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Optical Restoration of Lead Fluoride Crystals

A. Spilker,* P. L. Cole, T. A. Forest, M. Mestari, S. Naeem, and N. LeBaron.

Idaho State University, Pocatello, ID 83209, USA

P. Bertin and C. Muñoz Camacho.

Université Blaise Pascal, 63006 Clemont-Ferrand Cedex, France

J. Roche

Ohio University, Athens, OH 45701, USA

Abstract

Due to its relatively high resistance to high radiation, lead fluoride (PbF_2) crystals are becoming an increasingly popular material of choice for electromagnetic calorimetry, such as for experiments requiring the measurement of high-energy photons in Hall A of Jefferson Lab. For our studies we irradiated the PbF_2 crystals using an electron linear accelerator (LINAC) followed by exposing the crystals to blue light so as to restore the nominal optical properties. This technique of optical bleaching with blue light affords an efficient and low-cost means for reversing the deleterious effects of optical transmission loss in radiation-damaged lead fluoride crystals. Whereas earlier experiments irradiated the PbF_2 samples with 1.1 and 1.3 MeV gammas from ^{60}Co, we used pulsed beams of energetic electrons from the tunable 25-MeV LINAC at Idaho Accelerator Center of Idaho State University in Pocatello, Idaho. A 20-MeV beam of electrons was targeted onto four separate 19 cm length samples of lead fluoride over periods of 1, 2, and 4 hours yielding doses between 7 kGy and 35 kGy. Samples were then bleached with blue light of wavelength 410 - 450 nm for periods between 19.5 and 24 hours. We performed this process twice – radiation, bleaching, radiation, and then followed by bleaching again – for each of these four PbF_2 samples. We shall discuss the efficacy of UV curing on samples that had undergone the cycle of electron irradiation and optical bleaching.

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1) Introduction

1.1) Deeply Virtual Compton Scattering Experiment

The objective of the Deeply Virtual Compton Scattering (DVCS) experiment at Jefferson Lab (Hall A) is the construction of a three-dimensional image of nucleon structure. The study of this inner structure depends on the extraction of Generalized Parton Distributions from beam and target spin asymmetry measurements, which will introduce constraints in current models of nucleon structure. The current DVCS design probes the structure of protons through the scattering of a polarized electron beam off of a target of liquid hydrogen at 6 GeV. The reaction of interest is $ep \rightarrow pe'\gamma$.

High energy experiments require a fast response and radiation hardness in the detection material. An electromagnetic calorimeter of lead fluoride was chosen to meet both of these requirements for the DVCS experiment. The prototype for the DVCS calorimeter was originally designed in 2002 with 9 1.37 kg samples of PbF_2 [4]. It is currently being upgraded from 132 to 208 separate blocks of PbF_2 manufactured by the Shanghai Institute of Ceramics, Chinese Academy of Sciences (SICCAS), each optically coupled to a Hamamatsu R5900U photomultiplier tube. This design allows for an energy resolution of 2.4% and a timing coincidence of 0.60 ns at 4.2 GeV [4].

The 132 samples in the current design have already been exposed to high levels of radiation in experiments E-00-110 and E-03-106 [1,2]. The Ohio University group has designed a method for transmission measurements in optical bleaching studies [5] of samples from the DVCS calorimeter based on the work of Patrick Achenbach [3]. The objective of the Idaho State University group is to report the effects of optical bleaching on transmission losses after multiple exposures to a predictable source of radiation.

1.2) Lead Fluoride

Lead Fluoride is a Cherenkov radiator. It has a density of 7.77 g/cm^3, a radiation length of 0.93 cm, an index of refraction that ranges from 1.75 to 1.91 (this value is wavelength dependent) and an effective Moliere radius of 2.2 cm. It has been found to be radiation hard, lowly hygroscopic and responsive to optical bleaching. The UV absorption edge of lead fluoride is lower than that of most lead glasses at 300 nm.

Experimenters at Fermilab investigated the properties of lead fluoride in 1989 [6]. These trials led to the discovery of the material's affinity for the production of Cherenkov light. Since then 1,022 blocks of lead fluoride were used in the calorimeter for the A4 parity violation experiment at the Mainz microtron [3] and eleven dozen have been used in the DVCS calorimeter in Hall A at Jefferson Lab.

1.3) Optical Bleaching

Under exposure to radiation a crystal lattice accumulates absorbent centers that hinder the passage of particles through the medium. These centers are color centers or F-centers, from the German word *farbezentrum* meaning "color center." Radiation exposure produces anionic vacancies in the crystal lattice which attract electrons through local electromagnetic potentials. Since these electrons are bound to a positively-charged center they exhibit a spectrum of energy levels. Thus there is an emission of visible light in regions of negative ion vacancies in the lattice. In lead fluoride this discoloration is qualitatively yellow due to the color center absorption of red and blue photons.

Exposure of color centers to an outside source of energy releases trapped electrons from anionic vacancies in the lattice. This process has seen wide application in thermal annealing, where energy is absorbed by color centers from heat exposure. In optical bleaching energy is transferred from a light source at room temperature. Previous experiments in optical bleaching of lead halides [7] have found that the violet/blue region (410-495 nm) of the UV spectrum is the most effective range of frequencies for this process.

2) Experimental

2.1) The Method

Four 1.29 kg, 30x30x190 mm^3 blocks of PbF_2 from the DVCS calorimeter were exposed to high levels of radiation (7-35 kGy). The source of radiation was the tunable 25 MeV pulsed electron LINAC in the main hall of the Idaho Accelerator Center in Pocatello, Idaho [8] (see Fig. 3). The transmission was measured as a function of wavelength and as a function of transverse position along the block just after irradiation. Samples were then placed under an OSRAM [9] violet/blue light source (410 nm – 460 nm) for periods of 19-24 hours for the purpose of optical bleaching. Transmission measurements were taken again after blue light exposure. This irradiation/optical restoration cycle was repeated once more for all samples.

2.2) The Source

The 25 MeV electron LINAC is a reconditioned CLINAC with adjustable pulse widths and repetition rates. These tunable features range from 80 ns to 2 μs and 1 Hz to 1000 Hz under long pulse operation. The maximum instantaneous current for long pulse operation is up to 80 mA per pulse. For our purposes, the beam parameters were set at a pulse window of 100 ns, repetition rate of 30 Hz, a peak current of 50 mA per pulse and a beam energy of 20 MeV. Under these conditions DVCS calorimeter samples could be irradiated at a rate lower than the power threshold for temperature-induced damage, accounting for heat capacity and thermal expansion.

2.3) Determination of Dose

Dose for each irradiation was calculated from measurements of beam intensity taken several times over the duration of exposure. Oscilloscope traces were rendered from values of current registered on a Pearson Electronics [10] current monitor positioned downstream the exit window of the LINAC. These values were graphed as a function of time to ensure the uniformity of dose throughout exposure and then averaged together for extraction of a mean intensity. For energy E and beam intensity I the power deposition P is calculated by $P = E/e \times I$, where e is electron charge. A mass m exposed over time t gives a dose D of $D = P \times t/m$ [11].

2.4) Transmission Measurements

Transmission was measured on an Ocean Optics USB4000 spectrometer [12]. This device offers a frequency range of 200 nm to 1100 nm. The light source was a deuterium tungsten lamp that emits in the range of 200 nm to 2000 nm. Spectra for transmitted intensity were rendered at 1, 2, 6, 11, and 16 cm from the point of beam interface with each block at 0° and 90° to transverse position during irradiation. Measurements for background and unobstructed light were made for each set of spectra.

2.5) Analysis

Transmission T was calculated as a ratio of transmitted intensity I_t to unobstructed intensity I_u, $T = I_t / I_u$. [13]. This value was graphed as a function of wavelength. Transmission spectra at 0° and 90° that showed little variation were averaged together for better statistics. Spectra for all trials of irradiation and optical bleaching taken at the same transverse position on the sample were overlain for investigation of the restorative effects of optical bleaching. Transmission measurements were also made for one sample two hours after irradiation to investigate the effects of exposure the ambient background before optical curing.

Transmission data was used to extract the index of refraction as a function of wavelength. This calculation was based on the light attenuation and the relationship between reflectivity and transmission from Fresnel's Law [7]. The extracted values were compared to measurements made at FNAL [6] 20 years ago (see Fig. 4).

3) Results

All results reported herein are for evaluations of transmission made at 1 cm from the beam interface unless otherwise noted. Effects at further distances from the interface fit the same trends as described below with transmission loses and gains being less drastic (see Fig. 2).

Two samples (blocks 18 and 33) exhibited similar responses to optical curing (see Figs. 1a & 1b). Transmission measurements of these samples suggest that optical bleaching is effective in regions greater than 680 nm. Below this frequency transmission measurements made after optical curing are actually lower than those made just after irradiation for the first cycle and equivalent to measurements of transmission just after irradiation for the second cycle. This suggests that optical bleaching is ineffective at lower frequencies for these samples and bodes poorly since the quantum efficiencies for R5900U type -01 and type -04 PMTs peak around 410 nm.

Block 16 exhibits the same behavior as blocks 18 and 33 for the first cycle, with the exception of recovery in the region spanning 300 – 350 nm (see Fig. 1c). The transmission spectrum for optical bleaching after the second cycle shows a loss of 15% transmission throughout the spectrum. This sample also exhibits strange behavior in transmission at 16 cm from the beam interface (see Fig. 2c). The transmission spiked upward prominently at wavelengths of 437 nm (3.02 eV), 550 nm (2.25 eV), and 610 nm (2.03 eV) in the spectrum rendered after the first irradiation. This behavior is anomalous by comparison to the other blocks.

Block 39 shows the best response to optical bleaching (see Fig. 1d). Transmissions calculated just after irradiation show up to 70% loss in transmission in the violent/blue spectral region. Gains in transmission through the entire spectrum were made after both trials of optical bleaching.

4) Discussion

These four samples of the DVCS calorimeter show significant variations in response to optical bleaching. This behavior is suspected to be due to variations in crystal structure at the time of their production or accumulated from exposure to radiation during previous experimentation. In particular, different samples may contain different quantities of contaminants, doping agents and defects in the lattice [3]. Quantifying efficiency loss in the entire calorimeter will require a more extensive study of all blocks currently in use and those slated for inclusion in the current upgrade. This procedure will lend itself to a set of quality standards for the manufacturer(s) and a prescriptive method for optical bleaching so as to minimize variations in efficiency among the PbF_2 crystals.

5) Figures

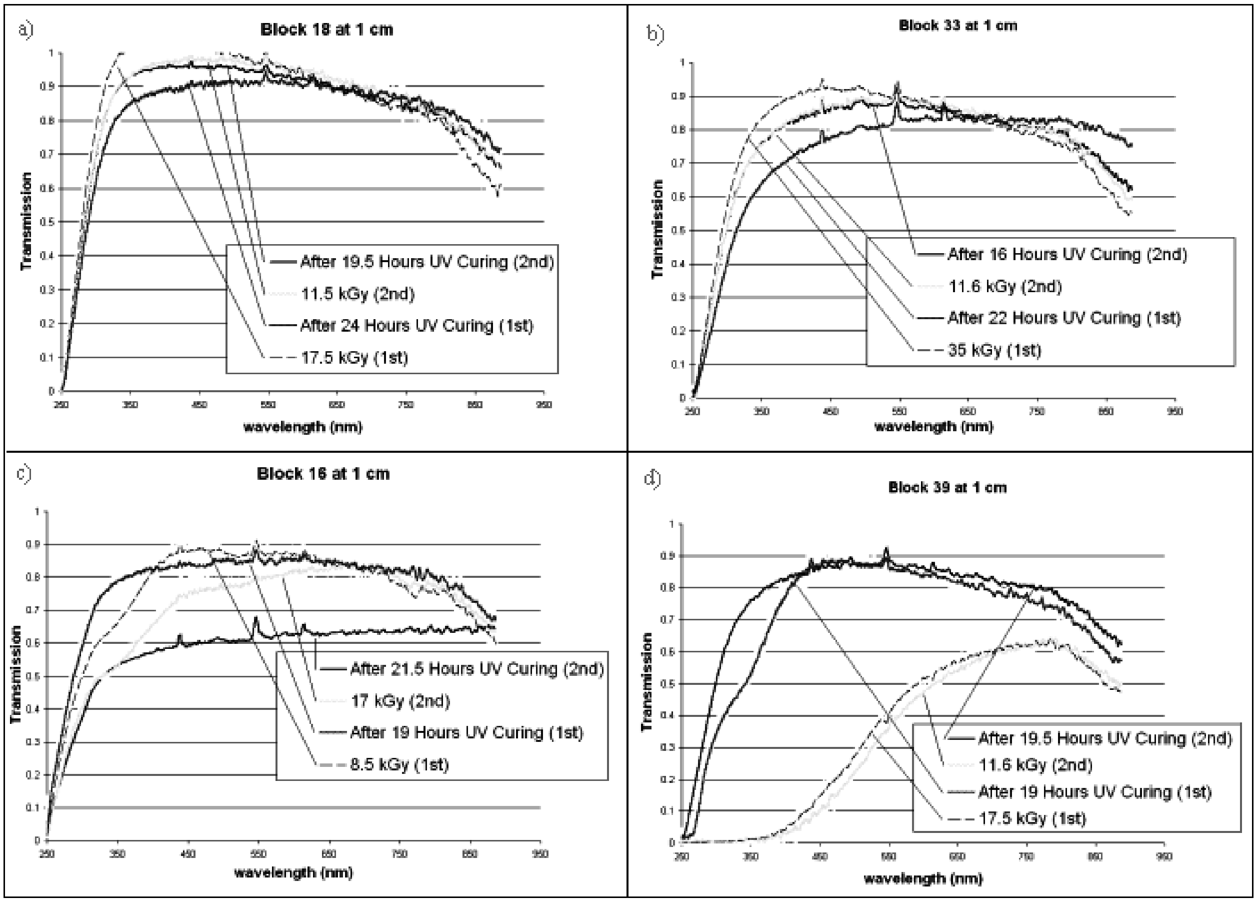

Figure 1. Transmission as a function of wavelength taken at 1 cm for both irradiation/restoration cycles in blocks 18 (a), 33 (b), 16 (c), and 39 (d).

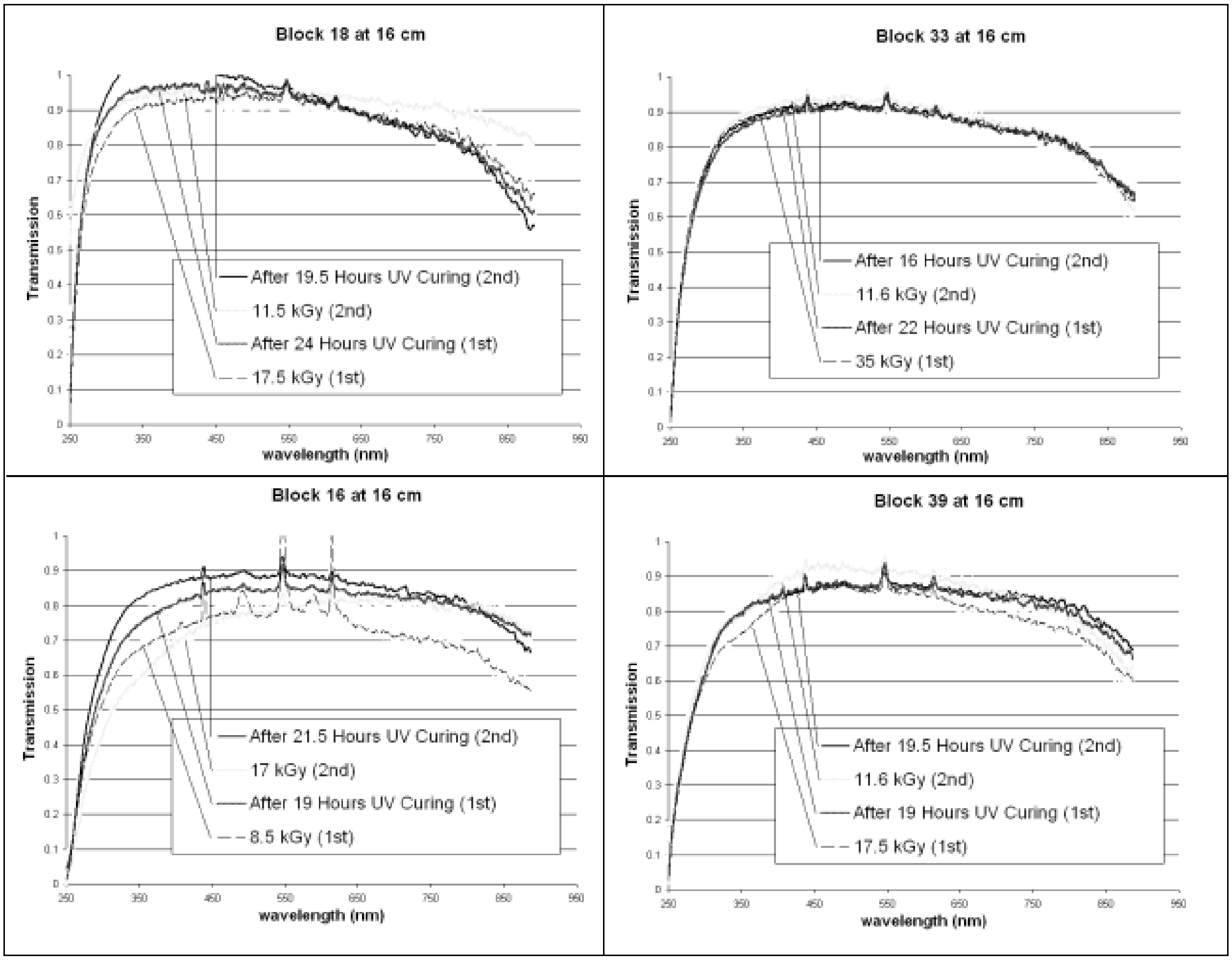

Figure 2. Transmission as a function of wavelength taken at 16 cm for both irradiation/restoration cycles in blocks 18 (a), 33 (b), 16 (c), and 39 (d).

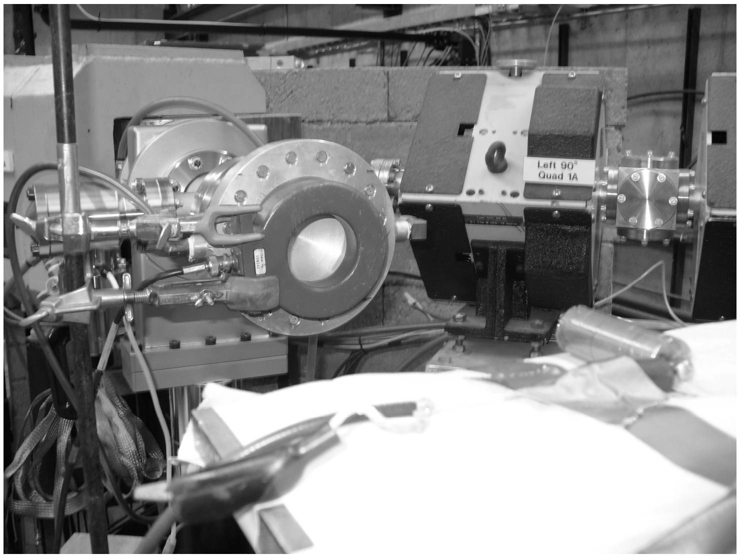

Figure 3. Photograph of experimental design. The current monitor (pick-up coil) is just downstream the exit window of the 25 MeV LINAC while a Faraday Cup holds the position of samples during irradiation.

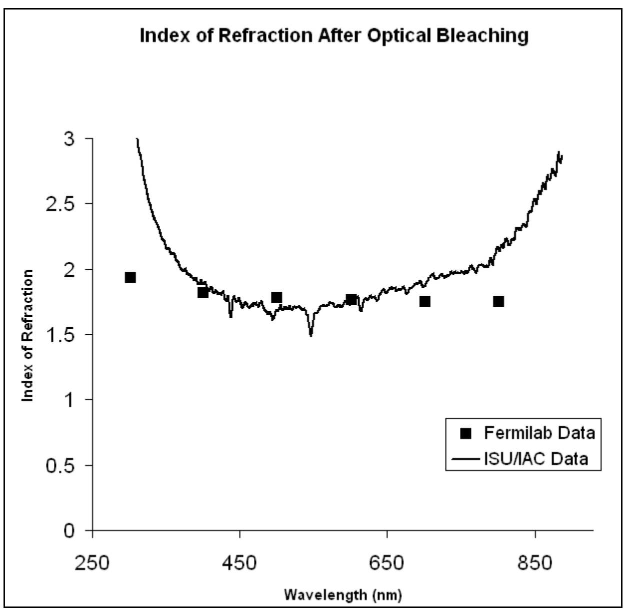

Figure 4. Index of refraction comparison. Squares: Fermilab data taken in 1989 (Anderson). Line: data from this work.

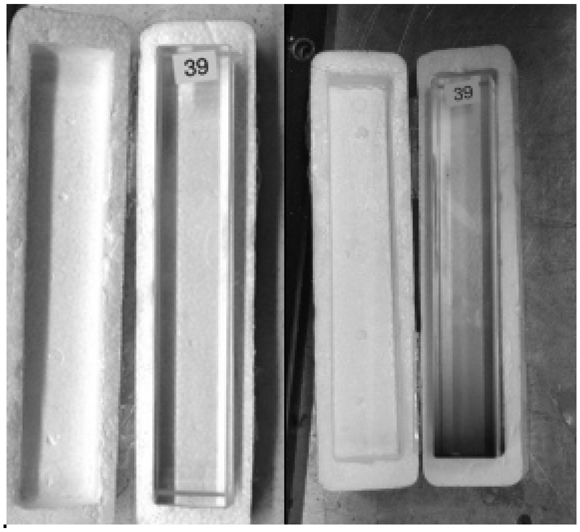

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Figure 5. Photographs of block 39 before (left) and after (right) irradiation.

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