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**RADIATION-DAMAGE MEASUREMENTS ON
PVT-BASED PLASTIC SCINTILLATORS**

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Abstract

Samples of PVT-based plastic scintillators produced by Nuclear Enterprise Technology Ltd. (NET) were irradiated up to 9 kGy, both with a gamma source and within a typical accelerator radiation field (CERN PS ACOL Irradiation Facility).

The consequent reduction of scintillating efficiency and light transmission were measured, as well as subsequent recovery, over a period of several months.

The main results show that irradiation affects more the light transmission than the light emission. The radiation type does not affect either the amount of transmission reduction or the recovery.

Observations were also made by means of polarized light. Non-uniformities and internal stresses were observed in scintillator bulks which were polymerized too quickly. These defects influence the light transmission.

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1. INTRODUCTION

The evolution of particle physics towards high-rate machines such as the Large Hadron Collider (LHC) and the Superconducting Super Collider (SSC), where the calculated radiation dose levels will be up to 100 kGy per year [1, 2], makes the radiation hardness a *sine qua non* condition in choosing detector materials, including scintillators.

Much scientific work is dedicated to the study, understanding, and improvement of the radiation resistance of plastic scintillators, which are—for the time being—those used in the largest quantities.

This paper presents the results obtained within CERN on plastic scintillators produced by Nuclear Enterprise Technology Ltd. (NET), with the main emphasis concentrated on radiation fields which are representative for future collider detectors.

2. GENERAL CONDITIONS

Plastic scintillators consist of a polymeric base (matrix), mainly used in the interaction with ionizing radiation, which contains, as solutes, one or two fluors (dyes) — at molar concentrations as low as 10^{-2} to 10^{-4} . The fluors can absorb the UV light emitted by the matrix, convert it, and re-emit it at longer wavelengths.

The results presented here refer only to scintillators based on polyvinyltoluene (PVT), which is, together with polystyrene (PS), one of the most radiation-resistant matrices, with good optical and mechanical properties. PVT has a pronounced tendency to crosslink when irradiated. The π electrons in its aromatic structure are primarily excited, making PVT an intrinsic scintillator and providing, up to a certain level, a protective effect. The radiation-induced undesirable effects are strongly dependent on the nature and dimensions of the scintillator and on the nature of the radiation, dose, dose rate, temperature, and atmosphere during and after irradiation, as well as on the nature and concentration of additives and the parameters of the fabrication process [3–5].

3. EXPERIMENT

3.1. Materials

The samples were supplied in the form of 24 mm diameter rods, of various lengths. They were cut to appropriate lengths [short samples (10 mm)

for scintillation-efficiency measurements, and long samples (10 cm) for transmission measurements].

All of the samples were hand-polished at both ends; their preparation and some preliminary test results have already been presented [6, 7]. The scintillating wavelengths were communicated by NET and are presented in Table 1. It should be noted that pure PVT is an intrinsic scintillator, but its emission wavelength is too short to be of direct use. The scintillation wavelength of Pilot-U is also short, this scintillator however is used for fast response at high speed.

3.2. Irradiation facilities and dosimetry

In order to approach realistic conditions and to reduce the number of parameters, tests were carried out in an atmosphere of air either in the radiation field of a pure gamma ^{60}Co source or in the radiation environment of an accelerator facility.

Gamma ^{60}Co irradiations were carried out by means of an industrial source (50 kCi) at the French company Conservatome; the 9 kGy were obtained at a dose rate of 80 Gy/h [8].

Irradiations in a mixed accelerator radiation field were mainly carried out at the PS ACOL Irradiation Facility (PSAIF) at CERN, where neutrons are produced from a target hit by a 26 GeV proton beam [9]. The NE 118 sample was irradiated behind the target T6, in the north area TCC2 of the CERN SPS [8].

Polymer-alanine dosimeters (PAD) and radiophotoluminescent (RPL) dosimeters were used to monitor the absorbed dose in both irradiation facilities [10]. The dose rate in the PSAIF was of the order of 80 Gy/h, but the irradiation was not continuous. The dose rate in TCC2 was of the order of 50 Gy/h; the irradiation lasted three weeks, with frequent interruptions.

The neutron fluences in the PSAIF and TCC2 were established by means of activation foils [9]. The neutron spectrum is an evaporation spectrum with a peak at about 1 MeV plus an important quantity of low-energy neutrons. For the purpose of this study, these two accelerator irradiations are considered as equivalent, except with regard to the dose rate.

Irradiations were carried out at several dose levels from a few hundreds of Gy up to 9 kGy in the accelerator facilities, and up to 29 kGy in the cobalt source (at a dose rate of 800 Gy/h). The results presented here refer mainly to the 9 kGy irradiations; accelerator and gamma irradiations will be compared at the same levels. The results with the long samples irradiated up to 29 kGy with gammas will also be presented in the graphs and will be discussed briefly.

3.3 Test methods

The scintillation yield was measured by means of a Philips XP 2212 photomultiplier working in the current mode, using a ^{90}Sr -Y excitation source. Corrections were made taking into account its spectral sensitivity. No optical coupling was used.

The radiation-induced modifications on transmission properties were measured, between 300 and 600 nm, by means of a PYE UNICAM SP 8800 spectrophotometer, taking air as a reference. Non-uniformities of rod scintillators which are linked to the fabrication process were observed by means of polarized light.

Between the measurements, the samples were kept in soft paper sheets and in plastic bags, stored at room temperature, only partly air- and light-tight.

4. RESULTS

4.1. Scintillation yield

The scintillating yield was recorded prior to and after various irradiation doses both in the PSAIF and in the ^{60}Co gamma source. The results showing only the maximum absorbed doses (9 kGy in ^{60}Co and 4.05 kGy in the PSAIF) are presented in Table 1 measured up to 25 days following the end of the irradiation. This data is given as a percentage of the light intensity measured prior to irradiation. The error is estimated to be a few per cent. Note that no correction was made with regard to the transmission-loss contribution, which may be considered as negligible for most of the samples but which may influence the values of pure PVT and Pilot-U with short emission wavelength.

4.2. Light transmission

Discrepancies in light transmission samples were observed in the non-irradiated samples; the percentage of transmitted light depends on the position and on the orientation of the samples in the holder. Non-uniformities and internal stresses were observed by means of polarized light (Fig. 1). The largest non-uniformities appear in the samples which were polymerized too rapidly (i.e. 24 hours instead of 7 days).

To reduce the error in light-transmission measurements, the best transmission was selected and the corresponding position of the sample was marked. The position was then the same for all measurements and this reduced the error of the values given in Tables 2 and 3 to a few per cent.

The transmission property of the pure matrix PVT is given for comparison together with that of all scintillators in Tables 2 and 3; absolute values are somewhat different because they were measured on two samples. Figure 2 shows the transmission curves as a function of the wavelength, prior to irradiation, after the cobalt irradiation up to 9 kGy, and after 310 days recovery. The transmission through the 10 cm long samples (mean value) is compared with that in air.

Table 2 presents the transmission percentages after the 9 kGy cobalt irradiation and the subsequent recovery over a period of 460 days. Table 3 presents the transmission percentages after the 9 kGy accelerator irradiation and the subsequent recovery over a period of 480 days.

The recovery of the transmission property of the materials NE 108, NE 110, NE 118, NE 122, and Pilot-U appears in Figs. 3-7 after three irradiations. The transmission values taken into account were those corresponding to the emission wavelengths for each type of scintillator.

5. DISCUSSION

The reason for the observed discrepancies in the light transmission and of the non-uniformities observed by means of polarized light is the fast polymerization process, which leads to a large dispersion of molecular weights, leaves unpolymerized monomers, and induces internal stresses, causing random variation of refractive index. A subsequent thermal treatment — 80°C for 60 hours — drastically reduced these internal stresses.

It should be noted that the fast polymerization process was only applied to some samples of this study, and that commercialized products were polymerized in a standard and much slower way. In order to minimize such undesired effects it is recommended to conduct the polymerization process as slowly as possible. For the plastic scintillating fibres an absolute constancy of the drawing process from the mechanical and thermal point of view is also highly recommended.

Measurements of both light emission and light transmission show, as expected [4, 5], that the level of radiation-induced damage is mainly related to dose and does not seem to depend on the type of radiation. At similar dose levels, pure gamma rays and mixed accelerator fields are therefore expected to produce similar levels of defects.

Comparison of results of pure PVT with those of other scintillator materials (Tables 2 and 3) clearly demonstrates the main contribution of the ageing of the matrix to the reduction of the transmission.

The transmission measurements carried out just after the 9 kGy irradiations have also confirmed the ones obtained by the measurements of scintillation yield (Table 1), and show that NE 108 is less affected than NE 110 and Pilot-U (Figs. 8, 9). Ne 118 irradiated in SPS-TCC2 appears to be less

affected, but it should be noted that this irradiation lasted for 3 weeks and that recovery occurred during that time (dashed line in Fig. 8).

The 29 kGy irradiation with ^{60}Co strongly affects the transmission of NE 110 and NE 122, whilst NE 108 keeps very good optical properties. The two former recover fairly well (Fig. 10).

If recovery occurs, a period of a few weeks is necessary for a plastic scintillator irradiated at 10 kGy to recover its transmission properties. With respect to the influence of the surrounding atmosphere during and after irradiation — which is linked to the recovery phenomenon — an atmosphere of air was chosen bearing in mind the previsible realistic conditions within future detectors. Owing to the fact that in plastic scintillators the fluorescent emission occurs at the molecular level, it is recommended to study the radiation-induced damage of matrices and fluors in separate ways by means of complementary methods, such as infrared (IR), electron-spin resonance (ESR), and radio-thermoluminescence spectroscopies (RTL). It must not be forgotten that a 'recovered' plastic scintillator might be chemically different compared to the unirradiated one. The transient radiation-induced species trapped in the polymeric matrix slowly react with the dissolved oxygen and/or the oxygen arriving by diffusion. Thus, a recovered plastic scintillator will contain macromolecular peroxides or hydroperoxides which are generally less strong optical absorbers. This might be the reason for the continuous degradation of some irradiated scintillators (NE 108).

6. CONCLUSION

Discrepancies in light transmission according to the sample orientation, and observation by means of polarized light, have shown the importance of a slow polymerization during the making of the scintillators.

As already observed in other studies, radiation affects more the light transmission than the scintillating efficiency.

For the doses and dose rates used in this study, no significant difference was observed between a pure gamma irradiation and an irradiation with a complex particle accelerator field, either concerning the transmission or the scintillation.

In the radiation-damage studies for future detector and collider components, the follow-up of the recovery is as important as the degradation measurements. If the scintillators studied had to be classified according to their radiation resistance, just after the 9 kGy irradiation, the order would be NE 108, NE 122, NE 118, NE 110, and Pilot-U. The follow-up of the recovery changes this order; whilst NE 122 recovers completely after six months, degradation of NE 108 seems to continue (Fig. 9).

The absolute value of the initial transmission must also be considered for detector applications. This reinforces the position of NE 122, whilst NE 108, NE 110 and NE 118 are at the same level, and Pilot-U appears to be

very low (Figs. 8,9). The high-dose irradiation (29 kGy) makes NE 108 a good candidate for high-energy physics applications.

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Table 1
Scintillation yields given in per cent of the non-irradiated samples

Material	λ_e (nm)	Irradiation date	Irradiation field	Dose (Gy)	Days after irradiation					
					1	3	6	8	14	25
PVT	300	16.01.91	PSAIF	4050	98.5	87	82		80	79
		11.06.91	GAMMA	9000			44.3	49	52.1	62.8
NE 108	570	16.01.91	PSAIF	4050	92.1	96.3	93		88.1	
		11.06.91	GAMMA	9000			97.9	78.8	82.9	82.9
NE 110	434	16.01.91	PSAIF	4050	95.3		92		93	97.2
		11.06.91	GAMMA	9000			86.7	87.4	92.7	97.6
NE 118	434	16.01.91	PSAIF	4050	97.5	97.9	98.1		96.3	98
		11.06.91	GAMMA	9000			99.3	92.3	94.2	95.5
NE 122	495	16.01.91	PSAIF	4050	95.2	99.2	93.3		94.4	94.3
		11.06.91	GAMMA	9000			88.5	86.9	94.4	93.5
Pilot-U	390	16.01.91	PSAIF	4050	90.4	94.3	91.6		96.5	90.5
		11.06.91	GAMMA	9000			74.5	75	79	87.1

Table 2
Relative transmission (compared to air) and recovery after
a 9 kGy irradiation with ^{60}Co

Material	λ_e (nm)	Irradiation date	Transmission (%)					
			Non- irradiated	After 6 d	After 20 d	After 40 d	After 310 d	After 460 d
NE 108	570	11.01.91	68.5	62.5	67.5	54		58
PVT		11.06.91	73	69			72	
NE 110	434	11.01.91	75	21	36	54		62
PVT		11.06.91	68	9			63	
NE 118	434	11.01.91	66	40	50		58	
PVT		11.06.91	68	9			63	
NE 122	495	11.01.91	73	53.5	62	75		72
PVT		11.06.91	73	50			70	
Pilot-U	390	11.01.91	54.5	9	21.5	33.5		37
PVT		11.06.91	45	3			37	

Table 3
Relative transmission (compared to air) and recovery after a 9 kGy irradiation with the PSAIF mixed field

Material	λ_e (nm)	Irradiation date	Transmission (%)						
			Non-irradiated	After 1 d	After 8 d	After 26 d	After 180 d	After 300 d	After 480 d
NE 108	570	19.12.90	68	63	65	68	60		56
PVT		23.06.91	68		66			68	
NE 110	434	19.12.90	77	13	35	45	57		60
PVT		23.06.91	63		26			58	
NE 118	434	23.06.91	68	57	60			60	
PVT		23.06.91	63		26			58	
NE 122	495	19.12.90	74	54	64		74		72
PVT		23.06.91	67		58			66	
Pilot-U	390	19.12.90	53	4.5	17	23	33		37
PVT		23.06.91	45		6			26

Fig. 1 : Scintillator samples seen with polarized light

Figure 2 : Radiation effect in PVT by NET

Fig. 3 : Light transmission in NE-108 ($L=10\text{cm}$, $\lambda=570\text{nm}$)

Fig. 4 : Light transmission in NE-110 ($L=10\text{cm}$, $\lambda=434\text{nm}$)

Fig. 5 : Light transmission in NE-118 ($L=10\text{cm}$, $\lambda=434\text{nm}$)

Fig. 6 : Light transmission in NE-122 ($L=10\text{cm}$, $\lambda=495\text{nm}$)

Fig. 7 : Light transmission in NE-PILOT U ($L=10\text{cm}$, $\lambda=390\text{nm}$)

Fig. 8 : Light transmission in NE scintillators (10 cm)

Fig. 9 : Light transmission in NE scintillators (10 cm)

Fig.10 : Light transmission in NE scintillators (10 cm)