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Radiation damage and recovery properties of common plastics PEN (Polyethylene Naphthalate) and PET (Polyethylene Terephthalate) using a ¹³⁷Cs gamma ray source up to 1.4 Mrad and 14 Mrad

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ABSTRACT: Polyethylene naphthalate (PEN) and polyethylene teraphthalate (PET) are cheap and common polyester plastics used throughout the world in the manufacturing of bottled drinks, containers for foodstuffs, and fibers used in clothing. These plastics are also known organic scintillators with very good scintillation properties. As particle physics experiments increase in energy and particle flux density, so does radiation exposure to detector materials. It is therefore important that scintillators be tested for radiation tolerance at these generally unheard of doses. We tested samples of PEN and PET using laser stimulated emission on separate tiles exposed to 1 Mrad and 10 Mrad gamma rays with a ¹³⁷Cs source. PEN exposed to 1.4 Mrad and 14 Mrad emit 71.4% and 46.7% of the light of an undamaged tile, respectively, and maximally recover to 85.9% and 79.5% after 5 and 9 days, respectively. PET exposed to 1.4 Mrad and 14 Mrad emit 35.0% and 12.2% light, respectively, and maximally recover to 93.5% and 80.0% after 22 and 60 days, respectively.

KEYWORDS: Radiation damage to detector materials (solid state); Radiation-hard detectors; Scintillators, scintillation and light emission processes (solid, gas and liquid scintillators)

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1 Introduction

Polyethylene naphthalate (PEN) and polyethylene teraphthalate (PET) are known organic scintillators with peak emission wavelengths of approximately 450 nm and 350 nm, respectively. General organic scintillators like those traditionally used in high energy physics experiments have light yields of roughly 10,000 photons per MeV. PEN and PET are found to have 10,500 and 2,200 photons per MeV respectively [1].

These are attractive additions to the arsenal of high energy physics experiments' organic scintillators because of their ease of manufacture and low cost. However, high energy physics experiments like those around the LHC [2] and future colliders [3] are creating increasingly problematic radiation zones for the detector equipment, therefore, before considering PEN or PET to replace existing scintillator technologies, it is important to understand how their scintillation light yields respond to high levels of radiation.

For example, the CMS Experiment [4] contains a hadronic calorimeter (HE) composed of layers of SCSN-81 scintillators within brass absorbers, and is considering options to replace the SCSN-81 scintillators as they approach the end of their expected lifetime. The scintillator tiles within the HE calorimeter can expect to receive between 0.4 Mrad to 10 Mrad [5] over 10 years, requiring that any tile selected to replace SCSN-81 must be able to survive doses up to 10 MRad.

With this in mind, two sets of PEN and PET tiles were uniformly irradiated to 1.4 Mrad and 14 Mrad doses of gamma radiation under a 1" diameter by 12" long rod of ¹³⁷Cs at the University of Iowa RadCore facility [6]. Their light yield levels were measured before irradiation and regularly after irradiation, for a period of more than 60 days. Our goal was to understand the light yield attenuation for low dose vs. high dose exposure to PEN and PET, as well as the subsequent recovery time of each sample at each dose.

The reported dose values of 1.4 Mrad and 14 Mrad were provided by the RadCore personnel. The dose is uniform across the samples.

We used 100 mm \times 100 mm \times 2 mm sheets of PET, and 100 mm \times 100 mm \times 1 mm sheets of PEN. The PET sheets were purchased from Goodfellow.com [7]. The PEN sheets were purchased from Teijin [8], under the brand name Scinterex [9], their scintillator formulation of PEN.

This experiment is not an attempt to recreate the CMS radiation environment, only a first indication of how well the plastics will perform in a radiation field. Their performance in this environment indicates whether or not they merit further study. A further study could be undertaken with in situ measurements of various scintillators within the experimental collision hall.

2 Experimental setup

Three tiles of each scintillator were cut to 100×100 mm squares, and the edges were polished. Each tile was tested prior to radiation damage and each tile had measurement results within 5% of each other, showing consistency between tiles. One tile was kept in the dark, not irradiated, for reference, and the other two were placed on the lucite tray used to install the tiles into the radiation source. One sample of the remaining two was placed on the top shelf, the other on the bottom shelf. The top shelf sits closer to the source, and therefore receives a 10x higher dose rate.

The lucite tray was then mounted against the source, and irradiated for 67.614 hours, for a total of 13.87 Mrad on the high shelf, and 1.387 Mrad on the low shelf.

Each tile was tested within an hour after removal from the radiation source. Then they were tested twice each day for the first 2 days after exposure, and then once per day for approximately 20 days, and then once per week for over 60 days after the initial exposure.

Before every test, a reference sample of a generic scintillator was used to ensure stability of measurements in order to reduce systematics from laser and PMT warm up effects.

Between tests, each sample was kept in a light-tight briefcase in order to limit exposure to ambient light, which might influence recovery. The samples were stored and tested in a climate controlled room kept at 22.5 °C, with constant a humidity level.

The tiles were tested by pulsing a 3 ns pulse-width 337 nm nitrogen laser perpendicular to the tile surface, and reading out the scintillated light from the edge of the tile using a Hamamatsu R7600 PMT [10], figure 1. A separate Hamamatsu R7525 PMT [11] was placed directly above the tile to be used as a trigger for the measurements.

The R7600 PMT used in this test is the same PMT used in the Hadronic Forward calorimeter within the CMS Experiment [12]. This PMT has a usable range for detecting photons between 300 and 500 nm, with a peak quantum efficiency near 400 nm, making the PMT well matched to test the scintillation light yields from both the PEN and PET tiles.

This experiment was designed to mimic a true calorimeter readout setup so that any light yield loss could be interpreted as radiation damage, which would also be interpreted as radiation damage by detector technicians. The nature of the light loss exhibited by the tiles is not within the scope of this study, however a good study of radiation effects on the photoluminescence of PEN shows that photon irradiation causes a uniform degradation and uniform recovery across the spectrum [13].

3 Measurement results

We measured the light output from each tile before irradiation, and began testing within 1 hour of removal from the RadCore facility and continued until maximum recovery of the tile was observed. Each measurement was an average of 100 laser pulses using a Tektronics TDS5034 oscilloscope [14].



Figure 1. Experimental test setup.



Figure 2. Average waveforms of the pre irradiation measurement, the 1st measurement immediately after irradiation, and the 22nd measurement after irradiation, taken over 40 days of the PET 14 Mrad dosed sample.

Figure 2 shows three examples of the average waveforms of each of 21 measurements taken over 40 days of the sample of PET irradiated to 14 Mrad.

Tables 1 and 2 summarize the measurement results for PEN and PET respectively. The first column in the table labels the sample, 1.4 Mrad or 14 Mrad. The 'initial light yield' column describes the measured light output compared to the undamaged tile immediately after the exposure.

 Table 1. Summary of PEN irradiation results.

Total Dose	Initial Light Yield	Recovered Light Yield	Recovery Time (days)
1.4 Mrad	$71.4 \pm 2.4\%$	$85.9 \pm 2.4\%$	5
14 Mrad	$46.7 \pm 2.7\%$	$79.5 \pm 2.7\%$	9

Table 2. Summary of PET irradiation results.

Total Dose	Initial Light Yield	Recovered Light Yield	Recovery Time (days)
1.4 Mrad	$35.0 \pm 2.4\%$	$93.5 \pm 2.4\%$	22
14 Mrad	$12.2 \pm 2.7\%$	$80.0 \pm 2.7\%$	60



Figure 3. PEN Results, 21 measurements over 40 days.

After removal from the radiation source, the tiles slowly increase their light yields and plateau to a maximum value. We refer to this period as the recovery period, and refer to the maximum value as the 'recovered light yield' in the third column of the tables. The time it takes to reach the plateau is listed in column 4, as 'recovery time'.

The 1.4 Mrad and 14 Mrad results can be seen in figures 3 and 4, left and right, for PEN and PET, respectively. Each data point in figures 3 and 4 is the integral of the averaged waveforms shown in figure 2, subtracted from 100%. Repeated testing of the reference tile yielded a standard deviation of 3%, which we have applied as an error to each measurement. The plots include fits of the form ' $ae^{-bx} + c$ ', where 'a + c' gives the initial percent damage, 'c' describes the permanent damage, and '1/b' characterizes the rate of recovery.

Figure 5 plots these values as a function of dose and compares PEN to PET. The leftmost plot shows the initial damage immediately after irradiation. The center plot compares the permanent damage between the tiles. The rightmost graph plots the recovery constant. These plots show that



Figure 4. PET Results, 21 measurements over 40 days.



Figure 5. Damage vs Dose characteristics for PEN and PET, comparing total dose with initial damage, permanent damage, and recovery time.

while PEN handles a high radiation environment better than PET, PET has a remarkable ability to recover. PET, however, takes a considerable amount of time to recover.

The damage also appears to be correlated with dose following a logarithmic trend, consistent with Oldham and Ware [15] and Buss et al. [16]. In other words, most of the damage occurs early in the exposure. For example, PET was damaged 65% in the first 1 Mrad of exposure, but increased only 23% further after another 9 Mrad.

4 Conclusions

Samples of Polyethylene Naphthalate (PEN) and Polyethylene Teraphthalate (PET) were dosed to two different amounts, 1.4 Mrad and 14 Mrad. Both exhibited a greater reduction in light yield with higher dose as expected. PET was significantly more sensitive to the radiation than PEN, by a factor of 2 for the 1.4 Mrad dose, and a factor of 3.8 for the 14 Mrad dose. PEN also reached maximum recovery more quickly than PET by a significant margin.

The lower sensitivity to radiation, and the shorter recovery period indicate that PEN is a better candidate for consideration in high radiation zones. Due to its low cost, ease of manufacturing, and nearly 50% light yield remaining after 14 Mrad of gamma radiation, PEN is a good candidate for consideration as a detector scintillator, and should be investigated further. PET is especially susceptible to radiation damage, and is not likely to be useful in collider experiments. A possible copolymer of PEN and PET might reveal superior/optimal radiation-hardness and recovery specifications when compared to the individual components.

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