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POSSIBLE REASONS FOR CRACKING OF COMPOSITE SCINTILLATORS AFTER IRRADIATION IN AN ELECTRON ACCELERATOR AT A HIGH DOSE RATE

A.V. Krech[1],*, A.Yu. Boyarintsev[1], Ia.V. Gerasymov[1], T.E. Gorbacheva[1], N.L. Karavaeva[1], L.G. Levchuk[2], V.F. Popov[2]

[1]Institute for Scintillation Materials, NAS of Ukraine, Kharkiv, Ukraine;
[2]National Science Center "Kharkov Institute of Physics and Technology", Kharkiv, Ukraine
****E-mail: antonkrech@gmail.com***

Possible reasons for cracking of composite scintillators after irradiation in an electron accelerator at a high dose rate are proposed. The main causes of cracking of composite scintillators, such as ionizing radiation and thermal expansion, are considered. Ionizing radiation can affect the polymer part of the composite scintillator, while thermal expansion mainly affects the crystal grains. A possible mechanism of cracking the composite scintillator during high dose rate irradiation is described.

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INTRODUCTION

Earlier [1–3] we investigated composite scintilla-tors based on grains of organic or inorganic single crystals trying to find radiation-resistant materials. Under the action of the accumulated radiation dose D, the amplitude of the scintillation signal, before irradiation $I(0)$, decreases to $I(D)$ after irradiation. As in our previous works, we will follow the classical definition of a "radiation-resistant scintillator" formula-ted by Birks [4]. A scintillator is considered radiation-resistant up to the accumulated dose D if the relative amplitude of the scintillation pulses $I(D)/I(0)$ is still greater than or equal to 1/2.

Earlier, we fabricated and studied composite scintillators based on the Sylgard-184 transparent dielectric gel composition containing inorganic grains GSO:Ce, GPS:Ce, Al_2O_3:Ti, YSO:Ce, and YAG:Ce [1–3].

In our previous work [1] we studied the radiation resistance of the composite scintillators based on the grains of inorganic single crystals (GSO:Ce and GPS:Ce) in the polydimethylsiloxane. The composite scintillators based on GSO:Ce and GPS:Ce is radiation-resistant at least to $D > 200$ Mrad when dose rate is 0.2 Mrad/h and $D > 250$ Mrad when dose rate is 1,500 Mrad/h. For higher doses, the samples of composite scintillators tend to crack. It results in a premature failure of the composite scintillator. In [2], we showed that composite scintillators containing Al_2O_3:Ti grains began to crack at dose rates of 1,500 and 0.2 Mrad/h for D values of 550 and 125 Mrad, respectively.

At first glance, a paradoxical situation arose. At a lower dose rate (0.2 Mrad/hour), when the intensity of the radiation exposure was lower, the scintillators lost their properties at lower values of the accumulated dose D than when irradiated with a high dose rate (1,500 Mrad/h). It should be noted that at a low dose rate, it takes a longer time to increase the dose D than at a high dose rate. In conducting these studies, we found the following unusual effect. As a result of prolonged exposure at a low dose rate (0.2 Mrad/h), in contrast to exposure at a high dose rate (1,500 Mrad/h), a small amount of liquid appeared on the surface and around the composite scintillator. We measured the pH of this liquid with a universal indicator and found that the liquid has a pH of about unity. This means that the liquid obtained on the surface of the scintillator as a result of prolonged irradiation is a concentrated acid solution.

To explain the cracking of composite scintillators in [5], we considered the following working hypothesis. Cracking occurs in an atmosphere in which radio-chemical reactions are possible. To analyze this hypo-thesis, we investigated several possible chemical reac-tions that could lead to cracking in the irradiated zone. We have performed the following experiments. Compo-site scintillators were placed in aqueous solutions (with pH = 1) of acids H_2SO_4, HCl, H_3PO_4, and HNO_3. The sulfuric acid dissolved the polymer base without cracking. After 72 h, the degree of dissolution reached 90%. We did not observe the effect of cracking in solutions of HCl and H_3PO_4 acids for 720 h. In the process of destruction of samples in a concentrated solution of nitric acid, four stages were observed, which differ from each other, namely: 1) retention of elasticity (120 h), 2) initiation of surface cracks during bending that does not lead to the destruction of the sample (240 h), 3) manifestation of brittleness, namely, the sample is partially destroyed by the sample as a result of weak mechanical stress (480 h), 4) spontaneous cracking of the sample (720 h).

To exclude the presence of sulfuric acid in a liquid that can appear after irradiation, in [5] we also carried out a qualitative analysis of the presence of SO_4 directly in the samples of this liquid. As a result, we showed that the acid H_2SO_4 does not form upon irradiation.

To confirm the presence of nitric acid in the liquid that appeared on the scintillator after irradiation, we performed a quantitative analysis of the content of NO_3 ions in it. In chemical reactions with copper and H_2SO_4, we observed the evolution of brown gas (NO_2), which

confirms the presence of nitric acid in the liquid (for more details, see [5]).

At the low rate of irradiation, the surrounding atmosphere in the irradiation zone has a significant effect on the cracking of composite scintillators due to radiochemical reactions occurring in it. Work [6] shows, that the main aggressive components, the action of which leads to the cracking of the composite scintillator, are nitric acid and possible products of its radiolysis. To avoid cracking of composite scintillators, they must be used in a vacuum or protected from corrosive substances formed in the air during irradiation [6]. However, the aggressive environment in the irradiated zone does not directly affect the relative light output as long as the grains are protected by the gel composition [6].

At the same time, these results do not answer the question: what is the nature of the cracking of composite scintillators at a high dose rate? To answer this question in this article, we propose the theory of the process of cracking radiation-resistant composite scintillators after electron irradiation of the LA-10 linear accelerator at a high dose rate.

1. EXPERIMENTAL DETAILS

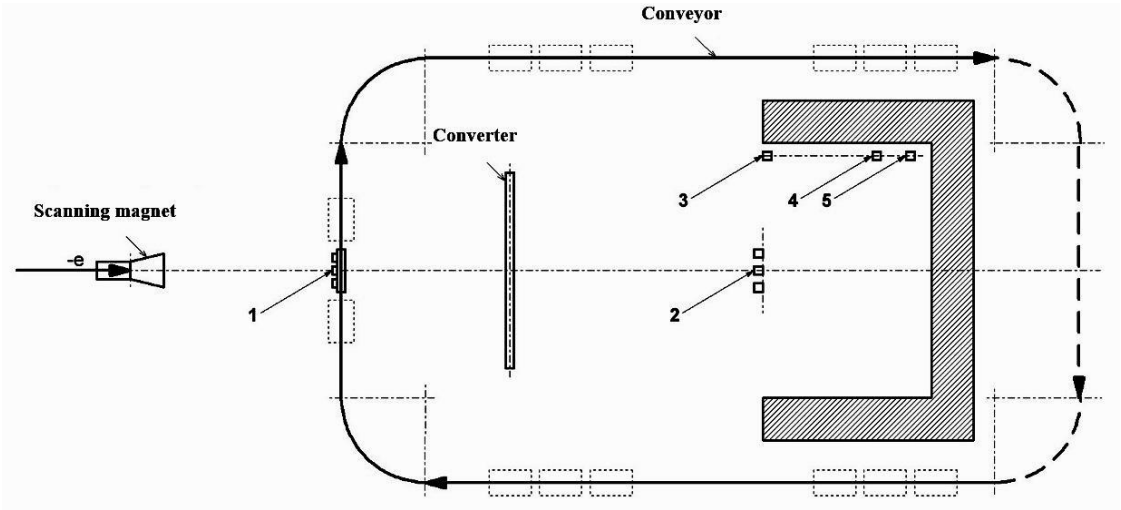
1.1. IRRADIATION OF THE SAMPLES

As in previous works [1–3, 6], electrons with an energy of 10 MeV from the linear electron accelerator of the NSC "Kharkov Institute of Physics and Technology" (Figure) irradiated scintillators at room temperature. The degree of inhomogeneity of the dose rate over the sample surface did not exceed 5%. Plastic dosimeters Harwell Perpex 4034 and radiochromic film dosimeters FWT-60-00 determined the dose rate (for more details, see [7]). The measurement error was ± 5%. These dosimeters are limited to the maximum dose. Therefore, we used a set of identical dosimeters. A single dosimeter accumulated a relatively small dose interval. We determined the integral dose D that the scintillator receives during irradiation by summing up the results of such successive measurements. We irradiated scintil-lators with two dose rates, namely (0.2 ± 0.01) Mrad/h (mainly bremsstrahlung photons) and $(1{,}500 \pm 5)$ Mrad/h (the electron beam directly scans the sample surface).

It is important to note that here and below all the dose values D refers to the absorbed radiation dose in the water equivalent of the irradiated material.

The process of irradiation of composite scintillators takes place in the surrounding atmosphere. As noted above, we irradiated scintillators with either a low ((0.2 ± 0.01) Mrad/h) or high ($(1{,}500 \pm 5)$ Mrad/h) dose rate. In the first case, it was a flux, mainly of brems-strahlung photons. In the latter case, the electron beam directly scanned the sample surface.

At a high dose rate, the accumulated dose D was 500 Mrad as a result of irradiation for 0.34 h (i.e., 20 min). At a high irradiation rate, the samples were on a conveyor belt that moved at a constant speed. The conveyor periodically moved the scintillators to the region of the direct electron beam. In one pass in front of the electron beam, which lasts 12 s, the sample received a dose of about 5 Mrad. In this case, the sample was quickly heated. One pass of the conveyor with a return to the same point lasted about 40…50 min. During this pass, the sample was cooled. Repeated repetition of this process led to the accumulation of the required dose D.



Scheme of the experimental hall of the linear electron accelerator LA-10

2. DISCUSSION

Radiation dose load map in the experimental hall of the linear electron accelerator LA-10, the scheme of which is shown in Figure. Since the samples were irradiated with electrons, the samples were placed on the conveyor (place No.1 on Figure).

Composite scintillators based on grains of inorganic crystals such as GSO, GPS, YSO, and YAG were taken as an example.

When the samples under study are irradiated with electrons, the irradiation dose rate is determined by the speed of movement of the conveyor moving along the circumference. This is also a condition for a constant (stable) value of the electron energy and the average beam current for one pass of the samples in front of the beam. In this case, the value of heating the irradiated samples should be taken into account. Thus, when a sample receives a dose of 0.1 Mrad (1 kGy), the temperature of the sample will increase by 1 °C.

The value of the dose rate of electron irradiation for one pass of the sample in front of the beam is 5 Mrad (50 kGy) for the selected place No.1 of the experimental hall of the accelerator (conveyor) at the selected speed of the conveyor. The time spent by the irradiated sample under the electron beam with the selected movement of the conveyor is ≈ 12 s, which corresponds to the dose rate of electron irradiation of (1,500 ± 50) Mrad/h. The heating of the sample at such a chosen speed of movement is ~ 50 °C, and one turnover of the conveyor will be ≈ 45 min, so the sample will have time to cool down before receiving the next dose of radiation.

Due to instantaneous heating (~ 4 °C per 1 s), thermal expansion of the composite scintillator can be observed. Composite scintillators consist of two components such as grains of inorganic crystals and a gel composition. Therefore, these components may have different coefficients of thermal expansion. At the same time, the gel composition is a polymer (elastomer), which, after stretching (up to several linear dimensions), can return to its previous shape without destruction. Why, then, is cracking observed in composite scintillators after irradiation at a high dose rate?

One possible explanation could be that ionizing radiation affects the polymer. As was shown in [8], the irradiation of the binder base for a composite scintillator (gel composition) led to the densification of polymers. Thus, ionizing radiation contributed to the crosslinking of the polymer chain of the gel composition, polymerized the residual monomer, and so on. As a result, the chemical structure of the polymer was disturbed and the neighbouring polymer chains were bonded, which led to a decrease in the elasticity of the gel composition. However, pure gel compositions without introducing crystalline grains into them were studied in [8]. Therefore, the effect of cracking was not observed, only during mechanical action on polymer samples.

In the case of irradiation of composite scintillators (gel composition + grains of inorganic crystals) with a high dose rate (1,500 Mrad/h), cracking is observed when doses greater than 200…250 Mrad. To obtain such a dose, the samples must go through 40…50 irradiation cycles on the conveyor. That is, the composite scintil-lators are heated and cooled 40…50 times. At the same time, ionizing radiation affects the polymer chains, making them less elastic and more brittle. Thus, a combination of factors such as a thermal expansion of crystal grains and polymer compaction leads to cracking after 40…50 irradiation cycles.

A possible cracking mechanism during high dose rate irradiation is as follows:

1 – after a certain number of cycles of irradiation, cross-linking of the polymer occurs;

2 – after a sufficient degree of crosslinking, the thermal expansion of crystal grains begins to affect;

3 – a slight increase in the linear dimensions of the grains of crystals in the polymer after compaction and a decrease in elasticity leads to small breaks in it;

4 – with each subsequent cycle, this influence increases, thereby increasing the number of defects in the polymer structure;

5 – after a sufficient number of irradiation cycles and accumulation of defects (damages), the composite scintillator is destroyed by cracking its polymer part.

CONCLUSIONS

When composite scintillators are irradiated with high dose rates (such as 1,500 Mrad/h), cracking is observed at doses greater than 200 Mrad. In this case, cracking may be affected by both ionizing radiation and thermal expansion of materials.

1). Ionizing radiation. Ionizing radiation leads to compaction and cross-linking of the polymer part of the composite scintillator, thereby reducing its elasticity.

2). Thermal expansion of materials. At a dose rate of 1,500 Mrad/h, a composite scintillator on a conveyor receives a dose of 5 Mrad in 12 s. This leads to the heating of the sample by 50 °C and a slight increase in the linear dimensions of the crystal grains.

Thus, a decrease in polymer elasticity and thermal expansion of crystal grains under the action of irradiation lead to cracking.

REFERENCES

1. A.Yu. Boyarintsev, N.Z. Galunov, Ia.V. Gerasymov, et al. Radiation-resistant composite scintillators based on GSO and GPS grains // *Nuclear Inst. and Methods in Physics Research, A*. 2017, v. 841, p. 124-129.
2. N.Z. Galunov, T.E. Gorbacheva, B.V. Grinyov, et al. Radiation resistant composite scintillators based on Al_2O_3:Ti grains and their properties after irradiation // *Nuclear Inst. and Methods in Physics Research, A*. 2017, v. 866, p. 104-110.
3. N.Z. Galunov, Ia.V. Gerasymov, T.E. Gorbacheva, et al. Composite scintillators based on single-crystal grains Y2SiO5:Ce (YSO) and Y3Al5O12:Ce (YAG) // *Problems of Atomic Science and Technology. Series "Nuclear Physics Investigations*". 2017, N 3(109), p. 35-39.
4. J.B. Birks. *The theory and practice of scintillation counting.* Oxford, London: "Pergamon Press Ltd". 1967, p. 205 211.
5. V.L. Cherginets, N.Z. Galunov, B.V. Grinyov, et al. Cracking of composite scintillators after significant doses of irradiation // *Functional Materials*. 2019, v. 26, N 4, p. 690-694; https://doi.org/10.15407/fm26.04.690
6. A.V. Krech, A.Yu. Boyarintsev, N.Z. Galunov, et al. Influence of irradiation conditions on cracking of radiation-resistant composite scintillators // *Problems of Atomic Science and Technology*. *Series "Nuclear Physics Investigations".* 2021, N 6(136), p. 30-33; https://doi.org/10.46813/2021-136-030
7. Available on http://www.harwell-dosimeters.co.uk/harwell-red-4034 and http://www.fwt.com/racm/fwt60ds.htm
8. A.Yu. Boyaryntsev, N.Z. Galunov, N.L. Karavaeva, et al. Study of radiation-resistant gel bases for composite detectors // *Functional Materials*. 2013, v. 20, N 4, p. 471-476; http://dx.doi.org/10.15407/fm20.04.471

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МОЖЛИВІ ПРИЧИНИ РОЗТРІСКУВАННЯ КОМПОЗИЦІЙНИХ СЦИНТИЛЯТОРІВ ПІСЛЯ ОПРОМІНЕННЯ НА ПРИСКОРЮВАЧІ ЕЛЕКТРОНІВ ІЗ ВЕЛИКИМ ТЕМПОМ ОПРОМІНЕННЯ

А.В. Креч, А.Ю. Бояринцев, Я.В. Герасимов, Т.Є. Горбачова, Н.Л. Караваєва, Л.Г. Левчук, В.П. Попов

Запропоновано можливі причини розтріскування композиційних сцинтиляторів після опромінення в прискорювачі електронів із великим темпом опромінення. Розглянуто основні причини розтріскування композиційних сцинтиляторів, такі як іонізуюче випромінювання та теплове розширення. Іонізуюче випромінювання може впливати на полімерну частину композиційного сцинтилятору, тоді як теплове розширення в основному впливає на кристалічні гранули. Описано можливий механізм розтріскування композиційного сцинтилятору під час опромінення із великим темпом.