Atomic Energy, Vol. 90, No. 4, 2001

OPTICAL CHARACTERISTICS OF γ-IRRADIATED POLYSTYRENE-BASED POLYMER FIBER

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UDC 535.8.666.189.2;539.107.8;666.11:621.039.553

The experimental results of an investigation of x-ray and photoluminescence and relaxation of the induced absorption spectrum of γ irradiated (0–10^7 Gy) polystyrene-based organic scintillation fibers are presented. Estimates are given for the limits of radiation resistance of the fibers investigated with respect to luminescence intensity and induced absorption. 3 figures, 5 references.

Fiber-optic technology is expected to be widely used for generation and transmission, to external devices, of diagnostic signals from relatively inaccessible regions of the plasma in a thermonuclear reactor [1] and regions of intense nuclear reactions accompanying collisions of high-energy nuclear particles in future accelerators [2]. This naturally leads to the problem of adequate identification of the useful signal, which depends on diverse processes occurring under specific conditions in the fibers employed as the active medium in detectors. The total radiation arising in a fiber in an ionizing radiation field contains, together with Cherenkov radiation, luminescence from activator and radiation-induced centers. In addition, the detected intensity of the light transported by a fiber is also strongly influenced by the nonunique behavior of the induced optical absorption centers. Therefore, these processes must be taken into account appropriately, which makes it necessary to study the optical characteristics of fiber lightguides during and after their irradiation with various types of radiation. With respect to practical applicability in various fields, organic scintillation fibers have a special place among other fiber lightguides. They are of interest because these fibers, though they are less radiation resistant than quartz fibers [3], possess certain advantages: fast response, flexibility, high light output, and low cost.

This work is devoted to an investigation of the optical characteristics of γ-irradiated polymer fiber based on polystyrene.

Object of Investigation and Experimental Procedure. The polymer-organic fiber, obtained from the E. Fermi Laboratory in the US and used in the experiment, consists of a polystyrene core surrounded by cladding consisting of polymethyl methacrylate and an outer protective coating consisting of fluoridated acryl. The diameter of the core/cladding/coating is 770/800/830 μm. The polystyrene core is activated by doping with 1% *p*-terphenal as the primary dye and 1500 ppm 3-hydroxyflavone as the secondary dye. The *p*-terphenal in the polystyrene core is a luminescence activator and the 3-hydroxyflavone is present as a spectrum-shifting additive. The polymethyl methacrylate cladding with a low, compared with the core, refractive index is the waveguide-fiber structure.

In order to absorb the photo- and x-ray luminescence, during sample preparation the polymer fiber was cut into approximately 2 mm long pieces, the ends were carefully polished and rigidly secured side by side on a metal base. Irradiation with ^{60}Co γ rays and measurement of the photo- and x-ray luminescence were conducted without removing the samples from the base. This ensured that the irradiation and measurement conditions were identical. The irradiation dose ranged from 10^3 to 10^7 Gy. All measurements were performed at room temperature at wavelengths 300–800 nm.

Institute of Nuclear Physics, Academy of Sciences of the Republic of Uzbekistan. Translated from Atomnaya Énergiya, Vol. 90, No. 4, pp. 287–291, April, 2001. Original article submitted May 22, 2000.

 1063-4258/01/9004-0304$25.00

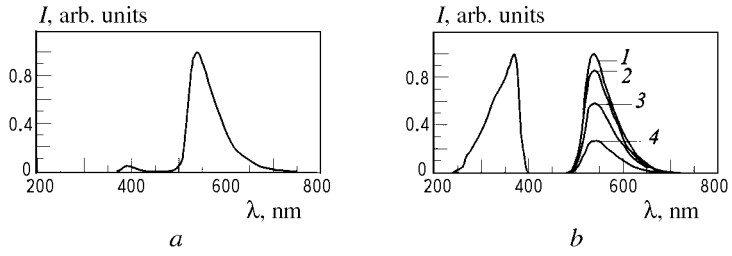

Fig. 1. x-Ray luminescence spectra of unirradiated (*a*) and photoluminescence of γ irradiated wtih dose 0 (*1*), 10^4 (*2*), 10^5 (*3*), and 10^7 Gy (*4*) of polymer fiber and its photoexcitation spectrum (*b*).

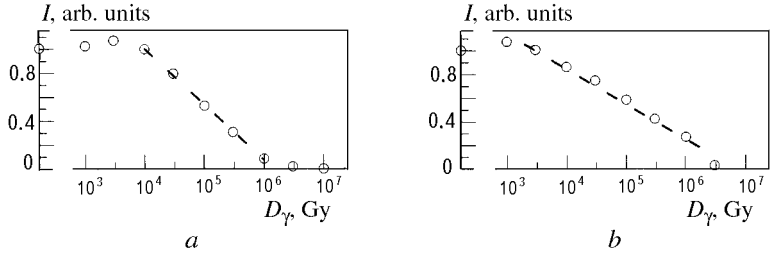

Fig. 2. Dose dependences of the x-ray (*a*) and photoluminescence (*b*) of a polymer fiber.

Two special cells were prepared to investigate the induced absorption spectrum of polymer fibers and the time relaxation of this spectrum (i.e., the restoration of transparency during annealing at room temperature in the dark). Each cell contained the same number of 40 mm long fibers secured side by side and made it possible to measure the irradiation-induced absorption in a Specord UV VIS spectrophotometer. The absorption spectrum was measured at room temperature in the wavelength range 400–750 nm and dose range 10–$3.5\cdot10^5$ Gy with γ irradiation dose rate 1.2 Gy/sec.

Experimental Results and Discussion. The experimentally measured x-ray luminescence spectrum of the nonirradiated polymer fibers consists of two well-resolved bands with maxima at the wavelengths 390 and 540 nm (Fig. 1*a*). In addition, the luminescence intensity at 390 nm is much weaker than the intensity of the main band at 540 nm. The lines in the spectrum of the main x-ray luminescence band at long wavelengths are flatter and their shape remains essentially the same in the entire experimental dose range. The initial γ irradiation with dose 10^3 Gy results in a negligible increase in the intensity of the x-ray luminescence of the main band; as the dose increases, it decreases monotonically and becomes undetectable at a dose of $3\cdot10^6$ Gy (Fig. 2*a*). The second weak luminescence band at 390 nm exists and remains unchanged with an accumulated dose up to $3\cdot10^5$ Gy, after which it vanishes.

The photoluminescence of the experimental polymer fibers is excited by UV light with λ = 370 nm. The band maximum falls at the position of the maximum of the main x-ray luminescence band (540 nm). The weak luminescence band with λ_{max} = 390 nm, observed under x-ray excitation, in this case is not observed either because it overlaps with the long-wavelength tail of the photoexcitation spectrum or it is simply not excited.

The photoluminescence and its excitation spectra for several doses are presented in Fig. 1*b*. The dose dependence of the maximum is presented in Fig. 2*b*.

The observed photo- and x-ray luminescence bands of a polymer fiber with a maximum at wavelength λ_{max} = 540 nm is due exclusively to the presence of a spectrum-shifting reemitter in the polystyrene core – 3-hydroxyflavone in the form of a scintillating additive. This correlates with the luminescence spectrum of pure 3-hydroxyflavone, whose maximum lies at 540 nm [4]. The smoothness of both spectra of the fibers at long wavelengths is also explained by the flatness of the spectrum

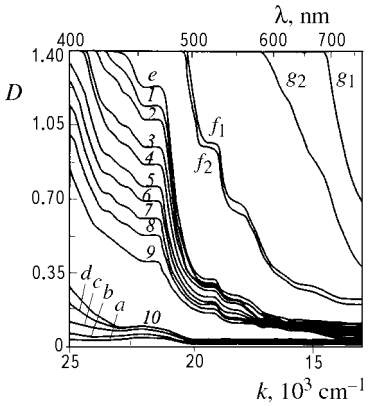

Fig. 3. Induced absorption spectra of a polymer fiber with irradiation dose 10 (*a*), 10^2 (*b*), 10^3 (*c*), $3\cdot10^3$ (*d*), 10^4 (*e*), $2.4\cdot10^4$ (f_1), $2.4\cdot10^4$ (f_2 in 2 h), $2.7\cdot10^5$ (g_1), and $2.7\cdot10^5$ Gy (g_2 in 3 h) and temporal relaxation of the spectrum. The numbers on the curves correspond to the time in which the given spectrum is measured after the spectrum (*e*) is measured: *1–9*) 2, 4, 8, 10, 14, 24, 35, 50, 80 min; *10*) 17 h.

of the 3-hydroxyflavone itself. The weak band (390 nm) in the x-ray luminescence spectrum is most likely due to the intrinsic luminescence of the base (i.e., polystyrene) and the primary scintillating additive – *p*-terphenal. The bands in the luminescence spectra with maxima near 330 nm extend to 400 nm and, together with changes in the transmission spectrum of polystyrene (at wavelength above 360 nm), they can explain the presence of the emission band itself at 390 nm.

The dose dependences of the spectra of the polymer fibers show that for initial γ irradiation with dose 10^3 Gy the maximum of the luminescence increases somewhat. This small increase in the intensity of luminescence can be attributed to the partial restoration of the optimal ratio of the primary and secondary dye concentrations, which is destroyed at the technological stage when the fiber is extruded (at least at the core – cladding interface) because of the formation of associative centers with low binding energy. The first effect of irradiation destroys these associative centers, thereby increasing the number of effectively operating luminescence centers, and as a result the luminescence intensity increases very little. At higher irradiation doses, the intensity decreases, and complete radiation destruction of the luminescence centers occurs at a high dose – greater than $3\cdot10^6$ Gy. For comparison, we note that the optical properties of fibers based on polyvinyl toluene irradiated with 16 MeV electrons are not restored even at $2\cdot10^5$ Gy [5].

The observed decrease in the intensity of luminescence with increasing dose is due to the combined action of the processes leading to the destruction of luminescence centers and the formation of color centers, which increase the induced absorption. The behavior of the dose dependence of the intensity of the luminescence maximum is described well by the expression

$$I/I_0 = 1 - A\lg(D_\gamma/D_0),$$

where I/I_0 is the relative intensity of the luminescence maximum, D_γ is the accumulated γ irradiation dose, and A and D_0 are constants, at least for the dose range 10^4–10^6 Gy. This dependence has been observed with the electronic irradiation of an organic scintillating fiber based on polyvinyl toluene [5].

Comparing the dose dependences shows that the x-ray luminescence intensity decreases more rapidly than the photoluminescence intensity (compare the slopes of the straight lines in Figs. 2*a* and *b*). In the case of x-ray excitation of luminescence, this is probably due to the large contribution of induced absorption as a result of the formation of short-lived (from fractions to several tens of seconds) color centers which do not occur with photoexcitation.

A characteristic feature of the processes occurring in polymers is that they are not instantaneous, but rather they require substantial time intervals, sometimes measured in months. This feature is also confirmed by investigations of the tem-

poral variations of the induced absorption spectra of γ irradiated polymer fibers. Figure 3 displays the spectrum of the induced absorption of a polystyrene fiber. This spectrum was obtained with different γ irradiation doses. The figure also shows the relaxation of the spectrum during annealing at room temperature. In these spectra, the contribution of short-lived color centers to the induced absorption is neglected, since the spectrum after each dose accumulation was measured after 5–6 min. A reliable spectrum, which is repeatable from one measurement to another, was obtained in the wavelength range 400–750 nm. It is evident from Fig. 3 that γ irradiation of polystyrene fibers with a low dose (up to $3\cdot10^3$ Gy, curves *a*, *b*, *c*, and *d*) contributes negligible additional absorption. When the next dose is accumulated, equal to 10^4 Gy (curve *e*), the absorption increases substantially (the optical density exceeds 1.4 in the range 400–450 nm). With time the induced absorption decreases (curves *e*, *1–10*) and in approximately 17 h it almost reaches the initial absorption. Further irradiation gives rise to more absorption bands (curves *f*, *g*), which at dose $\sim5\cdot10^4$ Gy merge, covering the entire experimental wavelength range, i.e., in the entire range the optical density becomes greater than 1.4. It should also be noted that once the γ-irradiation dose $\sim5\cdot10^4$ Gy and higher is accumulated the induced absorption spectrum changes irreversibly, i.e., it no longer returns to the initial level after a definite period of time has elapsed.

Conclusions. The experimental investigation of the x-ray and photoluminescence spectra and the relaxation of the induced absorption spectrum of γ irradiated, in the dose range $0–10^7$ Gy, organic scintillation fibers based on polystyrene showed that the decrease in the intensity of the luminescence is less than 15% up to accumulated dose 10^4 Gy. As the total dose increases further, the intensity decreases logarithmically, and complete destruction of luminescence centers occurs at a high dose – greater than $3\cdot10^6$ Gy. The radiation color centers responsible for the induced absorption and formed in a fiber irradiated by γ rays with dose less than 10^4 Gy are completely annealed at room temperature in 17 h. The γ ray dose 10^5 Gy and higher for the fibers changes the induced absorption spectrum of the fibers irreversibly.

We thank A. Zeminskii for a fruitful discussion of this work.

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