Spectral and dosimetric properties of multilayer structures of radiochromic absorbed dose reference materials

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Abstract. The paper considers a method for extending ionizing radiation absorbed dose measurement range by multilayer technology of one-, two-, three- and four-layer radiochromic films. It shows that this technology expands the AD measurement range to the small dose region of 10-1000 Gy.

1. Introduction
At the All-Russian Scientific Research Institute of Physicotechnical and Radio Engineering Measurements (VNIIFTRI) developed radiochromic, dosimetric RSF films (radiation-sensitive colored film) on the base of which the approved type reference materials of absorbed dose (RM AD) for use in an industrial radiation technology were created. However, due to the insufficient sensitivity of the dosimetric characteristics of the RSF composition at low radiation doses, their application is difficult in the field of radiation treatment of agricultural and food products [1, 3-5].

The response of a radiochromic RSF film to ionizing radiation (II) is determined by a change in its induced optical density (NetOD) at the wavelength of $\lambda=550$ nm (maximum sensitivity to the $^{60}$Co and $^{137}$Cs irradiation). Depending on the level of absorbed radiation dose, the color RSF composition based on 4-diethylaminoazobenzene varies from yellow to different shades of yellow in the spectrum. Currently radiochromic RM AD based on 4-diethylaminoazobenzene are mainly used for measuring the absorbed dose in the (1-10) kGy range («RM AD – 1/10»). The low sensitivity of these RSF has limited their use in the lower radiation dose field, such as dosimetry for food and agricultural products processing by ionizing radiation, including various objects irradiation studies in the (0.05 - 1.0) kGy range.

As demonstrated in [2], the layering of films increases the sensitivity of a radiochromic film for six MV X-rays. They also showed that increasing the number of layers increases sensitivity, but reduces their spatial resolution. The results of these studies were limited only for megavolt X-rays. As far as we know, the technique of multilayer films has not been studied for high rate photon and accelerated electron beams.

This report presents the study of the radiation sensitivity of multilayer structures made on the base of reference film materials «RM AD -1/10» under high rate of $^{60}$Co and $^{137}$Cs radionuclides radiation.

2. Materials and methods
Reference materials of absorbed dose «RM AD -1/10» are made of a single-use radiation-sensitive colored polymer film, manufactured ~28 microns thick according to the technical specifications TU 2379-006-1327176-00, deposited on a carrier made of ~100 µm thick polyethylene terephthalate. The long and wide radiochromic film executed in this way is cut into rectangular fragments with a size of (10-12) x (30-35) mm² and hermetically packed (thermally sealed) 3-6 pieces in a container.
made of paper ~ 50 microns thick, laminated with polyethylene HDPE brand 15803-020 or 10803-020 according to GOST 16377-77 [1]. This laminated paper was chosen due to its weak susceptibility to ionizing radiation. Each container with 5 multilayer identical film fragments was irradiated by doses ranging from 5 to 1000 Gy in a cylindrical aluminum phantom (height of 90 mm, a diameter of 30 mm and a wall thickness of 1.8 mm). The MRH-100 installation with $^{60}$Co radionuclides (dose rate of 2.5 Gy/sec) or on the LMB-γ-1M installation with $^{137}$Cs radionuclides (a dose rate of 0.5 Gy/sec) were processed by radiation. In these experiments, the phantom with films was located at a distance of 40 mm from the sources at the center of a cylindrical squirrel wheel (ø ~ 80 mm) with radionuclides.

To measure the optical density (OD) of multilayer films, a Specord-210 Plus spectrophotometer was used in the absorption measurement mode. The optical density (OD) in the absorption measurement mode is saturated at level eight, and its spatial resolution is no worse than 1mm (the size of the optical beam is ~7x1 mm$^2$). The wavelength range varies from 190-1000 nm in one nm increments. The maximum sensitivity of multilayer compositions is observed at a wavelength of 550 nm, as well as for a single-layer initial one, on which the NetOD of multilayer compositions were read. The NetOD of the irradiated films readout were ~ 8 minutes after the end of irradiation (the time spent for delivering and installing them to the spectrophotometer holder).

First, the absorption spectrum measured for single, two-, triple and four-layer combinations of films irradiated to 1000 Gy in order to establish the wavelength where multilayer films exhibit maximum sensitivity (the highest sensitivity is in the range of 540 - 560 nm with a maximum at 550 nm). Then the optical densities of multilayer films measured at a single wavelength of 550 nm after irradiation at a certain step in the dose range of 5-1000 Gy.

3. Results
Figure 1 shows the measured optical densities as a function of the optical radiation wavelength $\lambda$ for 1-, 2- 3-, 4-layer radiochromic film compositions exposed to ionizing radiation. Figure 2 shows the values of the induced optical density (NetOD) after irradiation of multilayer compositions. The results indicate that for each of these groups of multilayer structures, the maximum response to the absorbed dose occurs at a wavelength of 550 nm, as for single pores of the film. Note that for the selected dose, the peak response of the 4-layer film does not reach the saturation region for Specord 210 Plus spectrometer.

![Fig.1. The spectrum of absolute optical density of unirradiated one-, two-, three- and four-layer radiochromic films.](image)
Figure 2 shows the dependences of the induced optical density NetOD versus wavelength of the optical radiation of the spectrophotometer, calculated by the expression

$$NetOD = A_{\text{irrad}}^i - A_{\text{unirrad}}^i$$ (1)

where $A_{\text{irrad}}^i, A_{\text{unirrad}}^i$ - the absolute value of the optical density of irradiated and non-irradiated films of the i-layer structure, respectively.

![Figure 2](image1.png)

Fig. 2. The NetOD spectrum of one-, two-, three- and four-layer radiochromic films after irradiation with a 1000 Gy dose

Figure 3 shows a comparison of the sensitivity (i.e. optical density per dose unit) of three groups with different film layers measured at different wavelengths.

![Figure 3](image2.png)

Fig. 3. Sensitivity of multilayer compositions for different wavelengths relative to the NetOD for single-layer film at 1 kGy dose

These results show that near the peak at the wavelength (550 nm) the change of the radiochromic composition sensitivity of multilayer structures is insignificant.
Figure 4 is a measured NetOD of the multilayer films in the dose range of 10 - 1000 Gy under the gamma radiation. As can be seen from these results, in the low-dose region, the response of multilayer films linearly depends on the absorbed dose $D$.

From the data in Fig.4, it follows that the sensitivity absolute value for multi-layer structures is equal to $12\cdot10^{-5}$, $23\cdot10^{-5}$, $39\cdot10^{-5}$, $52\cdot10^{-5}$ NetOD/kGy, respectively. Thus, the sensitivity of layered films increases due to an increase in the number of layers. In addition, it follows from Fig.4 that the radiation dose required to obtain an induced optical density of 0.2 decreases from 850G for a single-layer film below 100G for three- and four-layer films. Thus, the multilayer technology increases the sensitivity of the film and expands its scope of application to an area with a lower dose.

The reproducibility of dosimetric properties and the lowest values of the measured dose by multilayer films in the range of 10-1000 Gy can be judged from the graph of relative uncertainty of the optical density $U_i/\text{NetOD}_i$ in Figure 5.
Here

\[ \text{Relative NetOD uncertainty} = \frac{U_i(\text{NetOD})}{\text{NetOD}_i} \]  \hspace{1cm} (2)

(i is the layers number) were obtained according to the results from measurements for five samples of each multilayer structure by the arithmetic mean estimating of the values NetOD\(_{i,k}\) as

\[ \text{NetOD}_i = \frac{1}{n} \sum_{k=1}^{5} \text{NetOD}_{i,k} \]  \hspace{1cm} (3)

and the standard uncertainty of \(U_i(\text{NetOD})\) according to

\[ U_i(\text{NetOD}) = s(\text{NetOD}_i) = \sqrt{\frac{1}{n(n-1)} \sum_{k=1}^{5}(\text{NetOD}_{i,k} - \text{NetOD}_i)^2} \]  \hspace{1cm} (4)

4. Conclusions

With the development of high-power radiation technology installations, such as self-protected installations based on radionuclides \(^{60}\)Co, \(^{137}\)Cs and installations with accelerated electrons up to 10 MeV, dosimetry of the absorbed radiation dose in irradiated products has become very relevant. Radiochromic film dosimetry is a well-established method of measuring the intensity and absorbed dose of radiation to ensure quality during sterilization of medical devices, food, agricultural products processing and the use of radiation techniques in other significant fields of science and technology. Radiochromic film has gained considerable popularity in recent years. They have been investigated for several years in order to circumvent the disadvantages of X-ray films, which exhibit an energy dependence of the response to PD. Moreover, this type of films does not require chemical treatment, allows data processing in daylight, and the measurement results practically do not depend on the temperature during irradiation, as well as in the measurement process. Despite the noted superiority, the low sensitivity of radiochromic films seriously limits its use in low-dose applications, such as dosimetry in the radiation treatment of food and agricultural production. In this study, we have shown that the scope of application of radiochromic films for radiation dosimetry can be expanded into a range of small doses. The results of these studies show that the dosimetric sensitivity of such films can be increased by the quantity of film layers. These results are in good agreement with the published data [2].

References

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