STUDY OF RADIATION AND NATURAL AGING OF PLASTIC SCINTIL-LATORS

N.P. Khlapova, V.G. Senchyshyn, V.N. Lebedev, A.F. Adadurov ISMA NTK «Institute for single crystals» NAN of Ukraine, Kharkov, Ukraine tel. (0572) 3410269, E-mail: sench@isc.kharkov.com

An influence of radiation and natural aging on scintillating characteristics of polystyrene scintillators (PS) produced by different technologies is studied. Influence of molecular-mass distribution on PS radiation hardness is also studied. It is shown that with increasing of polydispersion a radiation hardness decreases, which is connected with a change in a free volume of polymer and ultimately with amount of dissolved oxygen. By results of accelerated tests on radiation and thermal aging, assessed operation times are presented for PS of different types. Recommendations are proposed regarding PS selection for different operation conditions.

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INTRODUCTION

In modern high energy physics experiments (LHCb, SSC, CDF, OPERA) in the complex detecting systems with cell or plane structure, PS are often used in a tile, strip or bar shape, their weight being up to several ton. For detector satisfactory working during long time (~10 year) and continuous radiation impacts (~10... 50 kGy) PS must have rather high level of operational reliability. Light yield changing during the whole operational time must not exceed 20%, and light attenuation length – 50%. Severe requirements to PS long stability stimulate designing of scintillating materials resisting to long-term action of external factors (ionizing radiation, air oxygen exposure, humidity and temperature variation).

In the present work we studied influence of technological factors of PS manufacturing on the degree of their natural and radiation aging.

EXPERIMENTAL TECHNIQUE

Scintillators of UPS series were produced on industrial equipment by styrene bulk polymerization in aluminum ampoule (UPS-923A) and in a glass mold (UPS-97GC), as well as by injection molding (UPS-96M). The content of PS was: PST+1.5%p-TP+0.02%POPOP. Some samples were produced with introducing different diffuse enhancers and stabilizers. Produced PS samples were tested on radiation and thermal aging. Samples irradiation was made on a Co-60 unit up to 10...40 kGy under different dose rates. Thermal aging of PS was made in two air thermostats under 60 and 80°C during 3000 hours. At each stage of tests – before, immediately after irradiation and during post-irradiation period, light yield and light attenuation length in PS were measured [1]. Mean weight M_w molecular mass was measured by viscosimetry method. Besides, mean weight M_w and mean number M_n molecular masses were calculated according to thermal regimes of polymerization. On the base of temperature-temporal superposition with the results of thermal aging tests, estimated operational time of PS was calculated.

EXPERIMENTAL RESULTS

Fig.1-3 present the results of PS scintillating characteristics in the process of radiation aging. Immediately after irradiation, significant decreasing was observed of

light attenuation length L_R/L (more than 50%) and light yield I_R/I (up to 25%) (Fig.1,a,b).

After the termination of irradiation, a gradual recovering of samples characteristics is observed. The recovering time is about 80-300 hours dependent on sample content and cumulated dose. When dose rate is decreased from 0.25 kGy/h to 0.1 kGy/h, a partial recovering of samples characteristics is observed during irradiation time (100...400 hours). By transparency changing pattern, one can see (Fig.1,c) that designed composition can be used in such an environment, where the irradiation rate does not exceed 0.1 kGy. In the other case, a significant (more 50%) change in the attenuation length will be observed.

As experimental data show, the dose, which causes up to 20% of the light yield change, slightly depends on the production method and for PS of UPS is 57... 65 kGy. The dose, under which the light attenuation length is decreased by 50% is 18...50 kGy and is a maximum tolerance dose.

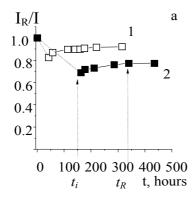
It is to be note that the radiation hardness rather strongly depends on regimes and methods of compositions obtained. The compositions with a high polydispersion $(M_w/M_n=4.5...6.0)$ and a low mean weight molecular mass $M_w=0.6\cdot10^5$ do not possess the high radiation hardness (14...20 kGy). PS produced by a specially chosen temperature regime allows reducing the residual monomer concentration, quantity of end groups and obtaining PS with more high molecular weight and with much less polydispersion $(M_w=(2...4)\cdot10^5, M_w/M_n=(2.2...3.2)$.

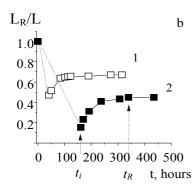
Samples with improving molecular-mass characteristics with significant increasing of the radiation hardness – up to 30 kGy are observed (Fig.2).

Plasticizers introduced promote increasing of molecular mobility and provide an active diffusion of oxygen and favor recombination of radiolysis products. The compositions with the plasticizers have the highest radiation hardness (40...50 kGy), their concentrations being from 5 to 15% (Fig. 2).

According to the results of accelerated tests on a long-term stability, the operational times of different PS were estimated (see Table). These results have good agreement with natural aging data of UPS-923A scintillator in a shape of a long plate of 200×30×2 cm. As can be seen in Fig.3, during four year storage, attenuation of

this plate was change by 7/8%, whereas analogous plate made of foreign scintillator NE 114 - by 25.8%.





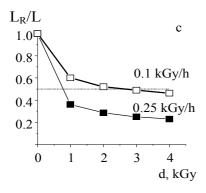


Fig.1. Change of characteristics of UPS-97GC samples irradiated by doses: 1...10 kGy; 2...40 kGy on recover-

ing time: a) light yield I_R/I_r ; b) technical attenuation length L_R/L_r ; c) technical attenuation length L_R/L_r as a function of dose d and dose rate P=0.1...0.25 kGy/h

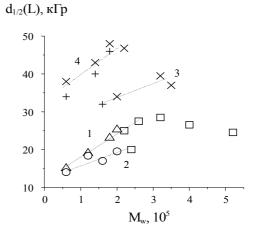
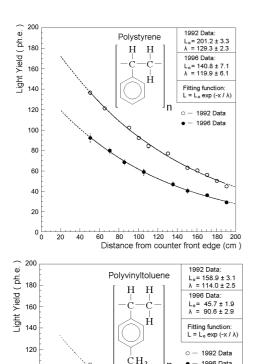


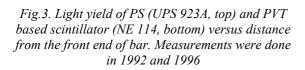
Fig.2. Dependence of half attenuation dose $d_{1/2}$ on production methods of polystyrene compositions producing and their molecular-mass characteristics: () – polymerization in thin glass molds; (o) – polymerization in an aluminum ampoule; (Δ) – injection molding. Samples with plasticizers: $3 - C_p = 5\%$, $4 - C_p = 15\%$. (x) – polymerization, (+) – injection molding

The Table presents the operational parameters, radiation hardness and estimated operational times of different scintillators of UPS series and their foreign analogues. One can see in the Table that scintillators of UPS series, obtained at optimal conditions have high transparency (TAL up to 150 cm, BAL to 400 cm) and high radiation hardness (50 kGy), and also meet the requirements for long-term stability.

Main parameters of plastic scintillators of UPS (Amcrys-H), BC ("Bicron") and SCSN-81 ("Kuraray")

PS type	Scint, eff,%	Light attenuation length, cm		Estimated operational	Dose of halve attenuation d _{1/2} ,	
		(BAL)	(TAL)	time, year	BAL	TAL
UPS-89	100	150	75			
UPS-923A	100	280400	120		5	25
UPS-97GC	108	150250	135150	810	7	25
UPS-98GC	108	180250	135150		7	25
UPS-98RH	92	110	7585		50	50
UPS-97M	95	3060	3060	11	6	25
UPS-98M	95	4070	5070	11	6	25
BC-408	130	370	130150	5	18	30
SCSN-81T	80	110	7085	10	5.5	30





100 120 140

Distance from counter front edge (cm.)

160

180 200

60 80

100

80

60

20

CONCLUSIONS

- 1. Contents are developed and production methods are designed for radiation-hard plastic scintillators of UPS series with diffuse enhancers, which exceed best foreign analogues BC-408 and SCSN-81 by the radiation hardness.
- 2. It is shown that under dose rates that are less than 0.1 kGy/h, samples of 4 mm thickness almost completely recover during irradiation time.
- 3. Dependence of scintillators radiation hardness on molecular mass and polydispersion is studied. It is shown that PS produced by regimes offering a composition forming with a high mean-weight molecular mass and low polydispersion ($M_w = 2...4 \cdot 10^5$, $M_w/M_n = 2.2...3.2$) have high radiation resistance.
- 4. Polystyrene scintillators obtained at optimal conditions have high transparency to the light of self-luminescention (100...150 cm) and high radiation hardness (25...50 kGy), and also meet the requirements of long-term stability of characteristics in operation during 10 years.

REFERENCE

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ИССЛЕДОВАНИЕ РАДИАЦИОННОГО И ЕСТЕСТВЕННОГО СТАРЕНИЯ ПЛАСТМАССОВЫХ СЦИНТИЛЛЯТОРОВ

Н.П. Хлапова, В.Г. Сенчишин, В.Н. Лебедев, А.Ф. Ададуров

Изучено влияние радиационного и естественного старения на сцинтилляционные характеристики полистирольных сцинтилляторов (ПС), полученных по различным технологиям. Изучено влияние молекулярномассового распределения на радиационную стойкость ПС. Показано, что с увеличением полидисперсности радиационная стойкость падает, что связано с изменением свободного объема полимера и, в конечном счете, с количеством растворенного кислорода. По результатам ускоренных испытаний на радиационное и термостарение приводятся прогнозируемые сроки службы различных типов ПС. Предложены рекомендации по выбору типа ПС для конкретных условий эксплуатации.

ДОСЛІДЖЕННЯ РАДІАЦІЙНОГО І ПРИРОДНОГО СТАРІННЯ ПЛАСТМАСОВИХ СЦИНТИЛЯТОРІВ

Н.П. Хлапова, В.Г. Сенчишин, В.Н. Лебедєв, О.Ф. Ададуров

Досліджено вплив радіаційного і природного старіння на сцинтиляційні характеристики полістирольних сцинтиляторів (ПС), отриманих за різними технологіями. Вивчено вплив молекулярно-масового розподілення на радіаційну стійкість ПС. Показано, що з підвищенням полідисперсності радіаційна стійкість падає, що пов'язано зі зміною вільного об'єму полімеру і, в загальному підсумку, з кількістю розчиненого кисню. За результатами пришвидшених тестів на радіаційне і термостаріння приводяться прогнозуємі строки служби різних типів ПС. Запропоновано рекомендації по вибору типу ПС для конкретних умов експлуатації.