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RADIATION HARDNESS STUDY ON MOLDED SCINTILLATION TILES AND WAVELENGTH SHIFTING FIBERS

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Abstract

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A comparative study of the radiation damage caused by γ-quanta and hadron irradiation in polystyrene scintillation tiles produced by the injection molding technique and in wavelength shifting fibers is presented.

Аннотация

Карюхин А.Н. и др. Исследование радиационной стойкости литьевых сцинтилляционных пластин и спектросмещающих волокон: Препринт ИФВЭ 95-97. – Протвино, 1995. – 15 с., 8 рис., 2 табл., библиогр.: 21.

Представлены результаты сравнительного исследования радиационной стойкости полистирольных литьевых сцинтилляционных пластин и спектросмещающих волокон,облученных в потоках γ-квантов и адронов.

1. Introduction

With the development of a new generation of particle accelerators the radiation hardness of materials used in modern calorimeters acquires a great importance. It is expected that some parts of future experimental setups will have to endure high doses of irradiation [1]. Many promising calorimeters [2,3,4], which are presently under research and development programs, contain a large amount of plastic materials (scintillation tiles, wavelength shifting (WLS) fibers etc.). Extensive application of these materials demands the availability of cheap production technologies.

It is known that scintillation tiles of any shape can be easily produced by the injection molding technique. Among the molded scintillator samples the tiles based on a granulated polystyrene (PS) of PSM-115 trade mark showed the highest level of radiation hardness as compared to PS or polyvinyltoluene samples produced by the bulk-polymerization [5,6,7,8,9,10,11]. Due to this fact the PS base has been chosen for further investigation of the influence of different scintillation dopants and production technology on their radiation properties.

This paper is an extension of our search for radiation-resistant scintillation tiles and WLS fibers planned to be used in the electromagnetic (EM) [12] and hadron calorimeters (ATLAS hadron calorimeter [13]) where the integral doses from 40 kRad to 1 MRad are expected.

2. Experimental procedure

All our scintillation tiles were produced by the injection molding technique at IHEP [5]. The granulated PS (PSM-115 trade mark, granules of about 3 mm in diameter and 4-5 mm long) was mixed with finely dispersed scintillation dopants. The mixture was loaded into an injection molder, the temperature was raised to about 200^0C, and the mixture melted. In order to improve the optical properties of large size scintillation tiles we elongated the duration of mixing procedure at high temperature.

Appropriate quantities of the melt were injected into the mold at a pressure of about 1000 atm where scintillation tiles were formed in several minutes. Large size tiles were cut mechanically to dimensions of 3×50×170 mm^3 to fit the available for irradiation volume and covered with aluminized Mylar films excluding the edges coupled to a FEU-84-3 photomultiplier (PM) having the green extended photocathode spectral sensitivity.

The samples were irradiated uniformely in a flux of γ-quanta from ^{137}Cs radioactive sources at a dose rate of 6 Rad/s (190 MRad/year) at room temperature air. Note that the dose rate in these irradiations was two orders of magnitude higher than that one expected in future experimental setups [1]. In such a case the irradiation time is close to the recovery time and this is the reason why the recovery processes after the irradiation should be taken into account in further examination of new materials for the calorimeters application.

The samples were coupled to the PM photocathode without optical contact and excited with a ^{90}Sr radioactive β-source. The PM photocurrent was measured. We measured the light output before I_0 and after irradiation I as well as the recovery processes. The accuracy of these measurements was about $\pm 2\%$.

Different types of particle detectors using scintillators or WLS fibers impose different levels of requirements on the maximum light output losses under irradiation. For example, for hadron calorimeters and vertex detectors the total loss of light output should not be more than 50% [14,15]. The most strict requirements on the light output losses are imposed for the CMS EM-calorimeter [12]. For long term exploitation (about 10 years), the total light output losses under irradiation should be less than 10%. The radiation hardness of any calorimeter is determined by its most radiation-sensitive element. This means that the maximum contribution to the light output losses of each radiation-sensitive element should also be less than 10%.

Let us define the radiation hardness of scintillation tiles or WLS fibers as a dose D at which they lose 10% of initial light outputs from their far ends. The main disadvantage of such a definition is that in this case it is very sensitive to the geometry of scintillator, wavelength region of operation, technology of plastic materials production etc. and it is very difficult to compare the data from different measurements.

3. Results

3.1. Scintillation tiles radiation hardness

At the first stage of a search for promising materials for calorimetry application it is necessary to estimate radiation properties of the main radiation-sensitive elements (scintillation tiles and WLS fibers) separately.

We measured the relative light output I/I_0 from far ends (x=13.5 cm, opposite to the PM) of available scintillation tiles samples as a function of the dose. Some of these data are presented in Figs. 1a,b. Experimental results had shown the presence of certain increase of I/I_0 ratio (measured immediately after the irradiation) near a dose of about 0.5 MRad for some samples: molded PS scintillators containing 1.5%PTP + 0.05%POPOP, 1.5% PTP + 0.05%DMPOPOP, 1.5%PTP + 0.05%BBOT and Kuraray-3HF scintillator (chemical formulae of used dopants and their peak absorption λ_{abs} and emission λ_{em} wavelengths are listed in Appendix). Similar characteristics were reported elsewhere [16,8,9]. This effect is not understood yet and needs more detailed investigations.

The radiation properties of PS based scintillator samples containing 1.5%PTP + 0.05%POPOP and 1.5%PTP + 0.05%DMPOPOP are very close to each other. Radiation characteristics of Kuraray-3HF and 1.5%PTP + 0.05%POPOP + 20%1MN bulk-polymerized PS scintillators are presented in Table 1 for comparison. Note that the maximum radiation hardness is provided by bulk-polymerized PS scintillators containing 1.5%PTP + 0.05%POPOP + 20%1MN and Kuraray-3H.

Experimental data allowed us to estimate the samples light yield $I_0(0)$ (as the linear approximation of I_0 to the zero length), radiation hardness D and scintillation tiles recovery time t_{max}. The light yield $I_0(0)$ of a bulk-polymerized PS scintillator containing 1.5%PTP + 0.05%POPOP was taken as 100%. Experimental results are summarized in Table 1.

Table 1. Scintillation and radiation properties of some PS based scintillators

N	Scintillators	$I_0(0)$, %	D, kRad	t_{max}, days
1	1.5% PTP + 0.05% POPOP	90	360	1-3
2	1.5% BO + 0.05% POPOP	83	30	8-30
3	1.5% PPO + 0.05% POPOP	95	130	4
4	1.5% PTP + 0.05% POPOP + 5% TPL	50	340	10
5	1.5% PTP + 0.05% DMPOPOP	91	360	2-3
6	1.5% PTP + 0.05% BBOT	84	220	8-30
7	1.5% PTP + 0.05% POPOP + 20% 1MN	80	1000	10
8	Kuraray-3HF[1]	45	1200	9

The addition of heavy metal containing high concentration additives (TPL) improves slightly the radiation hardness of scintillation tiles up to a dose of 340 kRad. Due to the presence of lead in plastic, γ-quanta can be detected effectively with this PS based scintillator. Due to this reason we are going to use such heavy metals loaded scintillators in promising EM calorimeters [3,4] to improve their energy resolutions.

[1]Kuraray Corp., Japan.

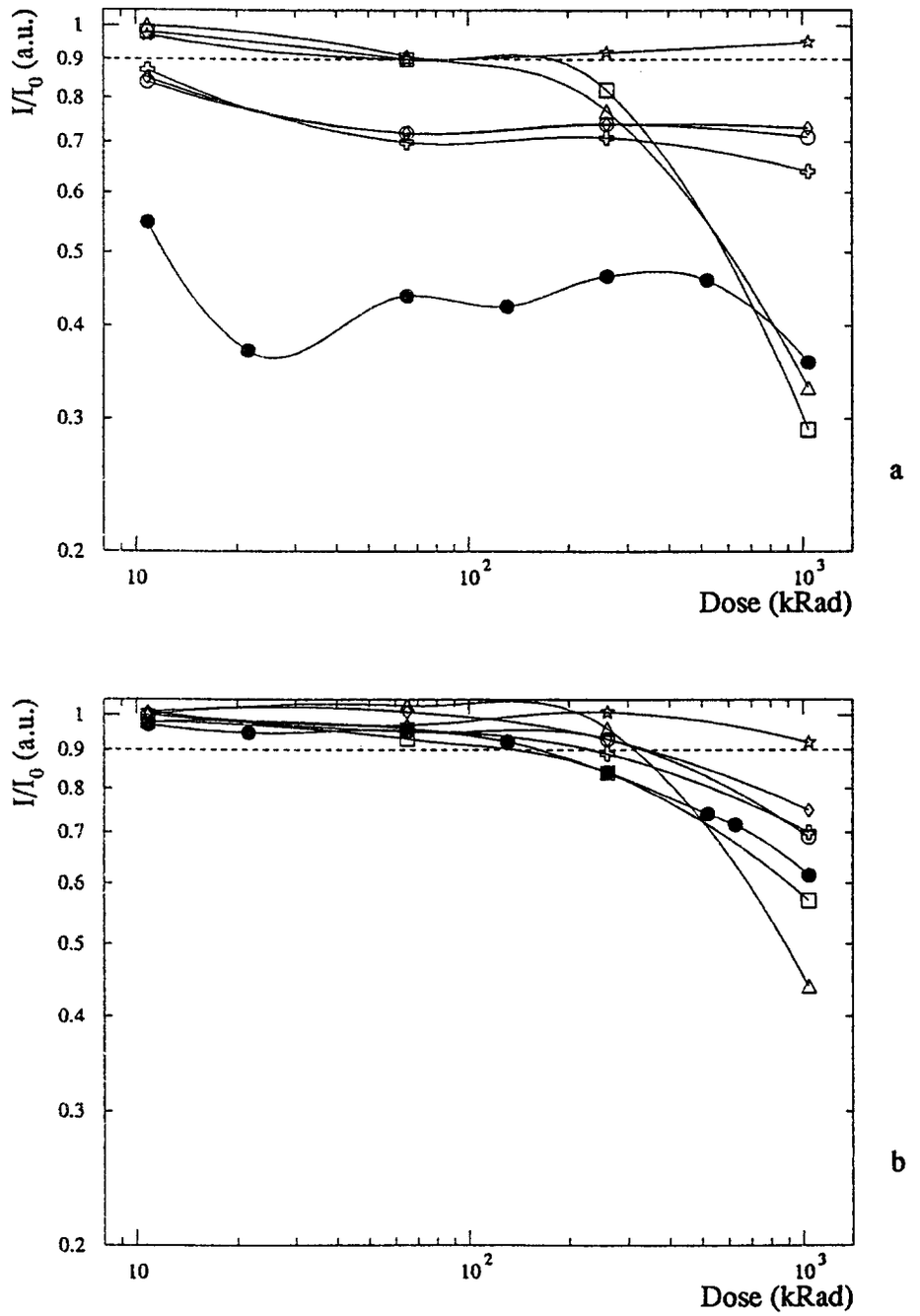

Fig. 1. Relative light output of scintillation tiles at x=13.5 cm versus the dose (a – immediately and b – in a month after irradiation): • – 1.5%PTP+ 0.05%POPOP (fiber readout); ☆ - 3HF; ✣ - 1.5%PTP+0.05%BBOT; △ – 1.5%PTP+0.05%POPOP+5%TPL; ◊ – 1.5%PTP+0.05%DMPOPOP; □ – 1.5%POPOP+0.05%POPOP; O – 1.5%PTP+0.05%POPOP.

At the second stage of the radiation hardness study, we used another experimental setup shown in Fig. 2 because the measuring scheme of this setup was more close to the layout of plastic elements in some calorimeters [3,4]. A sample of scintillation tile containing 1.5%PTP + 0.05%POPOP was excited by a ^{90}Sr radioactive β-source. The emitted light was absorbed by an unirradiated Y-11 WLS fiber. The light output from the far end of the fiber was measured. The accuracy of these measurements was about ±2.0%.

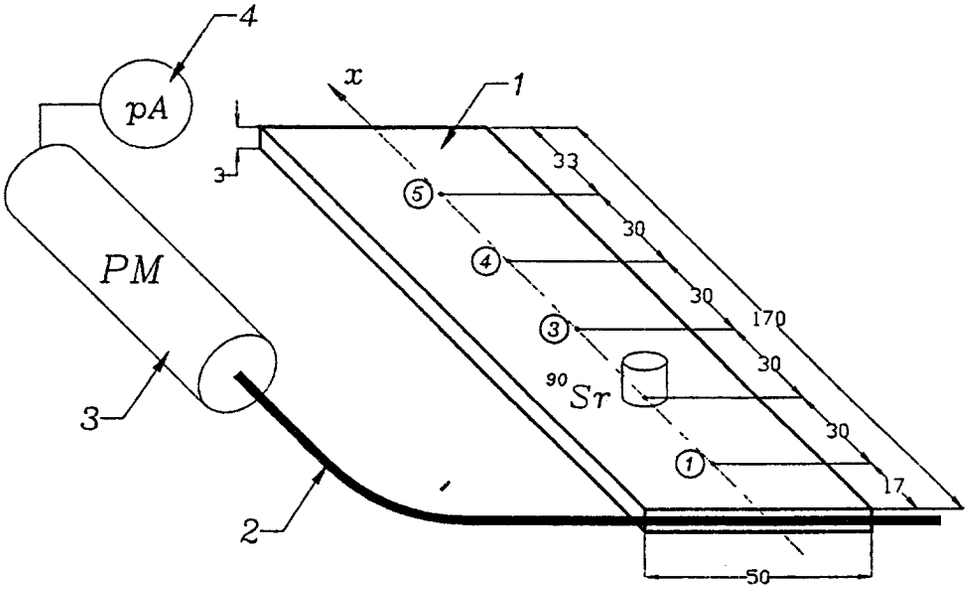

Fig. 2. Experimental setup layout: 1 – sample; 2 – Y-11 WLS fiber; 3 – photomultiplier FEU-84-3 and 4 – microampermeter.

These experimental results are also presented in Figs. 1a,b. The higher level of radiation damages in this case is explained by the fact that the WLS fiber absorbs only a short wavelength fraction of the light emission spectrum of scintillators, which is more sensitive to irradiation [5,17]. An estimated radiation hardness is about D=170 kRad which is 2.1 times lower in comparision with the measurements without the WLS fiber for the same material (sample #1 in Table 1).

It is also interesting to compare these data with the results when scintillation tiles were produced with a shorter duration of the mixing process at high temperature [18]. Test results for such materials reported in [7,8,9] showed that the radiation hardness of those scintillation tiles was about D=780 kRad. A comparison shows that the elongation of the mixing process lowered the scintillation tiles radiation hardness 4.6 times.

A detailed picture of the recovery processes for a PS sample containing 1.5%PTP + 0.05%POPOP is shown in Figs. 3a-c. One can see fast (7-30 hours) recovery processes immediately after the irradiation. The time interval between the end of exposure and the moment when I/I_0 reaches its maximum is defined as the scintillator recovery time t_{max}. Note that the recovery time is a dose dependent characteristics. The recovery times presented in Table 1 were determined for doses ranging from 65 to 260 kRad. For $t > t_{max}$ we observe the beginning of degradation of I/I_0. This fact needs more detail examination.

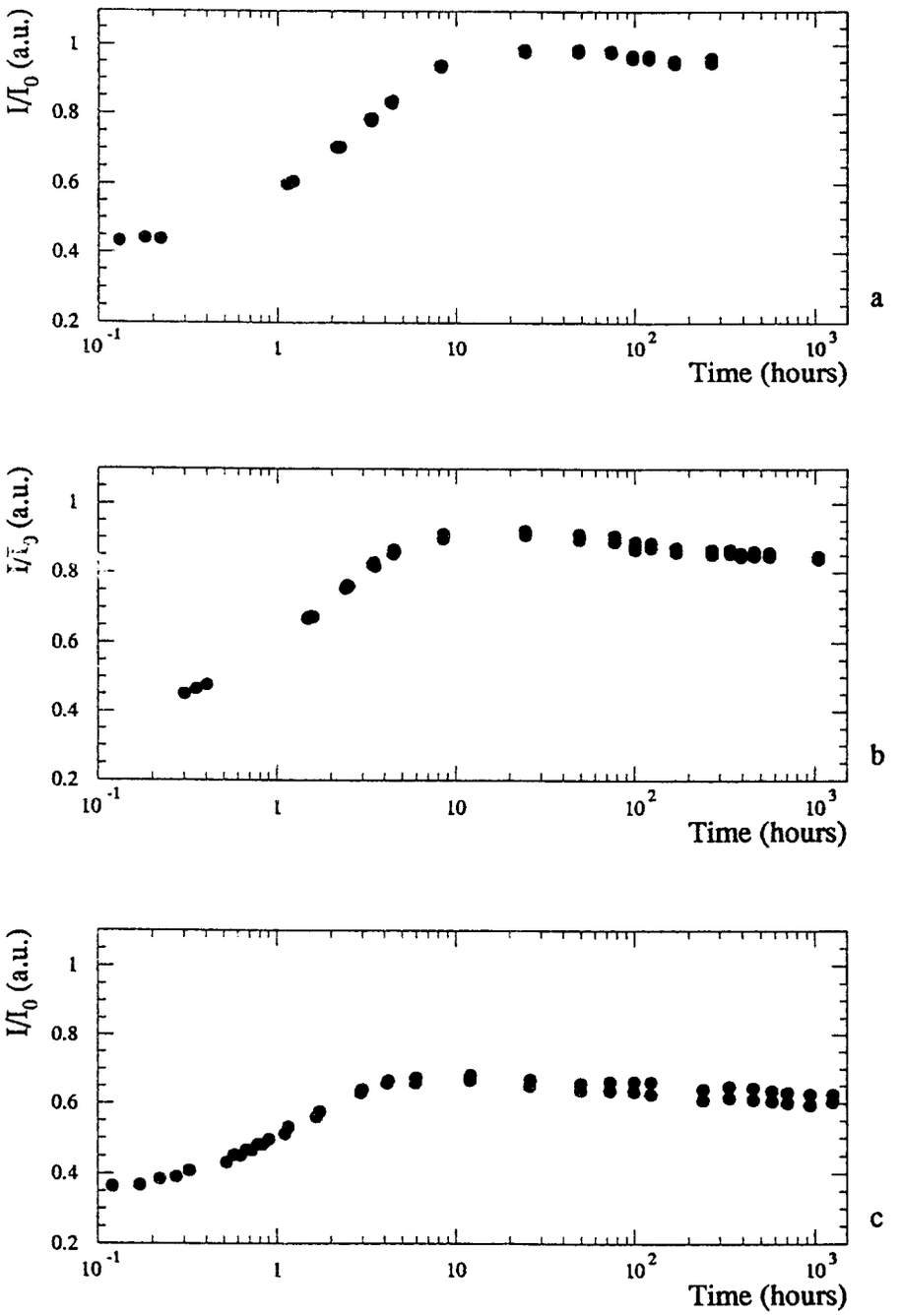

Fig. 3. Relative light output of a scintillation tile containing 1.5%PTP+0.05%POPOP at x=13.5 cm versus recovery time for different doses (a – 65 kRad; b – 260 kRad and c – 1037 kRad).

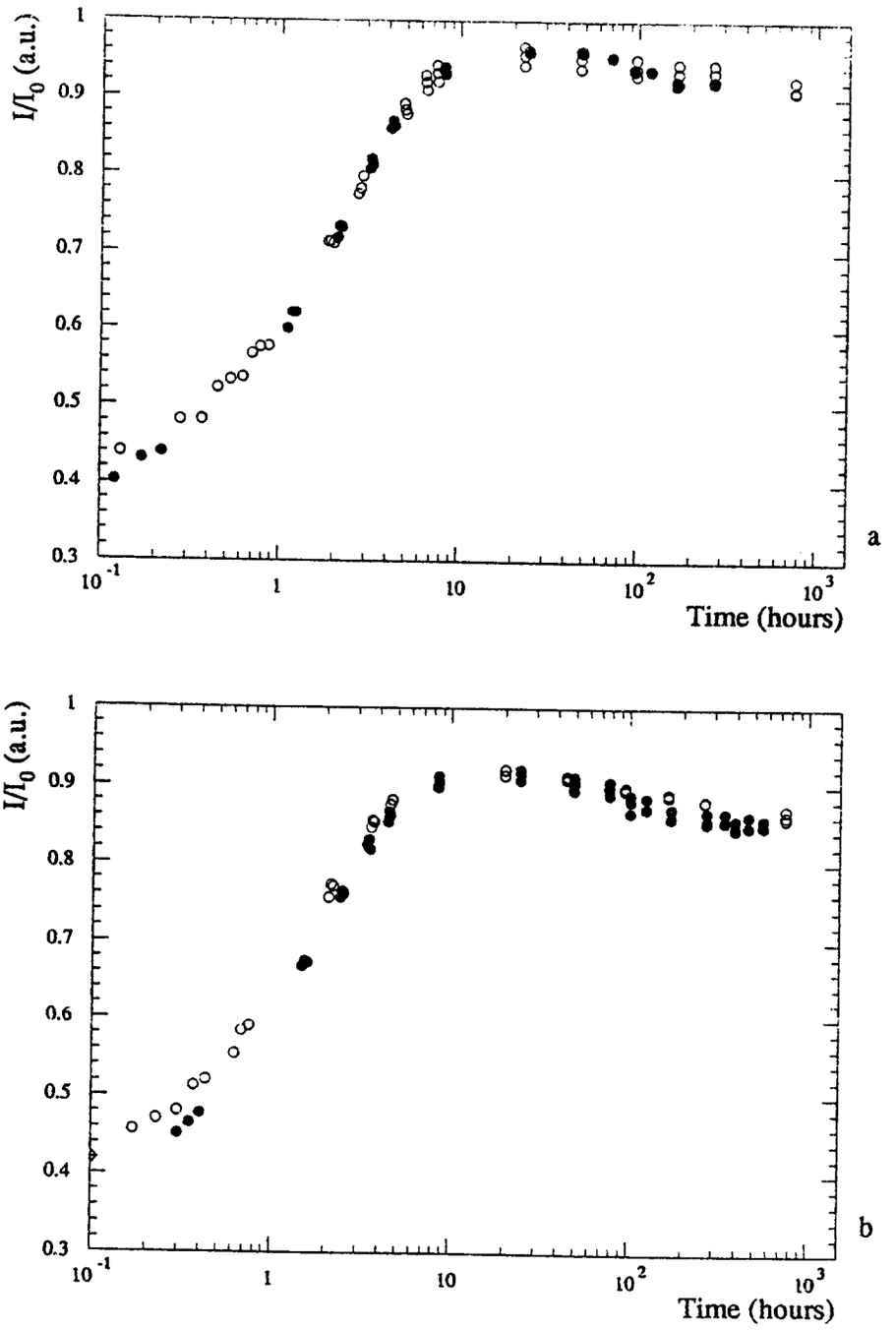

Fig. 4. Relative light output of a scintillation tile containing 1.5%PTP+0.05%POPOP at x=13.5 cm versus recovery time for different doses (a – 130 kRad, b – 260 kRad), accumulated in one step (•) and two steps (o).

Under real conditions of calorimeters operation the fibers and scintillation tiles will be exposed multiply with some breaks between irradiations. We tried to simulate this effect providing the given dose in one and two steps with fortnight's break for double

irradiation. The results presented in Fig. 4 show a negligable increase of the I/I_0 ratio for the two-step irradiation, but it is necessary to perform additional measurements for more exact conclusions.

Among the samples investigated the maximum radiation hardness for PS based molded scintillators has been reached for the tiles containing standard widely used dyes 1.5%PTP + 0.05%POPOP and 1.5%PTP + 0.05%DMPOPOP.

3.2. WLS fibers radiation hardness

We have chosen several promising WLS fibers for the radiation hardness tests. The experimental setup for WLS fibers measurements was similar to that presented in Fig. 2. The WLS fiber samples were about 1 mm in diameter and 180 mm in length. The tested fibers were excited with the use of an unirradiated PS based scintillator bar (containing 1.5%PTP + 0.05%POPOP), which in turn was excited with a ^{90}Sr radioactive β-source. Relative light attenuation curves for available WLS fibers were measured before and after γ-irradiation as well as the recovery processes. The measurements accuracy was about $\pm 5\%$. Some of experimental results are shown in Fig. 5.

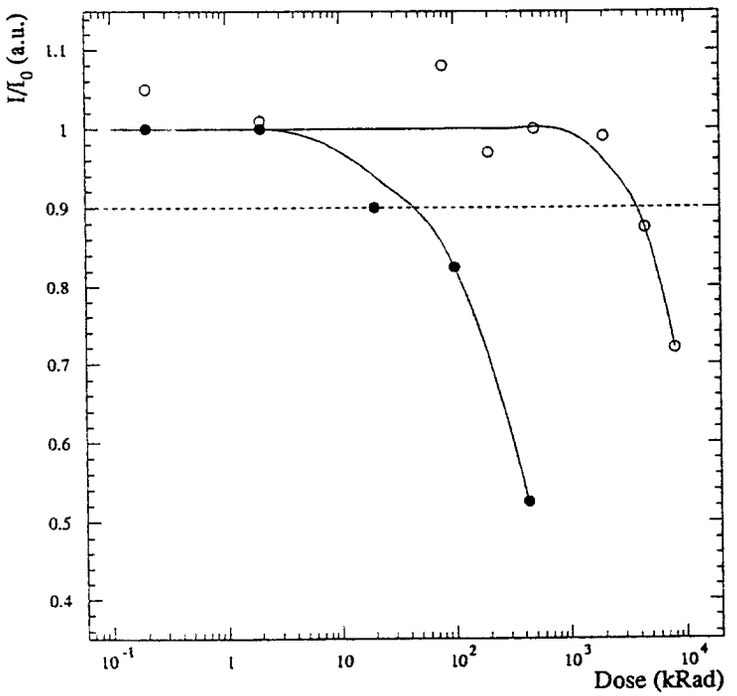

Fig. 5. Relative light output of some WLS fibers at x=17 cm versus the dose immediately after the irradiation (o – Y-8 and • – Pol.Hi.Tech 5948-4-100).

These experimental data allowed us to estimate the WLS fibers conversion efficiency $J_0(0)$ for the chosen scintillator (as the linear approximation of light output from a fiber to the zero length) and radiation hardness D. The conversion efficiency $J_0(0)$ of an Y-7 WLS fiber was taken as reference.

It was found that the ability of the tested WLS fibers to recover their luminescent characteristics was small (up to 20%) and their recovery times were about t_{max}=3-7 days. Experimental results are summarized in Table 2.

Table 2. Luminescent and radiation properties of some WLS fiber scintillator pairs (containing 1.5%PTP + 0.05%POPOP)

N	WLS fibers	$J_0(0)$, %	D, kRad
1	Y-7[2] s.c.	100	2000
2	Y-8, s.c.	100	3600
3	Y-11, d.c.	121	450
4	BCF 91a[3] d.c.	103	2000
5	BCF 92a, s.c	149	160
6	BCF 99-28, d.c.	140	120
7	BCF 99-36, d.c.	150	100
8	Pol.Hi.Tech 5948-4-100[4] s.c	100	45

The maximum radiation hardness is provided by Y7, Y8 and BCF 91a WLS fibers. WLS fibers having double claddings (d.c.) provide more light trapping efficiency in comparison with WLS fibers having single claddings (s.c.). Note that for fibers with poor mechanical properties, i.e. fragile or having mechanical defects, the radiation hardness is up to 10 times lower than it is presented in Table 2.

3.3. Radiation hardness of plastic materials in hadron fluxes

The presence of intensive high energy secondary particles irradiation will be a characteristic feature of many experimental setups for future particle accelerators [19]. The radiation hardness investigation of plastic materials in high energy hadron fluxes is also of great importance now. In our case we dealt with secondary particles (mostly hadrons) originating from an *Al*-target of the 70 GeV IHEP accelerator. PS based scintillator samples were exposed in a flux of secondary particles at 30^0 to the beam axis at an average dose rate of about 2-3 rad/s [20]. Hadron irradiation doses were measured with polymer-alanine dosimetry [21].

A comparative study of the influence of γ-quanta and hadron irradiation on scintillation tile samples containing 1.5%PTP + 0.05%POPOP is shown in Figs. 6a,b. The recovery processes for the same PS samples are shown in Figs. 7a-c. The experimental data show that the hadron irradiation causes more serious radiation damages. Moreover the relative (hadron to γ-quanta irradiation) damages increase with the absorbed dose. Note that the observed difference of radiation properties between these two types of irradiations (Figs. 6,7) may allow one to reveal the effect of atoms dislocation in materials which distinguishes hadron irradiation from γ-irradiation.

[2]Kuraray Corp., Japan.
[3]Bicron Corp.,Newbury, Ohio 44065, USA.
[4]Pol.Hi.Tech.s.r.l. S.P.Turanense km 44.400,67061 Carsoli (AQ) Italy.

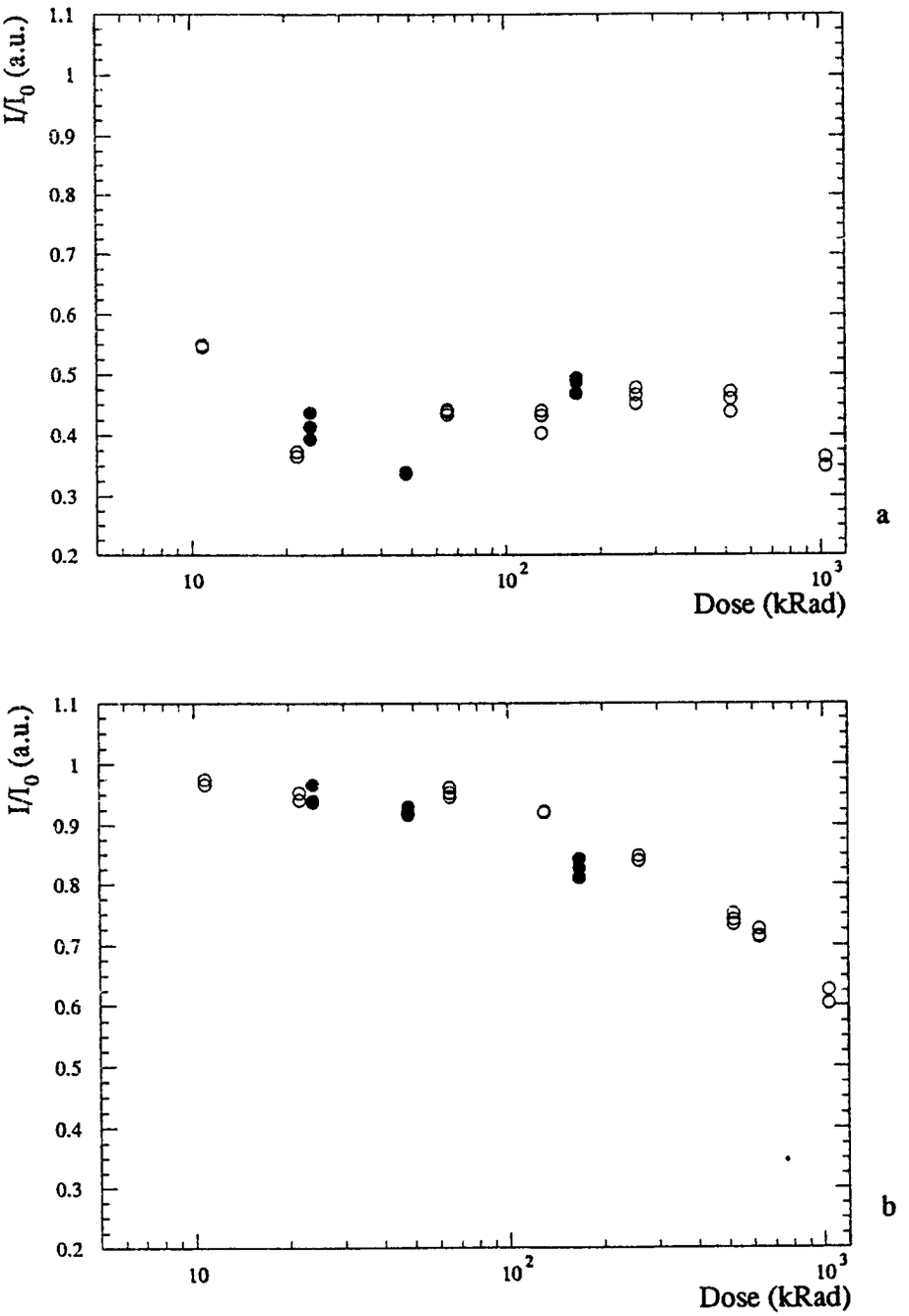

Fig. 6. Influence of γ-quanta (o) and hadron (•) irradiation on the relative light output at x=13.7 cm of a scintillation tile containing 1.5%PTP + 0.05%POPOP versus the dose (a – immediately and b – in a month after irradiation).

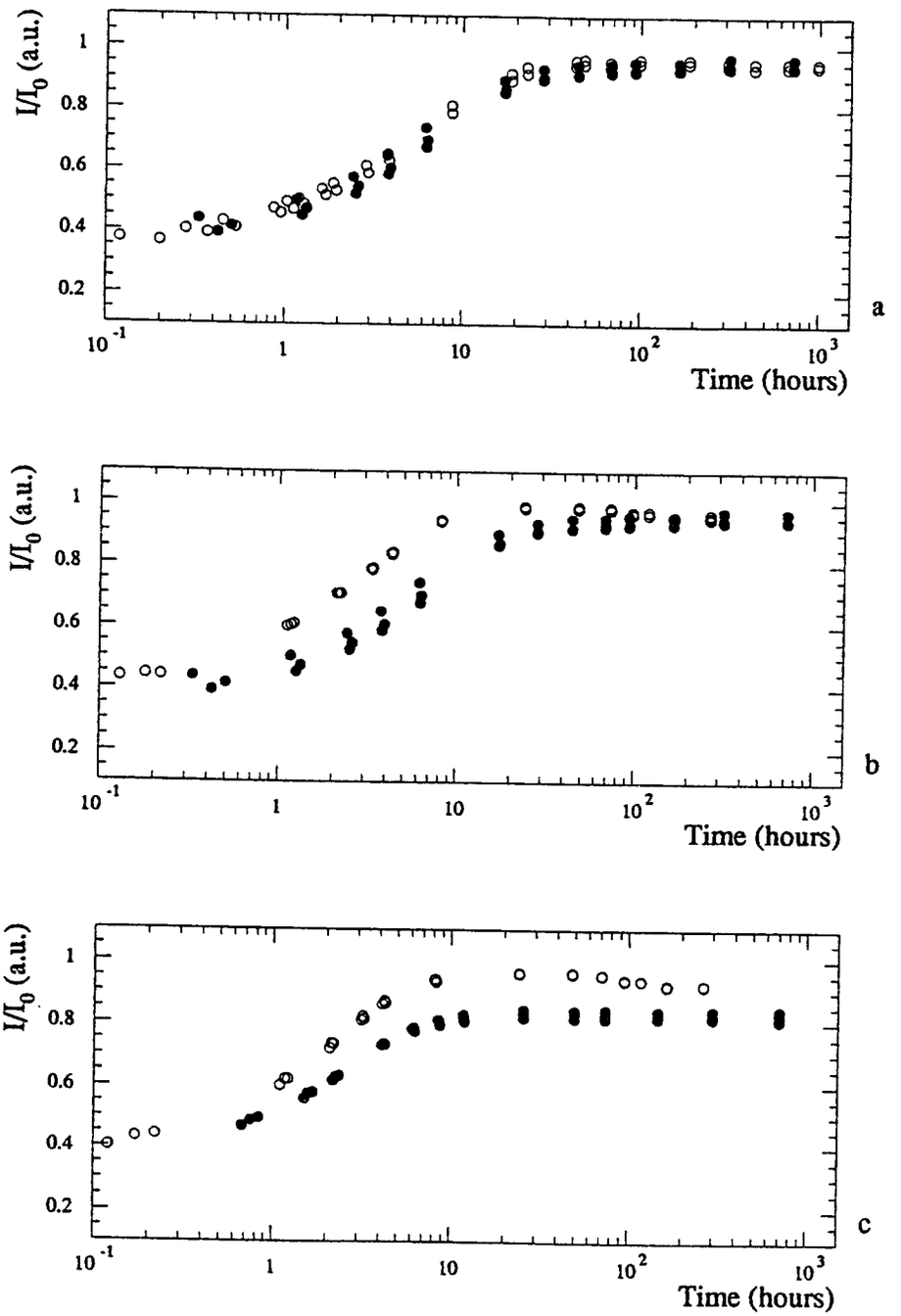

Fig. 7. Influence of γ-quanta (o) and hadron (•) irradiation on the relative light output of a scintillation tile containing 1.5%PTP + 0.05%POPOP at x=13.7 cm versus the recovery time (a – after 24 kRad of γ-quanta and 22 kRad of hadron irradiation, b – after 48 kRad of γ-quanta and 65 kRad of hadron irradiation, c – after 130 kRad of γ-quanta and 168 kRad of hadron irradiation).

During the hadron irradiation various nuclear reactions like $^{12}C(\pi^-,\pi^-n)^{11}C$, $^{12}C(n,2n)^{11}C)$, $^{12}C(p,pn)^{11}C$ etc. go in the plastic samples mainly with the production of β^+-radioactive isotope ^{11}C with the half-life time $T_{1/2}$=20.4 min. Positrons from ^{11}C almost immediately annihilate with electrons of materials producing of two 511 keV γ-quanta. Fig. 8 shows the "decay curve" of the light output I from PS based scintillation tiles samples caused by their induced radioactivity. As illustrated in Fig. 8 PS based scintillators produce only short-lived radioactive isotopes with half-life time of 20.5 minutes which is very close to the tabulated value for ^{11}C isotope.

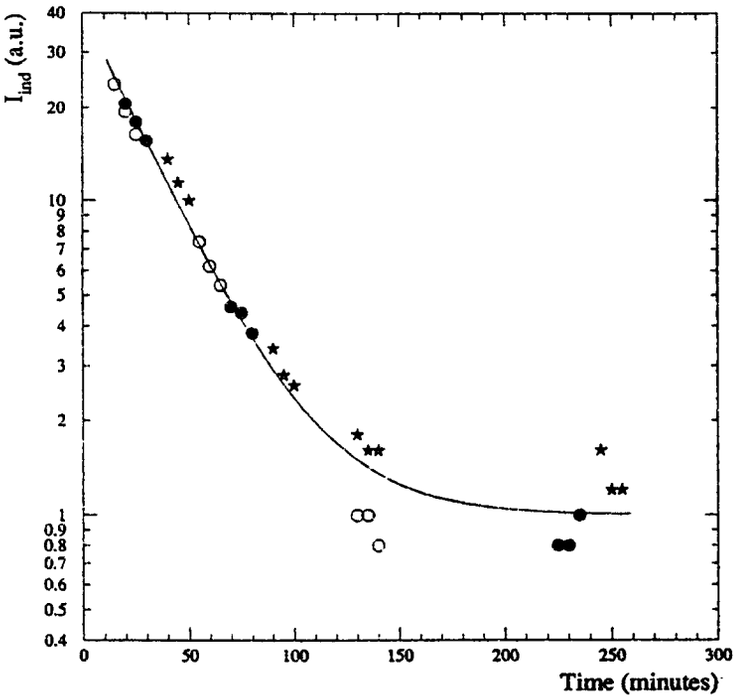

Fig. 8. "Decay curve" of the light output from scintillation tiles containing 1.5%PTP + 0.05%POPOP caused by induced radioactivity after receiving of different doses of hadron irradiation (• – 24 kRad; o – 48 kRad and ⋆ – 168 kRad).

4. Conclusions

A comparative study of radiation damage caused by γ-quanta and hadron irradiation in polystyrene scintillation tiles produced by the injection molding technique and wavelength shifting fibers is presented. The experimental results of this study may be summarized as follows:

- The maximum radiation hardness for molded PS based scintillators is provided when they contain the following dopants 1.5%PTP + 0.05%POPOP and 1.5%PTP + 0.05%DMPOPOP.

- The level of radiation hardness D is strongly dependent on measurements scheme and technology of PS production and ranges from 170 kRad to 780 kRad.
- The two-step irradiation causes less radiation damages than one-step irradiation for the same absorbed dose.
- In comparision with γ-irradiation the hadron irradiation reveals stronger radiation damage increasing with the absorbed dose in scintillation tiles.
- The hadron irradiation produces only short-lived radioactive isotopes (mainly ^{11}C) in PS based scintillation tiles.
- WLS fibers Y-7,Y-8 and BCF 91a have the highest light conversion capability and radiation hardness of about D=3600-2000 kRad among the tested fibers.

The above experimental data on the radiation hardness of plastic materials can be used as preliminary for estimating calorimeter radiation hardness. The exact values can be received after calorimeter modules radiation damage test under low dose rate irradiation only.

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The list of dopants used in this study:

1MN – 1-methylnathalene, λ_{abs}=275 nm, λ_{em}=330 nm
PTP – para-terphenyl, λ_{abs}=275 nm, λ_{em}=340 nm
PPO – 2,5-diphenyloxazole, λ_{abs}=310 nm, λ_{em}=365 nm
BO – 2-(4'-dimethylaminophenyl)-benzoxazole, λ_{abs}=330 nm, λ_{em}=400 nm
POPOP – 1,4-bis-[2-(5-phenyloxazolyl)]-benzene, λ_{abs}=365 nm, λ_{em}=420 nm
DMPOPOP – 1,4-di-2-(4-methyl-5-phenyloxazolyl))-benzene,
λ_{abs}=370 nm, λ_{em}=430 nm
BBOT – 2,5-di-(tret-butyl-2-benzoxazolyl)-thiophene, λ_{abs}=380 nm, λ_{em}=436 nm
3HF – 3-hydroxyflavone, λ_{abs}=350 nm, λ_{em}=530 nm
TPL – tetraphenyl lead.

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