Radiation study of Lead Fluoride crystals for the Crilin calorimeter

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ABSTRACT: Lead fluoride ($\text{PbF}_2$) crystals represent an excellent and relatively innovative choice for high resolution electromagnetic calorimeters with high granularity and fast timing. During the R&D stages of the Crilin calorimeter, three $\text{PbF}_2$ crystals sized $5 \times 5 \times 40$ mm$^3$ were irradiated with $\text{^{60}Co}$ photons up to $\sim 4$ Mrad and with 14 MeV neutrons up to a $10^{13}$ n/cm$^2$ total fluence. Their loss in transmittance was evaluated at different steps of the photon and neutron irradiation campaign, and two optical absorption bands associated with the formation of colour centres were observed at $\sim 270$ nm and $\sim 400$ nm. Natural and thermal annealing in the dark, along with optical bleaching with 400 nm light, were performed on the irradiated specimens resulting in a partial recovery of their original optical characteristics.

KEYWORDS: Calorimeters, Cherenkov detectors, Radiation-hard detectors
1 Introduction

Being modern tracking systems very precise, jet performance in particle flow-like reconstruction algorithms is usually limited by the calorimeter performance. In particular, a high granularity is required in order to distinguish signal particles from background and to solve the substructures necessary for jet identification. Time of arrival measurements in the calorimeter could play an important role in HL-LHC, since a high number of pile-up collisions is expected, and the timing could be used to assign clusters to the corresponding interaction vertex. In a Muon Collider, the timing could be used to remove signals produced by beam-induced background, asynchronous with respect to the bunch crossing. The calorimeter energy resolution is also fundamental to measure the kinematic properties of jets: a finely segmented calorimeter design should be favored in order to solve the jet substructure. However, this contrasts with the requirement for high timing resolution even for signal events involving low energy deposits, such as in the case of high impulse muons.

Our proposed design, the Crilin calorimeter, is a semi-homogeneous calorimeter based on Lead Fluoride (PbF$_2$) Crystals readout by surface-mount UV-extended Silicon Photomultipliers (SiPMs). PbF$_2$ was originally proposed in the late ’60s [1] as a candidate for EM calorimeters, but it was only during the ’90s that it was confirmed to be a pure Cherenkov radiator [2]. PbF$_2$ crystals represent a very interesting choice for new generation calorimeters; differently from the most common crystals used in HEP, PbF$_2$ is relatively cheap to produce and - being a Cherenkov radiator - its timing performance is not limited by the rise and decay times characteristic of scintillation. PbF$_2$ shows a very good transmittance in the UV region with a spectral cut-off edge at ~240 nm. Its optical transmittance is a key parameter, since the number of Cherenkov photons is evaluated as [3]:

\[ dN \propto d\lambda/\lambda^2, \]

therefore, good transmittance at shorter wavelengths allows a better yield of Cherenkov light [4]. A comparison with other high-density crystals frequently used in electromagnetic calorimetry is reported in Table 1.
Table 1. Properties of PbF$_2$ compared to those of other promising crystals

| Crystal     | PbF$_2$ | BGO    | BaF$_2$ | CsI | CeF$_2$ |
|-------------|---------|--------|---------|-----|---------|
| Density [g/cm$^3$] | 7.7     | 7.13   | 4.87    | 4.51| 6.16    |
| Radiation length [cm] | 0.93    | 1.1    | 2.1     | 1.9 | 1.7     |
| Molière radius [cm] | 2.2     | 2.7    | 4.4     | 3.8 | 2.6     |
| Decay constant [ns] | Ch 300  | 0.6, 620 | 26    | 5, 30 |

The radiation hardness of Crilin crystals is a crucial parameter to evaluate: for the case of a future Muon Collider [5], the requirements relative to the barrel electromagnetic calorimeter (EMC) involve a 1 Mrad total ionising dose (TID) and a $10^{12}$ n$_{1MeV}$/cm$^2$ equivalent neutron fluence. The individual and combined effect of TID and neutrons was evaluated by measuring the resulting deterioration in transmittance for three crystals sized $5 \times 5 \times 40$ mm$^3$, manufactured by SICCAS[6] using a melt growth process, thus resulting in a cubic form ($\beta$-PbF$_2$). The totality of the crystals used for the irradiation tests were randomly selected from the main production batch of the Crilin prototype.

The first crystal was tested without any kind of wrapping (in the following it will be referred to as the “naked” one), the other two were wrapped with a 100 $\mu$m thick Mylar foil. One of two crystals wrapped in Mylar was also cladded with an additional 100 $\mu$m thick Teflon layer.

2 Optical properties before the irradiation campaign

As previously stated, transmittance measurements are useful to investigate properties and quality of these type of crystals. Transmittance is defined as the ratio between the intensity of a light beam attenuated by its passage through the crystal, to its original intensity: $T = \frac{I_0 (1-R) e^{-\alpha d}}{I_0}$.

The transmittance of the three samples was measured longitudinally - i.e. through the crystal axis a- with PerkinElmer Lambda 950 UV/VIS dual-beam spectrometer [7]. In this case the optical transmittance was evaluated as follows:

$$T = \frac{S-D}{R e f - D} \frac{S_{0} - D_{0}}{R e f_{0} - D_{0}}$$

where $S$, $D$ and $Ref$ are respectively the measured, reference and dark signals, while the subscript 0 refers to the baseline measurement performed without the crystal inside the spectrometer.

Before starting the measurements, a repeatability test was performed by removing and replacing the crystal on the sample holder for each acquisition. The obtained spectra are reported in Figure 1 (Left). The spread at each wavelength can be evaluated as:

$$\sigma = \frac{T_{\text{max}} - T_{\text{min}}}{T_{\text{max}}}$$

Where $T_{\text{max}}$ and $T_{\text{min}}$ are respectively the maximum and minimum transmittance values relative to a single set of measurements. As shown in Figure 1 (Right), a $\sigma = 1.1\%$ is associated with the longitudinal transmittance measurement.
Figure 1. Longitudinal transmittance measurement on “naked” crystal (Left). $\sigma$ values for repeated longitudinal transmittance measurements performed removing and replacing the crystal on the holder (Right).

A comparison of the obtained transmittance spectra is shown in Figure 2. To avoid reflection losses the transmittance spectrum of the naked crystal was corrected as follows, with a reflection factor $P = (4n)^2/(1 + n)^4$ [8]:

$$T^i(\lambda) = (1/P) \times T(\lambda)$$

3 Optical properties after $^{60}$Co photons irradiation

After the reference transmittance measurements, the irradiation phase was started in Calliope[9], a pool-type gamma irradiation facility equipped with a $^{60}$Co radio-isotopic source array housed in a large volume ($7.0 \times 6.0 \times 3.9$ m$^3$) shielded cell. The source rack is composed of 25 $^{60}$Co source rods (with $41 \times 90$ cm$^2$ active area) arranged in a planar geometry, producing photons with $E_\gamma = 1.25$ MeV.

The activity of the plant was $1.97 \times 10^{15}$ Bq at the time in which the measurements were conducted. The crystals were positioned 70 cm away from the source, with their longitudinal axis perpendicular to it, yielding a 100 krad/h dose rate.

The transmittance measurements were performed at different steps of the irradiation, which took place over the span of three days for a total absorbed dose of 4.4 Mrad. In Table 2 the dose,
expressed in Gy(air), absorbed in each irradiation step are reported. In Figure 3 the spectra obtained

| Irradiation Step | Dose in air [krad] |
|------------------|---------------------|
| I                | 30.2                |
| II               | 89.88               |
| III              | 2082                |
| IV               | 4031.8              |
| V                | 4435.5              |

Table 2. Irradiation steps and corresponding total dose absorbed by the crystals

for the first two steps of irradiation are reported, as well as the spectra measured after the recovery
time. A transmission recovery of 10% is observed at 350 nm. Figure 4 shows the longitudinal
transmittance spectra obtained at step III, IV and V of the irradiation.

After a TID of approximately 80 krad, any subsequent irradiation step up to 4.4 Mrad did not result
in any significant further decrease in transmittance, suggesting a saturation effect associated with
the damage mechanism, as reported elsewhere [10]. The maximum degradation observed on all
three specimens is at the level of ~ 40%.

4 Natural annealing and optical bleaching of the crystals

After the irradiation phase, crystals were kept in a dark box to allow their natural recovery, according
to the procedure described in [11]. To further improve the recovery process, a light-bleaching run
on the irradiation crystals with 400 nm blue light was performed, according to the procedures and methods described in [8]. After 16 hours of optical bleaching, a modest recovery of a few percent was observed in the transmittance spectrum, indicating a small accelerating effect on the recovery process with respect to the previously described natural annealing case. A summary of the results obtained with natural annealing and the optical bleaching can be found in Figure 5.

![Figure 5](image-url) Transmission spectra obtained after 38 days of natural annealing and 16 hours of optical bleaching the naked crystal (left) and the crystal with Mylar wrapping (right).

5 Optical properties after neutron irradiation

The three crystals were subsequently irradiated at the Frascati Neutron Generator (FNG) facility of ENEA Frascati [12] using 14 MeV neutrons. Neutron generation at FNG is based on the T(d,n)α fusion reaction, producing 14 MeV neutrons with a flux up to $10^{12}$ neutrons/s in steady state or pulsed mode. In Figure 6, the neutron spectrum is shown as a function of the fluence. The crystals under test were placed 1 cm away from the source (relatively to their front face). Figure 7 reports the neutron flux map in the FNG facility. In order to better evaluate the expected neutron fluences, a simulation of the irradiation process was implemented on the McStas simulation package [13], using 1 cm bins along the crystal axes. As shown in Figure 7 (right), the TID associated with the neutron irradiation process resulted in a (negligible) 1 Gray dose for crystals.

The total fluence delivered in the first cm of the crystals during ~ 1 hour and 30 minutes of irradiation with 14 MeV neutrons was $10^{13}$ n/cm². Due to the technical time related to logistics and
Figure 7. Simulation of the neutron (left) and gamma (right) flux: crystals were placed 1 cm away from the neutron gun.

shipment of the crystals, transmittance measurements could only be performed 14 days after the irradiation and showed no alteration in the transmittance spectrum (Figure 8).

Figure 8. Transmission spectra obtained after the irradiation at FNG with 14 MeV neutrons for a total fluence of \(10^{13}\) n/cm\(^2\) compared with the results obtained before irradiation and after the 16 hours optical bleaching.

6 Conclusion

The optical transmittance of three PbF\(_2\) crystals, grown by SICCAS was studied before and after irradiation performed with photons and neutrons. Crystal with two different wrapping schemes (Mylar and Mylar plus Teflon) were tested, along with a naked crystal with no wrapping. The transmittance of the crystals under test were monitored during the gamma irradiation steps, up to a 4.4 Mrad TID, and after the subsequent neutron irradiation runs. Natural recovery of crystals, along with light-beaching and thermal annealing were carried out and compared in terms of their effect on the recovery process.

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