Results on New Scintillating Crystals from the "CRYSTAL CLEAR" COLLABORATION *

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Abstract

A large effort has been undertaken since nearly 2 years by 14 research institutes (the "Crystal Clear" collaboration, CERN RD project 18), in order to develop and investigate new dense, radiation hard and fast inorganic scintillators suitable for very good resolution electromagnetic calorimeters to be build at the new high luminosity accelerators. Other fields of interest like nuclear physics, gamma ray astronomy, medical imaging (PET), safety systems and non destructive industrial applications are also rapidly growing. Recent results on the most promising candidates are given with some comments about mass scale production and cost considerations.

I. INTRODUCTION

The Crystal Clear Collaboration[1] is in charge of one of the European research and development activities being initiated by CERN in view of preparing the experimental program of the future Large Hadron Collider in Europe (LHC). The task of the collaboration is to investigate new dense, radiation hard and fast inorganic scintillators to be used as the components of very good resolution electromagnetic calorimeters at the new high luminosity accelerators, as well as in other fields like Nuclear Physics, Space Research, Medicine for PET scanners (positron emission tomographs) and several other industrial applications.

This work is carried out in close collaboration with a large number of companies and research laboratories all over the world. The program of the Crystal Clear Collaboration has been described elsewhere[1]. This report will mainly concentrate on recent results on Cerium Fluoride (CeF₃), which is presently considered as the most promising candidate. The most significant results of our measurements on other scintillators will be shortly presented.

II. CERIUM FLUORIDE

With its high density (6.16 g/cm³) and fast decay time (2 components at 8 nsec and 25nsec), CeF₃ is one of the most promising candidates for electromagnetic calorimeters at high luminosity hadron colliders[2,3]. We will report here on a systematic study of the radiation induced effect on the optical properties of this crystal.

Several CeF₃ crystals produced by different manufacturers using different raw materials and different growing conditions were irradiated with a Co60 (4400Ci) radiation source in 250 Gy dose steps with 48 h breaks between steps, up to a total dose of 1250 Gy. Since in principle both production and transport of light might be affected by radiation, not only

optical transmission but also all the other parameters of interest in the scintillating process were measured during the breaks (luminescence spectrum, decay time, light output).

Some of the crystals are sensitive to the radiation and the damage is different for each crystal depending on the producer, the growth conditions and the raw material. For a crystal uniformly irradiated perpendicular to its axis, a gradient of the damage along the crystal growth direction is generally observed (see Fig.1.).

Fig 1 Gradient of damage along a CeF₃ crystal after 1000Gy

A. Radiation effect on the light production

In order to study the radiation effect upon the decay time and light yield at the light production centres, slices of typically 1-2 mm width were cut parallel to the growth axis from different crystals in which a variation of the optical transmission due to the radiation was noticed. The slice form was preferred in order to be able to neglect the effect of optical absorption during the transport from the emitting centres to the photodetector.

Measurements performed on such samples showed that there was no modification of the decay time and light yield (proportional to the integrated surface under the decay time curve) at both ends of the crystal, before and after irradiation.

Using a special positioning of samples, luminescence spectra were recorded before and after irradiation in a way that strongly reduced the optical absorption contribution in the sample. As can be seen in Fig 2, emission peaks have the same positions and relative amplitudes.

It is therefore quite clear that the characteristics of the light production in the cerium fluoride (scintillation efficiency, decay time, emission spectrum) are not modified by a gamma irradiation up to 1250 Gy.

Fig 2 CeF3 emission spectrum before and after 1250 Gy

B. Radiation effect on the light transport

For the study of the radiation effect upon the light transport, both optical absorption and surface effects were considered.

Optical transmission measurements were performed on CeF3 crystal slices of different thickness cut along the crystal axis of the same ingot, before and after irradiation. Extrapolating the results for a slice of "zero" thickness, an identical transmission value was found before and after irradiation, which is compatible to the transmission expected from the reflexion losses only (Fig. 3).

Fig 3 Transmission through slices of different thickness before and after 1000 Gy for a good and a bad crystal

This result indicates that in CeF3 crystals there are no observable radiation induced surface effects. The only observed damage concerns a reduction of the optical transmission in some specific absorption bands, which results in a slightly pink coloration of some crystals after irradiation.

It must be noticed that the positions and intensity of these absorption bands can be very different from crystal to crystal, depending on the manufacturer and the raw material quality.

Fig 4 Transmission in the middle of a CeF3 crystal with oxygen contamination for several irradiation doses

Fig 5 Transmission in the middle of a CeF3 crystal of the first generation before and after 1000 Gy

Fig 6 Transmission in the middle of a recent CeF3 crystal for several irradiation doses

Fig 4,5 and 6 show the lateral transmission curves (2 cm thickness) measured in the middle of 3 crystals about 10 cm long before and after increasing doses of irradiation. In one of

them an oxygen contamination took place which resulted in a strong absorption band around 500 nm (Fig 4). For another more typical crystal two absorption bands at 350 and 390 nm are produced by the irradiation. It is to be noticed on this sample that a rather strong contamination of Neodymium resulted in a series of sharp absorption lines in the initial spectrum (Fig 5). Starting from a raw material of improved quality, Cerium Fluoride can be quite radiation hard as it is shown on Fig 6.

When the radiation damage is present, a strong gradient of it along the crystal growth direction is seen. Most probably this is due to the segregation of impurities occurring during the growth process. Researches are on the way in order to establish the nature of the colour centres produced in CeF_3 crystals after irradiation, as well as the dynamics of their production and annihilation.

III. OTHER CRYSTALS AND GLASSES

Some other interesting candidates are being studied by the Crystal Clear collaboration[4]. Among them Cerium doped Yttrium Aluminate (YAP:Ce) which is a very promising laser material, has a high light yield (at least 50% more than BGO) and a decay time of about 30 nsec. Moreover its emission spectrum peaks at 370 nm, a wavelength matching well the efficiency of Si photodiodes. Unfortunately the damage after radiation results in a strong absorption band overlapping the emission region (Fig 7), but the first samples grown were of rather poor quality and it is expected that the radiation hardness will improve with a better control of the growing conditions and the raw material purity.

Fig 7 Transmission through a 3 cm long YAP:Ce crystal before, 6 hours and 6 days after 1000 Gy $\text{Co } 60$ irradiation.

Although the Moliere radius is small (2.8 cm), this crystal has a relatively long radiation length (2.8 cm) due to a rather low density (5.36 g/cm^3). In order to have a similar material with a higher density, we are trying to develop Ytterbium aluminate (YbAlO_3), or a mixed compound ($\text{Y,Gd} \text{AlO}_3$) with the company Monocrystal in Turnov, Czechoslovakia.

Cerium doped Gadolinium Silicate (GSO:Ce) is very attractive because of its very high density (6.71 g/cm^3), a very high light yield more than twice the light yield of BGO with an emission peak in the visible spectrum (440nm) [4] and an exceptional radiation hardness[5] as confirmed by our measurements on a 19 cm long crystal (Fig 8).

Fig 8 Transmission through a 19 cm long GSO:Ce crystal for several irradiation doses

Unfortunately the high melting temperature (1900°C) combined with a very low pulling rate ($\approx 1 \text{ mm/hour}$) to allow the Cerium to enter into the lattice, give little chance to cut the production price to an affordable level for a large detector in the immediate future. Nevertheless this crystal will certainly be a very good candidate for applications where a large light yield is necessary like positron emission tomography.

Thorium fluoride (ThF_4) and mixed compounds based on BaF_2 combined with lead, yttrium or ytterbium are also promising. The Crystal Clear collaboration is presently studying BaY_2F_8 and BaYb_2F_8 as a possible alternative to BaF_2 . The latter has a very high density (6.99 g/cm^3) but we have recently found that a high temperature phase transition could make the production of large crystals difficult. On the other hand, BaY_2F_8 has a modest density of 5.08 g/cm^3 but can be easily produced and is a well known laser material. When doped with 1.39% Thulium, it produces as much light as BGO, with 94% in two fast components of respectively 1.33 nsec and 7.6 nsec, and only 6% in a slow component of 100 nsec decay time[6]. This is of course a major improvement as compared to BaF_2 where 25% only of the light is going to a very fast component (0.6 nsec) and 75% in a very slow component (620 nsec).

The Crystal Clear collaboration has undertaken a systematic work to understand the optical properties of Lead Fluoride (PbF_2) which is a very attractive crystal because of its high density (7.77 g/cm^3). Crystals of 24 cm in length (already available today) would be sufficient to contain 100 GeV γ showers ($1 X_0 = 0.93 \text{ cm}$) and the Moliere radius is as small as 2.21 cm. The abundance of the raw material and the

low melting point (822 °C) should make this crystal one of the cheapest. This crystal is considered at the moment as a good Cerenkov material, as no scintillation has been detected so far. We have asked Optovac and the Shanghai Institute of Ceramics (SIC) to introduce up to 16 different scintillating ions in the PbF_2 lattice in order to understand the quenching mechanism of the scintillation. Table 1 summarizes the tests and the results obtained.

Producer	doping	band-edge	fluorescence
OPTOVAC	without	260 nm	no
OPTOVAC	Ba	330 nm	Weak 358 nm
OPTOVAC	Tb	260 nm	384,414,434,487,542nm, slow
OPTOVAC	Bi	260 nm	no
OPTOVAC	Co	350 nm	no
OPTOVAC	Ag	260 nm	no
OPTOVAC	Cu	305 nm	no
OPTOVAC	Cr	260 nm	no
OPTOVAC	Dy	260 nm	448nm,512nm,slow
OPTOVAC	Sm	?	564,594,600nm, slow
OPTOVAC	Yb	?	Weak 405 nm
OPTOVAC	Eu	?	467,510,589,619nm, slow
SIC	without	260 nm	no
SIC	Ce 100 ppm	315 nm	no
SIC	Ce 1 %	325 nm	no
SIC	Ba 10 % Ce 0.1%	325 nm	no
SIC	Ba 20 % Ce 0.1%	315 nm	no
SIC	Ce	325	no
SIC	Ce	325	no

Table 1

Similar studies are under way with Calcium fluoride (CaF_2) and Barium Fluoride (BaF_2), having the same crystalline structure as Lead fluoride, to compare the properties of the doping ions in these crystals. Tests are also on the way to compare the optical properties of lead fluoride in its 2 naturally occurring phases, cubic and orthorhombic. Mixed crystals ($\text{PbF}_2+\text{BaF}_2$) are under development as well as some other lead based structures like, Pb_2ZnF_6 , $\text{Pb}_3\text{Al}_5\text{F}_{19}$, PbCsF_3 .

Another very attractive solution for calorimeters could be the use of scintillating glasses, as their production cost may be much reduced as compared to crystals. Fluorozirconate and fluoroaluminate glasses seem to offer the best perspectives in terms of radiation hardness, according to our recent tests. Unfortunately, the density of these glasses was limited to 4 to 4.5 g / cm³ until recently. By some substitutions of heavy metals in fluorozirconate and fluoroaluminate glasses, densities of respectively 5.72 g / cm³ and 5.92 g / cm³ have been recently obtained by the French company "Le Verre Fluoré" and prospects to go up to 6.5 to 7 g / cm³ are encouraging. A very fast (≈ 10 nsec) and intense luminescence has been observed around 320 nm, unfortunately severely quenched when excited by high energy particles. A weak, 25 nsec scintillation could only be seen in a Cerium doped

fluorozirconate where a large proportion of Zirconium had been replaced by Hafnium (Fig 9).

Fig 9 Decay time curves for Hafnium glasses with and without Cerium doping.

The narrow peak represents the direct excitation of the photomultiplier by the source and is not associated with a scintillation. On the other hand the tail observed on the Cerium doped Hafnium glass represents the typical scintillation of Cerium, although severely quenched. Batches of 10 to 20 kg are feasible with the standard techniques in use for fluoride glasses. In addition the glasses offer the unique advantage over crystals, that moulding, hot pressing and extrusion techniques can be used to shape the blocks. This is of course very important if one considers relatively cheap materials, for which the mechanical processing will represent a significant, if not the dominant, fraction of the total cost.

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V. REFERENCES

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