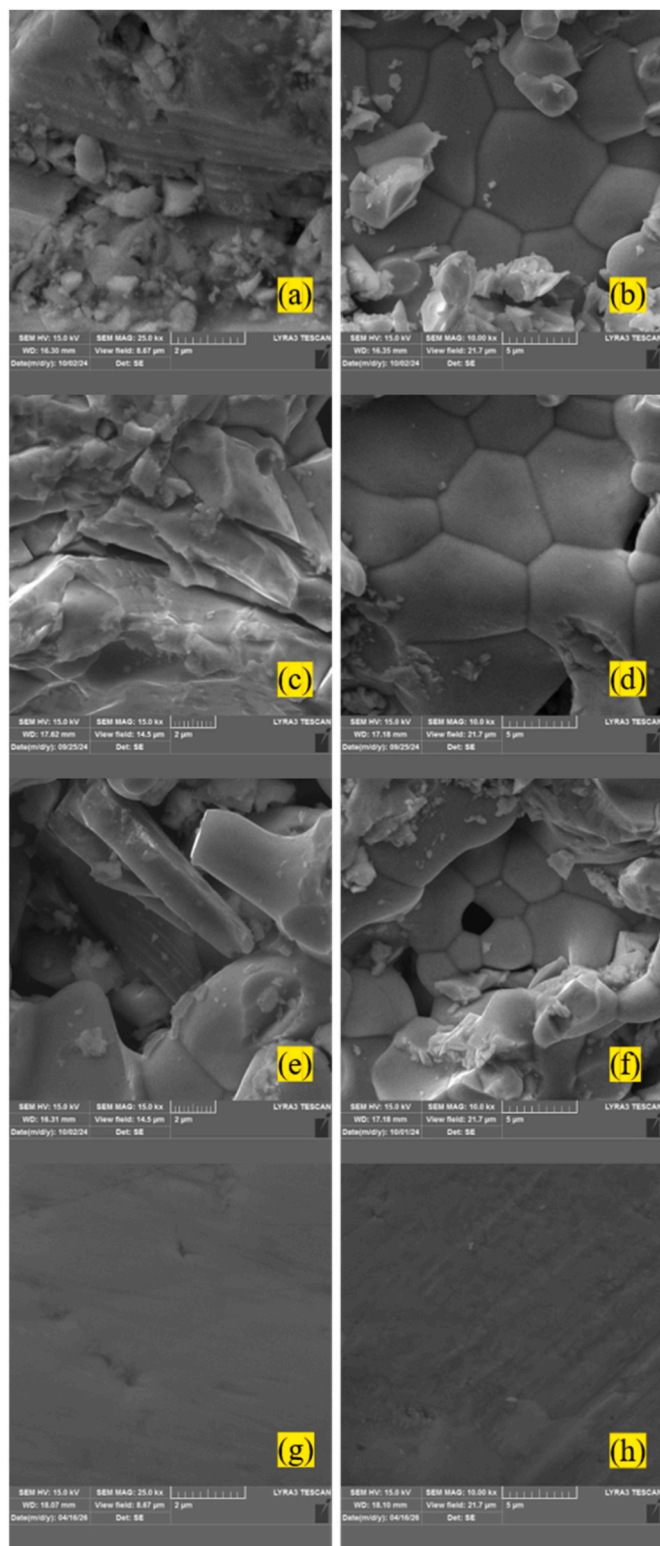


Fig. 5. SEM images of $\text{Gd}_3\text{Fe}_5\text{O}_{12}$ ceramics processed via magnetic slip casting: (a, b) 1T sample (a-surface, b-cross section); (c, d) $12\text{T}_\perp$ sample (c-surface, d-cross section); (e, f) $12\text{T}_\parallel$ sample (e-surface, f-cross section); (g, h) 0T sample (g-surface, h-cross section). CD is perpendicular to the sample surface.

on samples obtained by slip casting in magnetic field are well reproduced using three magnetic sextets, consistent with Fe^{3+} ions occupying the octahedral (16a), tetrahedral (24d), and a third minor spectral component associated by analogy with previous results also to Fe^{3+}

occupying the rare-earth dodecahedral (24c) sites of the garnet lattice. For each sample, the first sextet exhibits the largest hyperfine magnetic field (B_{hf}) of 54.76, 54.93, and 54.94 T and an isomer shift (IS) of 0.53, 0.50, and 0.52 mm/s for samples 1T, $12\text{T}_\parallel$ and $12\text{T}_\perp$, respectively. This sextet is characteristic of high-spin Fe^{3+} in octahedral coordination (Fig. 6-left, blue; Table 2). Its relative spectral area is in the range from 34 to 37%. The second sextet, associated with tetrahedral Fe^{3+} (Fig. 6-left, cyan; Table 2), shows a lower hyperfine field (B_{hf} of 48.18 T for $12\text{T}_\parallel$ and 1T and 48.28 T for $12\text{T}_\perp$) compared to the first components, an isomer shift (IS) of 0.26–0.27 mm/s, and it has a negligible quadrupole shift. Associated spectral area is in the range of 57 to 60%, being in good agreement with the occupation of the tetrahedral positions in the garnet structure. The third, i.e. weakest sextet (Fig. 6-left, green; Table 2), with a relative area in the range of 6 to 7%, is observed in all samples, and is attributed, in agreement with the previous discussion, to Fe^{3+} occupying the rare-earth (24c) site. This component shows a distinct local environment and presents intermediate values of $B_{\text{hf}} = 53.8\text{--}54.1$ T and $\text{IS} = 0.27\text{--}0.29$ mm/s. It is clear that the IS value of this component for the samples processed by slip casting in applied field are significantly lower than for the sample obtained by slip casting without applied field. The same applies with the values of quadrupole corrections which are all close to 0 for the sample obtained by slip casting without field, proving a much symmetrical anion configuration at the 24c site in the case of samples obtained by slip casting in magnetic field. In this context, the only explanation for lower IS values of Fe^{3+} on this site is related to larger Fe–O distances as compared to the sample obtained by slip casting without applied field in such a way to increase slightly the amount of 4s electrons of the cation [43]. The spectral linewidths of all sextets are in the range 0.34–0.38 mm/s, suggesting a uniform local environment for Fe^{3+} ions and quite good crystallization, especially for $12\text{T}_\parallel$ and $12\text{T}_\perp$ ceramics.

At nearly room temperature (300 K, Fig. 6-right), the Mössbauer spectra of $\text{Gd}_3\text{Fe}_5\text{O}_{12}$ ceramics remain magnetically split and are described by using two magnetic sextets corresponding to Fe^{3+} ions at the octahedral (Fig. 6-right, green) and tetrahedral (Fig. 6-right, cyan) sites. In contrast to the 7 K Mössbauer spectra (Fig. 6-left), the minor Fe^{3+} spectral component associated with Fe^{3+} in the dodecahedral (24c) site is no longer present as a separate sextet (Table 2). Instead, its contribution is mainly superposed over the more intense spectral component assigned to Fe^{3+} octahedral sites (Fig. 6-right, green, Table 2). The octahedral Fe^{3+} (16a) site gives rise at 300 K to a spectral component (Fig. 6-right, green) characterized by B_{hf} of 48.59–49.04 T and IS of 0.37 mm/s across all samples, while the spectral component of the tetrahedral Fe^{3+} (24d) (Fig. 6-right, cyan) shows B_{hf} of 39.8–40.1 T with a lower IS of 0.15 mm/s. The relative areas in the range of 40–42% for the sextet assigned to the octahedral sites and 58–60% for the sextet assigned to the tetrahedral site are close to the expected 2:3 ratio in the garnet structure. The linewidths of the 300 K sextets are larger (0.43–0.48 mm/s) than those of the sextets at low temperature (0.34–0.38 mm/s) due to magnetic relaxation phenomena, whereas the linewidths of the sextet belonging to the octahedral site are always 5% larger than those of the tetrahedral site because of the overlapped components in the first case.

The temperature dependence of magnetization measured under an applied field of $H = 300$ Oe (Fig. 7) unveils the characteristic ferromagnetic behavior of $\text{Gd}_3\text{Fe}_5\text{O}_{12}$ ceramics from 5 K up to 350 K, as obtained from ZFC–FC protocols. At low temperatures (5 K), the magnetization of all samples is the highest because the Gd^{3+} sublattice provides the dominant contribution to the net magnetic moment. This behavior was expected, consistent with the increased Gd moment ($3 \times 7 \mu\text{B}$) at low temperature in the garnet structure [44]. The magnetization hierarchy among the investigated samples (Fig. 7) correlates with the microstructural and morphological features addressed earlier. It is governed not only by crystallite size, but also by the spatial organization of the mesoscale assemblies observed by SEM. In the ceramic processed by slip casting under 1T, the grain organization forms

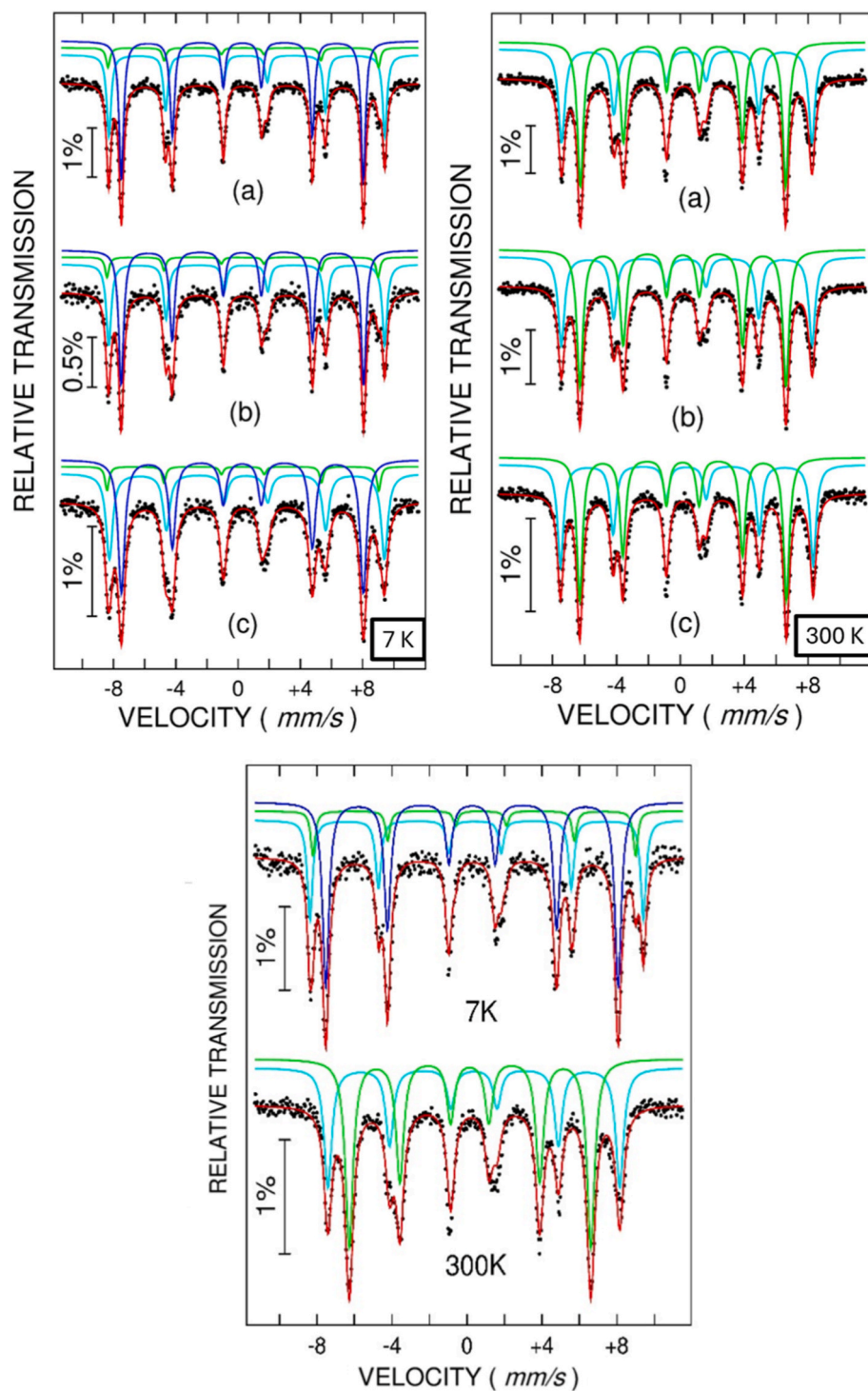


Fig. 6. Mössbauer spectra at 7 K (left) and 300 K (right) of 12T_{||} (a), 12T_⊥ (b), 1T (c). The image at the bottom shows Mössbauer spectra at 7 K and 300 K for the sample obtained in 0T.

a three-dimensional (3D) interconnected network with no preferred orientation, resulting in an isotropic magnetic response. Although these 3D mesoscale grain assemblies act as correlated magnetic regions, their random distribution results in reduced magnetization along the field direction. In contrast, the application of a strong magnetic field (12T) during slip casting promotes the formation of a two-dimensional (2D) mesoscale structure, resulting in magnetically correlated grain assemblies with a dominant planar organization. These elongated mesoscale agglomerates behave as magnetic entities with stronger ZFC-FC

irreversibility (Figs. 7 and 12T_{||} (black), 12T_⊥ (red)). Among the 12T samples, the perpendicular pressing geometry (sample 12T_⊥) preserves the planar organization more efficiently, explaining the highest magnetization observed (the magnetic field is applied along the casting direction, i.e., along the mesoscale grain assemblies). These results can indicate that the reduction of the elongated mesoscale objects dimensionality from 3D to 2D can be a microstructural component controlling the magnetic response, especially in low magnetic field, i.e., far from saturation.

Table 2

Magnetic hyperfine parameters (B_{hf} , IS, ε), the A23 intensity ratio, and relative area of the spectral components obtained by fitting the Mössbauer spectra from Fig. 6. IS is given relative to α -Fe at room temperature.

Sample	Temperature (K)	B_{hf} (T)	IS (mms^{-1})	ε (mms^{-1})	Area (%)	A23
0T	7	55.13	0.47	0.04	27.7(5)	2.07(5)
		48.32	0.26	0	61.8(5)	
		53.33	0.57	-0.15	10.5(5)	
	300	48.25	0.37	0	39.0(5)	1.95(5)
1T	7	34.94	0.15	0.01	61.0(5)	1.97(5)
		54.76	0.53	0.05	37.3(5)	
		48.18	0.26	0	56.7(5)	
	300	54.10	0.29	0	5.9(5)	2.05(5)
12T	7	49.04	0.37	0.03	42.5(5)	
		40.14	0.15	0.02	57.4(5)	
		54.93	0.50	0.08	34.2(5)	
	300	48.18	0.26	0	58.9(5)	2.06(5)
12T _⊥	7	53.84	0.29	0.03	6.8(5)	
		48.59	0.37	0.03	41.1(5)	
		39.88	0.15	0.01	58.8(5)	
	300	54.94	0.52	0.04	34.8(5)	2.01(5)
12T _⊥	7	48.28	0.26	0	58.5(5)	
		53.96	0.27	0	6.6(5)	
		48.67	0.37	0.03	40.3(5)	
	300	40.01	0.15	0.01	59.6(5)	2.08(5)

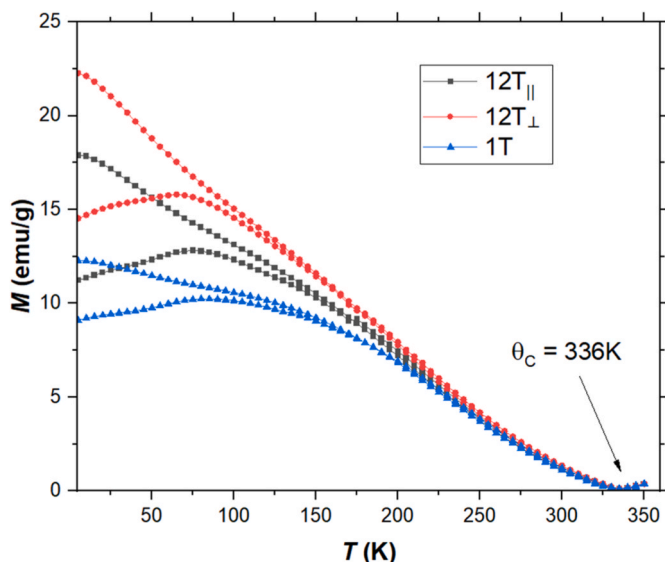


Fig. 7. ZFC-FC thermomagnetic curves for 12T_{||} (black), 12T_⊥ (red), and 1T (blue) under a magnetic field of $H = 300$ Oe applied along the casting direction. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

As the temperature increases, the magnetization of all samples decreases. This is a consequence of the antiferromagnetic coupling between the Gd and Fe magnetic sublattices, with the Gd sublattice relaxing much faster than the Fe sublattice. As stated in the Introduction section, a distinctive characteristic of rare earth iron garnets is the compensation temperature (θ_C), where the magnetization of Gd³⁺ at the 24c sites is equal and opposite to the magnetization of Fe³⁺ sublattice at the 16a and 24d sites, resulting in a net zero magnetization. Note that at the lowest temperature, the net magnetization of the Fe sublattices is $-5\mu_B$, and it relaxes much slower than that of the Gd one with an opposite orientation. Although the ZFC-FC curves of the three samples (Figs. 7 and 12T_{||} (black), 12T_⊥ (red), and 1T (blue)) show visible differences at low temperatures, they all present the same value of $\theta_C = 336$ K (Fig. 7), which is significantly higher than the θ_C values reported for bulk Gd₃Fe₅O₁₂ in the range of 290–317 K [32,45,46]. After the compensation

temperature, the Fe³⁺ sublattice becomes dominant and the net magnetization experiences a sign reversal. Up to 350 K, the magnetization of all samples remains close to zero, consistent with the much lower Gd³⁺ contribution and the increased relaxation of the Fe³⁺ sublattice. A detailed analysis of these effects and their connections to microstructural features will be presented. To evaluate whether the post-casting densification process occurring during pressing and/or sintering influences compensation temperature, additional ZFC-FC measurements were performed on the green compacts 0T green and 12T green, i.e. on material prior to pressing and sintering (Fig. 1, Supplementary). Compensation temperature and magnetic behavior around it are essentially unchanged after pressing and sintering. The 0T green compact and the corresponding sintered 0T ceramic exhibit the same compensation temperature of 363 K, while the 12T green compact shows a compensation temperature of 332 K which is close to the value of 336 K observed for the pressed and sintered 12T ceramics. Our analysis indicates that the pressing and sintering steps do not significantly modify the magnetic exchange interactions responsible for the compensation behavior.

The field-dependent magnetization curves up to 6T measured at 5K, 325K, and 350K, collected on all ceramics with the applied magnetic field along CD (Fig. 8), confirm the expected evolution, consistent with ferrimagnetic ordering in the Gd₃Fe₅O₁₂ ceramics across and above the compensation region. At 5K (Fig. 8, black curves), samples exhibit typical soft ferrimagnetic behaviour, opening hysteresis loops of low coercive fields and with rapid approach to saturation. These magnetization curves can be interpreted in terms of subtle differences in crystallinity and the distribution of local crystallite/grain orientations among the three samples. The slightly larger $M_S = 102 \text{ emu g}^{-1}$ in the 12T_⊥ sample suggests a more favorable statistical alignment of magnetic moments within crystallites/grains than in 1T, which displays the lowest saturation magnetization, $M_S = 91.5 \text{ emu g}^{-1}$. However, only 12T_{||} and 12T_⊥ display very low coercive fields H_C of about 15(3) Oe, whereas 1T shows a significantly larger coercivity ($H_C = 171(5)$ Oe, Fig. 8(c)). At 325 K (Fig. 8, red curves), close to the compensation temperature ($\theta_C = 336$ K) identified in the thermomagnetic curves presented in Fig. 7, the magnetization values in all samples are strongly reduced across the entire magnetic field range up to 6T, showing the near cancellation of the antiparallel Gd-Fe sublattice moments. In contrast to the low-temperature behaviour observed earlier, the coercive fields become almost identical for the three samples (12T_{||}, 12T_⊥, and 1T), with H_C values in the range of 120(5) Oe. This indicates that magnetization reversal near θ_C is controlled mainly by the Gd-Fe sublattices and it is no longer dominated by microstructural defects. At 350 K (Fig. 8, blue curves), above θ_C , the net magnetization changes, which is consistent with the weakening of the Gd contribution and the increasing contribution of the Fe sublattice at elevated temperature. Furthermore, the coercive fields decrease at 350 K to 70(2) Oe in all samples.

The clearest insight into the origin of the magnetic enhancement is provided by Mössbauer spectroscopy, which reveals an anomaly at the Gd³⁺ (24c) sites, where, in the case of the sample obtained by slip casting without field, almost 10% of the sites are partially substituted by Fe³⁺ ions from the octahedral (16a) sites, similar to the case of samples obtained by solid state reaction [32]. The Fe³⁺ ions on the Gd positions will be denoted as Fe³⁺(c) in the following section. However, for this sample, the IS values of the Fe³⁺(c) positions are relatively higher than for the sample obtained by solid state reaction (0.57 mm/s as compared to 0.42 mm/s), suggesting a lower density of 4s electrons, and hence a lower Fe-O distance assuming the same number of oxygen anions [43]. It should be noted that this also occurs with Fe³⁺ ions on 16a and 24d positions, and hence lower cation-anion distances are suggested for each position, in agreement with stronger Fe-O-Gd interactions and a higher compensation temperature for the sample obtained by slip casting without applied magnetic field. On the other hand, in case of the samples obtained by slip casting in the presence of the applied magnetic field, the IS values of the Fe³⁺(c) ions decrease substantially down to

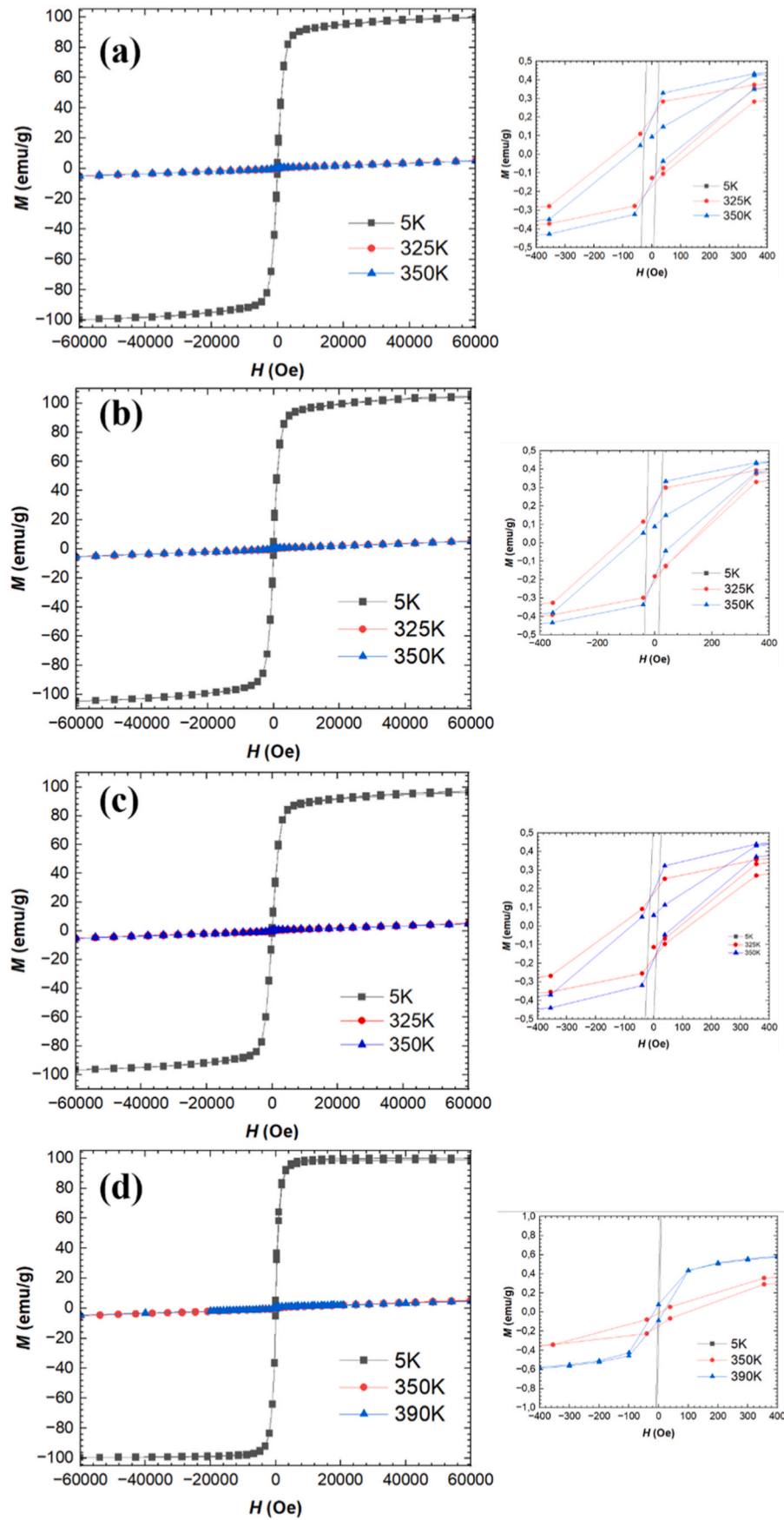


Fig. 8. $M(H)$ hysteresis loops collected at different temperatures (5K, 325K and 350K) for a magnetic field of 6T for 12T_{||} (a) 12T_⊥ (b), 1T (c) and 0T (d). The insets show enlarged views of the low-field region around the coercive field, highlighting the hysteresis behavior near magnetic compensation.

0.27–0.29 mm/s (Table 2) at low temperature (7 K), supporting larger Fe–O distances as compared to the sample obtained without applied field. Hence, weaker Fe–O–Gd interactions are expected for samples obtained by slip casting in the presence of the applied magnetic field as compared to the case of the sample obtained without magnetic field, this being in close agreement with the experimental observations of the relatively lower compensation temperatures. In order to check if the minor Fe^{3+} component resolved by Mössbauer spectroscopy at low temperature could be validated within a crystallographic model, additional Rietveld refinements were performed by introducing partial Fe deficiency on the tetrahedral site (24d), following the relative areas obtained from Mössbauer analysis (Table 2). For the 1T sample, the refined occupancies remain essentially unchanged, while for the 12T samples, the tetrahedral site occupancy converges to values close to 0.88. However, in all cases, this modification results only in a marginal increase in the weighted profile factor R_{wp} of around 0.1%, without a statistically meaningful improvement of the fit quality. This highlights a limitation of the XRD technique in resolving local site disorder in complex oxides, in contrast to the powerful Mössbauer spectroscopy technique.

In particular, the mesoscopic organization, as evidenced by SEM, provides a plausible structural origin for the observed anisotropic magnetic response. The hysteresis loops measured at 5 K for the 12T_{||} ceramic in the two measurement geometries (Fig. 9), with the applied magnetic field of 6T parallel to the casting direction (g1, Fig. 9, black line) or perpendicular to it (g2, Fig. 9, red line), clearly evidence magnetic anisotropy induced by the processing route. The slight differences between the two loops demonstrate that the mesoscale grain assemblies formed during magnetic slip casting are modifying how the material responds to an external magnetic field. In the geometry where the field is parallel to the dominant orientation (g1, Fig. 9, black line), the loop exhibits a slightly higher magnetization at intermediate fields compared with the perpendicular geometry (g2, Fig. 9, red line). This indicates that the mesoscale elongated grain clusters provide a preferred pathway for moment rotation and alignment. On the other hand, when the field is applied perpendicular to these structures, the loop takes longer to reach saturation, reflecting the additional energy required to rotate moments out of their alignment plane.

The field dependence of the ZFC–FC curves (Fig. 2, Supplementary)

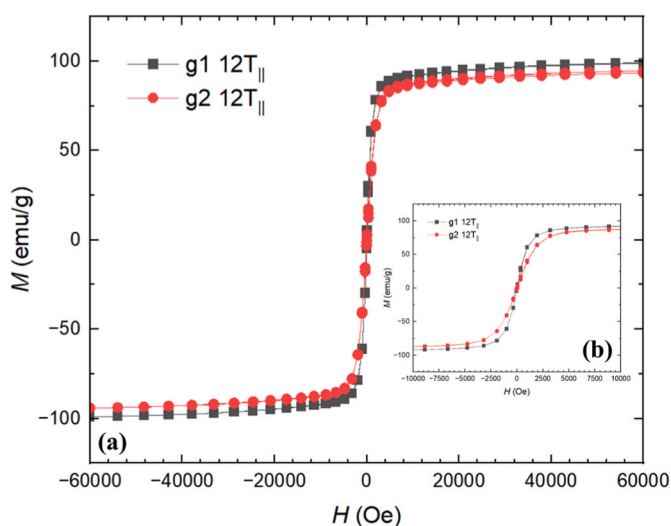


Fig. 9. (a) $M(H)$ hysteresis loops collected at 5 K in two geometries (g1: alongside the casting direction, dark curve; g2: perpendicular to the casting direction, red curve) for a magnetic field of 6T for 12T_{||} sample; (b) Inset: magnified view of the low-field (± 1 T) region. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

further highlights the role of the mesoscale organization induced by magnetic slip casting. For the 12T_{||} sample, low applied fields of $H = 300$ – 1000 Oe (Fig. 2, Supplementary, black and red curves) result in a pronounced separation between ZFC and FC curves over a large temperature interval. This behaviour indicates the presence of magnetic regions with preferential local orientation, associated with the identified mesoscale grain assemblies. In the ZFC protocol, the magnetic moments of these clusters are frozen in random orientations at low temperature, and the low applied field is insufficient to overcome the magnetic anisotropy. On the other hand, during the FC process, the magnetic field influences the orientation of these magnetically anisotropic clusters, resulting in a higher remanent alignment and a larger magnetization at low temperatures. As the applied field is increased to $H = 2$ T (Fig. 2, Supplementary, blue curve), the ZFC and FC curves nearly overlap in the entire temperature range.

Thus, while microstructure controls the magnitude and relaxation strength of the magnetic response in low and intermediate magnetic fields, the high compensation temperature θ_C originates indirectly from the processing route through its influence on local magnetic exchange. The larger grains produced by the magnetic slip-casting route shift θ_C upward. Despite their different relaxation characteristics, comparable crystallite and grain dimensions across all samples ensure that the local Fe–O–Gd superexchange integrals remain effectively identical, resulting in the same θ_C . To better understand this behaviour, a simulated thermal evolution of the Gd- and Fe-sublattice magnetizations for the three Gd₃Fe₅O₁₂ ceramics is presented in Fig. 3, Supplementary. These simulations are not intended to represent experimental data, but rather to offer a qualitative understanding of the underlying mechanisms. They show that while microstructural variations modify the magnitudes and slopes of the individual sublattices, the temperature at which the Gd and Fe curves cancel each other remains unchanged, thereby explaining the identical θ_C observed for all magnetic slip-casting processed ceramics.

4. Conclusions

Magnetic slip casting followed by sintering was demonstrated as an effective processing route for tailoring the microstructure and magnetic response of Gd₃Fe₅O₁₂ garnet ceramics. This approach leads to the formation of dense ceramics characterized by relatively large crystallite sizes (250–350 nm) and by the formation of a mesoscale grain structure when strong magnetic fields are applied. At the macroscale, no crystallographic texture was detected. The mesoscale structures form two-dimensional, magnetically correlated regions embedded in a randomly oriented polycrystalline matrix. Compared with the three-dimensional, isotropic assemblies observed in the 1T ceramic, this reduction in dimensionality has a pronounced impact on the low-field magnetic behaviour, giving rise to enhanced magnetization, stronger ZFC–FC irreversibility, and anisotropic magnetic behaviour. Mössbauer spectroscopy provided a detailed insight into the local magnetic environment, revealing partial occupation of the rare-earth (24c) sites by Fe^{3+} ions and a hyperfine signature consistent with enhanced Fe–O coordination. The elevated compensation temperature observed in all magnetic slip-casted samples is primarily associated with the large crystallite sizes achieved through the presented processing routes. Despite the differences in magnetic relaxation behaviour, the crystallite dimensions ensure that the local Fe–O–Gd superexchange integrals remain effectively similar, resulting in the same compensation temperature for the magnetic slip-casted ceramics. This indicates that magnetic slip casting offers an effective path towards engineering magnetic functionality in ferrimagnetic ceramics through microstructural and mesoscale design. This approach opens new possibilities for tailoring the properties of rare-earth iron garnet ceramics and related ferrimagnetic oxides for applications that require both directional magnetic response and temperature stability.

CRediT authorship contribution statement

Andrei Alexandru-Dinu: Conceptualization, Formal analysis, Investigation, Methodology, Visualization, Writing – original draft, Writing – review & editing. **Claudiu Locovei:** Formal analysis, Investigation, Methodology, Visualization. **Mihai Alexandru Grigorescu:** Formal analysis, Investigation, Methodology, Visualization. **Nicutor Iacob:** Formal analysis, Investigation, Visualization. **Cristian Radu:** Formal analysis, Investigation, Visualization. **Andrei Kuncser:** Formal analysis, Investigation, Methodology, Visualization. **Oleg Vasykiv:** Conceptualization, Funding acquisition, Investigation, Methodology, Project administration, Resources, Supervision, Validation, Writing – review & editing. **Tohru Suzuki:** Methodology, Resources, Supervision. **Victor Kuncser:** Conceptualization, Formal analysis, Investigation, Methodology, Resources, Supervision, Validation, Writing – original draft, Writing – review & editing. **Petre Badica:** Conceptualization, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Supervision, Validation, Writing – original draft, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this chapter can be found online at <https://doi.org/10.1016/j.ceramint.2026.07.297>.

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