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Behaviour of CeF₃ scintillator in a LHC-like environment *

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Abstract.

The neutron radiation resistance of a sample of cerium fluoride has been investigated with the reactor ULYSSE. The fast, thermal and epithermal neutron fluxes and the gamma dose rate at the irradiation position have been determined. The overall irradiation conditions are analogous to the condition foreseen in the future electromagnetic calorimeters for high energy physics. The effect of neutron radiation on the transmission and photoluminescence properties has been studied up to a fast neutron fluence of 10^{14} neutrons/cm². The respective damage effect of gamma rays, thermal neutrons and fast neutrons is discussed, as well as the induced radioactivity observed.

Résumé.

La résistance aux neutrons d'un échantillon de fluorure de cérium a été étudiée avec le réacteur ULYSSE. Les flux de neutrons rapides, thermiques et épithermiques ainsi que le débit de dose gamma au point d'irradiation ont été déterminées. L'ensemble des conditions d'irradiation est analogue à ce qui est attendu dans les futurs calorimètres électromagnétiques pour la physique des hautes énergies. L'effet des neutrons sur les propriétés de transmission optique et de photoluminescence a été étudié jusqu'à une fluence de 10^{14} neutrons rapides/cm². L'effet respectif des photons gamma, des neutrons thermiques et des neutrons rapides est discutée, ainsi que la radioactivité induite observée.

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1. Introduction.

Cerium fluoride is one of the possible crystal scintillator candidates for high rate electromagnetic calorimetry. A large research and development effort has been undertaken to evaluate precisely its properties, particularly within the «Crystal Clear» collaboration [1]. It has been presented as an option for the electromagnetic calorimeter of the CMS experiment proposed for the Large Hadron Collider in project at CERN [2]. Table 1 recalls its main characteristics compared to other scintillating crystals.

Among other specifications to fulfil for an electromagnetic calorimeter in the experiments proposed for the Large Hadron Collider, radiation resistance is imperative [3]. For example, in the CMS electromagnetic calorimeter, the highest ionisation damage will be observed at the maximum of the electromagnetic cascade. The yearly dose (assuming 1 year $\approx 10^7$ s of operation) is expected to vary from 1 kGy in the central part ($\eta = 0$) to 40 kGy in the forward region ($\eta = 2.6$) [2]. The hadron fluence per year will range from $2 \cdot 10^{12}$ 1 MeV equivalent neutrons/cm² at $\eta = 0$ to $4 \cdot 10^{13}$ neutrons/cm² at $\eta = 2.6$ [4].

We present here results on fast neutron resistance of a CeF₃ sample produced by Optovac (USA). Its size is 9.5 x 15.5 x 19.5 mm³. As reported in [5], this sample, (labelled CF128), has been irradiated in a first phase by ⁶⁰Co gamma photons, at a low dose rate, during about 6 months. The total absorbed dose was 5.3 ± 1.1 kGy (530 ± 110 krad) at an average rate of 1.2 Gy/h (120 rad/h). No damage was observed in the transmission properties after this irradiation. A crystal of the same production batch was irradiated at high dose rate at Frascati, up to 100 kGy (10 Mrad) with no noticeable damage [6].

Table 1. : Properties of CeF₃ and other dense scintillating crystals.

crystal	CsI	BaF ₂	CeF ₃	GSO	BGO	PbWO ₄	PbF ₂
density (g/cm ³)	4.51	4.89	6.16	6.71	7.13	8.28	7.77
radiation length (cm)	1.86	2.03	1.65	1.38	1.12	0.89	0.94
Molière radius (cm)	3.5	3.39	2.63	2.42	2.24	2.2	2.22
decay time (ns)	16 1000	0.6 620	5 30	60	300	2.5 11	Cerenkov
peak emission (nm)	315 565	210 310	300 340	440	480	420 520	300 (cut-off)
refractive index (at peak emission)	1.95 1.79	1.55 1.50	1.62	1.85	2.15	2.25	1.93
light output (photons/MeV)	2000	2500 6500	2000	10000	10000	200	1.3

2. Experimental Conditions.

After the low rate gamma irradiation, the sample CF128 was exposed in the nuclear reactor ULYSSE of the Institut National des Sciences et Techniques Nucléaires (INSTN) in Saclay. This small U-metal reactor is mainly dedicated to training purposes. Its great advantage is the possibility to monitor its power over 5 orders of magnitude (from 1 to 10^5 W) and thus to vary the neutron fluxes in the same proportions [7].

Figure 1. : Neutron spectra in a CeF_3 calorimeter with moderator, as calculated in ref. [4] and neutron spectrum in the channel HN1 of the reactor ULYSSE. The spectra are normalised to give the same total fluence between 100 keV and 1 GeV.

For our measurement we used the channel 'HN1', which allows to put the samples almost in contact with the fuel elements. In that position, the fast neutron spectrum is close to the uranium 235 fission one, which can be parametrized, for $0.1 \leq E \leq 12$ MeV, by $\phi(E) = 0.454 \exp(-E / 0.965) \sinh(\sqrt{2.29E})$, where E is the neutron energy in MeV [8].

During the same period of time, the fast neutron, thermal neutron and epithermal neutron fluxes were measured by a thin foil activation method [9], with various metals (Au, In, Ni). At the position of irradiation the measures give $2.4 \pm 0.5 \cdot 10^6$ neutrons/cm²/s/W for fast neutrons ($E > 100$ keV), $8. \pm 2. \cdot 10^6$ neutrons/cm²/s/W for epithermal neutrons ($100 \text{ keV} > E > 1 \text{ eV}$) and $7. \pm 2. \cdot 10^6$ neutrons/cm²/s/W for thermal neutrons ($E < 1 \text{ eV}$). The gamma dose rate was measured with an ionisation chamber and is estimated to $3. \pm 1. \cdot 10^{-4}$ Gy/s/W. Hence the ratio of gamma dose versus fast neutron fluence ($1.25 \text{ kGy} \cdot \text{cm}^2 / 10^{13}$ neutrons) in ULYSSE is well in the range of the one foreseen in the proposed CMS electromagnetic calorimeter.

The fast neutron fluence delivered in ULYSSE is similar in shape to the neutron fluence in a high energy electromagnetic calorimeter, with a peak around 1 MeV. For example, Figure 1 compares it to the neutron spectrum calculated by Aarnio and Huhtinen for the CeF₃ + moderator option of the CMS electromagnetic calorimeter (Figure 4 b in ref. [4]). The epithermal fluences, not shown in the figure, are also analogous, the ratio between fast and epithermal fluences being 0.30 in ULYSSE and 0.34 in the CeF₃ + moderator option of CMS.

Thus we can consider the irradiation conditions as well representative of the environment of the proposed crystal electromagnetic calorimeter.

A cumulated fluence of $1.0 \pm 0.2 \cdot 10^{14}$ fast neutrons/cm² was reached in 7 steps. The fast neutron flux ranged from $2.4 \cdot 10^6$ neutrons/cm²/s to $1.4 \cdot 10^{10}$ neutrons/cm²/s. For comparison, the flux is estimated to vary from $2 \cdot 10^5$ neutrons/cm²/s at $\eta = 0$ to $4 \cdot 10^6$ neutrons/cm²/s at $\eta = 2.6$ in the CMS electromagnetic calorimeter [4]. The irradiation conditions and the doses received are reported in Table 2. For radiation protection reasons, the transmission and photoluminescence spectra were recorded on average four days after irradiation. The sample was kept out of light during this time.

Table 2. : Characteristics of the irradiation steps.

date of irradiation	power (W)	duration (min)	fast neutrons fluence (n/cm ²) cumulated		thermal neutrons fluence (n/cm ²) cumulated		gamma dose (Gy) cumulated	
92/06-12			(low rate gamma irradiation)				5300	5300
93/03/18	1	14	2.0 10 ⁹	2.0 10 ⁹	5.9 10 ⁹	5.9 10 ⁹	0.25	5300
93/03/22	10	28	4.0 10 ¹⁰	4.2 10 ¹⁰	1.2 10 ¹¹	1.2 10 ¹¹	5.0	5300
93/03/26	100	70	1.0 10 ¹²	1.1 10 ¹²	2.9 10 ¹²	3.1 10 ¹²	126	5400
93/04/09	300	70	3.0 10 ¹²	4.1 10 ¹²	8.8 10 ¹²	1.2 10 ¹³	380	5800
93/04/22	600	70	6.0 10 ¹²	1.0 10 ¹³	1.8 10 ¹³	3.0 10 ¹³	760	6600
93/04/30	2000	70	2.0 10 ¹³	3.0 10 ¹³	5.9 10 ¹³	8.8 10 ¹³	2500	9100
93/05/07	6000	80	6.9 10 ¹³	9.9 10 ¹³	2.0 10 ¹⁴	2.9 10 ¹⁴	8600	17700

3. Results.

3.1. Transmission.

As shown in Figure 2, a small degradation in the transmission properties is observed after the first steps (curves a, b, c). Then no variation occurred up to 10^{13} neutrons/cm² (curves c to f). A more significant degradation is observed for the highest fluences (curves g, h). The radiation induced absorption coefficient $\epsilon(\lambda, \Phi)$ is defined by $T(\lambda, \Phi) = T(\lambda, 0) \exp(-\epsilon(\lambda, \Phi) x)$, where $T(\lambda, \Phi)$ is the optical transmission measured along the same path for the different wavelengths λ and fluences Φ (or doses in case of gamma irradiation) and x the path length. The radiation induced attenuation length is the inverse of the absorption coefficient : $l_{att}(\lambda, \Phi) = 1/\epsilon(\lambda, \Phi)$. Table 3 indicates the values of the absorption coefficient at the three emission peaks for the different fluences. Owing to the small length of the crystal, the values of the radiation-induced absorption coefficients are rather imprecise. Incertitudes are calculated assuming absolute errors of 0.008 in the transmission measurements and 0.2 mm in the path length.

Table 3. : Radiation-induced absorption coefficients at the wavelengths of the emission peaks.

wavelength	286 nm	303 nm	340 nm
after irradiation 1	$0.6 \pm 1.2 \text{ m}^{-1}$	$0.7 \pm 0.7 \text{ m}^{-1}$	$0.7 \pm 0.6 \text{ m}^{-1}$
after irradiation 2	$0.8 \pm 1.2 \text{ m}^{-1}$	$1.4 \pm 0.7 \text{ m}^{-1}$	$1.2 \pm 0.6 \text{ m}^{-1}$
after irradiation 3	$-0.6 \pm 1.2 \text{ m}^{-1}$	$1.3 \pm 0.7 \text{ m}^{-1}$	$1.2 \pm 0.6 \text{ m}^{-1}$
after irradiation 4	$1.8 \pm 1.2 \text{ m}^{-1}$	$1.3 \pm 0.7 \text{ m}^{-1}$	$1.1 \pm 0.6 \text{ m}^{-1}$
after irradiation 5	$2.6 \pm 1.3 \text{ m}^{-1}$	$1.3 \pm 0.7 \text{ m}^{-1}$	$1.3 \pm 0.6 \text{ m}^{-1}$
after irradiation 6	$1.8 \pm 1.2 \text{ m}^{-1}$	$2.1 \pm 0.7 \text{ m}^{-1}$	$2.0 \pm 0.7 \text{ m}^{-1}$
after irradiation 7	$5.1 \pm 1.3 \text{ m}^{-1}$	$3.8 \pm 0.7 \text{ m}^{-1}$	$3.7 \pm 0.7 \text{ m}^{-1}$
27 days after irradiation 7	$4.9 \pm 1.3 \text{ m}^{-1}$	$3.3 \pm 0.7 \text{ m}^{-1}$	$3.0 \pm 0.7 \text{ m}^{-1}$
after UV exposure	$3.3 \pm 1.3 \text{ m}^{-1}$	$2.5 \pm 0.7 \text{ m}^{-1}$	$2.4 \pm 0.7 \text{ m}^{-1}$

It is difficult to conclude if the effect observed for the lowest fluences is intrinsic and related to the irradiation or extrinsic and related to other causes like surface degradation. However this effect is small and saturates after the two first irradiation steps. At higher fluences, we can describe the evolution of the transmission as the sum of this constant term (either intrinsic or extrinsic) and a term proportional to the fluence, due to the irradiation (Figure 3).

Figure 2. : Transmission, along 1.95 cm, versus wavelength. (Note the magnified scale, the maximum effect being about 6 %) :

a : before irradiation, b : after $2 \cdot 10^9$ neutrons/cm², c : after $4.2 \cdot 10^{10}$ neutrons/cm²,
d : after $1.1 \cdot 10^{12}$ neutrons/cm², e : after $4.1 \cdot 10^{12}$ neutrons/cm²,
f : after $1.0 \cdot 10^{13}$ neutrons/cm², g : after $3.0 \cdot 10^{13}$ neutrons/cm²,
h : after 10^{14} neutrons/cm².

Figure 3. : Radiation-induced absorption coefficients at 303 nm, as function of the neutron fluence.

Nevertheless the sample suffers at the same time from gamma photons, from fast and thermal neutrons. We presume that these effects are independent. We know from previous gamma irradiation tests [5,6] that gamma radiation damage is negligible below 100 kGy. Two main effects can be attributed to the neutrons : activation and displacement. Activation is mostly due to the thermal neutrons, for which the absorption cross sections can be important. A very small proportion of the nuclei are affected at the fluence level reached here, and no effects are expected thereby in the optical or luminescence properties. Atomic displacement is related to the neutron energy, and may lead to the creation of color centres which induce light absorption, and so-called quenching centres, which affect the scintillator light yield. It is known that the fast neutrons are much more efficient than the less energetic epithermal neutrons.

Thus we conclude that in that case the effects of gamma photons, thermal and epithermal neutrons are negligible, and that the radiation induced optical absorption is only caused by the fast neutrons. Then the neutron-induced absorption coefficient on various isotopes can be estimated to be $0.27 \pm 0.08 \text{ m}^{-1}$ per 10^{13} fast neutrons/cm² at the peak of the emission spectrum (303 nm). This is equivalent to an induced absorption length of roughly 4 m at a fluence of 10^{13} fast neutrons/cm², or 40 cm at 10^{14} fast neutrons/cm².

A slight recovery (natural annealing) is observed four weeks after the last irradiation. Exposure to UV light increases this recovery but the sample does not fully recover it's initial transmission (Table 3).

Figure 4. : Excitation spectra for a 303 nm emission (left curves) and emission spectra for a 277 nm excitation (right curves), and transmission before irradiation, as function of the wavelength.

3.2 Photoluminescence.

The photoluminescence spectra are not particularly affected by the neutron irradiation, as shown in Figure 4. The excitation peak at 277 nm (for an emission at 303 nm) does not vary in position, width or intensity. The background of excitation below 270 nm seems slightly reduced for the highest fluences. The emission spectrum shows the three usual peaks, near 286, 303 and 340 nm. The first peak, at 286 nm, is just at the absorption edge, and its variation of intensity is not correlated with the irradiation but likely results from differences in the absorption due to surface quality variation. For the other peaks, above the absorption edge, no variation is observed.

3.3 Activation.

It should be noted that CeF_3 is rather little activated by neutrons. The most abundant isotopes are ^{140}Ce and ^{142}Ce , of natural abundance 88.43(10)% and 11.13(10)% respectively. They are activated in ^{141}Ce and ^{143}Ce by thermal neutron capture, with cross-sections of 0.58(4) b and 0.95(1) b respectively. ^{141}Ce decays in ^{141}Pr (stable) with a half-life of 32.5 days. ^{143}Ce decays with a half-life of 1.38 days in ^{143}Pr , which itself decays with a half-life of 13.57 days in ^{143}Nd (stable). They all decay by β -emission [10]. The β -dose rate at the contact of the sample [11] was 250 $\mu\text{Gy/h}$ 5 days after the last irradiation, 60 $\mu\text{Gy/h}$ after 27 days, and 1.4 $\mu\text{Gy/h}$ after 192 days. This decrease is in good accordance with the calculation based on the nuclear data above mentioned.

4. Conclusion.

The ULYSSE Saclay reactor is a very suitable tool to study LHC detector material behaviour under radiation, such as crystal or plastic scintillator, photodetectors, electronics, etc. We are now using it routinely for such tests. From both ^{60}Co gamma irradiation and neutron reactor irradiation, we can conclude that for gamma doses below at least 50 kGy (5 Mrad) and fast neutron fluences below at least to $5 \cdot 10^{13} \text{ n/cm}^2$, the CeF_3 crystals can be used for electromagnetic calorimetry.

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