Radiation damage of scintillator rods with different concentrations of dopants

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Abstract

The performance of plastic scintillator degrades when exposed to radiation. In this paper, the reduction in light output is studied for scintillators with varying concentrations of the primary dopant and secondary dopant, over a wide range of dose rates. The scintillators used polystyrene or polyvinyltoluene as the substrate, and produced blue or green light. some findings here

Keywords: organic scintillator, radiation hardness, calorimetry

1. Introduction

Plastic scintillator has long been an inexpensive way to detect charged particles produced in particle physics experiments. However, its light output decreases with accumulated dose. Designers of experiments in high radiations environments expected at future hadron colliders such as the Future Circular Collider at CERN[1] or the SppC in China[2] would benefit from an increased understanding of radiation damage in plastic scintillators.

Plastic scintillator consist of a substrate, often polystyrene (PS) or polyvinyltoluene (PVT), into which wavelength shifting primary and secondary dopants have been dissolved. When a charged particle traverses the scintillator, the molecules of the substrate are excited. This excitation can be transferred to

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the primary dopant radiatively in the deep UV at low concentrations or via the Förster mechanism [3] at concentrations above $\approx 1\%$ [4]. The primary dopant transfers the excitation radiatively to the secondary dopant. The visible light produced by de-excitation of the secondary flour must traverse the scintillator to reach a photodetector, and can be absorbed by "color centers" along its path. As the dopants are expensive, commericial scintillator typical has a 1% concentration of the primary dopant and a 0.1% concentration of the secondary.

When plastic scintillator is subjected to ionizing radiation, light self-absorption by the color centers increases ("yellowing") when the radicals created during irradiation are unterminated or re-terminate in ways that absorb visible light. The transfer efficiency of the initial excitation of the polymer to the dopants combined with the probability of radiative decays for the dopants ("initial light output") can lessen when bonds form that absorb in the ultraviolet (UV) or that decrease the relevent de-excitation mechanisms.

If the damage is primarily to the initial light output, increasing the concentration of the dopants may lessen the light reduction, as the amount of light transmitted via the Förster mechanism will increase, and the average path length from the primary to the secondary will decrease.

The light output as a function of dopant concentration has been studied at high dose rate [5, 6]. In Ref. [5], the authors studied light output of a variety of blue and green polystyrene-based scintillators after a dose of 10 Mrad accumulated at a high dose rate (400 krad/hr). They considered two concentrations of the secondary dopant, 0.01% and 0.02% after optimizing the concentration of the primary using unirradiated scintillators. They found the light output after irradiation did not depend on the dopant concentration. In Ref. [6], the radiation resistance of BC-408 [1], a blue scintillator, 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 (36000 krad/hr) and a total dose of 3 Mrad. They saw that varying the concentration of the secondary dopant did not

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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 this paper, we present measurements of the light output of blue and green scintillator rods before and after irradiation for various concentrations of dopants, and as a function of the material used for the substrate for dose rates from xxx to xxx.

2. Dose rate effects

The effect of radiation on plastics is known to depend both on dose and dose rate. [7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21] Two well-studied sources of dose rate effects in plastics involve oxygen. The penetration depth of oxygen during irradiation depends on the dose rate. Where oxygen is present, peroxides, which absorb in the UV, form almost instantaneously. Oxygen also allows the formation of different types of permenent color centers. For PS, the penetration depth goes as [9, 22].

$$z_0^2 = \frac{4mm^2\ krad/hr}{\dot{d}} \tag{1}$$

where $\dot{d}$ is the dose rate. There is an abrupt transition between areas with and without oxygen. The oxygen concentration in the oxidized regions is almost uniform [23]. For a tile with a thickness of 4 mm, oxygen permeates the entire sample at a dose rate of 1 krad/hr. For dose rates above this value, the damage to the scintillator will depend on the dose rate, as peroxides and color centers containing oxygen will form in this regions, but will not in the regions without. The rate of peroxide formation in the areas containing oxygen can show dose rate effects as well [23]. The rate of peroxide formation will scale as the square root of the dose rate if the bond formed is bimolecular.

3. Sample and irradiation details

The rods were supplied by Eljen Corporation, and are similar to EJ-200, a blue scintillator, and EJ-260, a green one, using either PS or PVT as the substrate. They are rectangular, with dimensions of 1x1x5 cm^3, and the edges were

diamond milled. The concentration of the primary dopant and the secondary dopant was 0.5, 1.0, and 2.0 times the nominal concentration. Fig. 1 [left] shows a photograph of some of the rods.

High dose rate irradiations (xxx–xxx krad/hr) were performed at the National Institute of Standards and Technology, Gaithersburg, MD, using their Cobalt-60 (Co-60) source. Intermediate dose rate irradiations (xxx–xxx krad/hr) were performed at Goddard Space Flight Center's Co-60 source. Very low dose rate were measured using the GIF++ facility[24] at CERN. The source is Cs-137? and the dose rate xxx krad/hr.

4. Measurement technique

The light output from the rods is measured before and after irradiation using an alpha source (^{269}Pu), as shown in Fig. 1 [right]. Before measurement, the rods were allowed to anneal at least 2 months, until all the temporary damage was gone. The rod is placed on a Hamamatsu R6091 photomultiplier tube, and the source is placed on the rod. An alignment jig ensures reproducibility of the alignment of the three pieces. The measurements were made using a textronics scope XXXXX, with a charge integration window of xxμs. A typical output spectrum is shown in Fig. 1 [right]. After pedestal subtraction, the distribution is fit to a Gaussian near the peak, and its mean is used as a measure of the light yield. The uncertainty in the light output is taken as the variation in this mean for measurements taken at different times, and after removing and replacing the rod from the jig, and is $\pm 2\%$.

The amount of radiation damage is quantified using D defined in Equation 2.

$$\frac{L(d)}{L_0} = \exp -d/D \tag{2}$$

where $L(d)$ is the measured light output after a dose d, L_0 is the measured initial light output, and D is the exponential dose constant from the fit.

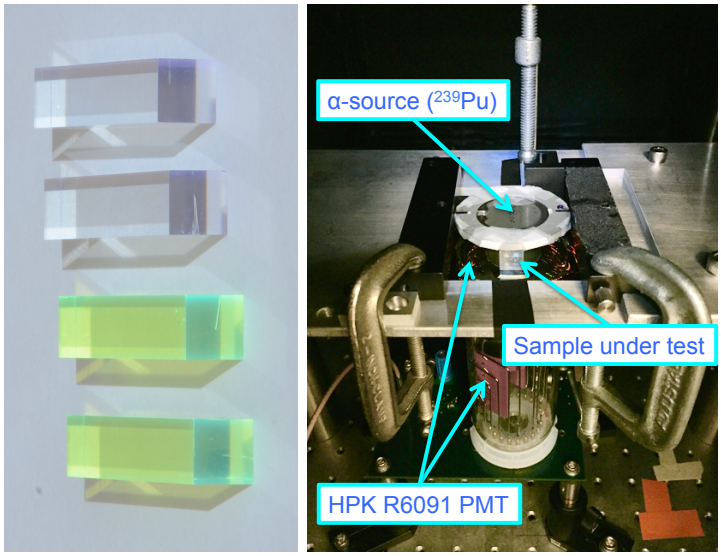

Figure 1: [left] photograph of some of the rods. From top to bottom: EJ200 with nominal dopings, EJ300 with twice the nominal concentration of the primary dopant, EJ260 with nominal dopings, EJ260 with twice the nominal concentration of the primary dopant. [right] Apparatus for measurements with alpha source.

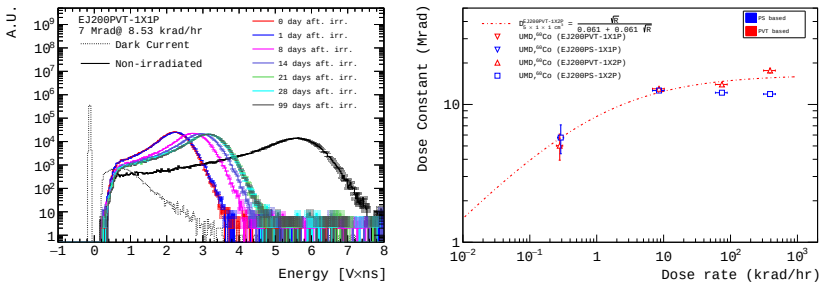

Figure 2: [left] A typical energy spectrum [right] resulting dose constants and comparison with the HE data. need version without HE data

Sample ID	Dose rate [krad/hr]	Dose constant [Mrad]
EJ-200-PS-1X2P	0.3	5.75 ± 1.36
EJ-200-PVT-1X2P		5.02 ± 1.08
EJ-260-PS-1X2P		5.10 ± 1.18
EJ-260-PVT-1X2P		5.03 ± 1.18
EJ-200-PS-1X1P	8.53	12.26 ± 0.26
EJ-200-PS-1X2P		12.62 ± 0.28
EJ-200-PVT-1X1P		12.46 ± 0.28
EJ-200-PVT-1X2P		12.99 ± 0.30
EJ-200-PS-2X1P	8.34	12.14 ± 0.23
EJ-200-PVT-2X1P		13.54 ± 0.30
EJ-260-PS-1X1P		14.80 ± 0.41
EJ-260-PVT-1X2P		12.53 ± 0.30
EJ-200-PS-1X1P	74.4	10.63 ± 0.11
EJ-200-PS-1X2P		12.17 ± 0.15
EJ-200-PVT-1X1P		13.64 ± 0.25
EJ-200-PVT-1X2P		13.97 ± 0.26
EJ-200-PS-1X1P	390.0	9.96 ± 0.21
EJ-200-PS-1X2P		11.90 ± 0.32
EJ-200-PVT-1X1P		16.06 ± 0.57
EJ-200-PVT-1X2P		17.62 ± 0.69
EJ-200-PVT-2X1P	390.0	16.92 ± 0.56
EJ-260-PS-1X1P		11.94 ± 0.31
EJ-260-PVT-1X2P		13.92 ± 0.43

Table 1: (*) Sample type, dose rate, and dose constant. In the naming convention EJ-XXX-YY-NXMP, XXX refers to blue (200) or green (260) scintilator, YY refers to the substrate (PS or PVT), NX refers to the concentration of the secondary relative to the nominal, and YP refers to the concenation of the primary dopant relative to the nominal.

5. Results

6. Conclusions

7. Acknowledgments

The authors would like to thank Chuck Hurlbut of Eljen Corporation for supplying many of the rods. The authors would like to thank the staff Goddard Space Flight Center and at the National Institute of Standards and Technology irradiation Facilities group for assistance with the irradiations. This work was supported in part by U.S. Department of Energy Grant DESC0010072.

References

[1] M. Benedikt, The global future circular colliders effort.

[2] J. Tang, *et al.*, Concept for a future super proton-proton colliderarXiv:1507.03224.

[3] T. Forster, Z. Naturforsch. a4 (1949) 321.

[4] J. Birks, The Theory and Practice of Scintillation Counting, International Series of Monographs on Electronics and Instrumentation, Volume 27, Pergamon Press, The Macmillan Company, New York, 1964. doi:https://doi.org/10.1016/C2013-0-01791-4.

[5] A. Bross, A. Pla-Dalmau, B. Baumbaugh, J. Godfrey, J. Jaques, J. Marchant, J. Piekarz, R. Ruchti, Development and characterization of new scintillation materials for fiber tracking and calorimetry, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 307 (1) (1991) 35 – 46. doi:http://dx.doi.org/10.1016/0168-9002(91)90128-D.

[6] S. Majewski, M. Bowen, C. Zorn, K. Johnson, V. Hagopian, J. Thomaston, H. Wahl, Radiation damage studies in plastic scintillators with a $2.5-$MeV electron beam, Nuclear Instruments and Methods in Physics

Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 281 (3) (1989) 500 – 507. doi:http://dx.doi.org/10.1016/0168-9002(89)91483-6.

[7] C. Zorn, Plastic and liquid organic scintillators, in: F. Sauli (Ed.), Instrumentation in High Energy Physics, 2nd Edition, World Scientific, 1993, Ch. 4, pp. 218–279. doi:10.1142/9789814360333_0004.

[8] U. Holm, K. Wick, Radiation stability of plastic scintillators and wavelength shifters, Nuclear Science, IEEE Transactions on 36 (1) (1989) 579–583. doi:10.1109/23.34504.

[9] K. Wick, D. Paul, P. Schrder, V. Stieber, B. Bicken, Recovery and dose rate dependence of radiation damage in scintillators, wavelength shifters and light guides, Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms 61 (4) (1991) 472 – 486. doi:http://dx.doi.org/10.1016/0168-583X(91)95325-8.

[10] B. Bicken, U. Holm, T. Marckmann, K. Wick, M. Rohde, Recovery and permanent radiation damage of plastic scintillators at different dose rates, Nuclear Science, IEEE Transactions on 38 (2) (1991) 188–193. doi:10.1109/23.289295.

[11] B. Bicken, A. Dannemann, U. Holm, T. Neumann, K. Wick, Influence of temperature treatment on radiation stability of plastic scintillator and wave-length shifter, Nuclear Science, IEEE Transactions on 39 (5) (1992) 1212–1216. doi:10.1109/23.173180.

[12] A. Bross, A. Pla-Dalmau, Radiation damage of plastic scintillators, Nuclear Science, IEEE Transactions on 39 (5) (1992) 1199–1204. doi:10.1109/23.173178.

[13] N. Giokaris, M. Contreras, A. Pla-Dalmau, J. Zimmerman, K. Johnson, Study of dose-rate effects on the radiation damage of polymer-based scsn23, scsn81, scsn81+y7, scsn81+y8 and 3hf scintilla-

tors, Radiation Physics and Chemistry 41 (12) (1993) 315 – 320. doi:http://dx.doi.org/10.1016/0969-806X(93)90069-7.
URL http://www.sciencedirect.com/science/article/pii/0969806X93900697

[14] K. Gillen, M. Celina, Predicting polymer degradation and mechanical property changes for combined radiation-thermal aging environments, RC&T 91 (2018) 27. doi:https://doi.org/10.5254/rct.18.81679.

[15] V. Khachatryan, et al., Dose rate effects in the radiation damage of the plastic scintillators of the cms hadron endcap calorimeter, JINST 11 (2016) T10004. doi:10.1088/1748-0221/11/10/T10004.
URL http://stacks.iop.org/1748-0221/11/i=10/a=T10004

[16] S. Shalaby, R. Clough, Radiation effects on polymers, Am. Chem. Soc. Symp. Ser. 475 (1991) 457.

[17] J. Bolland, Kinetic studies in the chemistry of rubber and related materials, J. L. Proc. R. Soc. A186 (1946) 218.

[18] Bolland, Kinetic studies in the chemistry of rubber and related materials. VII. –influence of chemical structure on the α-methylenic reactivity of olefins, Trans. Faraday Soc. 46 (1950) 358. doi:http://dx.doi.org/10.1039/TF9504600358.

[19] L. Bateman, Olefin oxidation, Q. Rev. Chem. Soc 8 (1954) 147.

[20] A. Cunlifee, Davis, Photo-oxidation of thick polymer samples - part II: the influence of oxygen diffusion on the natural and artificial weathering of polyolefins, Polym. Degrad. Stab. 4 (1982) 17. doi:https://doi.org/10.1016/0141-3910(82)90003-9.

[21] E. Biagtan, E. Goldberg, R. Stephens, E. Valeroso, J. Harmon, Gamma dose and dose rate effects on scintillator light output, Nuclear Instruments

and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms 108 (12) (1996) 125 – 128. doi:http://dx.doi.org/10.1016/0168-583X(95)00874-8.

[22] Gillen, Wallace, Clough, Dose-rate dependence of the radiation-induced discoloration of polystyrene, Radiat. Phys. Chem. 41 (1993) 101. doi:https://doi.org/10.1016/0969-806X(93)90046-W.

[23] Gillen, Clough, Rigorous experimental confirmation of a theoretical model for diffusion-limited oxidation, Polymer 38 (1992) 1929.

[24] D. Pfeiffer, G. Gorine, H. Reithler, B. Biskup, A. Day, A. Fabich, J. Germa, R. Guida, M. Jaekel, F. Ravotti, The radiation field in the gamma irradiation facility gif++ at cern, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 866 (2017) 91 – 103. doi:https://doi.org/10.1016/j.nima.2017.05.045.