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***Abstract*—Transparent polycrystalline ceramic scintillators based on the garnet structure and incorporating gadolinium for high stopping power are being developed for use in gamma spectrometers. Optimization of energy resolution for gamma spectroscopy involves refining the material composition for high stopping and high light yield, developing ceramics fabrication methodology for material homogeneity, as well as selecting the size and geometry of the scintillator to match the photodetector characteristics and readout electronics. We have demonstrated energy resolution of 4% at 662 keV for 0.05 cm^3 GYGAG(Ce) ceramics with photodiode readout, and 4.9% resolution at 662 keV for 18 cm^3 GYGAG(Ce) ceramics and PMT readout. Comparative gamma spectra acquired with GYGAG(Ce) and NaI(Tl) depict the higher resolution of GYGAG(Ce) for radioisotope identification applications. Light yield non-proportionality of garnets fabricated following different methods reveal that the fundamental shapes of the light yield dependence on energy are not intrinsic to the crystal structure, but may instead depend on trap state distributions. With exposure to 9 MeV Brehmsstrahlung radiation, we also find that GYGAG(Ce) ceramics exhibit excellent radiation hardness.**

***Index Terms*—Gamma-ray spectroscopy, Garnets, Scintillators**

I. INTRODUCTION

Advantages of the new Gd-based transparent ceramic garnet scintillators include: (1) high fast light yield of >40,000 Ph/MeV and principal decay of ~100 ns, (2) photopeak efficiency superior to NaI(Tl) and $LaBr_3$(Ce), (3) excellent light yield proportionality, (4) ease of uniform fabrication via ceramics processing, and (5) no intrinsic radioactivity. In our earlier reports [1-3], we measured energy resolution of 4.5% at 662 keV with PMT readout and 4.2% with Silicon photodiode readout, utilizing small (0.05-2 cm^3) GYGAG(Ce) ceramics.

Identification of the ideal garnet host for Ce-activation providing the highest light yield and bluest emission for PMT readout requires analysis of the conduction and valence band positions, as well as the positioning of the Ce transition within the band gap [4]. When compared to e.g. YAG or LuAG, the Gd-based garnets incorporating Ga provide favorable crystal field for Ce^{3+} emission at relatively short wavelength, as well as the ability for excitation to migrate on Gd^{3+} as a stoichiometric sensitizer for Ce^{3+}. Both Gd and Tb offer comparable stopping to Lu, without radioactivity. However, Tb-based garnets exhibit slower decay times than Gd-garnets making them less desirable for gamma spectroscopy [5].

Transparent ceramics fabrication allows production of monolithic, fully-dense optics without macroscopic Ce-doping gradients. Ce-doping gradients are well-known in single crystal garnets, due to the poor match in ionic radius between Ce^{3+} (103 pm) and that of the substitutional sites of e.g. Gd^{3+} (94 pm) and Y^{3+} (88 pm), leading to a segregation coefficient of Ce in GGAG of 0.36 and in YAG of 0.082 [6, 7]. Although high light yields and good energy resolution in small single crystal samples have been obtained for Ce-doped Gd-garnets recently, large size crystals exhibit significant Ce gradients, resulting in light yield non-uniformity and impacting the performance for gamma spectroscopy [8].

Even scintillators without doping gradients or material non-uniformity can suffer degradation in performance in large sizes due to optical light trapping, when activators with small Stokes' shifts are employed. This is well-documented for the single crystal scintillator, SrI_2(Eu) [9-11]. In contrast to SrI_2(Eu), self-absorption by Ce in GYGAG(Ce) results in red-shifting of the emission that diminishes the quantum efficiency of conversion with PMT readout, due to the loss in bialkali photocathode sensitivity at longer wavelengths. We have therefore tried to identify the minimum Ce-doping concentration that still provides high light yield in GYGAG.

We have fabricated GYGAG(Ce) in large sizes of > 35 cm^3 (2 in^3). The garnet ceramics exhibit excellent mechanical toughness and are easily cut into small cuboids for integration with silicon photodiodes. Gamma spectroscopy with large size GYGAG(Ce) using PMT readout and small size GYGAG(Ce)

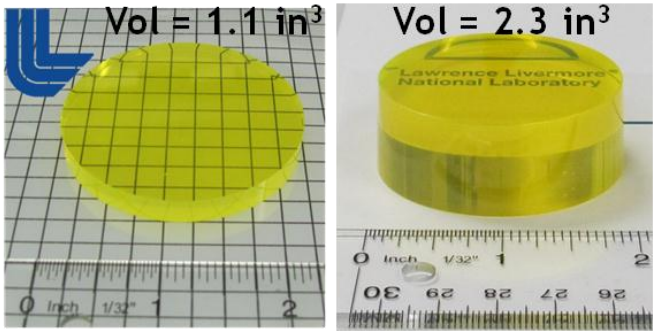

Fig. 1. Photos of the GYGAG(Ce) ceramics studied in this paper (left) 18 cm^3 (2.2" diameter x 0.3" thick) and (right) 38 cm^3 (2" diameter x 0.73" thick).

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with photodiode readout indicate both approaches can provide high resolution gamma spectroscopy. Initial results comparing small ceramic and single crystal GGAG(Ce) scintillation performance are also presented here.

The Scintillator Light Yield Non-proportionality Characterization Instrument (SLYNCI) [12-14] provides valuable feedback to compare various garnet structures and identify the best ceramics processing methods. We have studied a range of garnets formed by different methods to determine their impact on light yield proportionality.

II. Methods

A. Transparent Ceramics Fabrication

GYGAG(Ce), $Gd_{1.5}Y_{1.5}Ga_{2.2}Al_{2.8}O_{12}$(Ce), and GGAG(Ce), $Gd_3Ga_{2.2}Al_{2.8}O_{12}$(Ce), nanoparticles were synthesized via the flame spray pyrolysis (FSP) method by Nanocerox. Nanoparticles were suspended with organic binders in an aqueous solution, gel cast in a mold, and organics were removed by a heat treatment at 1050°C in air. Green bodies thus prepared were sintered under a vacuum of $<2\times10^{-6}$ Torr at 1600°C for 2 h in a tungsten element vacuum furnace, reaching closed porosity and densities of approximately 97%. The sintered samples were then hot isostatically pressed under 200 MPa of inert argon gas pressure at 1650°C for 4 h in a tungsten element HIP. Further details are available in [15].

B. Scintillator Absorption and Radioluminescence Spectra

Optical absorption spectra were obtained with a Thermo Evolution 220 UV-visible spectrometer. Radioluminescence spectra were acquired using a $^{90}Sr/^{90}Y$ source (~1 MeV average beta energy), and spectra were collected with a Princeton Instruments/Acton Spec 10 spectrograph coupled to a thermoelectrically cooled CCD camera.

C. Scintillation Characterization with PMT Readout

We measured pulse height spectra with ^{137}Cs and ^{60}Co sources with various scintillators. Hamamatsu R6231-100 and 6232-100 photomultiplier tubes, employed for all PMT measurements, were connected to an Ortec 113 preamplifier. The signals were shaped with a Tennelec TC 244 spectroscopy amplifier (shaping time typically 4 μs for Ce-doped garnets) and then recorded with an Amptek MCA8000-A multi-channel analyzer. Effective light yields were determined by reference to a standard YAG(Ce) ceramic from Baikowski, and energy resolution was obtained by the use of Gaussian peak fitting in Igor Pro. A Bridgeport Instruments eMorpho was used to record scintillation pulse traces, and those pulses which constituted a full-energy deposition were selected, and the scintillation decay obtained by averaging 10 traces from the selected pulses, and fitting the decay to a double exponential using Igor Pro.

D. Gamma Spectra with Silicon Photodiode Readout

Cuboids of GYGAG(Ce) were optically coupled to a silicon photodiode array, as described in [3]. We measured pulse height spectra with a ^{137}Cs source and employed a custom ASIC for signal readout and co-adding of coincident events.

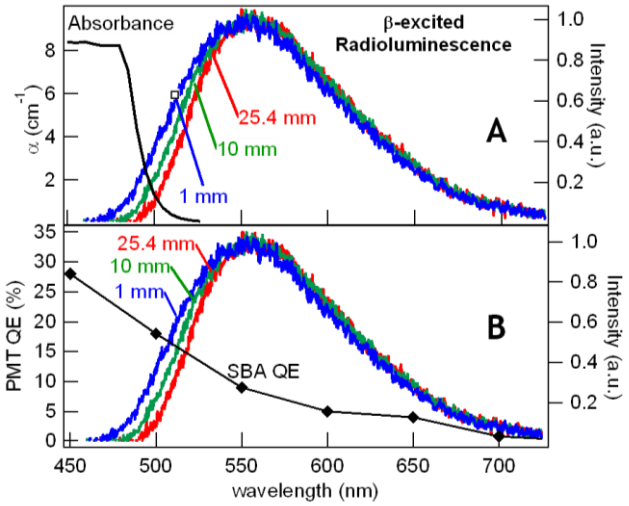

Fig. 2. (A) Absorption spectrum for GYGAG(Ce = 0.02) and beta-excited radioluminescence spectra acquired with 3 samples, 1, 10 and 25.4 mm thick. Significant red-shifting is observed as a function of thickness. Absorption cross-section is quantitative. (B) Super-bialkali photocathode responsivity shown in comparison with the same three radioluminscence spectra.

E. Light Yield Non-proportionality Measurements

The electron non-proportionality was obtained using the Scintillation Light Yield Non-proportionality Characterization Instrument (SLYNCI) using a Compton Coincidence technique as described in [16,17].

F. Radiation Hardness Study

Scintillator samples were exposed to radiation emitted by a Varian M9 LINAC Brehmsstrahlung source, nominal 9 MeV. Samples were placed as close as possible to the source and exposed to a flux of 10 kRad/min for a total dose of 53 MR, as characterized using an ion chamber.

III. Results and Discussion

A. Scintillator Absorption, Radioluminescence and Decay

The absorption spectrum measured for a 1 mm thick sample of GYGAG(Ce = 0.02) is shown in Figure 2, where it has been converted into the absorption cross section as a function of wavelength (signal saturates below 475 nm). The effect of the relatively small Stokes' Shift on the radioluminescence is observable when comparing the spectra obtained with identical samples of GYGAG(Ce = 0.02) of thicknesses 1 mm, 1 cm and 2.54 cm Figure 2). Light trapping (self-absorption and subsequent re-emission to longer wavelengths) and ultimately non-uniformity in light collection is the result.

Previous studies have detailed the effect of optical light trapping and methods for mitigation for Eu-doped Strontium Iodide [9-11, 18], however, that scintillator emits in the blue, where PMT quantum efficiency (QE) is high and nearly invariant. In contrast, between 475 and 650 nm, the PMT QE decreases by a factor of about 4, thus the net result of the light trapping and red-shifting emission as a function of size is decreased effective light yield, as well as position dependent

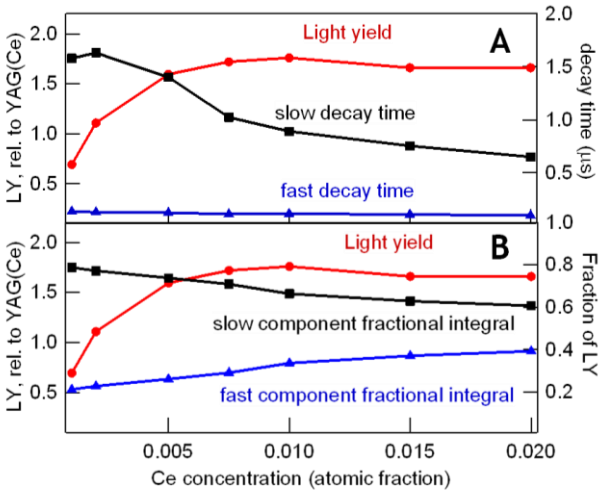

Fig. 3. (A) Seven GYGAG(Ce) samples fabricated with Ce = 0.001-0.02 reveal a consistent fast decay time (~100 ns) and a slow decay that decreases from 1.6 μs to 0.6 μs between Ce = 0.001 and 0.02. Light yield (8 μs shaping time) is nearly constant for Ce > 0.005. (B) The integrated fractional intensity of the two components indicate that the relative contribution of the fast component increases with Ce concentration.

inhomogeneity in the effective light yield.

In order to mitigate light-trapping and enable higher effective light yields and better light collection homogeneity, we undertook a Ce-doping study to identify the lowest Ce concentration that could provide high light yield with sufficiently fast decay for standard analog shaping amplifiers (<12 μs). GYGAG(Ce) exhibits a double exponential decay, as shown in [2]. Figure 3 shows that the light yield reaches its maximum just above Ce = 0.005 and does not decrease with concentration, up to Ce = 0.02. The fast decay, assigned to the Ce intrinsic lifetime in GYGAG, remains consistent at ~100 ns, though its relative amplitude increases steadily with concentration. The slow component, assigned to the effect of energy migration via Gd^{3+} as well as shallow traps becomes faster as the Ce concentration increases, while its relative

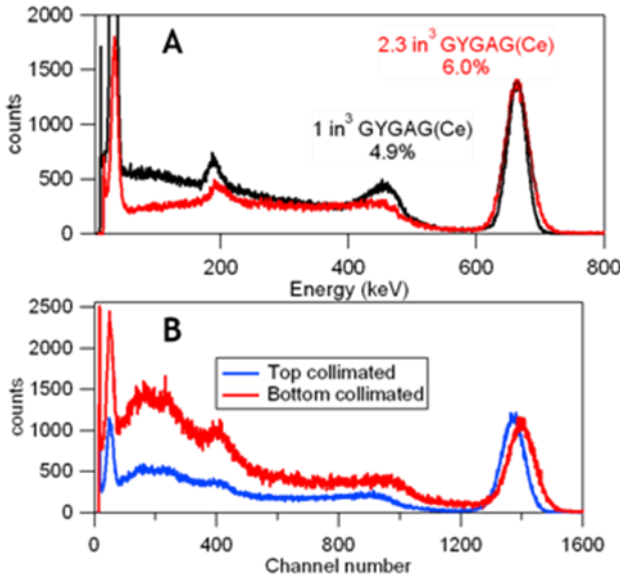

Fig. 4. (A) Pulse height spectra acquired with ^{137}Cs using the 1in^3 and 2.3 in3 GYGAG(Ce= 0.01) ceramics shown in Fig. 1. (B) Pulse height spectra obtained with a collimated ^{137}Cs source exhibits a 4% change in photopeak centroid, indicating significant light-trapping, leading to degradation of energy resolution.

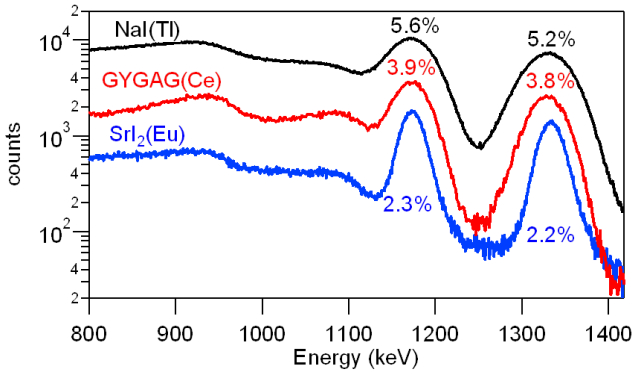

Fig. 5. Gamma spectra of ^{60}Co using 1 in^3 scintillators of SrI_2(Eu) GYGAG(Ce) and NaI(Tl).

amplitude decreases.

B. Gamma Spectroscopy

Scintillator-based gamma ray spectrometers are often used when large volumes for high sensitivity are required for detection of weak sources. We therefore are working to scale up GYGAG(Ce). Figure 4 shows the pulse height spectra with ^{137}Cs acquired with the 1 in^3 and 2.3 in^3 scintillators shown in Figure 1. The resolution is found to be degraded from 4.9% at 662 keV to 6% in the larger scintillator. It is notable that for the 2.3 in^3 GYGAG(Ce), instead of the expected Gaussian-shaped photopeak at 662 keV, an asymmetric tail to high energy is observed. This is due to light trapping, and is further revealed in Figure 4B, which shows that the effective pulse height is decreased by ~4% for the spectrum acquired at the

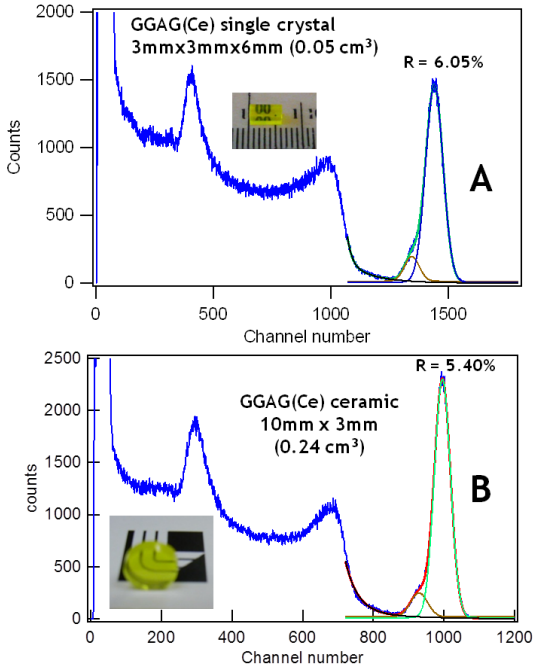

Fig. 6. (A) Pulse height spectrum acquired with ^{137}Cs with a 0.05 cm^3 GGAG(Ce) single crystal grown by Furukawa. (B) Pulse height spectrum acquired with ^{137}Cs with a 0.24 cm^3 GGAG(Ce) transparent ceramic fabricated at LLNL.

top of the sample, compared with when the collimated source irradiates the bottom of the scintillator near the PMT.

Comparative gamma spectroscopy obtained with three 1 in^3 scintillators using a ^{60}Co source is shown in Figure 5. The energy resolution obtained with GYGAG(Ce) is considerably better than that of NaI(Tl), but is surpassed by $SrI_2(Eu)$.

The photofractions (ratio of the photopeak efficiency over the total gamma stopping) of NaI(Tl) : GYGAG(Ce) : GGAG(Ce) are calculated to be 1 : 1.1 : 1.4, therefore GGAG(Ce) will offer higher detection efficiency for a given size. For this reason, we are also studying the fabrication and scintillation properties of GGAG(Ce). Pulse height spectra acquired with ^{137}Cs for a GGAG(Ce) single crystal from Furukawa and a GGAG(Ce) ceramic fabricated at LLNL are shown in Figures 6A and 6B, respectively. Comparable energy resolution and light yield to GYGAG(Ce) is obtained.

C. Gamma Spectra with Silicon Photodiode Readout

The Digirad gamma camera described in [3], is based on 2.8 x 2.8 x 6 mm scintillator cuboids coupled to a photodiode array, normally employing CsI(Tl). Here, we have studied in more detail the integration of GYGAG(Ce) as a possible improvement, due to its faster scintillation decay and better energy resolution over CsI(Tl). To further improve detection efficiency, we have configured the readout electronics with a custom ASIC to obtain the energy histogram comprised of both single events and double events arriving in temporal coincidence (within a 2 microsecond coincidence window). A further advantage of recording coincidence events is the ability to utilize Compton imaging to obtain directional information about the radioactive source.

Detection efficiency increased by ~35% by including double coincidence events, but energy resolution at 662 keV was degraded from 4.0% to 4.7% for GYGAG (see Figure 7). For CsI(Tl), studied in the same configuration, 5.0% was obtained for single hits, degrading to 5.6% for the summed spectrum of single and double hits. Resolution degradation observed in the co-added histogram is thought to be due to the doubling of both the electronic readout noise and the photodetector dark noise for co-added events. Additional

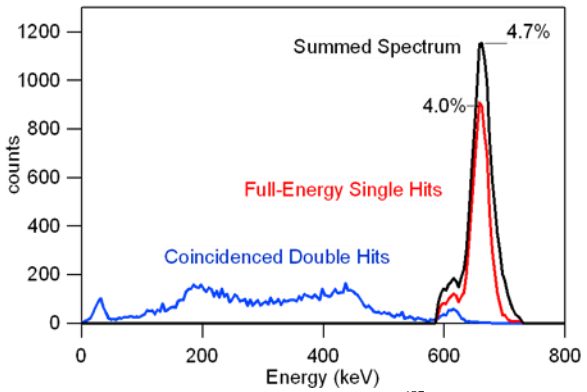

Fig. 7. Pulse height spectra acquired with ^{137}Cs with a 0.05 cm^3 GYGAG(Ce) ceramic fabricated at LLNL, using silicon photodiode readout. Energy resolution for photopeak events collected in single GYGAG(Ce) pixels is 4.0% at 662 keV (red trace), and when double hits arriving in coincidence (blue trace) are co-added, and then combined with the single event spectrum, the black trace is obtained.

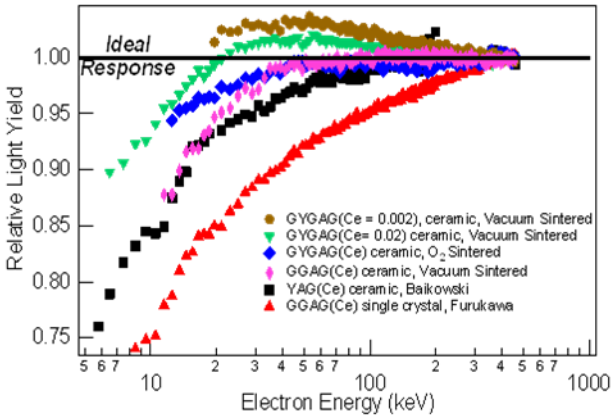

Fig. 8. SLYNCI data for GYGAG(Ce), GGAG(Ce) and YAG(Ce) samples indicate the a strong effect of the fabrication conditions, as well as changes with Ce-doping level. This suggests that the nature and concentration of defects and recombination centers differ markedly between samples.

contributions may arise from imperfect normalization of signals from each pixel and the difficulty of applying an exact correction for energy-dependent light yield non-proportionality.

D. Scintillation Light Yield Non-proportionality

It is well-known that the light output of scintillators is not rigorously proportional to the amount of energy deposited in the scintillator [12-14], and that scintillator energy resolution is poorer than that predicted by the photon statistics of the deposited energy. Figure 8 and Table I show the SLYNCI measurements acquired with several Ce-doped garnet transparent ceramics and single crystals.

We find that the methods of fabrication of the garnet scintillator have a strong effect on the energy dependent light yield proportionality, indicating that atomistic defects, resulting in trapping and de-trapping during carrier migration, exciton formation and transport substantially control the light

TABLE I
NON-PROPORTIONALITY MEASURED FOR SEVERAL CE-DOPED GARNETS

Scintillator	$\eta_{e/h}$ [a]	$(dE/dx)_{BIRKS}$ [b]	R_{NP}(%) predicted	LY (Ph/keV)
GYGAG(Ce=0.02) Ceramic, Vacuum sintered	18.7 %	625 MeV/cm	2.49	50
GYGAG(Ce=0.002) Ceramic, Vacuum sintered	18.7	909	2.15	33
GYGAG(Ce=0.02) Ceramic, O_2 sintered	10	1000	1.72	40
YAG(Ce) Ceramic	10	526	2.44	30
GGAG(Ce) Ceramic Vacuum sintered	21	400	3.08	50
GGAG(Ce) Single crystal	0	434	3.12	60

[a]Efficiency of exciton formation after carrier thermalization [14]
[b]Exciton-exciton annihilation via Birks' mechanism [14].

yield proportionality. Previously we described considerable variation among the light yield proportionality for multiple crystals of NaI(Tl) [12], but few other studies of the effect of fabrication conditions on light yield proportionality have yet been reported. In particular, for the transparent ceramic garnets, a fraction of the excitons that successfully migrate to Ce^{3+} are formed after thermalization, as is indicated by their large η_{eh} values in the 10-19% range. Exciton-exciton annihilation, thought to occur in the sub-nanosecond regime in the scintillation process, is mitigated in the oxygen sintered ceramic, while being most severe in the GGAG(Ce) single crystal.

E. Radiation Hardness Study

Color center formation upon exposure to ionizing radiation is a concern for medical and high energy physics applications. To address this, several scintillators were exposed to radiation from a 9 MeV Brehmsstrahlung source, and their optical absorption before and after exposure measured. Figure 9 shows that the GYGAG(Ce) ceramics did not darken with exposure, while a glass scintillator and a plastic scintillator exhibit marked new absorption bands. The total dose administered to the samples, all exposed in a single experiment, was 53 MR.

IV. Conclusion

GYGAG(Ce) transparent ceramics have been fabricated with excellent transparency and uniformity at sizes >2 in^3. Gd-based garnet scintillators offer energy resolution and stopping power superior to NaI(Tl) and are free of intrinsic self-radioactivity. Both PMT and silicon photodiode readout may be used for instrument development. Detailed studies of the trap state energetics and distribution will be useful to understand the strong dependence on fabrication conditions observed in the light yield non-proportionality, to optimize the composition and fabrication conditions in order to achieve high energy resolution, large volume Gd-garnet scintillators. Integration of GYGAG(Ce) transparent ceramics into high radiation rate experiments should be straightforward as no measurable darkening was observed even at high accumulated dose.

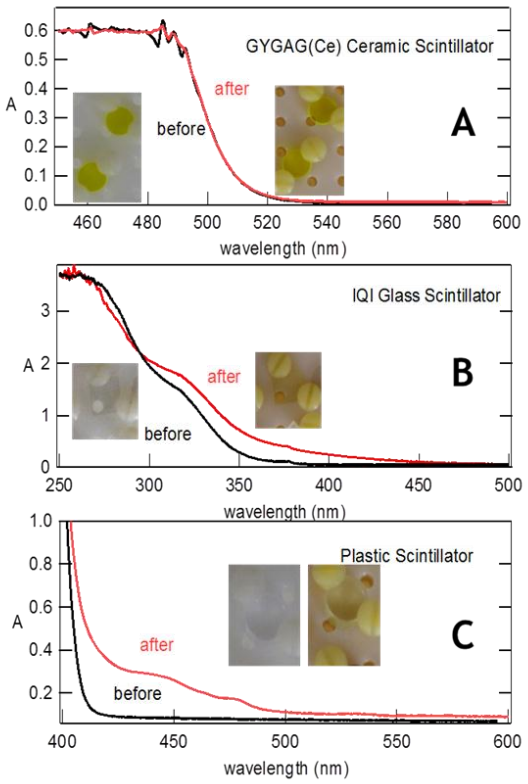

Fig. 9. (A) For GYGAG(Ce) ceramics, no radiation-induced darkening / color centers are observed In contrast, (B) glass and (C) plastic scintillators colored significantly.

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