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Use of poly(ethylene naphthalate) as a self-vetoing structural material

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Abstract. Poly(ethylene naphthalate), PEN, is an industrial polyester which has been shown to scintillate in the blue wavelength region. Combined with measurements of a high intrinsic radiopurity, this has sparked interest in the material for use in low-background experiments.

1. Introduction

Plastic scintillators are of great interest for low-background experiments. Special requirements such as high transparency, high structural integrity and high radiopurity significantly limit the choice of available materials. Poly(ethylene 2,6-naphthalate) or PEN, $[C_{14}H_{10}O_4]_n$, a transparent scintillating polymer, has been suggested as an alternative to commercial scintillators [1].

PEN is an industrial polyester which has been shown to scintillate in the visible blue region [1, 2] which makes the addition of wavelength shifting dopants unnecessary. Its emission-spectrum peaks in the deep blue region which makes it ideal for many photodetectors. The mechanical properties of PEN foils [3] and plates [4] have been measured both at room and cryogenic temperatures and proved that its tensile strength is even superior to copper. Therefore, PEN is a candidate to be used as structural material, for example, as support structure for detectors operated in cryogenic liquids such as liquid argon (LAr) or liquid nitrogen (LN_2).

In this article, the properties of PEN relevant to low-background rare-event physics experiments such as the *Germanium Detector Array*, GERDA [5] and the *Large Enriched Germanium Experiment for Neutrinoless $\beta\beta$ Decay*, LEGEND [6] are discussed.

2. Material properties

PEN is available as granulate from Teijin-DuPont in two variants: TN-8050 SC and TN-8065 S. In the past, PEN foil was already used for signal links and in the production of HV capacitors for the low-background experiments CUORE [7] and GERDA [8], respectively, where it was chosen for its radiopurity.

The radiopurity of the raw material in form of granulate was screened at the Gran Sasso National Laboratory (LNGS) [M. Laubenstein]. A high ^{40}K contamination was observed. This was likely to be due to surface contamination of the granulate. A new screening campaign of pellets is ongoing where the material went through a well-defined cleaning procedure to reduce among others the ^{40}K contamination. Apart from ^{40}K, which is not a limiting factor for neutrinoless double beta decay experiments, the result is comparable to the radiopurity of previously used materials in low-background physics. A more detailed table is available in [4].

Various objects such as plates and containers were made from PEN TN-8065 S using injection moulding. For this process, the optimal manufacturing parameters were investigated to prevent the produced parts from crystallisation and optical defects and thus, maximise transparency and light yield.

The Young's modulus, elastic modulus, and the yield strength, σ_{el}, of the custom-made scintillators were evaluated using the three-point bending flexural test according to DIN EN ISO 178:2013-09 [9]. Comparing the result at room temperature to electrodeposited copper, commonly used in low-background experiments [10], PEN shows a σ_{el} of (108.6 ± 2.6) MPa, around 20 % higher. Repeating the same measurement for PEN in LN_2 results in even higher tensile strength of (209.0 ± 2.8) MPa. The Young's modulus for PEN is lower than for copper, due to the higher flexibility of plastics in general. The measurement showed that the elastic modulus of PEN increased from (1.86 ± 0.01) GPa at room temperature to (3.71 ± 0.08) GPa when immersed in LN_2.

3. Optical properties

For the reproducible measurement of its emission spectrum, $30 \times 30 \times 3$ mm^3 PEN tiles were placed in a dark box and excited with 382 nm light. The excitation light was generated by guiding the light of a deuterium/halogen lamp through a monochromator. Here, a bandwidth of approximately ± 2 nm could be reached. The emission spectrum of PEN was then measured using a spectrometer. A detailed description including all the devices used can be found in [4]. For comparison, the same measurement was repeated with BC-408, a commonly used plastic scintillator, and Methacrylateor (PMMA), a non-scintillating acrylic glass. The blue emission from PEN can be seen in Figure 1 and the emission spectra for all three materials are shown in Figure 2. The wavelength of the peak emission is determined by fitting a second-order polynomial to the peak region. For PEN, this is at (445 ± 5) nm which matches well to the peak quantum efficiencies of many commercially available photomultipliers. A small shift of the peak emission can be observed depending on the path length of the light emitted in PEN. This is due to a "relatively poor" attenuation length the order of 5 cm. Generally, it has been found that the optical properties of PEN are dependent on the production batch. Comparing the amount of detected emission light by integrating the emission spectra in Figure 2 clearly indicates that PEN has a lower light output than BC-408.

The detection of LAr scintillation light with a peak emission wavelength of 126.8 nm [12] succeeds even with modern photomultipliers only with low efficiencies (< 15 %). In low-background experiments, the scintillation light of LAr is also used to veto background events. In order to increase the efficiency of such a veto system, for example, TPB coated materials are used to achieve a wavelength shift.

In order to investigate the wavelength-shifting properties of PEN, it was compared to a TPB-coated (200 μg/cm^2) acrylic tile. Excitation wavelengths from 116 nm to 300 nm were used and

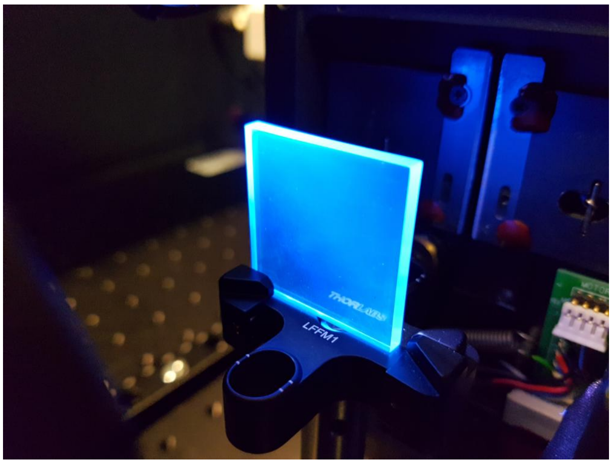

Figure 1. One of the PEN tiles used to obtain the emission spectrum placed in front of the spectrometer slit. For illumination, a 395 nm UV flashlight was used.

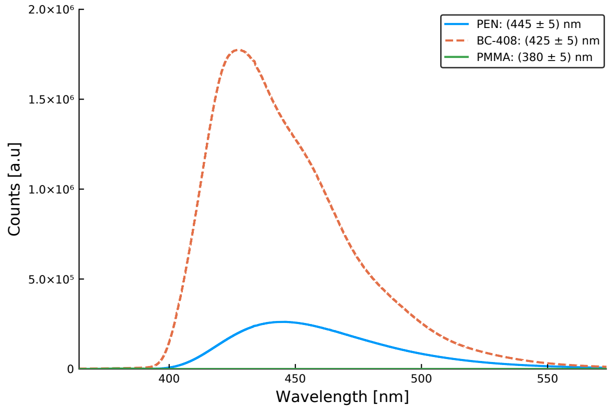

Figure 2. Emission spectra of PEN, BC-408 and PMMA tiles (all $30 \times 30 \times 3$ mm^3) obtained by 382 nm UV light excitation measured using a spectrometer. The wavelength of the peak emission is shown in the figures legend.

the emitted light was measured with a PMT. The ratio of the PMT anode currents for the PEN and the TPB-coated acrylic sample can be seen in Figure 3 (blue line). In addition, the emission spectrum of LAr at 85 K is plotted in orange with normalised intensity [12]. In the region of interest, PEN reaches about 60 % of the TPB-coated acrylic tiles wavelength-shifting efficiency.

Another measurement was performed to compare the light output of PEN to polystyrene (PS) when exposed to mono-energetic electrons. Here, two tiles of identical geometries were used. The emission light was detected using a 1 inch photomultiplier tube (PMT). To get monoenergetic electrons in the range of 0.4 to 1.5 MeV with a narrow energy spread (FWHM = (1.0 ± 0.2) % at 1 MeV) [11] a ^{90}Sr source in combination with an electromagnet was used. The resulting signal strength of PEN and PS for several electron energies, normalised to the emission of PS at 1 MeV, can be seen in Figure 4. The PEN sample emitted about 2.5 times less scintillation light than the PS sample.

4. Conclusions & Outlook

Due to it's high mechanical stability, transparency, relatively good radiopurity and it's various optical properties, PEN is suitable as an active structural material for low-background experiments. The commercially available raw material already shows a radiopurity well below 1 mBq/kg in ^{228}Th and ^{226}Rn without additional purification. With the goal of improving the radiopurity and reducing discoloration of PEN, research into the synthesis of the polymer is underway. For this, the purification of all ingredients and the choice of the catalysts plays a major role. This work in progress is done at Oak Ridge National Laboratory.

The shapes already cast from PEN have shown that scintillator-grade arbitrary shapes and structures can be created by injection moulding. The scintillation emission peaks at (445 ± 5) nm which makes it suitable for many commercially available photomultipliers. The wavelength-shifting efficiency of PEN compared to TPB shows that it can be used to shift the scintillation light of LAr to visible blue light. Furthermore, the scintillation pulses of PEN can be used to discriminate neutrons and gammas [4]. This combination of properties makes it a promising material for future applications in low-background experiments.

PEN could find its first application as structural material in LEGEND. A number of plates have already been produced under clean-room conditions. This production series is currently

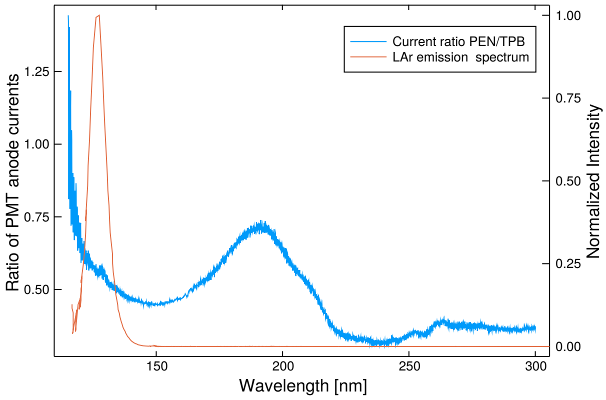

Figure 3. *Left axis:* ratio of the PMT anode currents (blue line) for a PEN tile and a 200 μm TPB-coated acrylic tile excited in the range of 116 to 300 nm with a tunable monochromator. *Right axis:* VUV/UV emission spectrum of LAr (85 K, orange line) with emission peak at 126.8 nm [12].

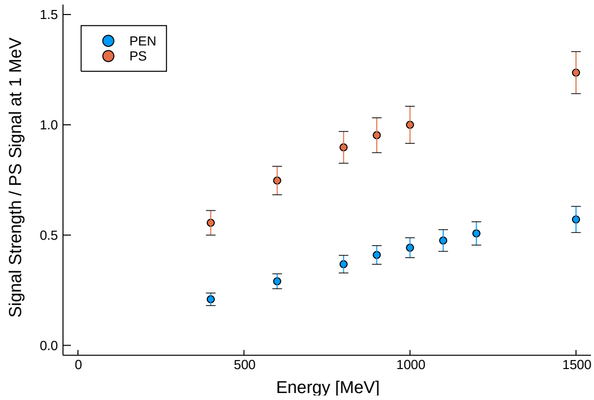

Figure 4. Light yield of PEN and PS tiles (both $30 \times 30 \times 3$ mm^3) excited using electrons from a ^{90}Sr source which, in combination with an electromagnet, produces electrons in the range of 0.4 to 1.5 MeV with a narrow energy-spread (FWHM $= (1.0 \pm 0.2)$ % at 1 MeV).

being screened for radiopurity. In addition, proof of principle tests are ongoing using PEN as support structure for Ge detectors. If these tests and the results of the radiopurity screening are successful, PEN can be used as a holding structure for detectors in LEGEND-200.

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