Hadron-Induced Radiation Damage in LuAG:Ce Scintillating Ceramics

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***Abstract*— Because of their potential low cost, bright light, and fast decay time, LuAG:Ce ceramic scintillators have attracted a broad interest in the high-energy physics community. One crucial issue for their application in future high-energy physics experiments is their radiation hardness against neutrons and protons expected at future hadron colliders. We report optical and scintillation performance of 1-mm LuAG:Ce ceramic samples doped with Mg^{2+} (and Ca^{2+}) and their radiation damage induced by hadrons. While Mg^{2+} co-doping improves their light output, Ca^{2+} co-doping improves their fast to total (F/T) ratio. LuAG:Ce ceramic samples were irradiated at the Los Alamos Neutron Science Center (LANSCE), Los Alamos, NM, USA, by neutrons up to 6.7 × 10^{15} n_{eq}/cm^2 and by 24-GeV and 800-MeV protons at CERN PS-IRRAD up to 1.2 × 10^{15} p/cm^2 and at LANSCE up to 2.3 × 10^{14} p/cm^2, respectively. All samples show excellent radiation hardness with more than 90% of light after irradiation. The RIAC values induced by neutrons are found to be a factor of 2 smaller than lutetium–yttrium oxyorthosilicate (LYSO:Ce) crystals. The RIAC values induced by protons are also found a factor of 2 smaller than LYSO:Ce crystals in LuAG:Ce ceramic samples with good optical quality. Research and development will continue to develop LuAG:Ce scintillating ceramics with improved optical quality for future investigation.**

***Index Terms*— Ceramics, neutrons, protons, radiation effects, scintillators.**

I. Introduction

BRIGHT and fast cerium-doped $Lu_3Al_5O_{12}$:Ce ceramic scintillators (LuAG:Ce) have attracted a broad interest in the high-energy physics (HEP) community. Thanks to the development of nanotechnology, by using high-purity nanoparticles as starting powders [1]–[3], polycrystalline cubic LuAG ceramics without intrinsic birefringence may be fabricated as fine-grained, pore-free, and thus transparent scintillator with optical quality comparable to their single-crystal counterparts [4], [5]. Our previous work revealed that LuAG:Ce ceramic samples show a high light output of 1400 photoelectrons/MeV (p.e./MeV) measured by a Hamamatsu R2056 PMT with bialkali cathode, a fast primary decay time of 50 ns, and are radiation hard against an ionization dose up to 2.2 MGy and a proton fluence up to 3 × 10^{14} p/cm^2 [6]. A slow scintillation component with a decay time of about 1 μs may also be suppressed by alkaline earth metal co-doping, such as Mg^{2+} [7], [8] and Ca^{2+} [6], [9] co-doping. The fast to total (F/T) ratio, defined as the ratio between light output in 200- and 3000-ns gate, may reach 90% with Ca^{2+} co-doping [6].

Because of a simpler production technology at a lower temperature, a higher raw material usage, and no need for after-growth mechanical processing, LuAG:Ce ceramic scintillators may be more cost-effective in mass production when compared to widely used cerium-doped lutetium–yttrium oxyorthosilicate (LYSO:Ce) crystals. This material, therefore, is promising for future HEP experiments in a severe radiation environment, such as the high-luminosity large hadron collider (HL-LHC) and the future hadron circular collider (FCC-hh). One possible application would be to replace LYSO:Ce crystal plates as the active material for a shashlik sampling calorimeter for future HEP experiments [6].

One crucial issue, however, is its radiation hardness against hadrons. With a 5 × 10^{34} $cm^{-2} \cdot s^{-1}$ luminosity and a 3000-fb^{-1} integrated luminosity, the HL-LHC will present a radiation environment, where up to 1-MGy ionization dose, 6 × 10^{14} charged hadrons/cm^2, and 3 × 10^{15} fast neutron/cm^2 are expected for the endcap calorimeter [10]. Although radiation hardness against ionization dose [6], [9], [11]–[13] and protons [6], [13] was reported for LuAG:Ce crystals and ceramics, investigations are ongoing to understand radiation damage induced by hadrons, including both neutrons and protons, in LuAG:Ce ceramic samples.

Mg^{2+} (and Ca^{2+}) co-doped LuAG:Ce ceramic samples were irradiated by neutrons at the East Port of the Weapons Neutron Research facility of Los Alamos Neutron Science Center (LANSCE), Los Alamos, NM, USA, up to a 1-MeV equivalent neutron fluence of 6.7 × 10^{15} n_{eq}/cm^2. LuAG:Ce ceramic samples were also irradiated up to 1.2 × 10^{15} p/cm^2 by 24-GeV protons at the CERN PS-IRRAD and up to 2.3 × 10^{14} p/cm^2 by 800-MeV protons at the Blue Room of LANSCE. Their optical and scintillation properties were measured before and after irradiation at the Caltech HEP Crystal Laboratory with corresponding degradations compared to LYSO:Ce crystals. The effect of Mg^{2+} and Ca^{2+} co-doping

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TABLE I

LuAG:Ce Ceramic Scintillator Samples Used in the Neutron and Proton Irradiation Experiments

Sample ID	Dimension (mm^3)	EWLT (%)	200 ns L.O. (p.e./MeV)	F/T ratio (%)	Experiment	Fluence (cm^{-2})	Cooling Time (days)
n-1	Φ14.4×1	52.7	1474	66.6	LANSCE-7638	1.7×10^{15}	180
n-2	Φ14.4×1	61.5	1479	65.6	LANSCE-7638	3.4×10^{15}	145
n-3	Φ14.4×1	62.8	1514	73.5	LANSCE-7638	6.7×10^{15}	85
p-1	Φ14.4×1	67.6	1486	74.2	CERN PS-IRRAD	7.1×10^{13}	339
p-2	Φ14.4×1	14.2	1305	62.5	CERN PS-IRRAD	3.6×10^{14}	341
p-3	Φ14.4×1	32.1	1283	61.6	CERN PS-IRRAD	1.2×10^{15}	351
p-4	Φ17×1	9.3	1013	88.0	LANSCE-8051	2.4×10^{13}	260
p-5	Φ17×1	20.3	1049	89.0	LANSCE-8051	2.3×10^{14}	259

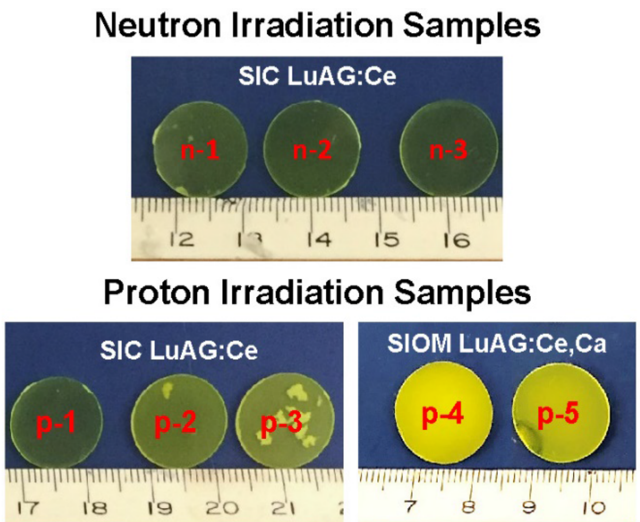

Fig. 1. Photos of three LuAG:Ce,Mg samples of Φ14.4 × 1 mm^3 from SIC used for the neutron irradiation (top), three LuAG:Ce,Mg samples of Φ14.4 × 1 mm^3 from SIC and two LuAG:Ce,Ca,Mg samples of Φ17 × 1 mm^3 from SIOM for the proton irradiation (bottom).

on slow component suppression and radiation hardness is also discussed.

II. Experimental Details

The top row of Fig. 1 shows three LuAG:Ce,Mg samples of Φ14.4 × 1 mm^3, n-1, n-2, and n-3, from the Shanghai Institute of Ceramics (SIC), Shanghai, China, for the neutron irradiation. The bottom row of Fig. 1 shows three LuAG:Ce,Mg ceramic samples of Φ14.4 × 1 mm^3, p-1, p-2, and p-3, from SIC and two LuAG:Ce,Ca,Mg samples of Φ17 × 1 mm^3, p-4 and p-5, from the Shanghai Institute of Optics and Fine Mechanics (SIOM), Shanghai, used in the proton irradiation. All samples were prepared by a solid-state reaction method. They were sintered above 1800 °C in vacuum, followed by annealing to eliminate point defects, such as the oxygen vacancies. The details for the fabrication process can be found elsewhere [14], [15]. As shown in the transmittance spectra, all samples show no obvious change of appearance after irradiation.

All available samples were used in this investigation without preselection. Development of transparent LuAG:Ce ceramics with excellent optical quality, however, requires extensive research and development. One crucial issue is optical quality. Porosity as low as 0.09% may results in a more than 30% drop in transmittance for 1-mm ceramic samples [16].

We notice that samples p2–p5 show poorer transparency when compared to other samples used in this investigation, which would lead to a larger uncertainty in the radiation-induced absorption coefficient data.

Three SIC LuAG:Ce,Mg samples were irradiated by neutrons in the Target four area in the neutron irradiation experiment LANSCE-7638. They were inside a sample holder at about 1.2 m away from the neutron production target in the East Port of LANSCE. The detailed description for the neutron irradiation site can be found elsewhere [17]. In addition, three groups of LYSO and BaF_2 crystals prepared by SIC were also irradiated at the same time together with the ceramic samples to facilitate a direct comparison. Two samples were used in each group for LYSO and BaF_2 crystals, so the corresponding results shown in this article are the average of two samples.

Three SIC LuAG:Ce,Mg samples were irradiated at the CERN PS-IRRAD Proton Facility by 24-GeV protons with a Gaussian shape and a full-width at half-maximum (FWHM) of about 12 mm. Two SIOM LuAG:Ce,Ca,Mg samples were irradiated in the Blue Room of LANSCE in the proton irradiation experiment LANSCE-8051 by 800-MeV protons with an FWHM of about 25 mm.

Table I summarizes the optical and scintillation performance for these samples before irradiation, the hadron fluence, and the cooling time after irradiation. Compared to the LuAG:Ce ceramic samples without co-doping, the Mg^{2+} co-doped LuAG:Ce samples show a higher light output, and the Ca^{2+} and Mg^{2+} co-doped samples show a higher F/T ratio. LuAG:Ce samples after hadron irradiation were radioactive. A cooling down time thus is required before they are safe to be shipped back to Caltech.

The neutron flux as a function of the neutron energy was calculated by using a Monte Carlo N-Particle eXtended (MCNPX) transport simulation package [18] developed by LANSCE. The corresponding 1-MeV equivalent neutron fluence was calculated according to the nonionizing energy loss (NIEL) cross section as a function of the neutron energy [19]. The details of these calculations and the neutron irradiation experiment can be found in our previous article on this subject [17]. The uncertainty of the neutron fluence is about 10%. The proton fluence at CERN PS-IRRAD was measured by dosimeters of 10 × 10 and 20 × 20 mm^3. The errors of the proton fluence at CERN and LANSCE are 7% and 10%, respectively.

All samples were wrapped with an aluminum foil to avoid optical bleaching. They were stored at room temperature after irradiation and were measured after cooling down. Since radiation damage in LuAG:Ce ceramic samples does not recover at room temperature, the measured damage level is not affected by the cooling time.

The transmittance was measured by using a Hitachi U3210 spectrophotometer with 0.2% precision. The emission-weighted longitudinal transmittance (EWLT) was calculated according to

$$\mathrm{EWLT} = \frac{\int T(\lambda)\mathrm{Em}(\lambda)d\lambda}{\int \mathrm{Em}(\lambda)d\lambda} \tag{1}$$

where $T(\lambda)$ and $\mathrm{Em}(\lambda)$ are the transmittance and emission spectra, respectively. The EWLT value provides a numerical representation of the transmittance over the entire emission spectrum. The RIAC value was calculated as

$$\mathrm{RIAC} = \frac{1}{l}\ln\left(\frac{T_0}{T_1}\right) \tag{2}$$

where l is the light path length along which the transmittance was measured. T_0 and T_1 are the transmittance values at 510 nm, which is the LuAG:Ce emission peak, measured before and after irradiation, respectively. The precision of the RIAC value depends on the light path length and its initial transmittance. It is about 5 m^{-1} for 1-mm-thick samples with good initial transparency, such as samples n-1, n-2, n-3, and p-1. For samples with poor initial transparency, such as p-2, p-3, p-4, and p-5, the precision of the RIAC data would be larger than 5 m^{-1}.

The light output before and after irradiation was measured by a Hamamatsu R2059 PMT with a grease coupling for 0.511-MeV γ-rays from a ^{22}Na source with a coincidence trigger. The systematic uncertainty of these measurements is about 1% [17].

III. Results and Discussions

A. Neutron Induced Radiation Damage

Fig. 2 shows the transmittance spectra measured before (black solid lines) and after (red dashed lines) a 1-MeV equivalent neutron fluence of 1.7, 3.4, and 6.7 × 10^{15} n_{eq}/cm^2, respectively, for the LuAG:Ce samples n-1 (top), n-2 (middle), and n-3 (bottom). Also shown in the figure are the LuAG:Ce emission spectra (blue dashes), the theoretical limit of LuAG:Ce transmittance (blue dots) calculated using the refractive index assuming multiple bounces and no internal absorption [20], and the numerical values of the EWLT for the three samples before (black) and after (red) irradiation. We note that no serious self-absorption in these LuAG:Ce ceramic samples with good optical quality. We also note that the transmittance of these ceramic samples does not approach the theoretical limit because of the scattering centers, such as the pores, inside the sample bulk. While scattering centers do not absorb scintillation light, they may affect the level of radiation-induced light output loss because of longer light path. The observed transmittance loss is very small, indicating excellent radiation hardness of the LuAG:Ce samples against neutrons up to 6.7 × 10^{15} n_{eq}/cm^2.

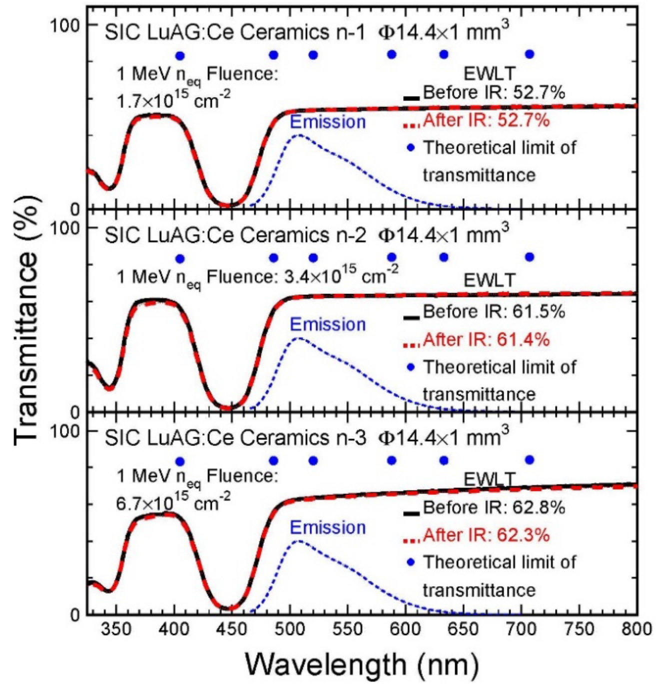

Fig. 2. Transmittance spectra are shown for three LuAG:Ce samples n-1 (top), n-2 (middle), and n-3 (bottom) before (black solid lines) and after (red dash lines) a 1-MeV equivalent neutron fluence of 1.7, 3.4, and 6.7 × 10^{15} n_{eq}/cm^2, respectively, at the East Port in the experiment LANSCE-7638.

Fig. 3 shows the light output as a function of the integration time measured before (black) and after (red) a 1-MeV equivalent neutron fluence of 1.7, 3.4, and 6.7 × 10^{15} n_{eq}/cm^2, respectively, at LANSCE for the LuAG:Ce samples n-1 (top), n-2 (middle), and n-3 (bottom). The observed light output loss is also small, confirming excellent radiation hardness of the LuAG:Ce samples against neutrons. In addition, the F/T ratio does not change up to 1-MeV equivalent neutron fluence of 6.7 × 10^{15} n_{eq}/cm^2.

Fig. 4 shows the RIAC values as a function of the 1-MeV equivalent neutron fluence for LYSO (black dots) and BaF_2 (blue triangles) crystals and LuAG:Ce (magenta triangles) ceramic samples irradiated at the East Port in the experiment LANSCE-7638 and the corresponding linear fits. Each RIAC value for LYSO and BaF_2 crystals is an average of two samples irradiated under the same condition, while only one LuAG:Ce sample was irradiated for each fluence. We note that the RIAC value of the LuAG:Ce ceramics against neutrons is about a factor of 2 smaller than that of the LYSO:Ce crystals.

Fig. 5 shows the normalized light output, defined as the light output after irradiation normalized to that before irradiation, as a function of the 1-MeV equivalent neutron fluence for SIC LYSO (black dots) and BaF_2 (blue triangles) crystal samples and three LuAG:Ce (magenta triangles) ceramic samples,

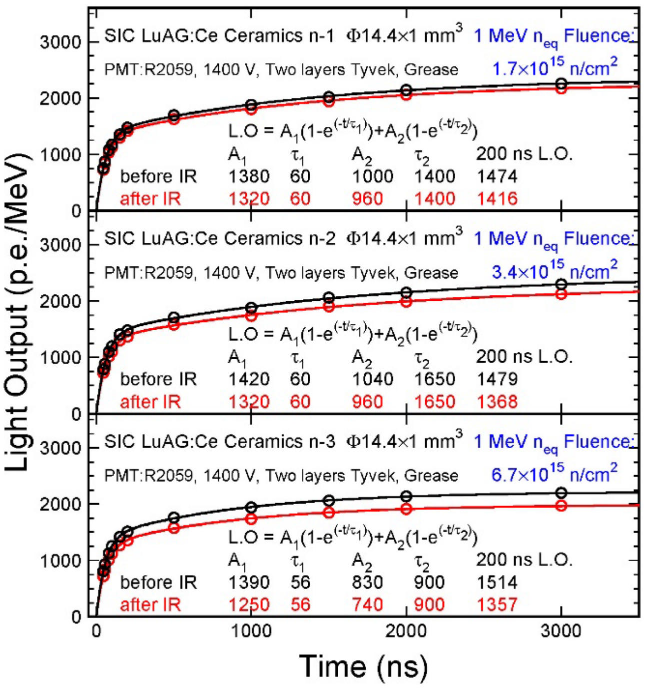

Fig. 3. Light output is shown as a function of the integration time for three LuAG:Ce samples n-1 (top), n-2 (middle), and n-3 (bottom) before (black) and after (red) a 1-MeV equivalent neutron fluence of 1.7, 3.4, and 6.7 × 10^{15} n_{eq}/cm^2, respectively, at the East Port in the experiment LANSCE-7638.

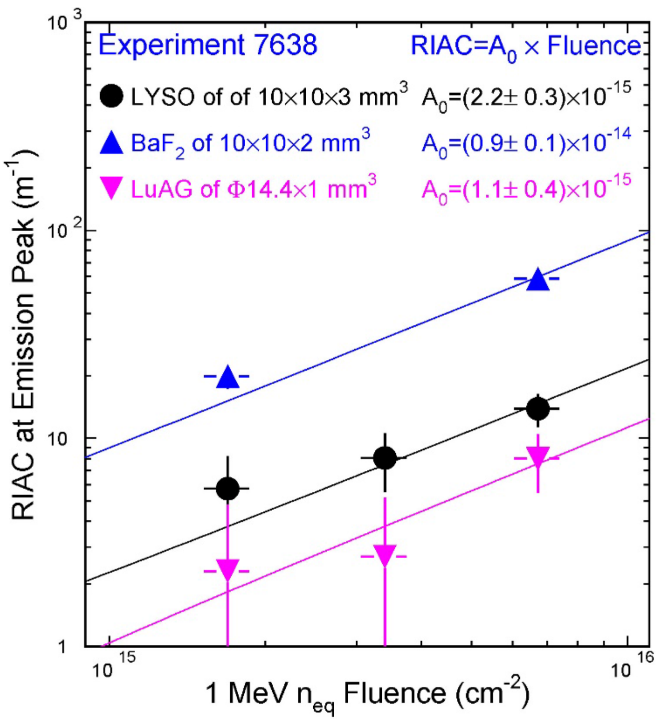

Fig. 4. RIAC values at the LuAG:Ce emission peak are shown as a function of 1-MeV equivalent neutron fluence for the SIC LYSO (black dots) and BaF_2 (blue triangles) crystal samples and three LuAG:Ce (magenta triangles) ceramic samples irradiated at the East Port in the experiment LANSCE-7638.

which were irradiated at the same time at East Port in the experiment LANSCE-7638. We note that all LYSO and BaF_2 crystals and LuAG:Ce ceramics survive a 1-MeV equivalent neutron fluence of 6.7 × 10^{15} n_{eq}/cm^2 well. The normalized light output values after a 1-MeV equivalent neutron fluence of

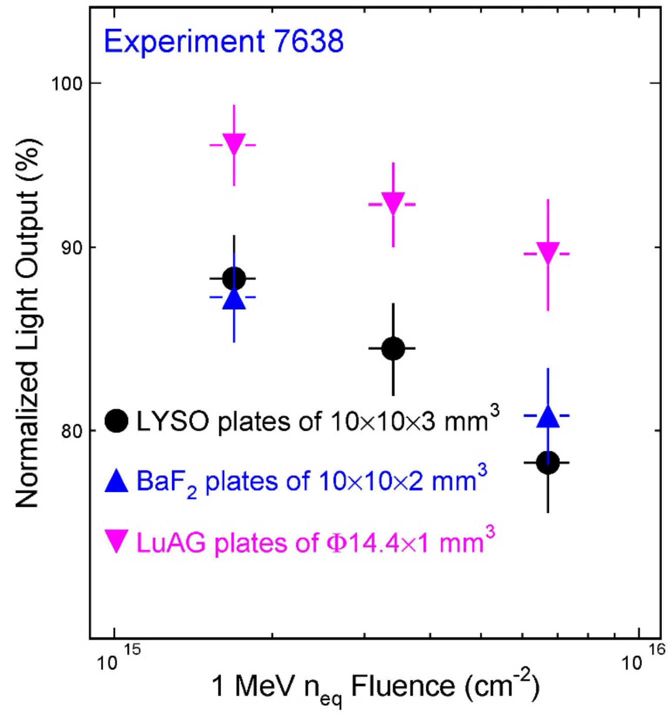

Fig. 5. Normalized light output is shown as a function of 1-MeV equivalent neutron fluence for SIC LYSO (black dots) and BaF_2 (blue triangles) crystal samples and LuAG:Ce (magenta triangles) ceramic samples irradiated at the East Port in the experiment LANSCE-7638.

6.7 × 10^{15} n_{eq}/cm^2 is 90% for the LuAG:Ce ceramic samples, which is higher than LYSO:Ce of 78% and BaF_2 of 81%. The larger light output degradation observed in the LYSO and BaF_2 crystal samples is partially due to their longer scintillation light path length because of their larger thickness.

B. Proton-Induced Radiation Damage

Fig. 6 shows the transmittance spectra measured before (black solid lines) and after (red dashed lines) a proton fluence of 1.2 × 10^{15} p/cm^2 at CERN PS-IRRAD (top) and 2.3 × 10^{14} p/cm^2 at LANSCE (bottom), respectively, for the Mg^{2+} co-doped LuAG:Ce ceramic sample SIC p-3 (top), and the Ca^{2+} and Mg^{2+} co-doped LuAG:Ce ceramic sample SIOM p-5 (bottom). Also shown in the figure are the LuAG:Ce emission spectra (blue dashes), the theoretical limit of transmittance (blue dots), and the numerical values of the EWLT before (black) and after (red) irradiation.

We note their poor optical quality shown as low transmittance caused by scattering centers in these LuAG:Ce ceramic samples, which affect more transmittance at shorter wavelengths.

Once again, the observed degradation of transmittance is very small, confirming excellent radiation hardness of the LuAG:Ce ceramic scintillators against protons up to 1.2 × 10^{15} p/cm^2.

Fig. 7 shows the light output as a function of integration time for the Mg^{2+} co-doped SIC LuAG:Ce ceramic sample p-3 (top), and the Mg^{2+} and Ca^{2+} co-doped SIOM LuAG:Ce ceramic sample p-5 (bottom) before (black) and after (red) a proton fluence of 1.2 × 10^{15} p/cm^2 at CERN PS-IRRAD

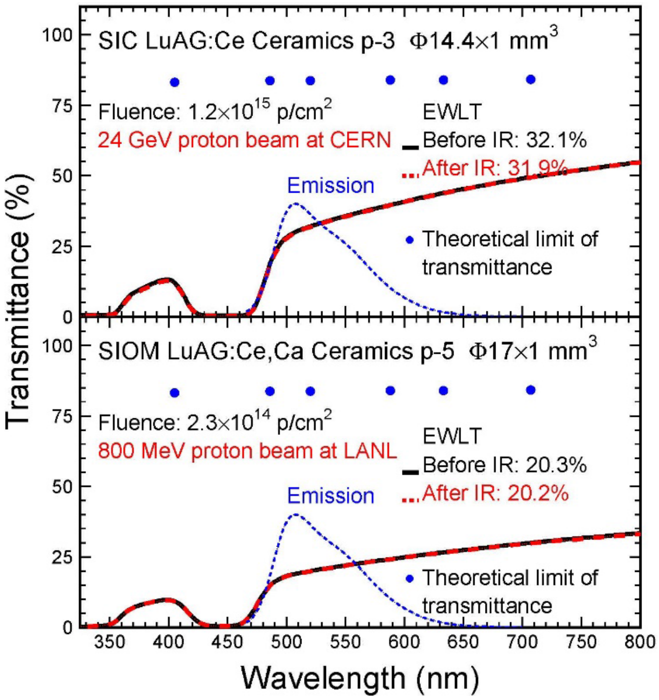

Fig. 6. Transmittance spectra are shown for the Mg^{2+} co-doped LuAG:Ce ceramic sample SIC p-3 (top), and the Mg^{2+} and Ca^{2+} co-doped LuAG:Ce ceramic sample SIOM p-5 (bottom) before (black) and after (red) a proton fluence of 1.2×10^{15} p/cm^2 at CERN PS-IRRAD (top) and 2.3×10^{14} p/cm^2 at LANSCE (bottom), respectively.

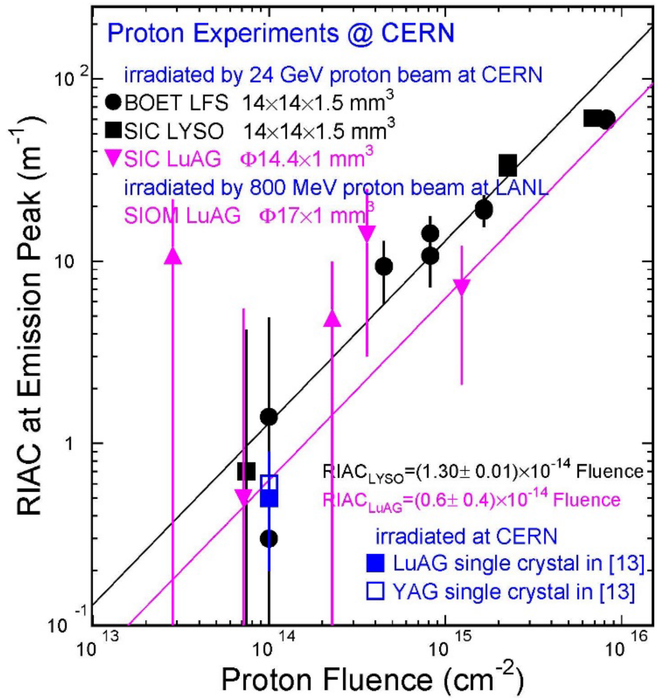

Fig. 8. RIAC values as a function of the proton fluence for the LYSO/LFS (black dots) crystal and LuAG:Ce (magenta triangles) ceramic samples irradiated by both 24-GeV protons at CERN PS-IRRAD and 800-MeV protons at LANL and the corresponding linear fits.

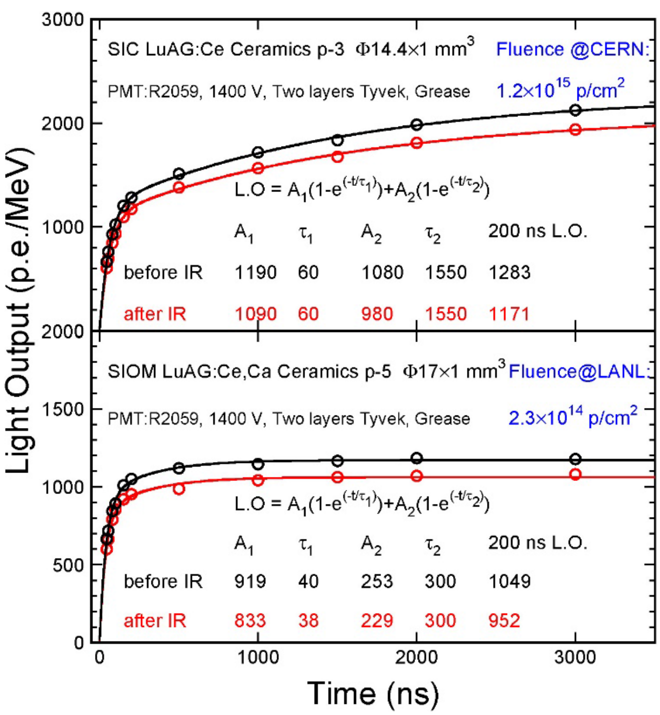

Fig. 7. Light output is shown as a function of integration time for the Mg^{2+} co-doped LuAG:Ce ceramic sample SIC p-3 (top), and the Mg^{2+} and Ca^{2+} co-doped LuAG:Ce ceramic sample SIOM p-5 (bottom) before (black) and after (red) a proton fluence of 1.2×10^{15} p/cm^2 at CERN PS-IRRAD (top) and 2.3×10^{14} p/cm^2 at LANSCE (bottom), respectively.

Proton Facility (top) and 2.3×10^{14} p/cm^2 at the blue room of LANSCE (bottom), respectively. Once again, about 90% light remains for both the samples. We note that the F/T ratio does not change for both samples. We also note that the Mg^{2+} co-doping improves the light output, and the Ca^{2+} and Mg^{2+} co-doping improves the F/T ratio.

Fig. 8 shows the RIAC values as a function of the proton fluence for the LYSO/LFS (black dots) crystal and LuAG:Ce (magenta triangles) ceramic samples irradiated by 24- and 800-GeV protons at CERN PS-IRRAD and the Blue Room of LASCE, respectively, and the corresponding linear fits. The results for the SIC LYSO and Beijing Opto-Electronics Technology Company Ltd. (BOET), Beijing, China, lutetium fine silicate (LFS) crystals were reported in our previous publications [21], [22]. Only one sample was irradiated for each data point.

We note that the proton-induced RIAC values for the LuAG:Ce ceramic samples with good initial transmittance, for example, samples p-1 and p-3, appear also about a factor of 2 smaller than that of the LYSO crystals. We also note that the proton-induced RIAC values are large and with large error bars for some LuAG:Ce ceramic samples, for example, p-2, p-4, and p-5, which is due to their poor optical quality. Also shown in the figure is the fit result of RIAC (m^{-1}) $= (0.6 \pm 0.4) \times 10^{-14}$ F_p which agrees well with 0.5 ± 0.3 m^{-1} reported in [13] for LuAG crystal samples after 10^{14} F_p by 24-GeV protons. This factor of 2 is consistent with the neutron irradiation data shown in Fig. 4.

Fig. 9 shows the normalized light output as a function of the proton fluence for the LYSO/LFS crystals (black dots) and LuAG:Ce ceramic samples (magenta triangles) irradiated by 24-GeV and 800-MeV protons at CERN PS-IRRAD and the Blue Room of LANSCE, respectively. Both LYSO crystals and LuAG:Ce ceramics survive a proton fluence of 10^{15} p/cm^2.

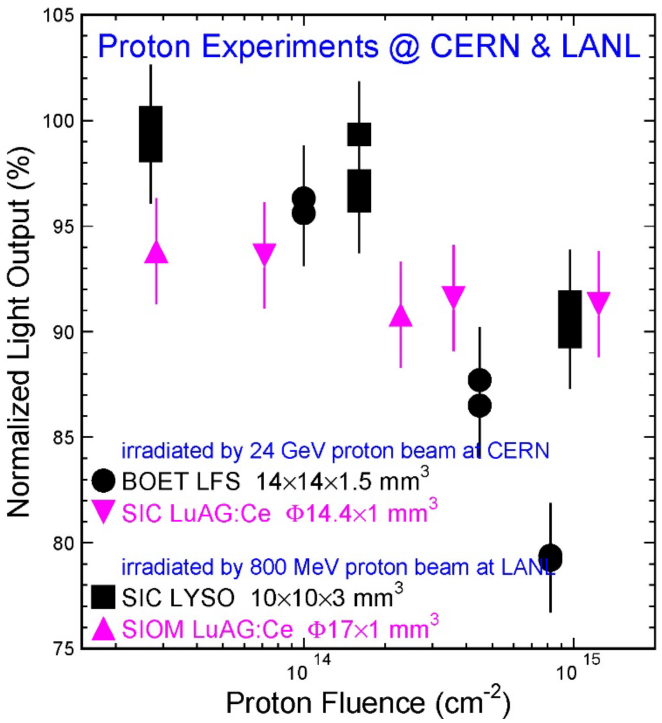

Fig. 9. Normalized light output is shown as a function of proton fluence for the LYSO/LFS crystals (black dots) and LuAG:Ce (magenta triangles) ceramic samples irradiated by 24-GeV protons at CERN PS-IRRAD and by 800-MeV protons at LANSCE.

The normalized light output value after a proton fluence of 1.2×10^{15} p/cm^2 is 91% for the LuAG:Ce ceramic samples, which is about the same values as the 90% observed for SIC LYSO crystals after a proton fluence of 9.7×10^{14} p/cm^2. LuAG:Ce ceramic thus may be considered as another promising inorganic scintillator for future HEP experiments in a severe radiation environment, such as the HL-LHC and the proposed FCC-hh.

IV. Summary

Ca^{2+} and Mg^{2+} co-doped LuAG:Ce ceramic samples were fabricated and irradiated by neutrons at the East Port of LANSCE up to 6.7×10^{15} n_{eq}/cm^2 and by 24-GeV and 800-MeV protons at the CERN PS-IRRAD up to 1.2×10^{15} p/cm^2 and at the Blue Room of LANSCE up to 2.3×10^{14} p/cm^2, respectively. Mg^{2+} co-doping in LuAG:Ce ceramics improves light output, while Ca^{2+} and Mg^{2+} co-doping improves the F/T ratio.

The RIAC values of LuAG:Ce ceramics were found to have a factor of 2 smaller than LYSO:Ce crystals against neutrons. Similar result was also obtained against protons for the LuAG:Ce ceramic samples with good optical quality. We plan to irradiate the LuAG:Ce ceramic samples of improved optical quality to reduce the uncertainties in the RIAC data in our future investigation.

More than 90% of the light output remains in 1-mm-thick samples after fluences of up to 6.7×10^{15} n_{eq}/cm^2 and 1.2×10^{15} p/cm^2 LuAG:Ce ceramic scintillators which makes this material promising for applications at the HL-LHC and the proposed FCC-hh.

Research and development will continue to develop Ca^{2+} and Mg^{2+} co-doped LuAG:Ce ceramic scintillators with improved optical quality, F/T ratio, and radiation hardness.

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