Fluorescence yield for plastic scintillators after irradiation

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Abstract

The ratio of the fluorescence yield versus wavelength for plastic scintillators EJ-408 and EJ-260 before and after irradiation by a ^{60}Co source for doses of 50, 30, 10, 4, and 2 Mrad at various dose rates and for different concentrations of the primary and secondary dopant. While the nominal dopant concentration gives the highest light output prior to irradiation, a higher concentration is found to be optimal for irradiated plastics.

Keywords: plastic scintillator, fluorescence, radiation hardness

1. Introduction

Organic scintillators such as polystyrene (PS) or polyvinyltoluene (PVT) containing wave-length shifting additives in solution have long been popular elements in detectors used in particle physics, nuclear physics, radiation safety, and heath physics applications due to their high light output, low cost, fast response, and versatility of physical construction. Prolonged exposure of plastic scintillator to ionizing radiation, however, can result in damage: light self-absorption (yellowing) increases and the transfer efficiency of the initial excitation of the polymer to the dopants combined with the probability of radiative decays for the

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dopants ("initial light output") can lessen. In this paper, we present measurements of ratio of the light output before and after irradiation for two different types of plastic scintillator manufactured by Eljen corporation, EJ-200 (similar to BC-408 from Bicron corporation) and EJ-260 (similar to BC-428), before and after irradiation by a ^{60}Co source for doses of 50, 30, 10, 4, and 2 Mrad at various dose rates and for different concentrations of the primary and secondary dopant. Dose rate effects are of interest because materials testing is typically done at much higher dose rates, because of the cost of reactor time, than the scintillator will experience in use.

The effect of dose and dose rate on radiation-induced yellowing have been the subject of many investigations. The results have been summarized in [1]. Immediately after irradiation at a high dose rate (Mrads/hr), the probability of absorption for visible light increases for wavelengths shorter than around 600 nm (yellow) due to color centers that form in the substrate[2]. After irradiation, interactions of oxygen that diffuses into the plastic reduces the color center density, and the plastic "bleaches". On time scales of orders of months, the bleaching stops, and some permenant damage (color centers) remains [1]. Oxygen is essential for this curing process, although samples irradiated in inert atmospheres have smaller permenant damage, even if the initial damage for high dose rate exposures is larger[3].

Dose rate effects for the initial yellowing, immediately after irradiation, are well documented [1],[4],[5],[6],[7],[8]. Here again the presense of oxygen plays an important role. The studies show that the penetration depth of oxygen into the substrate depends on the dose rate: at lower dose rates, oxygen penetrates more deeply. Because of the importance of the interaction of oxygen with radicals produced from the substrate during irradiation, this can lead to dose rate effects. Simulations based on oxygen diffusion have been shown to reproduce the time dependence of the induced attentuation length in scintillators based on polystyrene and PMMA[5]. As discussed in [9], the presense of oxygen increases the number of migration mechanisms for the radicals produced during irradiation.

Studies on the effect of radiation on the light yield are fewer than those on self-absorption. In an early study, Rosman and Zimmer[10] found that the light output for organic scintillators based on polystyrene versus dose is described, approximately and for relatively large dose rates, by a double exponential, with a small-dose component whose constant is approximately 100 Mrad. For a polystyrene scintillator doped with 1.5% TPB, they also used UV light to illuminate the samples and found that this showed less reduction of light than excitation via charged particles. Since polystyrene is transparent to UV, this indicates that the TPB was not damaged, and instead the damage was either to the polystyrene or the migration of excitation from the polystyrene to the dopant. Subsequent studies ([11] [8]) came to a similar conclusions for liquid scintillators and for various plastic scintillators respectively.

In another study by Bross, Pla-Dalmau, et al. from 1991[2], the authors look at light output for various primary (DAT, MOPOM, OLIGO408, OLIGO415A) and seconary (BBQ, K27, DMPOPOP, 3HF) dopants in PS for a dose of 10 Mrad accumulated with a relatively high (0.4 Mrad/h) dose rate. They looked at this for 2 different concentrations of the secondary dopant. They optimized the concentration of the first dopant using the light output before irradation. They found no change to the intrinsic light output, indicating at high dose rates the dopants were not damaged by this large dose.

In [12] (perhaps better documented in [1]), the authors show that for a PS fiber with primary dopant of PTP and secondary dopant of 3HF, increasing the dopant concentration from nominal to 20 times nominal continuously increases the light output after 100 Mrad (dose rate unknown).

In [13], the radiation resistance of BC-408 (from Bicron Corporation) was studied for different concentrations of the dopant from half to 3/2 the nominal concentration. The study was done at very high dose rate (36 Mrad/hr) and a total dose of 3 Mrad. They saw that varying the concentration of the secondary fluor did not affect the output as studied with a ^{207}Bi electron source. They found decreasing the secondary dopant made it less rad hard, but increasing did not help.

In [5], decreased light output for a plastic scintillator from Kuraray, SCSN-38, which is based on polystyrene with b-PBD primary and BDB secondary dopants, was seen. The main absorption wavelengths for polystyrene typically range between between 230 to 260 nm, for the primary dopant b-PBD between 270 to 330 nm, and for the wavelength shifting dopant BDB between 310 to 400 nm. They found that light loss was much stronger in the presence of oxygen. Note that while oxygen plays a beneficial role in regards to annealing of induced absorption length at the end of radiation, it plays a detrimental role in regards to light output. They also found the light output loss was independent of the wavelength of the light that was used to excite the scintillator when it was varied between 230 and 400 nm. From this they conclude that the damage is due to destruction of the second flour. This is different than what was found in the other studies, which indicated that damage to the dopants was small and that damage was mostly to the substrate, although different dopants were used in the studies. By looking at the damage as a function of the thickness of the scintillator, they concluded that the BDB molecules are mainly destroyed near the surface, which lends support to a mechanism involving oxygen diffusion.

If the damage is related to oxygen diffusion, a dose rate effect is expected. An interesting result, from 1996, is described in [14]. They look at light output reduction for two polystyrene-based scintillators (SCSN-38 and SCSN-81, from Kuraray) and a polyvinyltoluene-based scintillator (Bicron-499-35). Measurements were done as a function of time subsequent to irradiation, until the light output stabilized, using a ^{241}AM alpha source. They find a reduction in post-recovery light output that depends linearly on the log of the dose rate for dose rates ranging from 0.01 to 2 Mrad/hr. Note that the effect is not small: for a dose of 2 Mrad, the light loss is negligible for a dose rate of 2 Mrad/hr but 20% for a dose rate of 0.01 Mrad/hr for SCSN-81.

In order to understand the relative role of destruction of the dopants versus damage to the substrate for modern scintilating plastics, we have studied the light output for two plastic scintillators, EJ-408 and EJ-260, varying the concentration of the dopants, for different total doses and dose rates.

2. Measurements

Both EJ-408 and EJ-260 use PVT as a substrate. EJ-408 has a light output that is 60% of anthracene and a wavelength of maximum emission of 435 nm. EJ-260 also has a light output that is 60% of anthracene but a wavelength of maximum emission of 490 nm. Eljen prepared scintillator bars with dimensions of 1x1x5cm. For the EJ-408, bars were made with concentration of the primary scintillation dopant at 0.5, 1.0, 1.5, and 2.0 that of the standard concentration. For the EJ-260, bars with made with concentrations of the primary (x) and secondary (p) dopant of 1x1p, 1x2p,1x4p,2x1p,4x1p,2x2p, and 4x4p. The fluorescence output was measured using a fluoromax-4 fluorometer by Horiba Scientific using a right-angle configuration and an excitation wavelength of XXX nm. For the absorption measurements, a xxx by xxx was used. Air was used as the reference. In [2], the integration of the fluoresence spectra was shown to reproduce the results using a ^{207}Bi source to within a few percent for most dopants.

Radiations were done using a ^{60}Co source at the Univesity of Maryland with an activity of XXXXX. The dose was measured using...

Figure1 shows the spectra for EJ-408 with nominal doping before irradiation and after 30 Mrad at 1 Mrad/hr and after 50 Mrad at 1 Mrad/hr.

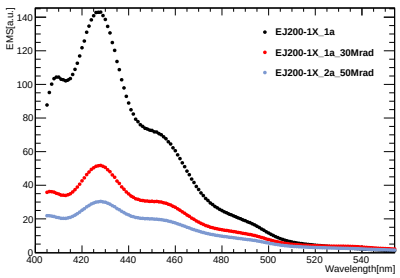

Figure 1: Emission spectrum for EJ-408 at nominal doping before irradiation, after 30 Mrad at 1 Mrad/hr and after 50 Mrad at 1 Mrad/hr.

3. Interpretation

If this dose rate effect is due to oxygen diffusion, then we expect it to be governed by the diffusion equation. Since the oxygen diffusion depth is greater at lower dose rates, we expect the damage for the same dose at different dose rates to increase up to the point where the dose rate is low enough that oxygen permeates the entire sample. The dose rate effect should plateau at this point. Specifically, we predict that the light output reduction should depend on both the dose and the dose rate as:

$$L(R, D) = 1 - [f(D)Z(R) + a(D)(1 - Z(R))]$$

where L is the % light yield, D is the dose, R is the dose rate, Z(R) is the fraction of the scintillator containing oxygen and is given by $min(\frac{2z_0(R)}{d}, 1)$ where d is the thickness of the scintillator and z_0 gives the depth of scintillator penetrated by oxygen, a(D) is related to the fraction of quenching in the part of the scintillator containing oxygen and is given by $1 - e^{-a_0 D}$, and f(D) is related to the fraction of quenching in the part of the scintillator not containing oxygen and is given by $1 - e^{-f_0 D}$. The diffusion depth $z_0(R)$ is given by diffusion theory as

$$z_0(R) = \sqrt{\gamma / R}$$

where γ is a property of the substrate and the radicals that are dissolved in that substrate during the diffusion process. The value for γ depends on the material, oxygen pressure, radical concentration (which is proportional to the dose) and temperature. The minimization in Z(R) occurs because the fraction of scintillator containing oxygen can not exceed one. Note that this equation assumes the temperature and the surrounding atmosphere is held constant, as this can affect diffusion.

4. Conclusions

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