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Radiation damage studies on the optical and mechanical properties of plastic scintillators

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Abstract

This paper describes the radiation damage studies on a large volume plastic scintillator based in polystyrene doped with PPO and POPOP. The consequences on their mechanical and scintillation properties were evaluated before and after irradiation with different dose rates of ^{60}Co gamma radiation, in several doses. The optical results show a significant difference in the radiation susceptibility, when the plastic scintillator is irradiated at low rate (0.1 kGy/h) with that irradiated at high dose rate (85 kGy/h). The losses in the optical and mechanical properties increase as the irradiation dose is increased. The damage evaluated by the transmittance, emission intensity, pulse height and tensile strength was normalized as a damage fraction and fitted by a bi-exponential function. It was observed that the damage for irradiation is not permanent and it obeys a bi-exponential function.

1. Introduction

The interest in radiation resistance of scintillators for application in scintillator-based particle detectors has been renewed, due to the new generation of particle accelerators, SSC, LHC and RHIC. It is expected that some parts of new experimental setups will have to work in a severe radiation environment. Among the devices most strongly affected by high radiation environment are electromagnetic (EM) calorimeter since they absorb the entire energy of the incoming particles. It is now realistic to expect that some components of EM-calorimeters will have to endure annual dose at least 1 Mrad or more [1,2]. Several particle detectors on the base of plastic scintillators with optical fibres for readout were reported in Ref. [1,3]. Most of the detectors used nowadays in High Energy Astrophysics are the same ones used in Nuclear Physics. However, for high energy astrophysics these instruments must be flown at high altitude onboard balloons and satellites in order to avoid photoelectric atmospheric absorption of cosmic X and gamma rays, and therefore are subjected to operate in a hostile

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environment and the detectors are submitted to an intense radiation flux [4,5]. So, radiation hardness is one of the parameters that should be considered.

Plastic detectors are widely employed in Astrophysics. In general, they are used as active shielding for the main detectors, since the atmospheric background is high compared to the cosmic sources fluxes. These detectors are often used to define the field of view of the telescope. Even though size is a limiting factor, for some experiments there is a need to build large volume plastic detectors [5]. A large volume plastic scintillator detector based on polystyrene containing PPO and POPOP has been developed by our laboratory [6] to be used by the MASCO telescope developed by National Institute for Space Research (INPE) [5,7].

The type of radiation damage in scintillators, usually classified as decreased light yield or decreased in transmission, have been studied with several variables, such as total dose, dose rate, temperature, gas environment [8–11]. Besides this, the fabrication process, the choice of fluorescent organic compounds and the choice of the polymer for detector matrix can also affect the radiation sensitivity of the plastic scintillator [12,13]. Due to this number of relevant variables, comparisons of results among different groups and several plastic scintillators have been made [8–13]. There are surprising, and sometimes, dismaying results in the literature [11]. In this work, we have performed radiation damage experiments for a large area plastic scintillator produced, in our laboratory, by polymerization of styrene solution containing 0.4% PPO and 0.04% POPOP. The radiation resistance of this plastic scintillator, in several radiation dose exposures at two different dose rates, has been evaluated, by analysis of its optical and mechanical properties. The recovery period of this plastic scintillator was analyzed during a month.

2. Experimental procedure

Large plastic scintillators of 1.20 m of length with a cross section with sides of 19 cm × 29 cm and 19 cm thick (55 1) were prepared on the matrix of the styrene monomer distilled in vacuum. The styrene solution containing 0.4% PPO and 0.04% POPOP was polymerized induced by catalyst 1-1Bis(terc-peroxibutil)ciclohexane in N_2 atmosphere. The polymerization cycle was performed in one week at 80°C and 30 h at 120°C. To carry out the experiments, several samples of the plastic scintillators were machined in desired dimensions and polished.

The samples were irradiated by ^{60}Co gamma rays sources in two different dose rates using two kinds of ^{60}Co sources, a panoramic one with activity of 114×10^{12} Bq (3900 Ci) and dose rate fields of 0.1 kGy/h (10 krad/h) and the other source is a gammacell type with activity of 432×10^{12} Bq (11 700 Ci) and average dose rate of 85 kGy/h (8.5 Mrad/h). The total delivered dose ranged from 1 to 10 kGy in air at room temperature. For optical characterization, systematic measurements of transmittance spectrum, emission spectrum and light output have been made in all samples before and immediately after irradiation.

The transmission spectra were measured with a Shimadzu spectrophotometer model.1601. The same measurements were carried out for the polystyrene prepared with a similar technique to the one used for plastic scintillator. The emission spectrum and the light output were measured using circular cylinders of 2.5 cm diameter and 3.0 cm thickness with all sides highly polished and the ^{137}Cs gamma-source were used to excite the plastic scintillators. The evaluation of luminescence emission spectra was carried out with the monochromator (JASCL, FP550A). The relative light output was evaluated by comparison of the pulse height.

The mechanical properties have been evaluated by the modification of the hardness and tensile properties of plastic. Tensile properties determinations were carried out using plastic form of standard dumbbell-shaped test specimens, according to the ASTM Standard Test Method D638. The tests were made in dynamometer Instron model 5567. The hardness was measured by using D Shore method, using a weight of 5 kg, according to the ASTM Standard Test Method D2240.

All measurements X (transmittance, emission intensity, pulse height and tensile strength) was normalized to a damage fraction $DF = (X_0 - X_{\mathrm{irrad}})/X_{\mathrm{irrad}}$. A biexponential function $\mathrm{DF} = A\,\mathrm{e}^{-\alpha D} - B\,\mathrm{e}^{-\beta D}$ was used to fit the damage fraction

versus the dose irradiation, where α and β are parameters related to the formation velocity of the damages as the dose increases. A and B are linear parameters that represent the proportional contribution of each exponential component. The program AnaComp version 4.1 was used to determine the four parameters α, β, A and B [14].

The recovery of the plastic scintillator damage was evaluated by its transmittance measurements during 1 month. The samples were stored in air and room temperature.

3. Results and discussion

The results of the transmittance, emission intensity and pulse height of the samples irradiated at high dose rate are presented in Fig. 1. For all measurements the damage decreases as the radiation dose increases. As Fig. 1a shows the decrease in the transmittance is greater in the blue region, or more accurately in the 440 nm vicinities, where the plastic scintillator emits. Fig. 1b shows that the luminescence intensity decreases as the radiation dose is increased. However, no significant difference was observed in the position of the emission peak at 440 nm. As shown in the Fig. 1c, as the radiation dose is increased the Compton edge decreases in amplitude, indicating an overall loss in pulse height.

These losses in transmittance, emission intensity and light yield observed in the developed plastic scintillator, may be due to either damage to the fluorescent organic compounds (PPO) or/and the plastic base itself. A similar behavior was observed at a low dose rate, as the radiation dose is increased the damage is also increased. However, the damage fraction curves from the low and high dose rates show always difference in their profile. At a low dose rate, the detectors are systematically more susceptible to the damage.

This same tendency was found in the results from the tensile strength measurement: the damage decreases as the irradiation exposure increases as compiled in Table 1. On the other hand, no significant differences were observed in the hardness of the plastic scintillators related to the effects of the irradiation doses and dose rates.

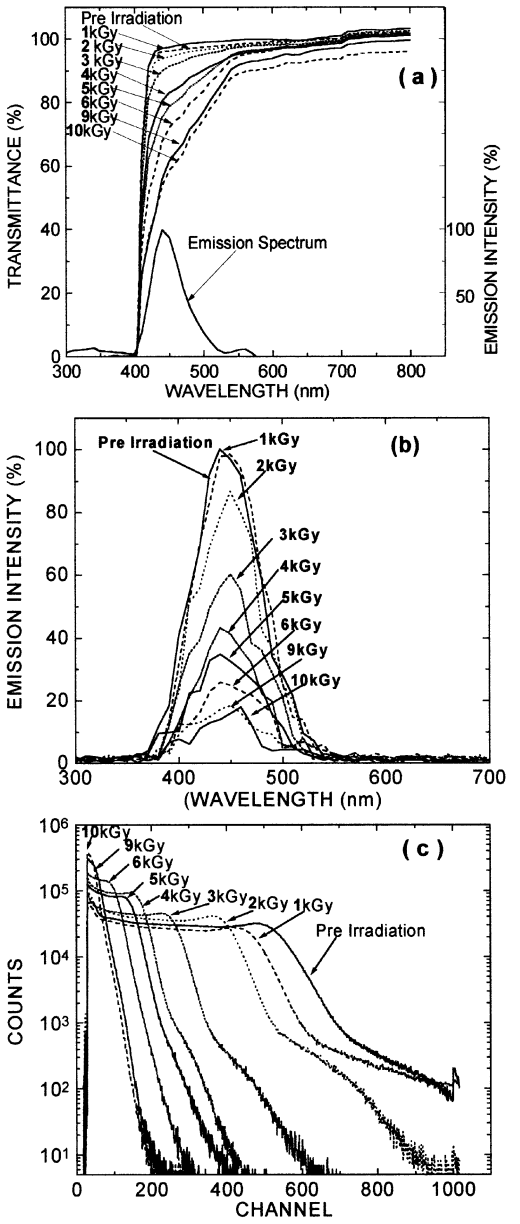

Fig. 1. Transmittance spectra (a), emission spectra (b) and pulse height spectra (c) of the plastic scintillator, before and immediately after different dose irradiation at 85 kGy/h.

Table 1 summarizes the results of the irradiation damage determined by the experimental data of transmittance (T) at 440 nm, emission intensity (I) at 440 nm, pulse height (Channel) and

Table 1
Evaluation of the losses in the quality of the plastic scintillator (damage) after its irradiation exposure by measurements of the transmittance and emission intensity at 440 nm the relative pulse height and tensile strength

Dose	Damage evaluation							
	Transmittance		Emission		Pulse height		Tensile	
	$\frac{T_0 - T_{irrad}}{T_0}$		$\frac{I_0 - I_{irrad}}{I_0}$		$\frac{Channel_0 - Channel_{irrad}}{Channel_0}$		$\frac{kg\ cm_0^{-2} - kg\ cm_{irrad}^{-2}}{kg\ cm_0^{-2}}$	
	Low	High	Low	High	Low	High	Low	High
0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
1.000	0.000	0.000	0.155	0.192	0.029	0.000	0.000	0.008
2.000	0.029	0.032	0.300	0.297	0.416	0.079	0.120	0.170
3.000	0.123	0.063	0.697	0.563	0.581	0.036	0.135	0.150
4.000	0.204	0.170	0.802	0.722	0.704	0.540	0.150	0.310
5.000	0.246	0.214	0.831	0.770	0.685	0.649	0.225	0.192
6.000	0.225	0.300	0.864	0.813	0.789	0.737	0.261	0.220
9.000	0.429	0.424	[a]	[a]	0.811	0.803	0.333	0.256
10.000	0.486	0.438	[a]	[a]	0.833	0.808	0.287	0.257

[a]The peak position is below of the threshold level.

tensile (kg f/cm^2). The experimental damage data were fitted using a bi-exponential function and plotted as a function of dose, as shown in Fig. 2. Table 2 summarizes the parameters estimated from the fitting data as a function of the irradiation dose for all experiments. A good correlation in the fitting of the experimental data was found.

In Fig. 2 and Table 2, it can be observed that the damage has always increased to the samples irradiated at a low dose rate. Dose is the energy deposited in the sample, so it was expected that the plastic scintillator damage were not affected to different dose rates, only due to the total dose delivered in the sample. In the plastic scintillator, damage is some chemical reaction that occurs in its structure and this reaction should be proportional to the dose (energy). However, the experimental results are not in agreement with this hypothesis as shown in Fig. 2. The greater susceptibility of the plastic scintillator at a low dose rate obtained in this work, may be due to the oxidation products formed during the irradiation, which decrease the transmittance of the plastic scintillator. When the samples were irradiated in the air, the oxygen present on the sample surfaces can react with some of the species formed under irradiation [8]. As at a low dose rate it is necessary more time than at a high dose rate to obtain the same dose, so a greater amount of oxidation products can be produced, increasing the damage in the detector. After the high dose irradiation the detector shows yellowish coloration, which tends to disappear with elapsing of the time. This yellow color disappears more quickly in the surface compared to the center of the detector.

Fig. 3a shows the transmittance spectra and Fig. 3b shows the damage fraction for polystyrene doped with PPO and POPOP and pure polystyrene. It is known that the peroxides are strong quench agents. As it was discussed above, peroxides can be produced during the irradiation, then acting as quenchers. This effect is present only in the scintillator plastic. The presence of the PPO and POPOP scintillators restricts the transparency of the plastic block to the wavelengths of its fluorescence spectra. As a consequence, the peroxide acts as a quencher, decreasing the light output, i,e., reducing the transmittance. Another hypothesis is that the scintillators are degraded, changing their chemical structure, so that they are able to capture the light photons but they no re-emit them.

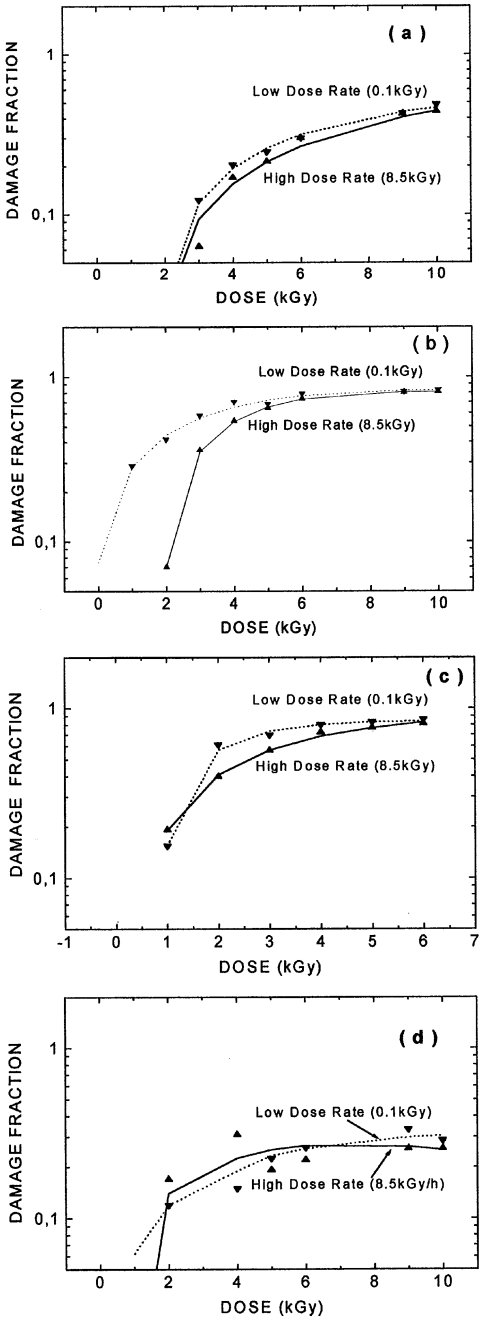

Fig. 2. Loss in transmittance (a) and emission intensity (b) at 440 nm, pulse height (c) and tensile strength (d) as a function of the dose. The continuous lines were fitted using a bi-exponential function $DF = Ae^{-\alpha D} - Be^{-\beta D}$.

Table 2
Parameters calculated by fitting data of the plastic scintillator damage fraction as a function of the irradiation dose

Para-meters	Transmittance		Emission intensity		Pulse height		Tensile strength	
	Low	High	Low	High	Low	High	Low	High
A	$0.922 \pm 1.01E-03$	$0.939 \pm 1.18E-03$	$1.348 \pm 7.65E-03$	$1.075 \pm 2.41E-03$	$0.851 \pm 8.97E-03$	$1.103 \pm 4.11E-03$	$0.462 \pm 5.13E-03$	$0.901 \pm 2.08E-03$
B	$1.114 \pm 1.28E-03$	$1.071 \pm 1.77E-03$	$1.273 \pm 1.11E-02$	$2.052 \pm 5.07E-03$	$1.701 \pm 2.34E-03$	$1.203 \pm 6.13E-03$	$0.471 \pm 8.56E-03$	$1.038 \pm 2.43E-03$
$\alpha(Gy^{-1})$	$0.018 \pm 4.70E-04$	$0.001 \pm 7.33E-04$	$0.033 \pm 1.63E-03$	$0.023 \pm 9.46E-03$	$0.001 \pm 4.94E-03$	$0.009 \pm 1.71E-03$	$0.019 \pm 2.33E-03$	$0.098 \pm 2.24E-03$
$\beta(Gy^{-1})$	$0.130 \pm 7.32E-04$	$0.078 \pm 5.51E-04$	$0.221 \pm 4.50E-03$	$0.382 \pm 1.21E-03$	$0.896 \pm 1.37E-02$	$0.288 \pm 3.65E-03$	$0.186 \pm 6.29E-03$	$0.249 \pm 2.32E-03$

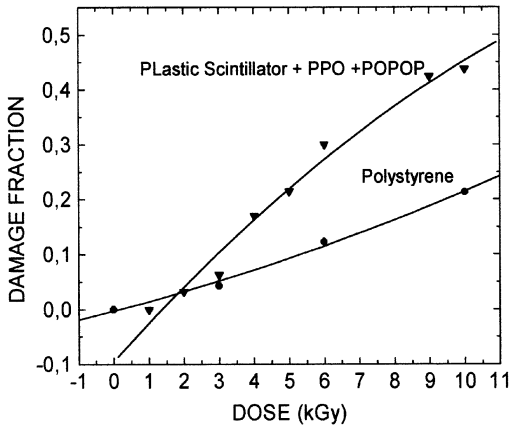

Fig. 3. Loss in transmittance of the polystyrene and plastic scintillaltors as a function of the dose. The continuous lines were fitted using a fitting polynomial.

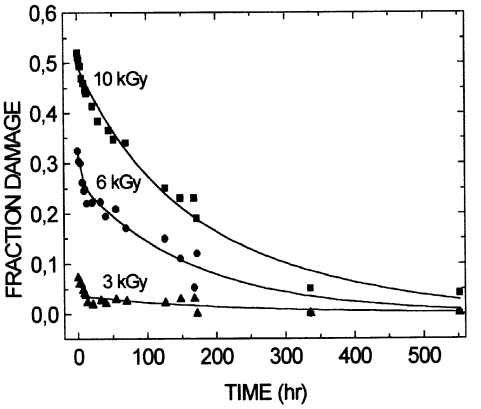

Fig. 4. Recovery of the plastic scintillator as a function of the time. These results refer to the transmittance results at 440 nm.

The damage due to the irradiation is not totally permanent. Fig. 4 shows the recovery of the plastic scintillator evaluated during one month. The recovery of the damage follows an exponential function. One month after the irradiation, approximately 5% of damage residue stays, which is a much lower percentage from that verified immediately after the irradiation.

4. Conclusions

It was found that the developed large plastic scintillator is less resistant when it is submitted to a low dose rate. In this work a bi-exponential model was proposed to explain these results. The presence of some scintillators, such as PPO and POPOP, which are strongly susceptible to the peroxides that act as quenchers in the polystyrene base, make the detectors more sensitive to the irradiation damage. The damage is not totally permanent. It is recovered, in course of time, and after one month the damage residue falls exponentially to approximately 5% of the damage generated immediately after being exposed to the irradiation dose. Finally, to carry out experiments where the plastic scintillator detectors have to work in an intense radiation environment, the total dose and their dose rates are parameters that should be considered.

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