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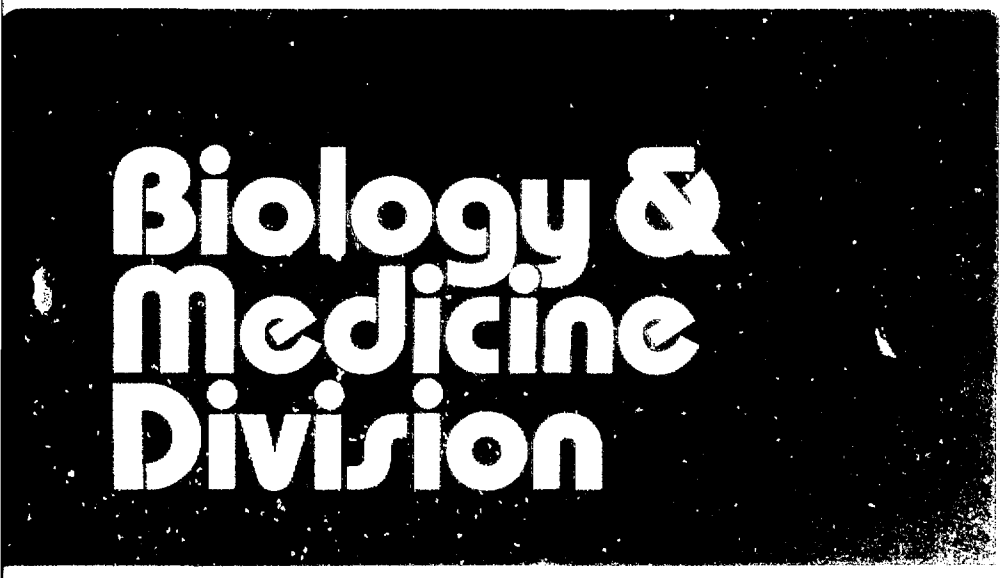

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CERIUM FLUORIDE, A NEW FAST, HEAVY SCINTILLATOR

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Abstract

We describe the scintillation properties of Cerium Fluoride (CeF_3), a newly discovered, heavy (6.16 g/cm^3), inorganic scintillator. Its fluorescence decay lifetime, measured with the delayed coincidence method, is described by a single exponential with a 27 ± 1 ns time constant. The emission spectrum peaks at a wavelength of 340 nm, and drops to less than 10% of its peak value at 315 nm and 460 nm. When a 1 cm optical quality cube of CeF_3 is excited with 511 keV photons, a photopeak with a 20% full width at half maximum is observed at approximately half the light output of a Bismuth Germanate (BGO) crystal with similar geometry. We also present measurements of the decay time and light output of CeF_3 doped with three rare-earth elements (Dy, Er, and Pr). The short fluorescence lifetime, high density, and reasonable light output of this new scintillator suggest that it would be useful for applications where high counting rates, good stopping power, and nanosecond timing are important, such as medical imaging and nuclear science.

1 Introduction

This paper describes the scintillation properties of Cerium Fluoride (CeF_3), a newly discovered inorganic scintillator. It begins with measurements of the fluorescent decay time, light output, emission spectrum, and coincidence timing resolution for undoped CeF_3, then explores the effect that several rare-earth dopants have on these scintillation properties. The paper concludes with a description of a technique that uses synchrotron radiation x-rays to rapidly measure the scintillation decay time and light output of powdered compounds. All measurements described in this paper were made with crystals provided by Optovac, Inc. of N. Brookfield, MA.

The physical characteristics of CeF_3 are well suited for use as a radiation detector. It has a density of 6.16 g/cm^3 [1], is not hygroscopic [1], and has an index of refraction of 1.62 [2]. The crystal structure is hexagonal [1], and it is colorless, transmitting wavelengths between 5000 nm and 300 nm [2].

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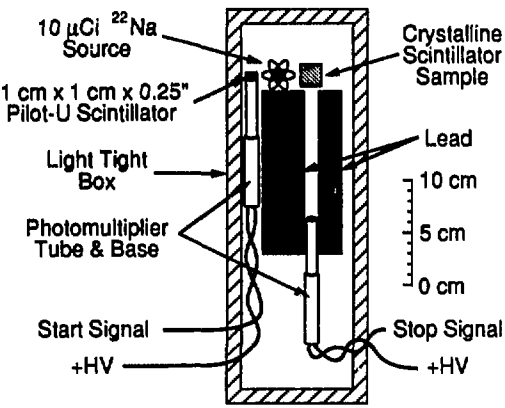

Figure 1: Delayed-Coincidence Apparatus

2 Undoped Cerium Fluoride

2.1 Fluorescent Decay Time

The fluorescent decay lifetime was measured using the delayed-coincidence method of Bollinger and Thomas [3], as modified by Moszyński and Bengtson [4]. A diagram of this set-up is shown in Figure 1. A piece of Pilot-U plastic scintillator coupled to a Hamamatsu R-2055 photomultiplier tube provides a start signal, and another quartz-windowed Hamamatsu R-2055 photomultiplier tube placed 13 cm away from the CeF_3 sample provides the stop signal. A 10 μCi ^{22}Na source provided the 511 keV photon pairs that excited both the plastic scintillator and the scintillator sample. Timing signals from both photomultiplier tubes are generated using Ortec 437A constant fraction discriminators, and the time difference between the start and stop signals was digitized with an Ortec 457 time to amplitude converter and a Tracor-Northern TN-1705 multi-channel analyzer.

The results of this measurement are shown in Figure 2.

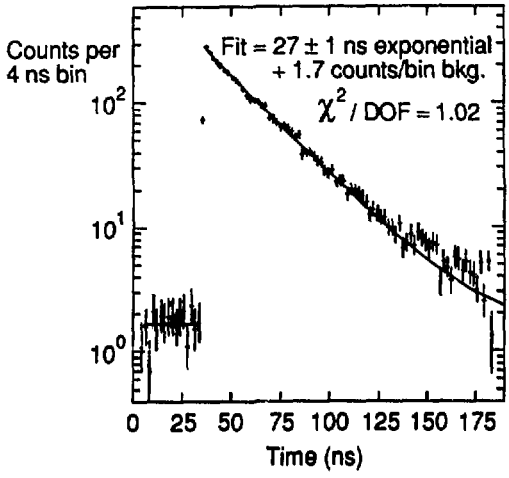

Figure 2: Scintillation Intensity of CeF_3 vs. Time

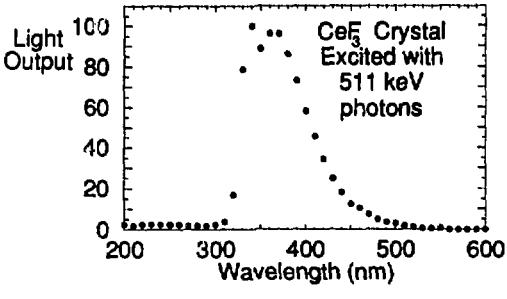

Figure 3: Emission Spectrum of CeF_3

A good fit to the data (the chi-squared per degree of freedom is 1.02) is obtained with a single exponential with a 27 ns decay constant plus a constant background. The error in this measurement is dominated by a systematic error of approximately 1 ns, which is estimated by varying the background level and the data points that are included in the fit.

2.2 Emission Spectrum

The emission spectrum of CeF_3 was obtained using a 0.25 meter Jarrell-Ash model 82-410 monochromator. The 511 keV photons from a 1.7 mCi ^{68}Ge source were used to excite a 1 cm cube of CeF_3 that was covered on 5 sides with reflective coating of white Teflon tape. The sixth side was placed at the entrance slit of the monochromator, and a quartz windowed Hamamatsu R-2055 photomultiplier tube (spectral range 200 nm to 600 nm) was placed at the exit slit. The resulting photomultiplier count rate is plotted, after background subtraction, as a function of monochromator wavelength in Figure 3. This

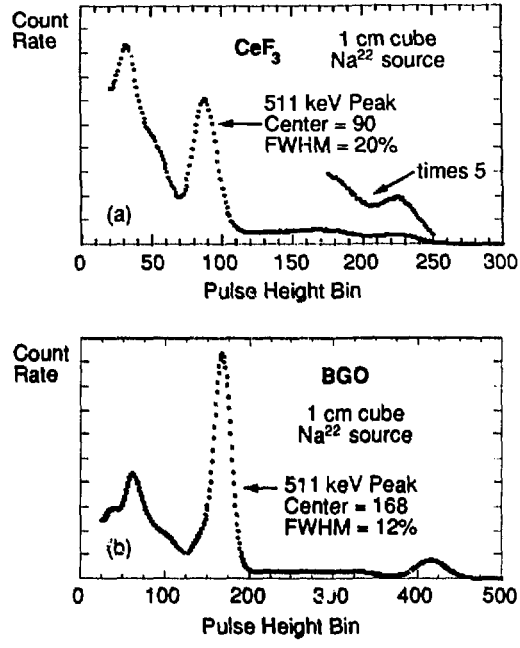

Figure 4: Light Output of CeF_3 and BGO

emission spectrum peaks at 340 nm, and drops to 10% of its maximum intensity at 315 nm and 460 nm. Note that absence of light below the 300 nm cutoff of borosilicate glass implies that fused silica or UV glass windowed photomultiplier tubes are not necessary in order to collect all of the CeF_3 scintillation light.

2.3 Light Output

The light output of CeF_3 was measured by comparing its response to 511 keV photons to the response of a Bismuth Germanate (BGO) crystal under the same conditions. A 1 cm optical quality cube of CeF_3 was coated on five sides with a reflective coating of white Teflon tape, then optically coupled to a quartz-windowed Hamamatsu R-1306 photomultiplier tube with General Electric Viscasil 600M silicone fluid. The crystal was irradiated with 511 keV and 1.27 MeV photons from a ^{22}Na source, and the output of the photomultiplier tube amplified with a Tennelec TC-222 amplifier with 1.2 μsec shaping time and digitized with a Tracor-Northern TN-1705 multi-channel analyzer. The resulting pulse height spectrum is plotted (after pedestal subtraction) in Figure 4(a). The CeF_3 crystal was removed and the same experiment was performed on a 1 cm cube of BGO, and the resulting spectrum is shown in Figure 4(b).

The photopeak corresponding to the 511 keV photon is clearly seen in each plot in Figure 4, as is a small

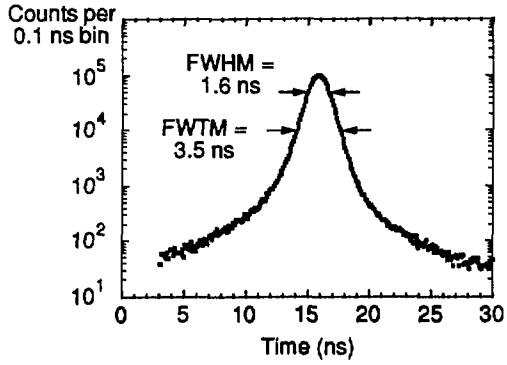

Figure 5: Coincidence Time Resolution

peak due to the 1.27 MeV photon. The 511 keV photopeak in CeF_3 is centered at a pulse height that is 54% of the 511 keV photopeak pulse height in BGO. Using the BGO light output of 8200 photons/MeV reported by Holl, et al. [5], this implies that the light output of CeF_3 is approximately 4400 photons/MeV. The full width at half maximum (FWHM) in CeF_3 of the 511 keV photopeak is 20% and the FWHM of the 1.27 MeV photopeak is roughly 13%, which is consistent with an energy resolution proportional to the square root of the incident gamma energy.

2.4 Coincidence Timing

The coincidence resolving time of CeF_3 was measured by exciting two 1 cm cubes of CeF_3, each coupled to a quartz windowed Hamamatsu R-2055 photomultiplier tube, with 511 keV photons resulting from positron annihilation from a ^{22}Na source placed between the two crystals. A timing signal from each photomultiplier tube was generated using a Ortec 437A constant fraction discriminator, and the time difference between the two timing signals was digitized with an Ortec 457 time to amplitude converter and a Tracor-Northern TN-1705 multi-channel analyzer. The resulting timing distribution, which has a FWHM of 1.6 ns and a full width at tenth maximum (FWTM) of 3.5 ns, is plotted in Figure 5. The same apparatus measures a timing distribution FWHM of 0.5 ns for Barium Fluoride (BaF_2).

3 Doped Cerium Fluoride

Cerium Fluoride crystals with three rare earth dopants (0.1% DyF_3, 1.0% ErF_3, and 0.12% PrF_3) were measured to determine whether they enhanced the scintillation properties of CeF_3. The fluorescent decay time of these doped crystals was measured with the same method used in Section 2.1. For two dopant elements, Erbium and Praseodymium, the resulting decay time distribution

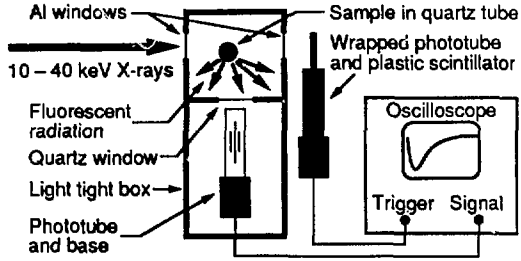

Figure 6: Apparatus for Screening Powdered Samples

could not be fit with a single exponential, and a second, faster decay component was necessary to achieve a good fit. The resulting fit decay times and fraction of total light emitted with each decay time are given in Table 1.

Due to their poor optical quality, none of the doped crystals showed a clear photopeak when irradiated with 511 keV photons, making accurate light output measurements impossible. For comparison purposes, a coarse estimate of the relative light output was made by selecting a relatively clear 0.375 inch diameter by 0.4 inch high right circular cylinder of each doped crystal and coating it on all but one end with reflective Teflon tape. The relative light output was estimated by coupling each crystal to a quartz windowed Hamamatsu R-2059 photomultiplier tube, irradiating it with 511 keV photons from a ^{68}Ge source, and measuring the output current of the photomultiplier tube with a Kiethley model 410 micro-microammeter. If all crystals are the same size, are in the same position relative to the source, and are equally transparent, then this output current will be directly proportional to the light output. The results of this measurement are included in Table 1. Although this method of estimating the relative light output is not very accurate, it does suggest that each of the three dopants significantly reduces the light output of CeF_3.

4 Fast Screening Technique

The scintillation properties of CeF_3 were discovered during an extensive search for new scintillators performed in November of 1987 at the Stanford Synchrotron Radiation Laboratory (SSRL). The apparatus used for this search, shown in Figure 6, is able to rapidly (within a few minutes per sample) measure the scintillation properties of powdered samples, avoiding the costly and time consuming task of preparing optical quality crystals. A nanosecond burst of 10 to 40 keV X-rays from a synchrotron beam passes into a vacuum chamber through a thin aluminum window. A portion of the beam is absorbed by a powdered sample in a thin walled (0.37 mm) quartz cuvette. The resulting fluorescent emanations, if any, pass through a quartz vacuum window onto the quartz face of a Hamamatsu R-2055 photomultiplier tube. The output

Crystal Composition	Fast Component Fraction & Decay Time	Slow Component Fraction & Decay Time	Relative Total Light Output
CeF_3 – undoped	none	100% @ 27 ns	100
CeF_3 – 0.1% DyF_3	none	100% @ 25 ns	35
CeF_3 – 1.0% ErF_3	9% @ 3.3 ns	91% @ 25 ns	15
CeF_3 – 0.12% PrF_3	26% @ 6.5 ns	74% @ 23 ns	50

Table 1: Effect of Dopants on Decay Time and Light Output

of this photomultiplier tube was measured by a Tektronics 2465 oscilloscope (350 Mhz bandwidth, 1 ns rise/fall time). The oscilloscope was triggered by a Pilot-U plastic scintillator attached to a photomultiplier tube, which was excited by the remainder of the X-ray beam after it exits the vacuum chamber via a second aluminum window. This apparatus is able to measure the decay time with 5 ns resolution (determined by the fall time of the photomultiplier tube) and the scintillation light output within an order of magnitude.

5 Conclusions

Cerium Fluoride is a newly discovered, heavy, inorganic scintillator. Its density of 6.2 g/cm^3 lies between that of Barium Fluoride (BaF_2, 4.9 g/cm^3) and BGO (7.1 g/cm^3). It has a decay time of 27 ± 1 ns, which is slower than the 0.8 ns "fast" component of BaF_2, but considerably faster than BGO (300 ns) or the "slow" component of BaF_2 (620 ns). The CeF_3 emission spectrum peaks at 340 nm, and the scintillation light output is approximately half that of BGO. Cerium Fluoride crystals with several rare-earth dopants have been measured to determine whether they enhanced the scintillation properties. Two of the dopants add a second, faster decay component, but each dopant decreases the total light output.

The combination of high density, short fluorescence lifetime, and reasonable light output suggest that CeF_3 would be useful for applications where high counting rates, good stopping power, and nanosecond timing are important, such as medical imaging and nuclear science. The absence of a "slow" fluorescent decay component implies that CeF_3 would be well suited for applications where counting rates as high as 10 Mhz are expected.

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