

Fluorescence yield for plastic scintillators after irradiation

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Abstract

The ratio of the fluorescence yield versus wavelength for plastic scintillators EJ-200 and EJ-260 before and after irradiation by a ^{60}Co source for various doses and dose rates and for different concentrations of the primary and secondary dopant was measured. 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
5 health 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

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10 the dopants (“initial light output”) can lessen. In this paper, we present mea-
 surements of the 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 various doses and dose rates
 15 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. Dose rate effects may be responsible for the factor of three
 larger light loss with integrated luminosity in the CMS endcap calorimeter [1, 2]
 20 than expected based on high dose rate exposures using ^{60}Co sources.

The effect of dose and dose rate on radiation-induced yellowing have been
 the subject of many investigations. The results were summarized in [3]. Im-
 mediately after irradiation at a high dose rate (Mrads/hr), the probability of
 absorption for visible light increases for wavelengths shorter than around 600
 25 nm (yellow) due to color centers that form in the substrate [4]. After irradiation,
 interactions with oxygen that diffuses into the plastic reduces the color center
 density, and the plastic “bleaches”. On time scales of the order of months,
 the bleaching stops, and some permanent damage (color centers) remains [3].
 Oxygen is essential for this curing process, although samples irradiated in inert
 30 atmospheres have smaller permanent damage, even if the initial damage for high
 dose rate exposures is larger [5].

Dose rate effects for the initial yellowing, immediately after irradiation, are
 well documented [3, 6, 7, 8, 9, 10, 11]. Here again the presence of oxygen plays
 an important role. The studies show that the penetration depth of oxygen into
 35 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 radi-
 cals 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 attenuation length in scintillators based
 40 on polystyrene and PMMA [7]. As discussed in [12], the presence 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 [13] found that the light
45 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
50 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 ([14, 10]) came to a similar conclusions for liquid scintillators and for various plastic scintillators respectively.

55 In another study, by Bross, Pla-Dalmau, *et al.* from 1991 [4], the authors looked at light output for various primary (DAT, MOPOM, OLIGO408, OLIGO415A) and secondary (BBQ, K27, DMPOPOP, 3HF) dopants in PS for a dose of 10 Mrad accumulated with a relatively high dose rate (0.4 Mrad/h). They looked at this for 2 different concentrations of the secondary dopant. They
60 optimized the concentration of the first dopant using the light output before irradiation. They found no change to the intrinsic light output, indicating at high dose rates the dopants were not damaged by this large dose.

In [15] (perhaps better documented in [3]), the authors show that for a PS fiber with primary dopant of PTP and secondary dopant of 3HF, increasing the
65 dopant concentration from nominal to 20 times nominal continuously increases the light output after 100 Mrad (dose rate unknown).

In [16], 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
70 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 [11], from 1993, SCSN-81, SCSN-23, and 3HF (based on PS) were studied
75 at doses between 10 krad to 1 Mrad and three dose rates, 1.2 Mrad/hr, 0.01 Mrad/h, using a long piece of plastic and a ^{90}Sr beta source. They also looked at 0.0003 Mrad/hr with doses of 1.3 and 0.57 Mrad. They saw a small decrease in light output after recovery.

In [7], decreased light output for a plastic scintillator from Kuraray, SCSN-
80 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.
85 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
90 destruction of the second fluor. 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
95 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 [17]. 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
100 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
105 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 scintillating plastics, we have studied the light output for two plastic scintillators, EJ-200 and EJ-260, varying the concentration of the dopants, for different total doses and dose rates.

110 2. Measurements

Both EJ-200 and EJ-260 use PVT as a substrate. EJ-200 has a light output that is 60% of anthracene and a wavelength of maximum emission of 435 nm (blue). EJ-260 also has a light output that is 60% of anthracene but a wavelength of maximum emission of 490 nm (green). Eljen prepared scintillator bars
115 with dimensions of 1x1x5cm. For the EJ-200, 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 were 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
120 wavelength of xx nm. For the absorption measurements, a xx by xx was used. Air was used as the reference. In [4], the integration of the fluorescence spectra was shown to reproduce the results using a ^{207}Bi source to within a few percent for most dopants.

125 Radiations were done using a ^{60}Co source at the University of Maryland with an activity of xxx. The dose was measured using i don't know

Figure 1 shows the spectra for EJ-200 with nominal doping before irradiation and after 30 Mrad at 1 Mrad/hr and after 50 Mrad at 1 Mrad/hr.

Table 1 shows the ratio of the light output at xxx nm for the EJ-200 to that
130 before irradiation for different doses, dose rates, and dopant concentrations. Figure 2 shows the ratio of the light output at xxx nm for the EJ-200 to that



Figure 1: Emission spectrum for EJ-200 at nominal doping before irradiation, after 30 Mrad at 1 Mrad/hr and after 50 Mrad at 1 Mrad/hr.

before irradiation as a function of dose rate for several total doses.

Tabl 2 shows the ratio of the light output at **xxx nm** for EJ-260 to that before irradiation for different doses, dose rates, and dopant concentrations.

135 Figure 3 shows the ratio of the light output at **xxx nm** for EJ-260 to that before irradiation as a function of dose rate for several total doses.

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

Table 1: Ratio of average light output at **xxx nm** to that before irradiation for EJ-200 scintillator. For the concentrations of the dopants, NpMs refers to N times the default concentration of primary and M times the default concentration of the secondary dopant the commercial version of the product.

Dose (Mrad)	Dose rate (Mrad/hr)	light output ratio for dopings			
		1p1s	0.5p1s	1.5p1s	2.0p1s
0	—	1.0	xx	xx	xx
2	0.1	xx	xx	xx	xx
2	0.3	xx	xx	xx	xx
2	1.0	xx	xx	xx	xx
4	0.1	xx	xx	xx	xx
4	0.3	xx	xx	xx	xx
4	1.0	xx	xx	xx	xx
10	0.3	xx	xx	xx	xx
10	1.0	xx	xx	xx	xx
30	1.0	xx	xx	xx	xx
50	1.0	xx	xx	xx	xx

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



Figure 2: Ratio average light output for EJ-260 at **xxx nm** to that before irradiation versus dose rate for several total doses.

140 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.

145 4. Conclusions

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Table 2: Ratio of light output at **xxx nm** to that before irradiation for EJ-260 scintillator. For the concentrations of the dopants, NpMs refers to N times the default concentration of primary and M times the default concentration of the secondary dopant the commercial version of the product.

Dose (Mrad)	Dose rate (Mrad/hr)	light output ratio for dopings						
		1p1s	2p1s	4p1s	1p2s	1p4s	2p2s	4p4s
0	—	1.0	xx	xx	xx	xx	xx	xx
2	0.1	xx	xx	xx	xx	xx	xx	xx
2	0.3	xx	xx	xx	xx	xx	xx	xx
2	1.0	xx	xx	xx	xx	xx	xx	xx
4	0.1	xx	xx	xx	xx	xx	xx	xx
4	0.3	xx	xx	xx	xx	xx	xx	xx
4	1.0	xx	xx	xx	xx	xx	xx	xx
10	0.3	xx	xx	xx	xx	xx	xx	xx
10	1.0	xx	xx	xx	xx	xx	xx	xx
30	1.0	xx	xx	xx	xx	xx	xx	xx
50	1.0	xx	xx	xx	xx	xx	xx	xx

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Figure 3: Ratio average light output for EJ-260 at **xxx nm** to that before irradiation versus dose rate for several total doses.

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