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Alkali-free Ce-doped and co-doped fluorophosphate glasses for future HEP experiments

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ABSTRACT

We report status of alkali-free cerium-doped and co-doped fluorophosphate glasses as a potential inorganic scintillator for future high energy physics experiments. Optical and scintillation properties, such as emission, transmittance, light output, decay time and their degradation after γ-ray irradiations, are measured for glass samples produced at AFO Research Inc. Further developments are needed for this potential cost-effective glass scintillator to be used for the HHCAL detector concept.

Contents

1. Introduction

Because of their relatively lower fabrication cost as compared to inorganic crystals, scintillating glasses have a potential for future high energy physics (HEP) experiment where a large volume of scintillators are needed. One example is the Homogeneous Hadronic Calorimeter (HHCAL) detector concept featured with unprecedented jet mass resolution by measuring both scintillation and Cherenkov light, where up to 100 m^3 scintillators are required [1–4]. Table 1 compares basic properties of alkali-free cerium-doped and co-doped fluorophosphate (Ba 44 wt%, P 13 wt%, Mg 5 wt%, Al 2 wt%, O 20 wt%, F 16 wt%) glasses [5–8] with commonly used inorganic and plastic scintillators. With a density of 4.6 g/cm^3 and a nuclear interaction length of 26.4 cm, this material provides a cost-effective solution for the HHCAL detector concept.

In this investigation, we measured optical and scintillation properties of alkali-free cerium-doped and co-doped fluorophosphate glasses fabricated at AFO Research Inc. (AFO glasses), including photo-luminescence (PL), PL pulse shape, X-ray excited luminescence (XEL), transmittance, pulse height spectrum (PHS), and light output (LO) before and after irradiations by γ-rays. We also discuss possible applications in future HEP experiments.

2. Samples and measurements

Fig. 1 shows two batches of 10 × 10 × 10 mm^3 glass samples produced in July (AFO-1607) and November (AFO-1611), 2016. These samples have a cerium doping level ranged from 0.5 to 5 wt% and various co-doping. Table 2 lists detailed information.

PL and time-resolved PL were measured by an Edinburgh Instruments FLS 920 fluorescence spectrometer. For the XEL measurements, samples were placed in the sample compartment of a HITACHI F-4500 spectrophotometer. X-rays generated by an Amptek Eclipse-III X-ray tube were used to excite the sample. Transmittance was measured by a PerkinElmer Lambda 950 spectrophotometer with 0.15% precision. LO was measured by a Hamamatsu R2059 PMT with a grease coupling for

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Table 1
A Comparison between AFO glass and commonly used scintillators.

	LSO/LYSO	CsI	BaF_2	CeF_3	$CeBr_3$	$LaCl_3$	$LaBr_3$	Plastic scintillator (BC 404) [9]	AFO glass
Density (g/cm^3)	7.4	4.51	4.89	6.16	5.23	3.86	5.29	1.03	4.6
Melting Points (°C)	2050	621	1280	1460	722	858	783	70[e]	–
X_0 (cm)	1.14	1.86	2.03	1.70	1.96	2.81	1.88	42.54	2.96
R_M (cm)	2.07	3.57	3.10	2.41	2.97	3.71	2.85	9.59	2.89
λ_I (cm)	20.9	39.3	30.7	23.2	31.5	37.6	30.4	78.8	26.4
Z_{eff}	64.8	54.0	51.6	50.8	45.6	47.3	45.6	5.82	42.8
dE/dX (MeV/cm)	9.55	5.56	6.52	8.42	6.65	5.27	6.90	2.02	6.84
λ_{peak}[a] (nm)	420	420 310	300 220	340 300	371	335	356	408	365
Refractive index[b]	1.82	1.95	1.50	1.62	1.9	1.9	1.9	1.58	–
Normalized light yield[a,c]	100	4.2 1.3	42 4.8	8.6	99	15 49	153	35	0.66
Decay time[a] (ns)	40	30 6	650 0.6	30	17	570 24	20	1.8	40
d(LY)/dT[a,d] (%/°C)	−0.2	−1.4	−1.9 0.1	~0	−0.1	0.1	0.2	–	–

[a]Top line: slow component, bottom line: fast component.
[b]At the wavelength of the emission maximum.
[c]Relative light yield normalized to the light yield of LSO/LYSO.
[d]At room temperature (20 °C).
[e]Softening point.

Table 2
Sample information for the samples investigated in this work.

Sample ID	Doping	Host Composition ID
AFO-1607-1	Ce 0.5 wt%	F2035
AFO-1607-2	Ce 0.5 wt%	F2035
AFO-1607-3	Ce 1.0 wt%	F2035
AFO-1607-4	Ce 1.0 wt%	F2035
AFO-1607-5	Ce 1.0 wt%, Pt 1.0 wt%	F2035
AFO-1607-6	Ce 1.0 wt%, Pt 1.0 wt%	F2035
AFO-1611-1	Ce 5.0 wt%, Gd 5.0 wt%	F2035
AFO-1611-2	Ce 5.0 wt%, Gd 5.0 wt%, Yb 1.0 wt%	F2035
AFO-1611-3	Ce 3.0 wt%, Gd 5.0 wt%, Yb 5.0 wt%	F2035
AFO-1611-4	Ce 1.0 wt%, Mn 0.5 wt%	WA40

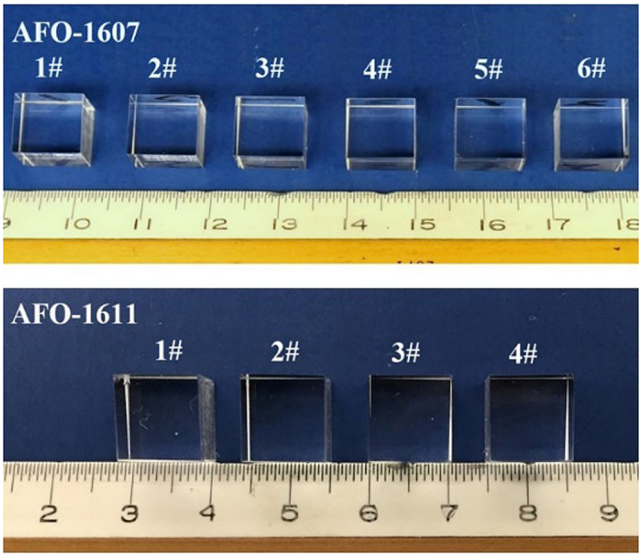

Fig. 1. Two batches of ten AFO glass samples of 10 × 10 × 10 mm^3.

0.662 MeV γ-rays from a ^{137}Cs source with self-trigger and 0.511 MeV γ-rays from a ^{22}Na source with a coincidence trigger. Since the γ-ray peaks are not well distinguished, LO was also measured by using 5.486 MeV α-rays from a ^{241}Am source, and compared to a lead tungstate ($PbWO_4$ or PWO) crystal. The systematic uncertainty of the LO measurement is about 1%. Four AFO-1611 samples went through γ-ray irradiations in ^{60}Co and ^{137}Cs γ-ray irradiation facilities at Caltech in two steps to reach a total dose of 10 and 100 krad.

3. Experimental results

3.1. Basic scintillation performance

Figs. 2 and 3 show PL and XEL spectra respectively for four glass samples. Consistent Ce^{3+} emission peaks at about 365 nm are observed in both PL and XEL spectra. The broad excitation bands peaked at 335 nm can be attributed to the Ce 4f-5d transition. The asymmetric shape of the emission peaks is due to overlap of two emission bands from the transitions between the 5d level and two split 4f levels of Ce^{3+} ($^2F_{5/2}$, $^2F_{7/2}$). The large stokes shift between excitation and emission spectra indicates no self-absorption.

Fig. 4 shows transmittance spectra measured along 10 mm path (solid lines) and emission spectra (dashed lines) together with the numerical values of the emission weighted longitudinal transmittance (EWLT) and the cutoff wavelength for four AFO glass samples. All samples show good transmittance with EWLT values higher than 80%, indicating excellent optical property. The absorption edges observed in the transmittance spectra are due to Ce^{3+} absorption, which are consistent with Ce^{3+} excitation bands shown in Fig. 2. The low cutoff wavelength at ~340 nm is excellent for Cherenkov light. With emission peaked at 367 nm and cut-off wavelength at 340 nm, this material shows no self-absorption.

Fig. 5 shows the cut-off wavelength and the EWLT values as a function of the Ce doping level. The cut-off wavelength increases from 340.6 nm to 344.1 nm when Ce concentration increases from 1% to 5%, confirming that the absorption edge is Ce induced. Although cut-off wavelength increases, the EWLT value remains stable with increased Ce concentration due to the large stoke shift of the Ce 5d–4f transition in AFO glasses.

Figs. 6 and 7 show the pulse shapes of PL at 355 nm for eight AFO glasses under 325 nm excitation. Because of their weak luminescence, we could not measure pulse shapes for the other two Pb doped samples. Single exponential function with a decay time of about 40 ns fits most samples well, which is consistent with the decay time of Ce 5d–4f transition. Ytterbium co-doping in the sample AFO-1611-3 introduce an additional fast decay component of 25 ns.

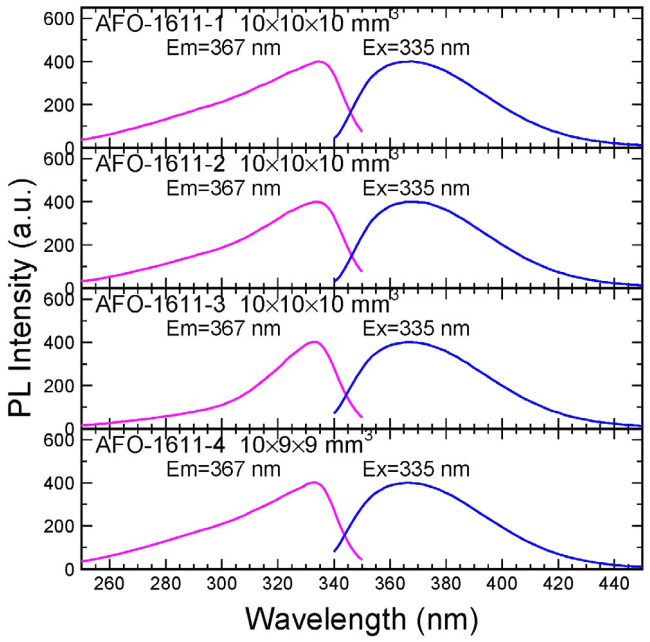

Fig. 2. Photoluminescence and excitation spectra for four AFO glass samples.

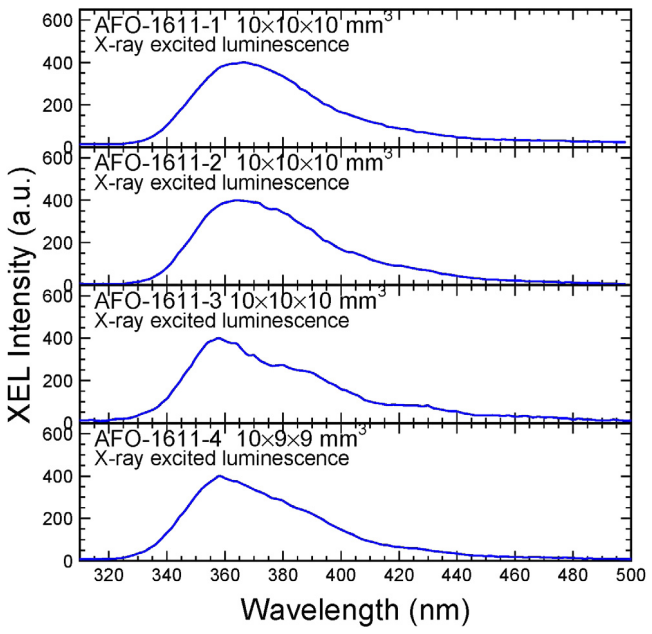

Fig. 3. X-ray excited luminescence spectra for four AFO glass samples.

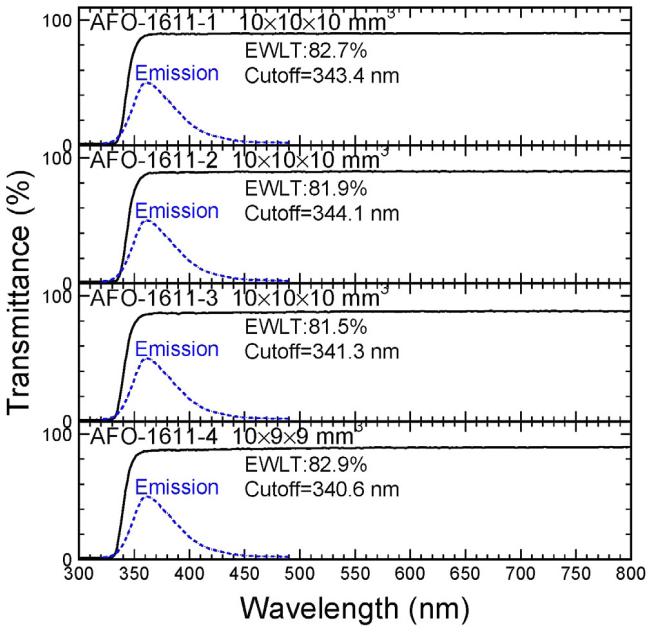

Fig. 4. Transmittance spectra measured along 10 mm for four AFO glass samples.

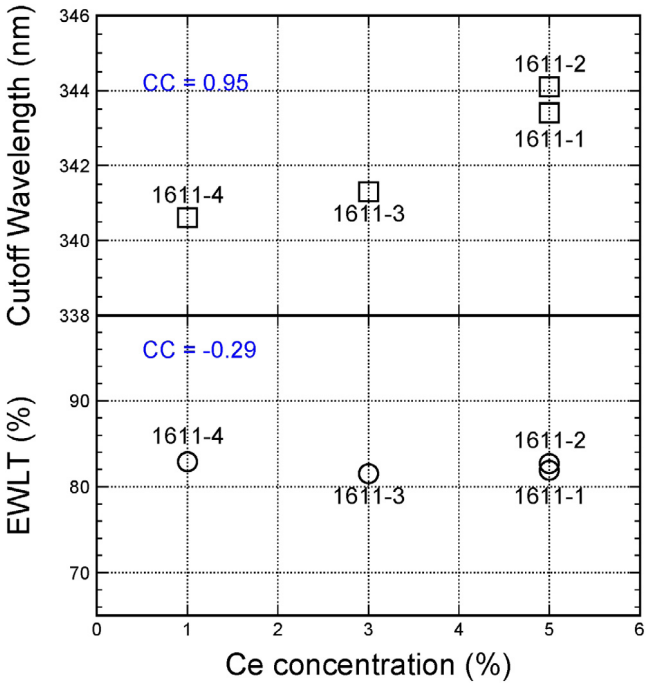

Fig. 5. Cut-off and EWLT wavelength values shown as a function of the Ce doping level.

Figs. 8 and 9 show the PHS excited by ^{137}Cs and ^{22}Na respectively for four AFO samples, and compared to a PWO sample of the same size. The numerical values of their LO and full width at half-maximum (FWHM) energy resolution are also shown in the figures. The γ-ray peaks are not well distinguished because of poor material uniformity and low effective Z value. Samples 1611-1 and 1611-4, however, show consistent LO values of more than 80 p.e./MeV measured by using ^{137}Cs and ^{22}Na sources, which is higher than PWO.

The LO values for all four glasses are also measured by using 5.486 MeV α-rays from a ^{241}Am source. Fig. 10 shows the PHS excited by ^{241}Am for four glass samples, and compares to the PWO sample, together with the numerical values of their LO and FWHM energy resolution. While α-ray peaks were clearly observed, their numerical LO values are lower than that measured by using γ-ray sources, which is well known for inorganic scintillators [10].

Table 3 compares the LO values and the α/γ response ratios for 4 glass and one PWO samples. The LO values for samples 1611-1 and 1611-4 are higher than samples 1611-2, 1611-3 and PWO. The α/γ response ratio is ~13% for the AFO glasses and ~30% for PWO.

3.2. Radiation hardness

Fig. 11 shows significant degradation in transmittance for four AFO glass samples before and after γ-ray irradiations of 10 and 100 krad

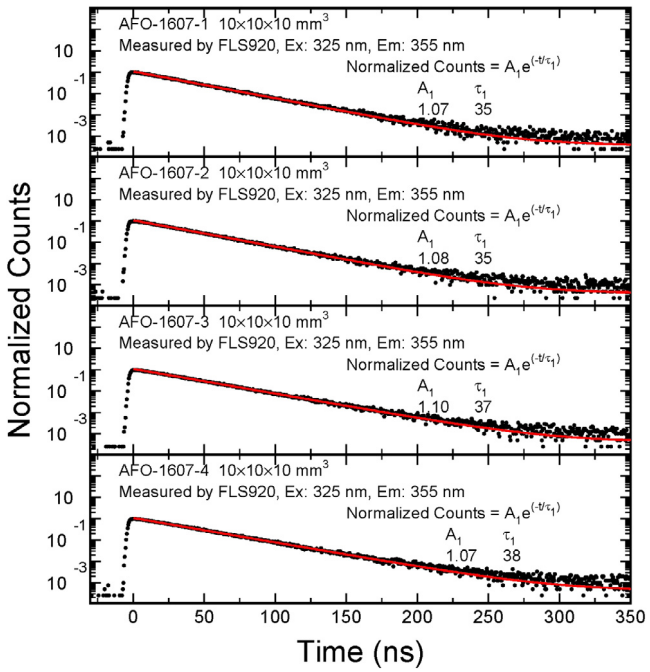

Fig. 6. PL pulse shapes of four 1st batch AFO samples with various Ce doping levels.

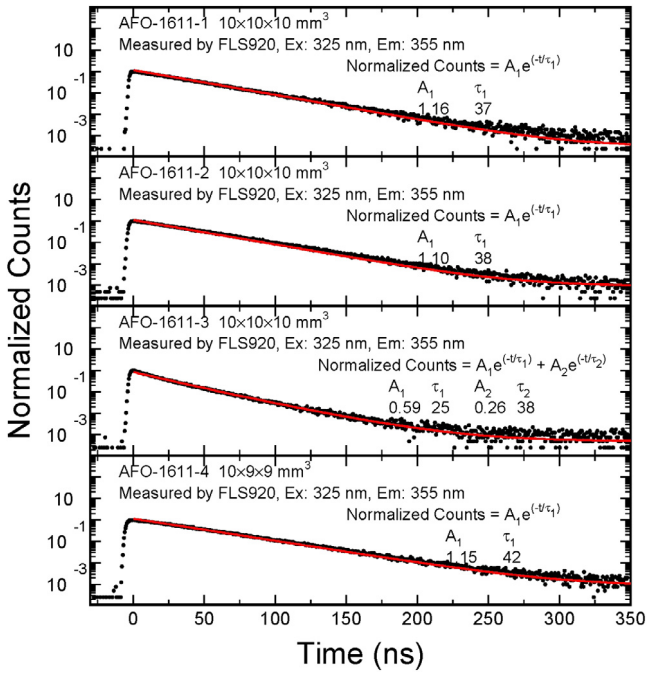

Fig. 7. PL pulse shapes of four 2nd batch AFO samples with various co-doping.

Table 3
Summary of light output and α/γ ratios for four glass and a PWO samples.

	^{137}Cs	^{22}Na	^{241}Am	α/γ (^{137}Cs)	α/γ (^{22}Na)
1611-1	97.8	97.2	13.0	13.3%	13.4%
1611-2	–	–	7.8	–	–
1611-3	–	–	1.8	–	–
1611-4	85.7	84.9	11.8	13.8%	13.9%
PWO	28.8	29.5	8.8	30.6%	29.8%

together with the numerical values of EWLT and cutoff wavelength. While this material is seriously damaged after 100 krad, it seems useable up to 10 krad with an EWLT value more than 60%.

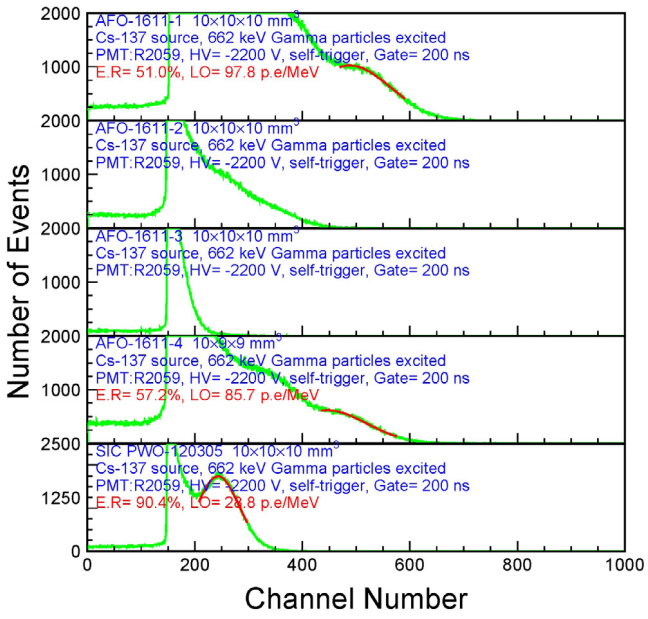

Fig. 8. Pulse height spectra for four glass and a PWO samples, excited by ^{137}Cs.

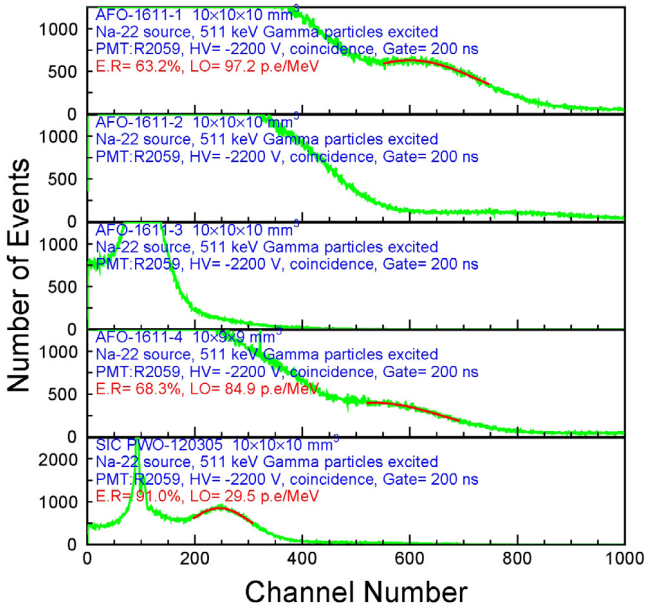

Fig. 9. Pulse height spectra for four glass and a PWO samples, excited by ^{22}Na.

Fig. 12 shows the PHS measured by using 5.486 MeV α-rays from a ^{241}Am source for four AFO samples after 10 krad γ-ray irradiation together with the numerical values of LO and FWHM energy resolution. While both LO and energy resolution are degraded, distinguished α-ray peaks are observed in samples 1611-1, 1611-2, and 1611-4, indicating that this material may be used up to 10 krad.

Fig. 13 summarizes the normalized EWLT (top) and LO (bottom) values as a function of the integrated dose for four AFO glass samples. Significant damages in both transmittance and LO were observed in all samples. Requiring more than 50% light, the glass samples of this size may be used in a radiation environment up to 10 krad. It was also found that samples 1611-1 and 1611-4 are radiation harder than their ytterbium-doped counterparts 1611-2 and 1611-3.

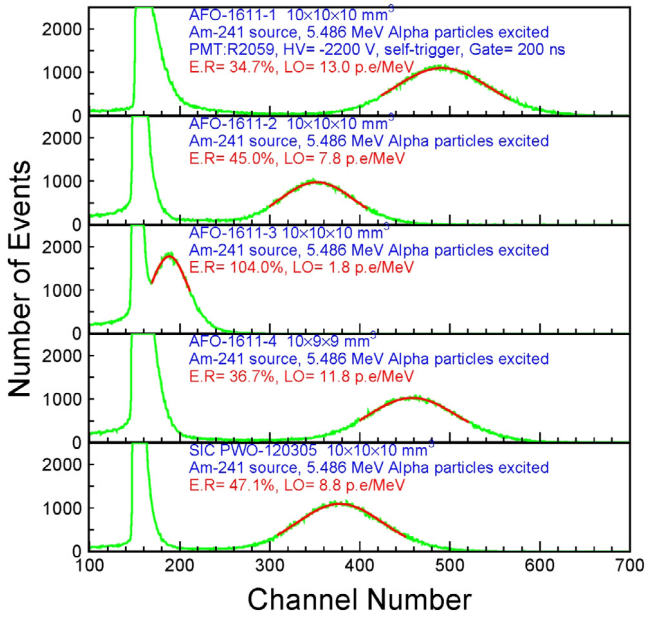

Fig. 10. Pulse height spectra for four glass and a PWO samples, excited by ^{241}Am.

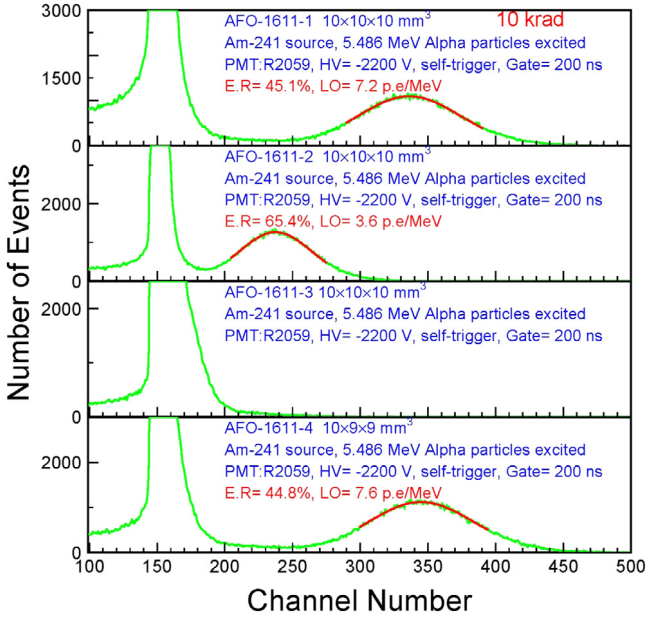

Fig. 12. 5.486 MeV α-ray PHS after γ-ray irradiation of 10 krad for 4 AFO samples.

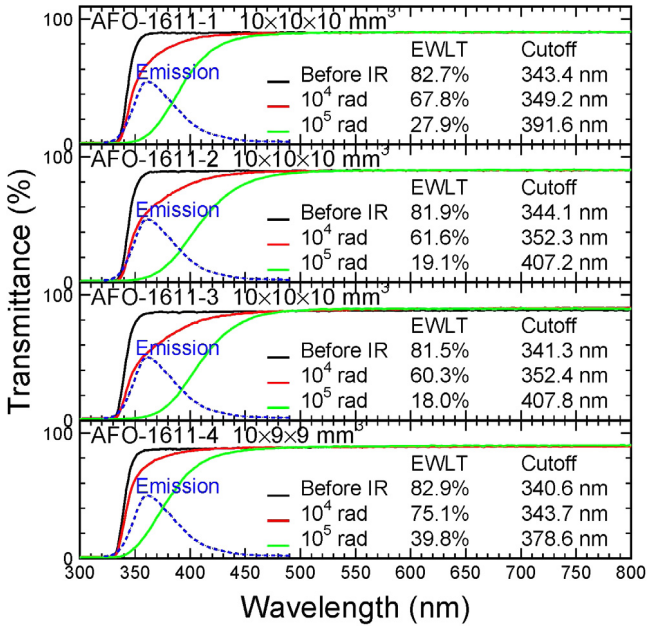

Fig. 11. Transmittance before and after γ-ray irradiation of 10 and 100 krad for 4 AFO samples.

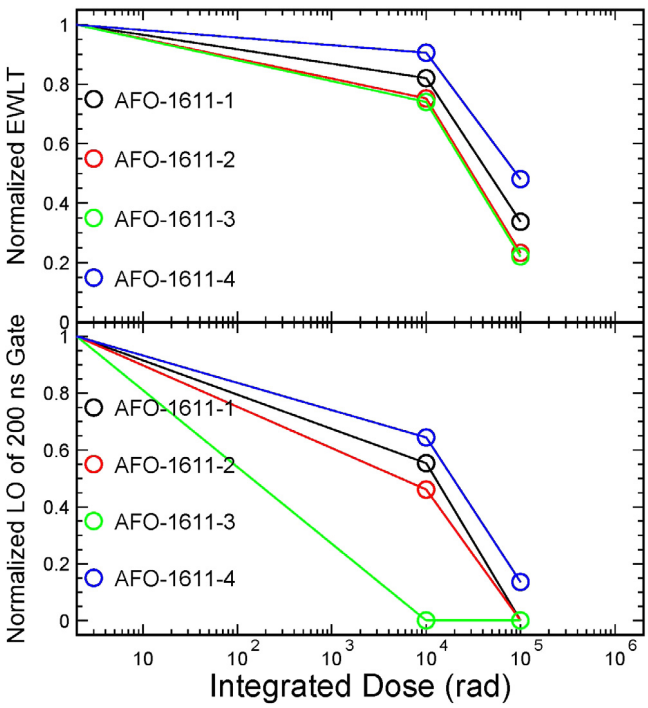

Fig. 13. Normalized LO and EWLT as a function of dose for 4 AFO samples.

4. Conclusion

Alkali-free cerium-doped and co-doped fluorophosphate AFO glasses show a good transmittance with EWLT over 80%, a cutoff wavelength at 340 nm excellent for Cherenkov light and an emission at 367 nm with no self-absorption. All cerium-doped samples have a decay time of about 40 ns. An additional faster decay component with 25 ns decay time is observed in a sample with ytterbium co-doping. While γ-ray peaks are not well distinguished, peaks from 5.486 MeV α-rays are clearly observed. Two AFO samples show a LO compatible with PWO crystals. Significant degradation in both transmittance and LO is observed in all samples. Requiring more than 50% light, AFO fluorophosphate glasses of this size may be used in a radiation environment up to 10 krad. Further development is required to improve performance of AFO glass scintillators, such as light output and radiation hardness, and to reduce mass production cost for this material to a level much lower than commercial available inorganic scintillators. If so, this material may be used to construct an HHCAL for a future lepton collider, where radiation environment is not as severe as hadron colliders.

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