Development and prospects of garnet ceramic scintillators: A review

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Abstract: Garnet ceramic scintillators are a class of inorganic scintillation materials with excellent overall performance. The flexibility of cation substitution in different lattice positions leads to tunable and versatile properties and a wide range of applications. This paper starts with an overview of the development history of the inorganic scintillation materials, followed by a description of major preparation methods and characterization of garnet scintillation ceramics. Great progress obtained in recent years consisting in applying the band-gap and defect engineering strategies to the garnet scintillation ceramics is reviewed. Finally, the respective problems in the preparation and performance of multicomponent garnet single crystals and ceramics and the effective solutions are discussed. The garnet scintillation ceramics with the highest application potential are summarized, and the future development directions are proposed.

Keywords: garnet ceramics; inorganic scintillators; single crystals; scintillation performance

1 Introduction

Scintillators help to detect high-energy ionizing radiation, which is invisible to the human eye, including swift charged particles, γ/X-rays, neutrons, and so on. Scintillation materials absorb and react with high-energy photons or particles and emit ultraviolet/visible (UV/Vis) light, and the photons of which are detected by a photodetector for counting, imaging, or other analysis. Therefore, scintillation detectors serve as the “eyes” of mankind to understand high-energy rays and particles in the process of exploring the world and the universe. Currently, the scintillation materials are widely used in nuclear medical imaging, radiation dosimetry, high-energy physics (HEP), homeland security, geological exploration, space exploration, and many other fields. Also, the underlying scintillation mechanism has been described in detail as well [1–3]. Scintillation phenomena have been found in many states of matter: solid, liquid, and gas. According to the difference in chemical composition, the scintillators can be divided into organic and inorganic ones. The inorganic scintillators have been widely concerned and applied due to their high density, high effective atomic number ($Z_{\text{eff}}$), and

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high light output [4–6]. A wide variety of their forms includes single crystals [7,8], optical ceramics [9–11], glasses [12,13], and thin films [14,15] to suit different application scenarios. In recent years, with the improvement of the preparation technology and advanced equipment, the scintillation ceramics have especially shown rapid development and achieved many practically promising results, occupying a pivotal position in the research and development (R&D) of inorganic scintillators, which is the focus of this paper.

1.1 Development of inorganic scintillation materials and garnet scintillators

The development history of the scintillator has been continued for more than 120 years since Röntgen [16] discovered X-rays in 1895. In 1896, CaWO₄ phosphor was used in X-ray intensifying screen as the first kind of scintillation materials. Hereafter, ZnS-based phosphors have received attention and were developed to detect X-rays and charged particles in alpha scattering experiment [17,18]. These powder scintillation materials were used in film detectors based on the phosphor screen. This period of phosphors ended with the development of a photomultiplier tube (PMT) and the discovery of the scintillation phenomenon in alkali halide crystals [19]. After NaI:Tl was discovered in 1948 [20], a burst of exploration on the alkali halide crystals was conducted for their high luminescence efficiency [21]. In the following years, other inorganic scintillation material systems have been developed in the 1950s including CdWO₄ crystals for γ-ray detection [22], LiI:Eu crystals for neutron detection [23], and the first glass scintillator, i.e., cerium-doped silicate glass [24]. Since CdWO₄, oxide scintillators received attention and further development occurred. Bi₄Ge₃O₁₂ (BGO) [25] crystals were fabricated later and became widespread scintillators, which were often used as “standard samples” for performance comparison. Since the 1980s, there has been a new period of the R&D of the scintillation materials with the rapid development of the HEP, medical diagnosis and therapy, and modern industrial field [26–29]. Derenzo and Moses [30] and Sreebunpeng et al. [31] are vigorously concentrated on the scintillators with response within tens of nanoseconds, high density, and high efficiency. The inorganic materials activated with rare-earth ions having allowed d–f transitions have become an imperative research direction. The oxides scintillation materials including Lu₂SiO₅:Ce (LSO:Ce) [32–34], (Lu/Y)₃Al₂O₁₂:Ce/Pr (Lu/YAG:Ce/Pr) [35–41], Lu/YAlO₃:Ce/Pr (Lu/YAP:Ce/Pr) [42–47], and Lu/GdAlO₃:Ce (LuGdAP:Ce) [48] and the halides including LaCl₃:Ce [49] and LaBr₃:Ce [50] were successfully developed and commercialized. Among them, the LSO:Ce and LYSO:Ce single crystals have become widely used in advanced medical imaging such as positron emission tomography (PET) [51]. Another material worth mentioning is BaF₂ single crystal, which, due to its cross-luminescence, is the fastest bulk inorganic scintillator among the existing materials. It has dominant decay time of 0.6–0.8 ns, providing a solid foundation for an ultrafast scintillator [52–55].

Among the oxide scintillation materials, the garnet-type scintillators are undoubtedly worth discussing. The scintillation materials with a garnet structure are promising candidates in radiation detection because of their good optical transparency, adjustable host composition, and flexible doping. In 1974, Pawley [56] proposed that YAG:Ce was a very promising scintillation material, and then polycrystalline powders and small crystals have been used. In 1978, the YAG:Ce single crystal was successfully fabricated to be a new fast scintillator for an electron detector in scanning electron microscopy (SEM) [38]. After that, the fabrication of the YAG:Ce single crystals and optical ceramics was performed, and the optical, luminescence, and scintillation properties were fully characterized and discussed [57–62]. In addition, the temperature dependence of the YAG:Ce ceramic scintillators was also studied [63,64]. However, YAG has a relatively low density of 4.57 g/cm³ and Z_eff of 35, so researchers’ attention was gradually drawn to LuAG with a much higher density of 6.72 g/cm³ and Z_eff of 63. Cerium- and praseodymium-activated LuAG single crystals were prepared by Czochralski method and micro-pulling down method, and the scintillation performance and the defect states in the crystals were discussed [35,37,65–69]. In the same period, LuAG:Ce transparent ceramics were fabricated by solid-state reaction method [70,71] and coprecipitation method [72–74]. Except for the famous doped ions such as Pr³⁺ and Ce³⁺, the scintillation properties of Yb³⁺ in the garnet structure have also been reported. Yb-doped YAG and LuAG have also been developed for solid-state lasers. Charge transfer (CT) luminescence of Yb³⁺ has very fast lifetime (90–100 ns) comparable with the 5d–4f one of Ce³⁺, which has aroused widespread research interest [75,76]. However, the severe luminescent quenching at room temperature
inhibits practical applications of the Yb-doped scintillators despite very fast scintillation response of around a few nanoseconds.

The cations in garnet are not limited to those mentioned above, but many elements with appropriate ionic radii can act as constituent elements of the matrix. Thanks to the development of “band gap engineering” strategy, in 2006, Ga $^{3+}$ was admixed into the LuAG:Pr single crystal [77]. The obtained LuGAG:Pr showed very fast scintillation decay time below 20 ns without any slower components. In 2007, similar attempts have been made in polycrystalline ceramics: Gd$^{3+}$ was introduced into YAG:Ce to partially replace Y$^{3+}$ to achieve higher stopping power to X/$\gamma$-rays, which significantly increased the $Z_{\text{eff}}$ from 35 to 50 and the density from 4.57 to 6 g/cm$^3$ [78]. Due to the attractive high stopping power and high light yield (LY), gadolinium-based garnets came under attention. The gadolinium aluminum garnet (GAG, Gd$_3$Al$_5$O$_{12}$) structure is thermodynamically unstable due to the mismatch caused by a large radius difference between two kinds of cations. However, the incorporation of other trivalent cations with intermediate radii such as Y, Sc, and Ga, can stabilize the garnet structure [79,80]. Through this composition-stabilization method, multicomponent garnet scintillators represented by (Gd,Ce)$_3$(Ga,Al)$_5$O$_{12}$ (GGAG):Ce single crystals [81–85] and (Gd,Y,Ce)$_3$(Ga,Al)$_5$O$_{12}$ (GYGAG):Ce transparent ceramics [86–89] have been fully developed and shown much higher LY of above 60,000 ph/MeV [90,91].

The garnet single crystal scintillators have been developed for decades, and large-size crystals with high quality were produced [92–94]. However, influenced by the intrinsic segregation coefficients of different ions in the melt, the composition distribution inside the single crystals is not homogeneous, which seriously affects the luminescent uniformity of the single crystals [95–98]. On the other hand, the volatilization of Ga$_2$O$_3$ above 1300 K [99] is also a severe problem in the preparation of the gallium garnet single crystals, which causes the Ga content to vary along the crystal growth direction [97,100]. Because of the development of the advanced transparent ceramic preparation technology [101–104], bulk optical ceramics have become another very important candidate for the inorganic scintillators. Especially for the garnet ceramics with a cubic structure, high optical quality comparable to that of the single crystals could be achieved, which provides more possibilities for improving the performance of the scintillation ceramics [105]. In the last few decades, YAG:Ce [63], LuAG:Ce [106], LuAG:Pr [107], and GGAG:Ce [108] ceramic scintillators exhibited better scintillation performance compared to single crystal counterparts because of homogeneous dopant concentrations and less anti-site defects generated due to high preparation temperatures in the case of the melt-grown crystals [109]. The garnet ceramic scintillators are thus increasingly showing an important significance and future application development prospects.

1.2 Fabrication of scintillation ceramics

As the garnet structure is cubic, it is optically isotropic and chemically stable, which enables its preparation into the transparent ceramics. Since Ikesue et al. [110] prepared the YAG:Nd transparent ceramics with high optical quality by the solid-state reaction method in 1995 and successfully achieved its lasing, the preparation of the garnet-structured transparent ceramics has received wide attention in Refs. [111–115]. While the translucent scintillators also have their applications, for HEP and high-tech medical devices, high transmittance is necessary to reduce self-absorption and optical scattering. The preparation of the transparent ceramics includes the steps of powder preparation, molding, sintering, and post-treatment, and the addition of the sintering aids also has to be considered. The refractive index of the material determines its theoretical transmittance, but for the transparent ceramics, impurity ions, residual pores, and secondary phases are the main causes of absorption and optical scattering losses, which have to be avoided during the preparation process. Depending on whether the phase formation reaction occurs during the sintering process, the preparation methods of the garnet ceramics can be divided into non-reactive sintering method and reactive sintering method.

1.2.1 Non-reactive sintering method

In the non-reactive sintering method, ceramics are prepared by directly sintering of pure-phase ceramic powders, and the preparation of the powders with the pure phase, high purity, and high sintering activity is the key to this method. Co-precipitation method is one of powder synthesis methods, which can meet the above requirements. By controlling the appropriate conditions, the mixed metal ion solution reacts with precipitants and produces insoluble precipitates. After separation, drying, and heat treatment, the pure-phase ceramic powders are obtained. Although the powders
prepared by the co-precipitation method have been calcined into the garnet phase before sintering, the sintering process under vacuum or reducing atmosphere can still lead to the volatilization of gallium and stoichiometric deviations. Ningbo Institute of Materials Technology and Engineering, Chinese Academy of Sciences (CNITECH, CAS) has done a lot of work in this area [116–119], and the traditional co-precipitation method was upgraded to ultrasonic-assisted co-precipitation method to obtain powders with a higher specific surface area and sintering activity [120]. Shanghai Institute of Ceramics (SIC, CAS) has also devoted many years of research on the co-precipitation method [72,121,122]. The GYGAG:Ce powders prepared by the co-precipitation method are combined with pre-sintering under an oxygen atmosphere and hot isostatic pressing (HIP) post-treatment to obtain the transparent ceramics [123]. The SEM micrographs of the synthesized precursors and the calcined powders are shown in Figs. 1(a) and 1(b), respectively. The garnet phase is formed completely in the process of calcination, as shown in Fig. 1(c), and the GYGAG:Ce nanopowders still have good sintering activity. The HIP post-treatment promotes further compression and elimination of intergranular pores, but is ineffective for intracrystalline pores, as shown in Fig. 1(d).

Flame spray pyrolysis (FSP) method is another efficient technique for synthesizing the nanopowders with high specific surface area, high sphericity, and almost no agglomeration, which is very important for the preparation of raw materials for ceramics by powder metallurgy [124–126]. Lawrence Livermore National Laboratory (LLNL) has prepared many high-quality garnet transparent scintillation ceramics by the FSP method including LuAG [127], GGAG, and GYGAG [89,128–130], especially GYGAG, which will be described in more detail in Section 2.3 dealing with quaternary garnet ceramics. The excellent micromorphology of the GYGAG powders prepared by the FSP method can be seen in Fig. 2.

![Fig. 1](image1.png)

Fig. 1  SEM micrographs of (a) precursor and (b) GYGAG:Ce powders calcined at 1100 °C for 4 h; (c) schematic illustration of precursor calcination for GYGAG:Ce powders; (d) schematic illustration of pore elimination in ceramics during pre-sintering and HIP post-treatment. Reproduced with permission from Ref. [123], © Elsevier B.V. 2020.

![Fig. 2](image2.png)

Fig. 2  TEM micrographs of GYGAG:Ce powders: (a) as-received from FSP method and calcined at (b) 600 °C, (c) 900 °C, and (d) 1050 °C. Reproduced with permission from Ref. [130], © Elsevier B.V. 2012.
1.2.2 Reactive sintering method

Solid-state reactive sintering is another traditional method of preparing the garnet ceramics [131,132]. Commercial oxide raw powders are mixed and crushed by ball milling in a certain ratio, undergo phase transformation during the sintering process of the ceramics, and eventually form the garnet phase. The solid-state reaction method is relatively simple and easy to realize the mass production of large-size ceramics. The preparation of composite structural GGAG/YAG-layered scintillation ceramics were achieved by the solid-state reaction method to get a tunable emission band and overall density [133]. The photographs of the green body and sintered samples of the Ce:GGAG/Cr:YAG composite ceramics are shown in Fig. 3. Depending on the sintering activity of the powder raw materials and the limited crushing in the ball-milling step, the reactive sintering can sometimes have an obstacle in the sintering densification process, requiring the addition of the sintering aids like tetraethyl orthosilicate (TEOS) [134] or ZrO2 [135] to achieve dense and even transparent ceramics.

2 Garnet ceramic scintillators

The compounds with a garnet structure can be expressed as \([A^{3+}]_3[B^{3+}]_2[C^{3+}]_3O_{12}\), where \(A = Y, Lu, Gd, Tb, \) etc., and \(B/C = Al, Ga, Sc, \) etc. This is an artificial structure, different from the garnets in nature that are formed by 2+ and 4+ cations. The garnet composed of trivalent cations enables to introduce many transition metal ions and rare earth ions as luminescence centers into the matrix, and obtain high stopping power to ionizing radiation. Garnet is an optically isotropic cubic structure and has high symmetry. In a garnet cell, the occupancy of different cations follows the rules: \(B^{3+}\) forms a \(BO_6\) octahedron with a coordination number of 6; \(A^{3+}\) occupies the center of the dodecahedron formed by the interconnecting tetrahedrons and octahedrons with a coordination number of 8. As shown in Fig. 4, in the LuAG crystal structure, \(Lu^{3+}\) occupies the dodecahedral center, 60% of \(Al^{3+}\) occupies the tetrahedral center, and 40% of \(Al^{3+}\) occupies the octahedral center [136]. \(Lu^{3+}\) could be replaced by many rare-earth ions to achieve a variety of luminescence characteristics [137–139]. In the Lu-based garnets, \(Y^{3+}\) and \(Gd^{3+}\) of a high concentration can be incorporated [140,141], and the \(Al^{3+}\) lattice site could be replaced by ions like Ga3+ and Sc3+ [77,142–144].

Band gap engineering strategy took advantage of the extended possibility of cation substitution to change the energy band structure and crystal field of materials, and to achieve the optimization of scintillation properties [145,146]. The introduction of Ga into LuAG hosts can lower the conduction band (CB) minimum, immerse some shallow traps, especially
those related to the anti-site defects into it, and thus diminish the delayed luminescence caused by trapping of the electrons at shallow traps [147,148]. The obtained Lu₃(Ga,Al)₅O₁₂:Ce (LuGAG:Ce) single crystals show a less slow scintillation component and a higher LY value compared with LuAG:Ce. Similar fast scintillation response was also found in LuGAG:Pr [77,149]. With the increase of Ga contents, the 5dᵢ energy level of activators shifts to higher energy, whereas the CB minimum shifts to lower energy, getting closer to each other. This result helps the thermal ionization energy barrier of the 5dᵢ excited level and makes the thermal ionization process more likely to occur around room temperature, which reduces the prompt part of the luminescence response. Fortunately, the introduction of Gd into the garnet system can lower the 5dᵢ energy level, diminishing its thermal ionization at room temperature [97]. Due to the energy transfer from Pr³⁺ towards Gd³⁺, this strategy is not suitable for praseodymium-doped garnets [150,151], but it is very encouraging in cerium-doped gadolinium–gallium–aluminum garnets and promotes the development of the so-called multicomponent garnet scintillators. In this material, the energy transfer from Gd-sublattice towards 5d₂ state of Ce³⁺ is realized, thus contributing to fast and effective excitation of Ce³⁺ emission center [152]. Figure 5 shows the energy level scheme and “band-gap engineering” in the cerium-doped aluminum garnet scintillators with Ga and Gd [97]. This schematic diagram depicts the movement direction of the 5dᵢ and 5d₃ energy levels of Ce³⁺ and CB minimum but does not represent the accurate energy levels of Gd³⁺ and Ce³⁺. For more details refer to Refs. [97,146,153].

In a garnet system, in addition to the substitution of matrix cations, the scintillation performance can also be improved by co-doping aliovalent ions, which is called “defect engineering”. Usually, lower-valence co-dopants like Mg²⁺, Ca²⁺, Li⁺, and others will change the defect structure of the materials by altering the charge compensation mechanism or affecting the vacancy concentration in the materials, thereby affecting the scintillation performance [154–158]. Especially in the cerium-doped materials, the low-valence ions can stabilize some of the cerium ions at the tetravalent valence. Ce⁴⁺ can act as a fast recombination luminescence channel during the scintillation process and compete with other shallow electron traps, as shown in Fig. 6. Mg-doped LuAG:Ce single crystals showed a much higher LY, faster scintillation decay, and lower afterglow intensity. The existence of Ce⁴⁺ was confirmed by its characteristic CT absorption band [159] and X-ray absorption near-edge spectroscopy (XANES) [160,161]. These two strategies, band-gap engineering and defect engineering, have many applications in the garnet ceramics, which will be described in Sections 2.1.2, 2.2, and 2.3.

2. 1 Binary garnet ceramics

2.1.1 YAG:Ce

In 1997, highly dense YAG:Ce transparent ceramics were firstly fabricated for scintillation application [58]. Although the YAG ceramics have the disadvantages of low stopping power and limited applications, they are very cheap scintillation materials for preliminary exploration. The YAG:Ce ceramics were fabricated by hot-pressing and air-annealing. The unoptimized preparation method resulted in the ceramics with poor optical and

![Fig. 5 Energy level scheme and “band-gap engineering” in cerium-doped aluminum garnet scintillators with Ga²⁺ and Gd³⁺ mixture. Note: VB is the abbreviation of valence band. Reproduced with permission from Ref. [97], © American Chemical Society 2011.](image-url)

![Fig. 6 Schematic diagram of scintillation mechanism at stable Ce³⁺ and Ce⁴⁺ centers in aluminum garnet host. Ce⁴⁺ acts as a faster recombination luminescence channel during scintillation process and can compete more effectively with shallow electron traps. Reproduced with permission from Ref. [157], © American Chemical Society 2014.](image-url)
scintillation quality, with a myriad of internal defects and the best sample only had a light output of about 50% of that of the single crystals. In 2005, the quality of the YAG:Ce ceramics improved significantly. The transmittance of the YAG:Ce ceramics was comparable to that of single crystals in a wavelength longer than 500 nm, which is about 80% (the theoretical transmittance calculated by the refractive index of YAG is 83.54% [162]). In addition to better optical quality, the best ceramic samples with 0.5 and 0.05 mol% Ce-doping also showed about 15% higher LY compared to the YAG single crystals. As for scintillation decay kinetics, the ceramic samples had a good performance with major decay time constants of about 80 and 300 ns [59]. A more comprehensive comparison of YAG:Ce ceramics and single crystals showed the complete absence of a 300 nm emission band in the YAG:Ce ceramics and single crystals showed the complete absence of a 300 nm emission band in the YAG:Ce ceramics, demonstrating the absence of anti-site defects in the ceramics prepared at a much lower temperature than the YAG melting point [163]. Due to the inherent limitations of low density and $Z_{\text{eff}}$ studies on YAG as scintillation materials gradually decreased. Researchers focused on heavier garnets like LuAG or Gd-admixed GdYAG [78,164].

2.1.2 LuAG:Ce

After the discovery of high stopping power, high light output, and fast decay of LuAG:Ce, research on the fabrication and properties of the transparent ceramics, in addition to the single crystals, was rapidly carried out [165]. The properties of LuAG:Ce are making it more interesting for the HEP community and moving towards radiation-resistant, brighter, and faster materials [166,167]. In the early preparation of the LuAG:Ce ceramics, although the transparent ceramics were successfully fabricated by the solid-state reaction method [168], coprecipitation method [72], or FSP method [79,169], the scintillation performance of the LuAG:Ce ceramics was poor and could not reach the level of the single crystals. Although there were no anti-site defects in the produced ceramics, there were deeper electron traps at grain surfaces or interfaces, which caused very slow decay components [109]. In 2011, the high-performance LuAG:Ce ceramics with an LY value of 14,800±1500 ph/MeV, higher than that of the Czochralski-grown single crystals, were fabricated by Konoshima Chemical Co. Ltd. [106]. The further breakthrough in the LuAG:Ce ceramics was mainly through low-valence-ion doping, i.e., by defect engineering. In 2014, the LuAG:Ce,Mg scintillation ceramics were fabricated by the solid-state reaction method, and Mg$^{2+}$ was introduced in the form of MgO [159]. The addition of MgO as a sintering aid was helpful for the densification process and the elimination of residual pores; more importantly, Mg$^{2+}$ could stabilize Ce$^{4+}$, especially after annealing in an oxidizing atmosphere.

The presence of O$^-$ centers as hole traps in the LuAG:Ce,Mg ceramics was confirmed by theoretical calculation and experiment [158,170], which compete with the activators to capture holes and act as a hole source for Ce$^{3+}$ to convert to Ce$^{4+}$, as shown in Fig. 7 [154]. The created Ce$^{4+}$ ions act as the fast luminescence centers under high-energy excitation in the luminescence process of Ce$^{3+}$-doped scintillators described by Reactions (5)–(7) [171]:

\[
\begin{align*}
\text{Ce}^{3+} + h_v & \rightarrow \text{Ce}^{4+} \\
\text{Ce}^{4+} + e^- & \rightarrow (\text{Ce}^{3+})^* \\
(\text{Ce}^{3+})^* & \rightarrow \text{Ce}^{3+} + h_v
\end{align*}
\]

where $h_v$, $e^-$, and $(\text{Ce}^{3+})^*$ represent the VB hole, the CB electron, and the excited Ce$^{3+}$ ion, respectively. $h_v$ refers to the emitted photon. The fabricated LuAG:Ce,Mg ceramics showed a high LY of 21,900 ph/MeV measured at 1 μs shaping time. More importantly, the ratio of LY as measured at 1 μs to that measured at 10 μs (LY$_{1\mu s}$/LY$_{10\mu s}$) of 79% obtained for the LuAG:Ce,Mg ceramics is higher than that of 67% achieved in the LuAG:Ce,Mg single crystals, indicating a larger contribution of fast scintillation component (i.e., faster scintillation response) for the former. After further optimization, the LuAG:Ce,Mg ceramics exhibited a higher LY$_{1\mu s}$ of 25,000 ph/MeV together with faster scintillation decay and lower afterglow signal. With the increase of the Mg concentration, the Ce$^{4+}$ absorption peak markedly increased in the XANES spectra, showing the increasing proportion of stabilized Ce$^{4+}$, as shown in Fig. 8 [160]. Subsequently, the effects of doping concentration and annealing condition on the scintillation performance of the LuAG:Ce,Mg ceramics were also studied [172,173].
In a new report in 2020, the LuAG:Ce,Mg ceramics prepared by the solid-state reaction method exhibited a high LY equivalent to 4.63 times that of the BGO single crystals and good radiation hardness under the irradiation of 45 MeV protons, the latter as demonstrated by an almost constant absorption spectrum in 200–800 nm range before and after irradiation [174]. Figure 9 shows the photographs of laser-cut LuAG:Ce,Mg ceramic arrays with a dimension of 50 μm × 50 μm and a distance of 10 μm and the testing of its flat panel X-ray imaging ability. The good quality images with a high spatial resolution were obtained under the evaluation by line-pair pattern method, which proved that the LuAG:Ce,Mg ceramics are promising scintillators for flat panel X-ray imaging [175].

In addition to Mg2+, Ca2+ is another effective co-dopant ion for defect engineering and has many successful examples [176]. The LuAG:Ce,Ca scintillation ceramics fabricated by the solid-state reaction method exhibited

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**Fig. 7** Schematic diagram of role of O− centers in scintillation process of LuAG:Ce,Mg ceramics. Reproduced with permission from Ref. [154], © WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim 2015.

**Fig. 8** Room-temperature XANES measurements on cerium LIII-edge (5723 eV) spectral region of (a) Ce3+ and Ce4+ reference samples and (b) as sintered LuAG:0.3 at% Ce ceramic samples with different Mg co-dopant concentrations. Reproduced with permission from Ref. [160], © WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim 2016.

**Fig. 9** Photographs of laser-cut LuAG:Ce,Mg ceramic arrays taken by (a) optical microscope and digital camera and X-ray imaging of standard line-pair pattern by ceramic samples (b) without and (c) with cutting. Reproduced with permission from Ref. [175], © Editorial Board of Journal of Inorganic Materials 2015.
fast scintillation decay time of 42 ns and a \( {\text{LY}}_{1\mu s} / {\text{LY}}_{12\mu s} \) ratio of 85.5%, which indicates faster scintillation response and lower content of slow scintillation components [177]. In 2018, the LuAG:Ce,Ca ceramics achieved the highest ratio of the fast component intensity to the overall intensity with the value of 90%. It is defined as the ratio of light yield values at 200 and 3000 ns shaping time [178]. The LuAG:Ce,Ca ceramics also showed excellent radiation hardness under an ionizing radiation dose of up to 2.2 MGy and a proton fluence of up to \( 3 \times 10^{14} \) p/cm\(^2\). In 2022, Zhu et al. [179] systemically investigated a wide range of Ca\(^{2+}\) co-doping concentrations in the LuAG:Ce,Ca ceramics. In a suitable range of co-doping concentrations, Ca\(^{2+}\) acted as an effective sintering aid to avoid the rapid grain growth and the formation of the intracrystalline pores and obtained good optical transmittance, as shown in Fig. 10. The fast-to-slow component ratio represented by \( {\text{LY}}_{0.5\mu s} / {\text{LY}}_{3.0\mu s} \) was optimized from 79% to 97%, which has important implications for the applications that require fast scintillation response with suppressed slower tails. In Ref. [166], the LuAG:Ce ceramics co-doped with Ca\(^{2+}\) or Mg\(^{2+}\) appeared more than twice higher radiation hardness with respect to the LYSO single crystals under both neutron and proton irradiations. Ultrafast scintillation decay and excellent radiation hardness of the LuAG:Ce,Ca/Mg ceramics show great potential in HEP applications.

Monovalent ions such as lithium appeared as a new choice of the co-doped ions for the LuAG:Ce scintillation ceramics. Doping Li\(^+\) brought great positive effects in LY, scintillation decay, and afterglow. The obtained LuAG:Ce,Li ceramics displayed a high LY of 29,200 ph/MeV at 10 \( \mu s \) shaping time, and it was the highest value ever reported in the LuAG:Ce-based scintillators [180].

In addition to the ion co-doping, another strategy for the LuAG:Ce ceramic optimization is to use its solid solution range around the stoichiometric ratio for the modulation of matrix components. According to the phase diagram of LuAG, a slight excess of Lu does not cause a second phase, but rather acts as a self-sintering aid in the ceramic sintering [181,182]. The excess of Lu was helpful for the improvement of optical transparency, but the overmuch Lu contents could increase the Lu\(_{3}\)Al\(_2\) anti-site defects and appeared harmful for the fast scintillation [136]. A similar phenomenon was found in the praseodymium-doped ceramics. The excess Lu in the LuAG:Pr ceramics was in the form of the second phase to pin the grain boundary at the initial stage of sintering and to avoid the rapid growth of the grains and facilitate the discharge of the pores from the grain boundary. At the later stage of sintering, the excess Lu was re-dissolved into the lattice as Lu\(_{3}\)Al\(_2\) anti-site defect without reducing the optical quality of the ceramics [183]. On the contrary, the non-stoichiometric Lu\(_{3-x}\)AG:Ce ceramics with deficient Lu\(^{3+}\) exhibited poor optical transmittance due to the existence of Al-rich second phase as the optical scattering centers, while the improved scintillation performance was observed. The

![Fig. 10](https://www.springer.com/journal/40145)
Lu$_{3-x}$AG:Ce ceramics showed faster decay and lower afterglow intensity compared to that with excess Lu, which was due to the suppression of the Lu$_{AI}$ anti-site defects [184].

2.1.3 LuAG:Pr

Pr$^{3+}$ has the allowed 5d–4f transition like Ce$^{3+}$, so the LuAG:Pr ceramics show even faster scintillation decay due to the UV shift of the Pr$^{3+}$ luminescence. Besides, among host materials like YAG, LuAG, YSO, and LSO, only LuAG presents neither Pr$^{3+}$ 5d$_{1}$–4f luminescence nonradiative quenching nor Pr$^{3+}$ ionization at room temperature, which demonstrates a significant advantage of this system [185]. After just a few years of development, since it was first reported in 2005, the LuAG:Pr single crystal has gained an application in positron emission mammography [186]. On the other hand, the preparation of the LuAG:Pr transparent ceramics has also been reported. The quality of the ceramics prepared in the earlier work was much lower than that of the single crystals [187–189]. However, in 2012, the LuAG:Pr transparent ceramics prepared by the coprecipitation method showed an LY of 21,800±1100 ph/MeV and an energy resolution of 4.6% under the $\gamma$-ray excitation, which was beyond the level of the LuAG:Pr single crystals [105]. After that, the effect of different kinds of the sintering aids in the LuAG:Pr transparent ceramics preparation was thoroughly explored. Although the transmittance of the ceramics was improved to a certain extent by adding the sintering aids, the scintillation performance deteriorated seriously due to the introduction of charge compensation and lattice defects [190,191]. Although isovalent sintering aids did not cause the charge compensation, they still caused local lattice distortion and had a negative impact on the scintillation performance [192].

In a study of $\gamma$-radiation hardness of the LuAG:Pr ceramics, it was shown that the defects introduced by the sintering aids and impurities such as Fe and Yb can cause severe deterioration of the optical transmittance and LY after the irradiation [193]. This makes the application of the LuAG:Pr ceramics in the field of high-energy photon detection under harsh irradiation conditions troublesome. The optimization of the Pr$^{3+}$ doping concentration has also been carried out in the LuAG:Pr ceramics. Higher doping concentrations were beneficial for the competition of the activators with defects and impurity ions, suppression of the slow component of the scintillation response, and increase of the LY [194]. In terms of imaging performance testing for practical applications, the processed LuAG:Pr ceramic pixels produced a clear and legible two-dimensional (2D) mapping that was superior to that of commercial BGO under the same coupling conditions [195].

Attempts have also been made to apply the defect engineering strategy to the LuAG:Pr ceramics [196]. However, in contrast to Ce$^{4+}$, Pr$^{4+}$ is completely detrimental to the scintillation properties of the LuAG scintillators. Pr$^{4+}$ CT absorption causes severe self-absorption and decreases the light output [197]. As shown in Fig. 11(a), the CT band of Pr$^{4+}$ has an onset of about 600 nm, which results in the absorption of the 5d–4f emission of Pr$^{3+}$ [198]. While the CT band of Ce$^{4+}$ has an onset of about 340 nm, thereby avoiding the adverse effects of self-absorption [159]. Therefore, it seems that any co-doping examined so far is detrimental to the LuAG:Pr ceramics, which limits this strategy for the scintillation performance improvement through defect engineering.

![Fig. 11](image-url) Absorption spectra of (a) LuAG:Pr$_2$Mg and (b) LuAG:Ce$_2$Mg. The insets in (b) are difference absorption spectrum in 200–340 nm and photograph of ceramic samples with a thickness of 2 mm. Reproduced with permission from Ref. [198] for (a), © Elsevier B.V. 2016; Ref. [159] for (b), © WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim 2014.
2.1.4 Tb₃Al₅O₁₂ (TAG):Ce

The TAG is commonly used in Faraday optical isolators due to the magneto-optic properties of Tb³⁺ [199,200]. When doped with cerium in a pure TAG host, it also exhibits the scintillation properties [201,202]. The TAG:Ce ceramics from the LLNL showed much higher β-excited radioluminescence intensity compared to the LuAG:Ce ceramics and single crystals, but very slow decay [169]. The slow principal decay of Ce in the TAG is the result of the energy migration in Tb-sublattice and Tb–Ce energy transfer [79]. On the other hand, the incongruent melting characteristic of the TAG leads to its being very sensitive to chemical components when prepared as the ceramics, and the tendency to have a second phase makes the preparation of highly transparent TAG ceramics very difficult.

2.2 Ternary garnet ceramics

2.2.1 LuYAG:Ce/Pr

The introduction of Y in the LuAG host shows positive effects of the increase of the light output and the decrease of slow scintillation component, which have been confirmed in both Ce³⁺-doped and Pr³⁺-doped LuYAG ceramics [141,203,204]. Due to the admixture of Y, the CB minimum decreases, and the shallow electron traps are partially inactivated. Adding 25% Y into the LuYAG:Pr ceramics increased the LY at 3 μs shaping time to 28,500 ph/MeV and fast primary decay time of 21 ns. The density of the ceramics decreases with an increase in the Y concentration, and the radiation hardness of the LuYAG ceramics was also lower than that of the YAG:Ce sample (20,000 ph/MeV). The afterglow level of the YAG:Ce ceramics was 13 ppm at 20 ns after X-ray radiation cutoff and was comparable to that of the commercial BGO single crystals, which was a very encouraging result. On the other hand, in a more refined transmission electron microscopy (TEM) structural analysis study of the YAG:Ce ceramics, it was found that Ce³⁺ has an evident inhomogeneous distribution within the grains and at grain boundaries. The Ce³⁺ concentration increased at ceramic grain boundaries [211]. This phenomenon led to a decrease in the energy resolution when applying optical ceramics to the scintillators, and the preparation technology needed to be further optimized.

2.2.2 LuGAG:Ce

It was first confirmed in the single crystals that replacing part of Al with Ga in the LuAG host can lower the CB minimum to bury shallow electron traps in it and improve scintillation efficiency [206]. Subsequently, the preparation of the LuGAG:Ce ceramics was also successfully realized by the solid-state reaction sintering method [142]. Although the ceramics had many residual pores and poor optical quality due to severe agglomeration of commercial powders and the absence of sintering aids, the LuGAG:Ce ceramics still showed inspiring fast scintillation decay. The fast component represented by $LY_{0.5\mu s}/LY_{6\mu s}$ value was up to 92.5%, while this value of reference LuAG:Ce ceramics was only 42.5%.

2.2.3 (Gd,Y)₃Al₅O₁₂ (GYAG):Ce

Gadolinium was originally introduced into the YAG to improve X-ray stopping power. However, the replacement of Gd has its limits. The GAG is an incongruently melting compound and is an unstable phase [207]. When the concentration of Gd³⁺ exceeds 75%, the garnet phase cannot be stabilized and turns into a perovskite phase [208,209]. In 2009, Cherepy et al. [79] fabricated a series of garnet ceramic scintillators including GYAG:Ce, which had an LY of 16,500 ph/MeV under γ-excitation and an energy resolution of 11.2% at 662 keV γ-rays. Highly transparent Ce-doped (Gd₀.₃₂Y₀.₆₈)₃Al₅O₁₂ ceramics were synthesized by Konoshima Chemical Co. Ltd. in 2017 [210]. The best GYAG:Ce sample had an LY of 18,000±1800 ph/MeV at 0.5 μs shaping time, which was slightly lower than that of the YAG:Ce sample (20,000 ph/MeV). The afterglow level of the GYAG:Ce ceramics was 13 ppm at 20 ns after X-ray radiation cutoff and was comparable to that of the commercial BGO single crystals, which was a very encouraging result. On the other hand, in a more refined transmission electron microscopy (TEM) structural analysis study of the GYAG:Ce ceramics, it was found that Ce³⁺ has an evident inhomogeneous distribution within the grains and at grain boundaries. The Ce³⁺ concentration increased at ceramic grain boundaries [211]. This phenomenon led to a decrease in the energy resolution when applying optical ceramics to the scintillators, and the preparation technology needed to be further optimized.

2.2.4 GGAG:Ce

Due to a relatively high density of 6.2 g/cm³, the GAG was an attractive candidate for a scintillator host. To solve the problem of unstable GAG garnet phase when growing the single crystals from the melt, it was found that Gd₃(Ga,Al)₅O₁₂ with partial substitution of gallium for aluminum can be prepared into the single crystal materials by the melting method, and GGAG:Ce had an even higher density of 6.63 g/cm³ [212]. What was even more exciting was that Czochralski-grown GGAG:Ce single crystals showed a very high LY for about 46,000 ph/MeV, higher than those of all previous garnet-based scintillation materials. Consequently, GGAG:Ce quickly became a hot topic in the research on the scintillation materials. Naturally, studies on the synthesis and
properties of the GGAG:Ce scintillation ceramics were also carried out. In fact, as early as 2008, the GGAG:Ce ceramics were prepared by the solid-reaction method. The effect of the non-stoichiometric ratio of the Gd and the (Ga,Al) sites on the LY and afterglow performance was investigated [213]. However, the lack of more detailed reports on the performance of other scintillations did not attract sufficient attention. In 2013, in a comparative study on the GGAG:Ce ceramics and single crystal scintillators, the fabricated GGAG:Ce ceramics were absolutely opaque but showed an extremely high LY of 70,000 ph/MeV, as shown in Fig. 12 [108]. This high LY was related to factors such as higher doping concentrations in the ceramics relative to the single crystals, optical scattering effect, and energy transfer due to an impurity phase. Later studies did not surpass this result in terms of LY, but new advances have been made in other aspects. For example, the GGAG:Ce scintillation ceramics co-doped with Mg$^{2+}$ and Ca$^{2+}$ obtained ultra-fast decay time of 31 ns for the PET and time of flight (TOF)-PET applications [214].

It is worth noting that although the co-precipitation method is considered capable of achieving homogeneous mixing at the atomistic scale during the precursor preparation process, activator segregation at the grain boundaries and inhomogeneous micro-luminescence were found in the GGAG:Ce ceramics, as shown in Fig. 13 [215]. The concentration of Ce showed more than five times enrichment at grain boundaries, causing a significant increase in the photoluminescence (PL) intensity. This reveals a difficult problem faced in the process of enhancing the luminescence uniformity of scintillation ceramics by optimizing grain boundary structure, which is worthy of attention and needs further studies.

### 2.3 Quaternary and higher garnet ceramics

With the advanced development of the band-gap engineering and phase stability expansion theory, the quaternary or higher garnets emerged. It embodies the highest flexibility of cation substitution on different lattice sites of multicomponent garnets and implies a

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**Fig. 12** (a) Photographs of single crystal (left) and ceramic (right) GGAG:Ce scintillators and (b) their emission under UV (365 nm) excitation. (c) $^{137}$Cs-excited pulse height spectra of ceramics and single crystal GGAG:Ce scintillators. The absolute LY values were determined based on $^{55}$Fe 5.9 keV X-ray peak. Reproduced with permission from Ref. [108], © Elsevier B.V. 2013.

**Fig. 13** Images of spatial distributions of PL intensity under excitation at (a) 405 nm and (c) 442 nm; images of spectral center of mass under excitation at (b) 405 nm and (d) 442 nm. Reproduced with permission from Ref. [215], © Elsevier B.V. 2021.
The high degree of tunability of the properties controlled by the compositions. It is noteworthy that the compositional disorder in the atomistic scale increases with the number of cation number in the host structure. In crystalline materials, this suggests that the migration length of the non-equilibrium carriers formed by ionizing radiation decreases, and the probability of the carriers reaching the emission centers increases, resulting in faster decay and a higher LY [216,217]. Recently, a novel term for the multicomponent garnets has also been referred to as high-entropy ceramics [218]. This is a broader meaning of the high-entropy ceramics, which is not quite the same as the traditional high-entropy ceramics in Ref. [219]. However, it also provides a new way of thinking to delve into the specificity of the multicomponent garnets. It is worth mentioning that in addition to the application in the scintillators, it is also easier for the multicomponent garnet to achieve ultrashort pulse lasers with wavelength tunability due to the wider emission band [220–223]. Moreover, due to the higher brightness and tunable luminescence, the multicomponent garnets can also be prepared into phosphors and ceramics for high-power white LEDs by introducing different luminescent centers [224,225]. As the number of elements increases, the delineation of properties and applications of the garnet scintillators becomes less clear. (Gd,Y,Lu)(Ga,Sc,Al)O₃ scintillators have promising applications in nuclear medical imaging [226–228], homeland security [86], neutron detection [229,230], and many other fields. High LY, fast decay, and high radiation hardness have become common development goals for the multicomponent garnets.

Although GGAG:Ce can be prepared in the single crystal form by the melting method, which also enables the preparation of the transparent ceramics, its structure still suffers from thermodynamic instability. When Gd³⁺ is substituted with smaller cations like Y³⁺ [128], Lu³⁺ [97], and Yb³⁺ [118], the degree of the lattice distortion is reduced, and the structural stability is improved. In the preparation of the scintillation ceramics, this is manifested easier to get good transparency. The partial substitution of Gd³⁺ with Y³⁺ is one of the most widely studied structures, namely GYGAG:Ce. In 2008, Cherépy et al. [80] reported the GYSAG:Ce ceramics fabricated using the powders prepared by the FSP method in combination with vacuum pre-sintering and HIP post-treatment. The GYSAG:Ce transparent ceramics showed a high LY of more than 50,000 ph/MeV with principal decay time of 100 ns. Also, the modulation of the composition showed that the transmittance of the ceramic sample was significantly increased with suitable Y³⁺ and Sc³⁺ concentrations. When the significant effect of gallium on the regulation of energy level positions was discovered, the GYGAG became the mainstream of research. In the following years, LLNL prepared high-quality GYGAG:Ce ceramics for γ-ray spectroscopy by the same method and further optimized the annealing process to reduce afterglow while expanding the size of the ceramics [87,231–234]. A high LY of 55,000 ph/MeV and an energy resolution of 4.59% under a 662 keV γ-ray excitation were obtained from the Gd₁.₄₉Y₁.₄₉Ce₀.₀₂Ga₂Al₂O₁₂ [130]. The study of phase stability is to meet the needs of large-scale commercial preparation. Mutual substitution of cations with different radii between three lattice sites in the garnet structure generates a phase stability region that can maintain a high degree of transparency despite reduced fine chemical control, reflecting the advantages of GYGAG:Ce [128]. In 2014, the large-size GYGAG:Ce transparent ceramic sample with 5.6 in³ was prepared and still maintained an excellent energy resolution of 3%, whose performance on γ-ray spectroscopy was much better than that of NaI:Tl [129]. In 2022, Jarrell et al. [235] proved that the GYGAG:Ce ceramic samples from the LLNL had high β radiation hardness. No significant decrease in the LY was observed after the irradiation with electrons for 3100 MGy, confirming its potential for the applications in indirect conversion nuclear batteries. Figure 14 shows the photographs of the GYGAG:Ce ceramics from the LLNL at different stages and their microtopography.

In addition to the FSP method, Zhang et al. [90,236] from the CNITECH used co-precipitation to prepare raw GYGAG:Ce powders, combined with oxygen pre-sintering and HIP post-treatment to prepare the transparent ceramics with different Y³⁺ concentrations. The ceramic sample with a chemical formula of Ce₀.₀₂Y₀.₉Gd₁.ₐ₀₅Ga₂Al₂O₁₂ had the best performance with an LY of 61,000 ph/MeV and the in-line transmittance of 77% at 545 nm. Subsequently, the traditional co-precipitation method was improved to ultrasound-assisted co-precipitation to obtain the nanopowders with better homogeneity and sinterability. The in-line transmittance of the GYGAG:Ce ceramic sample was up to 81%, reaching a level comparable to that of the single crystals [237]. With the improvement of ceramic fabrication, it is inevitable to move towards the application. Recently, the CNITECH characterized the mechanical properties.
of the GYGAG:Ce ceramics and processed them into arrays to prepare for future use in computed tomography detectors, as shown in Fig. 15 [238].

$\text{Ce}^{3+}$ as the activator of the garnet scintillator is undoubtedly the representative of high light output and fast scintillation response. However, in some other applications requiring higher brightness such as indirect energy converters of $\alpha$- and $\beta$-particle voltaic, $\text{Tb}^{3+}$ has a better performance. On the other hand, $\text{Tb}^{3+}$-activated gadolinium-based garnet scintillators have a promising application in neutron detection. Therefore, research on GYGAG:Tb has emerged in recent years and has yielded some results. In 2021, Korzhik et al. [239] fabricated Gd$_x$Y$_{2.85-x}$Tb$_{0.15}$Al$_2$Ga$_3$O$_{12}$ and Gd$_x$Y$_{2.97-x}$Ce$_{0.03}$Al$_2$Ga$_3$O$_{12}$ ceramics with different Gd contents by the co-precipitation method. The LY under $\gamma$- and $\alpha$-excitation of GYGAG:Tb/Ce were tested, and the highest LY of Tb$^{3+}$-doped samples was four times that of Ce$^{3+}$-doped samples when $x = 1$–1.2. The good irradiation tolerance and temperature stability of the GYGAG:Tb/Ce ceramics were also subsequently confirmed, which was strong evidence of their great potential for nuclear batteries [240].

$(\text{Gd}_{x-\gamma}\text{Lu}_{\gamma})(\text{Ga}_{1-\gamma}\text{Al}_\gamma)\text{O}_{12}:\text{Ce}$ (GLGAG:Ce) is considered to be the result of applying a band-gap engineering strategy in LuAG:Ce. It has a $Z_{\text{eff}}$ of 50–63 and a density of 6.7–7.1 g/cm$^3$ depending on Gd/Lu stoichiometric ratios, which are higher than those of GYGAG:Ce [241]. A comparative study on the GGAG:Ce single crystals and GLGAG:Ce optical ceramics revealed that the latter, prepared from coprecipitation powders, had a higher LY of 48,200 ph/MeV and is an order of magnitude lower afterglow level compared to the former [242]. In single crystalline films of GLGAG:Ce prepared by liquid phase epitaxy, the atomistic compositional disorder, i.e., the local variation of the Gd–Lu and Ga–Al concentrations, was evidenced in spectral shifts of Ce$^{3+}$ excitation and emission bands, which certainly must have a reflection in a local variation of the electronic band structure (CB and VB edges) [243].

The most important feature of the multicomponent garnets is the diversity of the ionic substitution at various lattice sites. As mentioned at the beginning of Section 2, the combined effect of Ga and Gd on the energy level structure is believed to be responsible for
the ultra-high LY obtained for the multicomponent garnet scintillation materials. In GLGAG:Ce, the characterization of the XANES showed that the introduction of Ga also had an effect on the valence states of cerium ions and was able to regulate the ratio of Ce$^{3+}$/Ce$^{4+}$, which was due to the change of separation in the 4f ground state energy of Ce$^{3+}$ and the Fermi energy level position ($E_F$) after Ga$^{3+}$ doping [244]. Another characteristic of the multicomponent garnets is the increased phase stability of the compound due to the flexibility of inter-ionic substitution, that is, the ability to maintain a single garnet phase to a certain extent from the stoichiometric ratio. The nonstoichiometric (Ce,Lu,Gd)$_{3+\delta}$(Ga,Al)$_{5-\delta}$O$_{12}$ transparent ceramics prepared by the solid-state reaction method could maintain a single phase and good transmittance with $\delta = 0.1$ [245]. In Ref. [246], Wang et al. fabricated the GLGAG:Ce transparent ceramics from nano-sized powders by the FSP method. The nonstoichiometric GLGAG:Ce ceramics with a slight (Lu,Gd) excess possessed small grains, fewer pores, pure garnet phase, and a high LY of about 60,000 ph/MeV.

The high LY of the multicomponent garnet ceramics makes it a promising scintillator in high-end medical imaging applications like CT and PET scans. In addition to the LY, afterglow is also of great interest. Khanin et al. [247,248] introduced 3d transition metal ions (Ti, V, Cr, Mn, Fe, Co, Ni, Cu, or Zn) into GLGAG:Ce to simulate the effect of transition metal impurities on afterglow level in the ceramic preparation. The causes of the afterglow caused by different transition metal ions and their elimination methods were also discussed. On the other hand, Mg and Ca were co-doped in GLGAG:Ce to optimize the decay characteristic and reduce the undesired afterglow [249–251].

3 Comparison of garnet ceramics and single crystals

The garnet scintillation single crystals and ceramics have great prospects in many applications. They may have significant differences in the performance due to different fabrication principles. The main issues that need to be addressed when facing challenges are also different between them. In the following, a brief comparison of the current development of the GGAG:Ce garnet single crystals and ceramics will be presented, and their current obstacles and possible effective solutions will be described.

In 2011, Kamada et al. [97] found that when Ga partially replaces Al, the GGAG:Ce structure can be prepared as single crystals by the micro-pulling down method, and subsequently prepared as 2-inch size single crystals by the Czochralski method [212]. The LY value of 46,000 ph/MeV and the energy resolution of 4.9% at 662 keV $\gamma$-rays were reported. In 2012, by optimizing the crystal growth conditions, the LY of GGAG:0.1% Ce single crystals was improved to 62,000 ph/MeV with an energy resolution of 4.7% at 662 keV $\gamma$-rays [91]. However, the composition distribution along the crystal growth direction was not perfectly uniform, and the increase of the Ga concentration in the crystal growth direction will cause a decrease in the LY [98,252]. Due to the variation of the ion concentration in the melt resulting in differences in cell parameters, different crystalline phases may be produced at the beginning and end parts of the crystals. Sometimes mixed phases of garnet and perovskite may occur [100,253]. In the preparation of the gallium-containing garnet crystals, the volatilization of the gallium oxide is often inhibited by adding an appropriate amount of O$_2$ or CO$_2$ to the inert atmosphere [254,255]. Usually, the proportion of O$_2$ is 1%–5%, and the proportion of CO$_2$ is up to 30%. Another strategy is to add a few percent of excess Ga$_2$O$_3$ raw materials to balance the portion lost by volatilization [256,257].

The melting point of the GGAG is around 1800 °C. To prepare the GGAG single crystals by the melting method, the crucible needs to be heated to over 2000 °C. This temperature is much higher than the sintering temperature of the GGAG ceramics, which is about 1500–1700 °C depending on the sintering activity of the raw powders. The lower preparation temperature will reduce the degree of gallium oxide volatilization, and without the limitation of the iridium crucible, an atmosphere with a higher oxygen partial pressure can be applied to better suppress the gallium oxide volatilization [258]. In addition to the regulation of the sintering atmosphere, it is also possible to provide a local oxidizing atmosphere during the sintering process of the ceramics by embedding it in the oxide powders such as Y$_2$O$_3$ or ZrO$_2$ [130]. The HIP is essential in the preparation of the garnet scintillation ceramics as an important means to further eliminate residual porosity and achieve transparency in the ceramics. Usually, the HIP devices use graphite heaters and an Ar atmosphere, and the completely reducing atmosphere at high temperatures introduces the oxygen vacancy in the
ceramics, which is an electron trap that can seriously deteriorate the scintillation performance [259]. Thermal annealing in an oxidizing atmosphere is an effective way to eliminate the oxygen vacancies and increase the number of the Ce\(^{4+}\) fast luminescence centers, resulting in higher LY, faster scintillation decay, and lower afterglow level [105,260]. Thermal annealing is very commonly used in the ceramic preparation and is also applied to enhance the scintillation performance of the single crystals [261,262]. When combining the HIP post-treatment with annealing, the compressed pores produced in the HIP process may swell in the annealing, causing a decrease in the optical transmittance of the ceramics. The balance between the scintillation performance and the optical quality is what needs to be taken care of in the ceramic preparation.

4 Conclusions and prospects

After decades of development, the garnet-based scintillation ceramics were prepared by a variety of methods, and the matured preparation processes show their own advantages. For the binary garnet system, which has been studied earlier, LuAG:Ce ceramics have a great future in HEP due to their high density, bright scintillation, fast scintillation decay, and high radiation hardness. In future research, the faster scintillation response of the LuAG:Ce ceramics will be further improved while reducing the preparation cost to replace the LYSO:Ce single crystals for the proposed shashlik calorimeter detector concept for future HEP experiments. Co-doping with the low-valence ions is an effective strategy for the LuAG:Ce ceramics. For the multicomponent garnet scintillation ceramics, represented by the chemical formula of \((\text{Gd},\text{Y},\text{Lu})_3(\text{Ga,Sc,Al})_5\text{O}_{12}\), its field of applications has greatly expanded. In the field of nuclear medical imaging, it is expected to be a high-end CT/PET scanner. In such applications, further reduction of afterglow levels and improvement of optical uniformity are essential to improving the performance of the imaging device. Excellent gamma LY and energy resolution make it the material of choice for gamma spectroscopy and radiography. The presence of gadolinium cations also makes it an effective neutron-detection material that can be used in the field of neutron radiography. The flexible and adjustable composition and properties of the multicomponent garnet ceramics offer great potential for both basic scientific research and industrial applications. High optical quality (transmittance and homogeneity) and low-cost production of the multicomponent garnet ceramics while expanding their size are major challenges for researchers.

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Declaration of competing interest

The authors have no competing interests to declare that are relevant to the content of this article.

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