

The Temperature Dependence of Gamma-Ray Responses of YAG:Ce Ceramic Scintillators

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Abstract—The temperature dependence (from -20 to $+20$ degrees C) of gamma-ray irradiated light outputs, energy resolutions, and decay time profiles of three YAG:Ce poly-ceramic scintillators are studied. The Ce concentrations are 0.5, 0.05, and 0.005 mol%. The relative light yield of the YAG:Ce with 0.5 mol% with a $2 \mu\text{s}$ shaping time was measured as 1 : 1.08 : 1.14 at $+20$, 0, and -20 degrees C, respectively, including the temperature dependence of the phototube (-0.2% /degree). The energy resolution stays almost constant at 7.2% for 662 keV gamma-rays. The ceramic with 0.05 mol% shows the almost same properties, while the light yield of that with 0.005 mol% is 2–4 times lower (hence the energy resolution becomes 14–19%). All the scintillators exhibit good linearities within $\sim 1\%$ between the light output and the irradiated gamma-ray energy from 59.5 keV to 662 keV. The decay time constants of the dominant decay components are about 80 ns and 300 ns at $+20$ degrees C. As the temperature increases from -20 to $+20$ degrees C, the effective decay of all the ceramics becomes faster, because the decay time constants and fractions of the slower components shorten and decrease, respectively. This result suggests that carriers which are captured in shallow traps before transferring excitation to Ce ions can escape the traps more easily at higher temperatures. Considering the decrease of the total light yield toward higher temperatures, it is thought that thermal quenching starts to dominate the temperature dependence of the ceramic YAG:Ce around the room temperature. The 0.5 mol% scintillator shows a lower quenching energy than the 0.05 mol% one. This can be explained in terms of self absorption of Ce emission.

Index Terms— Ce^{3+} , ceramics, quenching, scintillators, temperature dependence, $\text{Y}_3\text{Al}_5\text{O}_{12}$ (YAG).

I. INTRODUCTION

RECENTLY, Konoshima Chemical has developed a new sintering method for poly-crystalline ceramics and succeeded in producing highly homogeneous YAG (yttrium aluminum garnet, $\text{Y}_3\text{Al}_5\text{O}_{12}$) ceramics [1], [2]. With the effective atomic number and density 35 and 4.57 g/cm^3 , respectively, single-crystal YAG was known to scintillate when doped with

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Color versions of Figs. 1–3, 5, and 6 are available online at <http://ieeexplore.ieee.org>.

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TABLE I
SIZES OF CERAMIC YAG:CE AND CSI:TL

Sample	Diameter [mm] \times Thickness [mm]
YAG:Ce (0.5 mol%)	$\phi \sim 20 \times t \sim 2.0$
YAG:Ce (0.05 mol%)	$\phi \sim 20 \times t \sim 1.5$
YAG:Ce (0.005 mol%)	$\phi \sim 20 \times t \sim 1.4$
CsI:Tl	$15 \times 15 \times 15$

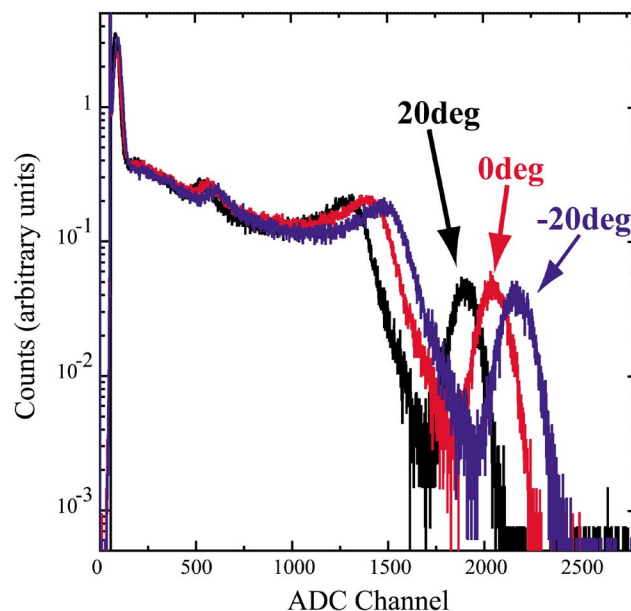


Fig. 1. Energy spectra of ^{137}Cs obtained with the ceramic YAG:Ce (0.5 mol%) at $+20$, 0 and -20 degrees C in a direction from left to right.

Ce, but the doping was possible only to very shallow depths from the surface. The newly developed ceramics can be doped uniformly without relying on the thickness. Moreover, they have a higher mechanical strength, larger design flexibility and almost the same transmittance as single crystals, and is expected to be a good scintillator with Ce doping, YAG:Ce. The YAG crystal also has a possibility that Y^{3+} is replaced with the heavier Gd^{3+} or Lu^{3+} atoms.

In our previous work [3], we studied basic properties of these YAG:Ce poly-crystalline materials, and found that they show scintillation performance as good as their single-crystal counterpart. In the present paper, we further evaluate temperature dependence of gamma-ray responses, namely light yields, linearities, energy resolutions and decay time profiles, of the YAG:Ce poly-ceramic scintillators.

Table I lists samples of ceramic YAG:Ce with different Ce concentrations, namely 0.5, 0.05, and 0.005 mol%. They are

TABLE II
LIGHT YIELDS AND ENERGY RESOLUTIONS OF THE YAG:Ce CERAMICS

YAG:Ce [mol%]	degrees C		
	+20	0	-20
0.5	0.37 / 7.2%	0.40 / 7.2%	0.42 / 7.3%
0.05	0.38 / 7.4%	0.41 / 7.3%	0.42 / 7.5%
0.005	0.10 / 19%	0.14 / 14%	0.17 / 14%
CsI	1 / 7.0%	1.02 / 6.2%	0.94 / 8.2%

Left column is light yields relative to that of the reference CsI at +20 degrees C, and right one is energy resolutions for 662 keV gamma-rays.

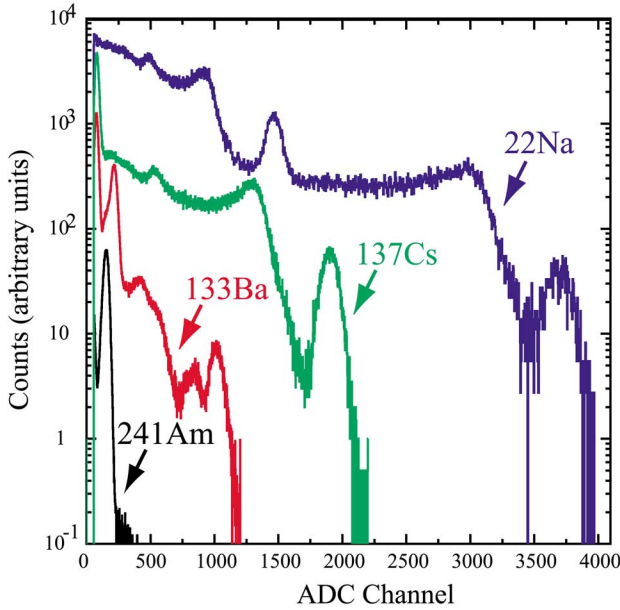


Fig. 2. Energy spectra of the ceramic YAG:Ce (0.5 mol%) at +20 degrees C, obtained by irradiating with radioactive sources ^{241}Am , ^{133}Ba , ^{137}Cs and ^{22}Na .

typically 20 mm in diameter and 2 mm in thickness. For comparison, we measured one commercial CsI:Tl ($15 \times 15 \times 15 \text{ mm}^3$), of which the maximum emission wavelength is close to that of YAG:Ce [3]. These ceramics are all the same as those measured in our previous study, and more detailed properties are described there [3].

II. LIGHT YIELD AND ENERGY RESOLUTION

In measuring gamma-ray responses of these ceramic YAGs, we mounted them on a 2-inch PMT (Hamamatsu R6231), and acquired data using a shaping time of $2 \mu\text{s}$. The scintillators were wrapped with GORE-TEX for light reflection, and attached to the PMT with optical grease (OKEN 6262 A). To control the temperature, the scintillator and PMT were located in a heat bath.

Fig. 1 shows gamma-ray energy spectra of ^{137}Cs obtained with the ceramic YAG:Ce (0.5 mol%) at temperatures of +20, 0, and -20 degrees C. The features from 662 keV gamma-rays (photoabsorption peaks, Compton edges and backscatter peaks) are clearly seen in all the spectra. The ratio of the light yield at +20, 0, and -20 degrees C is 1:1.08:1.14. Table II summarizes the temperature dependence of the light yield and energy resolution of each YAG:Ce, comparing with the CsI crystal. All

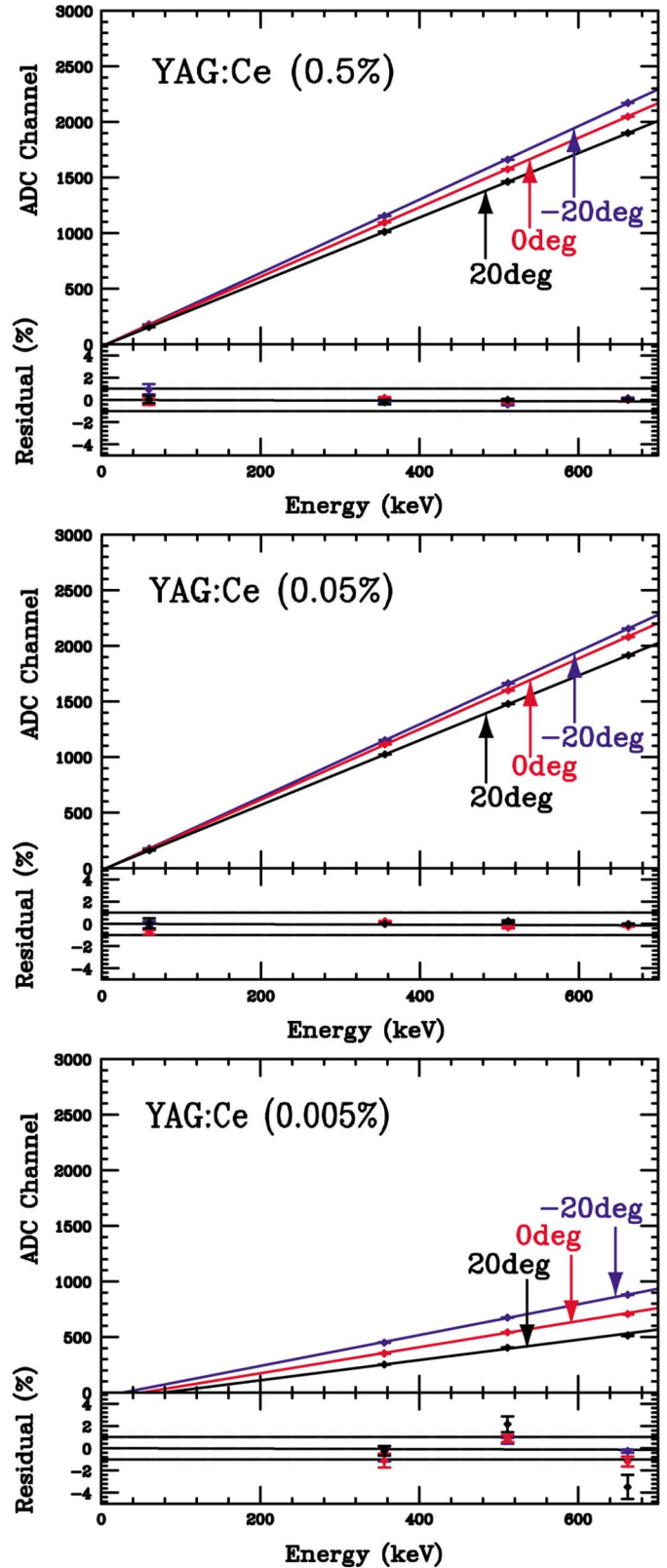


Fig. 3. Pulse-height versus energy of the three YAG:Ce (0.5 mol%, 0.05 mol% and 0.005 mol% from top to bottom). Light yields of all the ceramics decrease continuously from -20 to +20 degrees C.

the results include the temperature dependence of the PMT gain ($-0.2\%/degree$) [4]. As previously reported, the light yield of

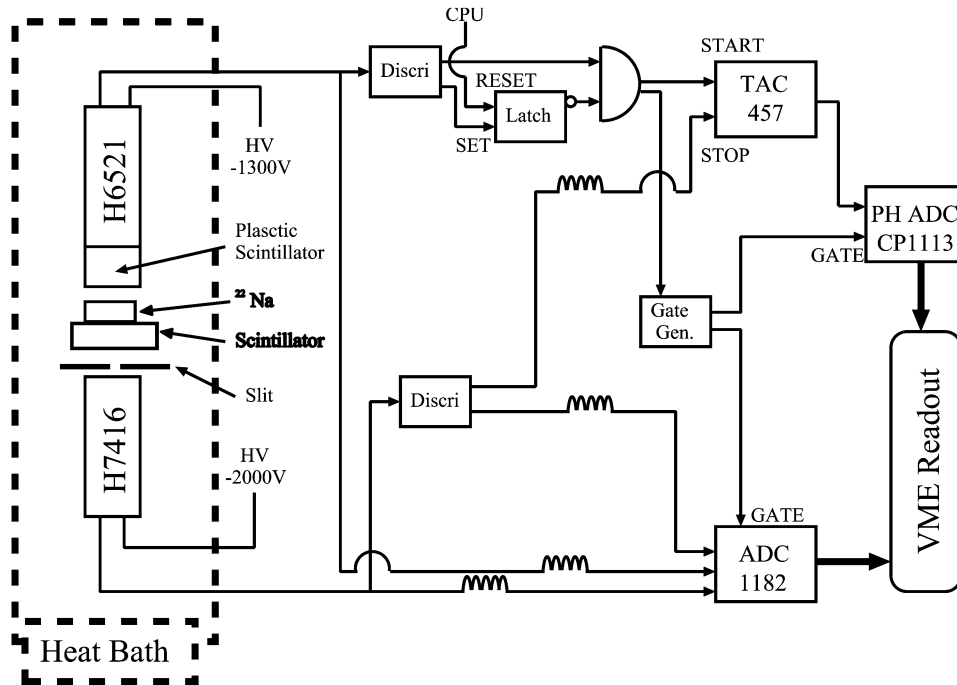


Fig. 4. Experimental configuration of the delayed coincidence method.

the 0.5 mol% Ce doped ceramic YAG at +20 degrees C reaches about 40% of that of CsI [3]. As the temperature decreases, the light yields of all the YAG:Ce thus increase, especially that with the 0.005 mol% Ce concentration.

Fig. 2 shows the spectra of ^{241}Am , ^{133}Ba , ^{137}Cs and ^{22}Na , measured with the YAG:Ce (0.5 mol%). We utilized the lines 59.5, 356, 511 keV and 662 keV lines to evaluate the pulse-height versus energy linearity. Because of a low photopeak efficiency in higher energies, the 1275 keV line from ^{22}Na were not utilized. As shown in Fig. 3, YAG:Ce (0.5 mol%) exhibits a good linearity within $\sim 1\%$ at each temperature. The energy resolution stays almost constant at 7.2% for 662 keV gamma-rays. The other two YAG ceramics with the lower Ce concentrations were measured in the same manner, and were confirmed to show similarly good linearities (Fig. 3). Here, the YAG:Ce (0.005 mol%) cannot detect the 59.5 keV gamma-rays, because the light yield is 30–40% lower than that of the 0.5 mol% one.

III. DECAY TIME PROFILES

Employing the delayed coincidence method in our previous setup [3], we measured scintillation decay time profiles of the YAG:Ce ceramics. The experimental setup is illustrated in Fig. 4. One data acquisition cycle is triggered by two 511 keV gamma-rays emitted in the opposite direction from ^{22}Na . One gamma-ray is detected by the plastic scintillator (BC-408 from BICRON) with the PMT (H6521) and “starts” the TAC. The other gamma-ray hits the ceramic YAG:Ce to be measured. Through a slit placed behind the scintillator, only one scintillation photon is sampled by the PMT (H7416) and “stops” the TAC. After many acquisitions, we obtained the $2 \mu\text{s}$ decay time profiles of YAG:Ce. Accidental background is negligible in the method. The results at +20, 0, and -20 degrees C of all the YAGs are shown in Fig. 5.

By previous studies [5]–[7], the intensity I of the decay curves at the time t was modeled as

$$I(t) = I_0 e^{-t/\tau_0} + I_1 (e^{-t/\tau_1} - e^{-t/\tau_0}) + I_2 (e^{-t/\tau_2} - e^{-t/\tau_0}) + I_3 (e^{-t/\tau_3} - e^{-t/\tau_0}) \quad (1)$$

where, τ_0, τ_1, τ_2 and τ_3 are decay time constants and I_0, I_1, I_2 and I_3 are the respective intensities at $t = 0$. The first term τ_0 represents the emission from directly excited Ce^{3+} centers [8], and the second one comes from the energy transfer from excited Y^{3+} to the Ce^{3+} . The others with τ_2 and τ_3 model other processes such as shallow trapping [9]. The obtained fit results are listed in Table III, where the intensity ratios are calculated by integrating the light yield of each term from $t = 0$ to ∞ with typical errors of a few percents. The derived values of $\tau_1 \sim 80$ ns and $\tau_2 \sim 300$ ns agree well with previous studies by other authors [8]–[12].

As the temperature increases from -20 to $+20$ degrees C, the decay time constants and fractions of the slower components shorten and decrease, respectively, and hence the effective decay of all the scintillators becomes faster. This result is also consistent with that of a single YAG:Ce crystal by Zych *et al.* [9], who conclude that this behavior is caused by the existence of many shallow traps, namely carriers which are captured in shallow traps before transferring excitation to Ce ions can escape the traps more easily at higher temperatures. Considering the decrease of the total light yield toward higher temperatures (§2), it is thought that there are effects of thermal de-excitation without luminescence as well as those of the above shallow traps, especially for the slower components of which the intensity fractions decreases toward higher temperatures compared with the faster ones.

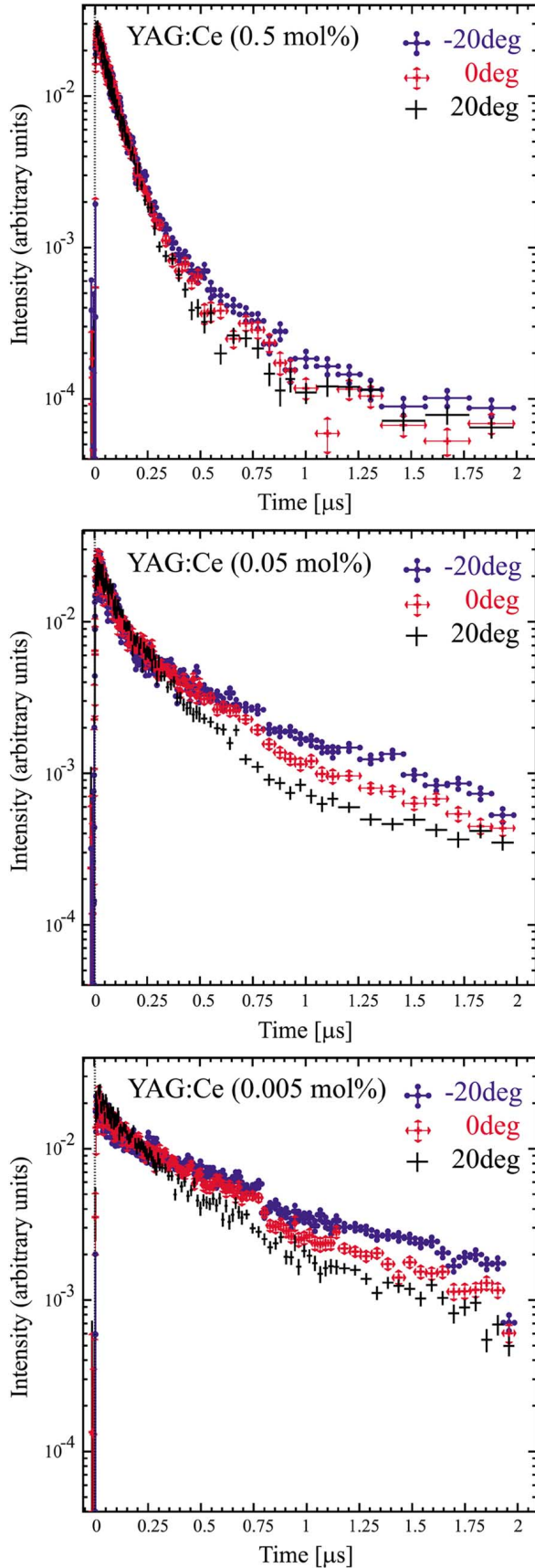


Fig. 5. Decay time profiles at +20, 0, and -20 degrees C of the ceramic YAG:Ce with 0.5 mol%, 0.05 mol% and 0.005 mol%. Decay of all the scintillators becomes faster continuously when temperatures increase from -20 to $+20$ degrees.

TABLE III
FIT RESULTS OF DECAY TIME PROFILES OF THE YAG:CE CERAMICS WITH (1)

YAG:Ce [mol%]	Temperature [degrees C]	$\tau_{0,1,2,3}$ [ns] (Intensity ratio [%])
0.5	+20	< 2 (0) / 78 ± 2 (64) / 294 ± 51 (12) / > 5000 (24)
	0	2 ± 1 (0) / 77 ± 2 (62) / 294 ± 28 (19) / > 5000 (19)
	-20	4 ± 2 (2) / 77 ± 3 (53) / 306 ± 27 (20) / > 5000 (25)
0.05	+20	4 ± 2 (1) / 81 ± 14 (17) / 263 ± 32 (48) / > 5000 (34)
	0	8 ± 1 (1) / 55 ± 10 (14) / 360 ± 27 (53) / 2600 ± 1400 (32)
	-20	5 ± 4 (1) / 76 ± 6 (20) / 418 ± 75 (24) / 1053 ± 107 (55)
0.005	+20	6 ± 4 (1) / 108 ± 26 (9) / 299 ± 21 (43) / 1217 ± 86 (47)
	0	5 ± 4 (0) / 46 ± 16 (2) / 486 ± 19 (41) / > 5000 (57)
	-20	< 2 (0) / 36 ± 9 (1) / 585 ± 120 (32) / > 5000 (67)

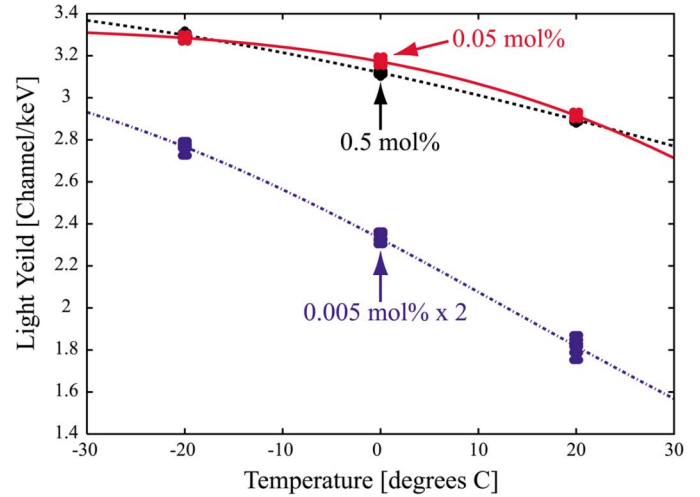


Fig. 6. Temperature dependence of light yields of YAG:Ce 0.5 mol%, 0.05 mol% and 0.005 mol% (multiplied by two) fitted with (2).

IV. DISCUSSION AND CONCLUSION

We have measured the temperature dependence (from -20 to $+20$ degrees C) of the light yields, linearities, energy resolutions, and decay time profiles of the poly-ceramic YAG:Ce scintillators produced by the new sintering method. As a result, the good linearities (within $\sim 1\%$ at every temperature) and the good energy resolutions ($\sim 7\%$ especially for YAGs with 0.5 and 0.05 mol% Ce) were observed. Toward the higher temperatures, their effective decay times became shorter and the light yields decreased. These results are consistent with those of single YAG:Ce crystals [9], and can be explained by considering the increases of the probabilities of both the escape from the shallow traps and the thermal de-excitation without luminescence.

The anti-correlation between the light yield I and the temperature T is usually attributed to the effect of thermal quenching, and is represented as

$$I = \frac{I_0}{1 + C \exp\left(-\frac{E_q}{k_B T}\right)}, \quad (2)$$

where I_0 is the initial intensity before affected by the quenching, C is a constant, E_q is quenching energy, and k_B is Boltzmann constant [13]. In Fig. 6, the measured light yield (i.e., pulse-height divided by energy) at each of 356 keV (^{133}Ba), 511 keV (^{22}Na) and 662 keV (^{137}Cs) gamma-rays of our sample ceramics are plotted as a function of temperature. While the result depends significantly on the sample, its energy dependence

TABLE IV
BEST FIT PARAMETERS OF (2) FOR YAG:Ce CERAMICS

YAG:Ce [mol%]	I_0 [channel/keV]	C	E_q [eV]
0.5	3.58	1.5×10^2	0.16
0.05	3.33	1.8×10^5	0.35
0.005	1.64	8.0×10^3	0.23

is negligible. The superposed curves are the best-fit models of (2), with their parameters summarized in Table IV.

The quenching energy E_q of YAG:Ce (0.05 mol%) is higher than that of the 0.005 mol% sample. This is probably because the average distance between the excited Y and Ce ions gets shorter as the dopant concentration increases, and hence the excited electrons have less probability to be trapped. However, E_q of the sample with the highest doping decreases again, presumably due to self absorption by Ce ions. This process makes the effective decay time longer, and hence the probability of the de-excitation without luminescence increasing. Actually, previous studies [3], [8] reported that the light yield saturates and even decreases when the Ce concentration exceeds 0.1–0.2 mol%, because the emission and absorption spectra of Ce-doped YAG overlap at around ~ 500 nm. Then, we expect that the slight quenching of the 0.5 mol% sample around the room temperature is caused through the self absorption, rather than the shallow traps which are important for lower Ce concentrations.

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