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DOSIMETRIC PROPERTIES OF TUNGSTEN-CONTAINING COMPLEXES OF CARBOXYMETHYL CELLULOSE

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A study is made of the change in the optical properties of polymer compositions based on W-containing complexes of highly substituted carboxymethyl cellulose under the action of ionizing radiation (α-particles, ^{60}Co γ-rays). It is shown that film layers based on the complexes mentioned can be used as radiochromic materials for α-radiation dosimetry in the dose range of $4 \cdot 10^2$–$5.1 \cdot 10^5$ Gy.

In radiation chemistry and especially in radiation technology use is often made of dosimeters based on thin polymer films. Film dosimetry possesses some practical advantages over other measurement methods: in addition to determination of doses, it allows recording of the topography of their distribution over the surface and depth. In particular, it provides the possibility of obtaining the total dose distribution in a plane in one exposure. It should be noted that the films possess a high resolution, which is particularly useful in measuring regions with large dose gradients. Polymer detectors based on transparent plastics (polymethyl methacrylate, polycarbonate, polyvinyl chloride, cellulose nitrates or diacetates, etc.) are widely described in the literature [1-6]. Their operation is most often based on the change in the absorption spectra in the ultraviolet (UV) or visible spectral region under the influence of ionizing radiation. The irradiation-initiated changes in the optical properties allow measurement of absorbed doses within the range of 10^3–10^5 Gy. However, the optical properties of materials based on transparent plastics depend significantly on the presence of impurities in their composition (plasticizers, monomers, etc.) and on a number of external factors: humidity, temperature, radiation aftereffect. Furthermore, these materials do not allow visualization of the absorbed doses or the picture of the dose distribution. From the viewpoint of practical use, systems where the optical properties change in the visible spectral region rather than in the UV region (as in the case of transparent polymer materials) are the most interesting. They make it possible to use simpler, inexpensive, small-sized devices with a simple light source, light filters, and a detector rather than UV spectrometers for quantitative evaluation of the dose or to evaluate it visually by comparing the color (or its change) to a standard calibration scale. At present radiative and chemical transformations in polymer films containing different additives that lead to the occurrence of or a change in the color under the action of radiation are being studied extensively [7-14]. These films are characterized by a lower sensitivity to the external conditions, including surface oxidation, mechanical damage, and so on, and by an ability to change their sensitivity with variation of the concentration of the additives. As the latter, use is commonly made of dyes that either are bleached under the action of radiation [1, 9, 10, 14] or are colored under irradiation [1, 11-13]. Among such dyes are, for instance, colorless forms of leucocyanides of aminotriphenyl methane dyes, which under irradiation switch to a strongly colored form. Radiochromic film dosimeters (RFDs) based on leucocyanides of aminotriphenyl methane dyes are characterized by a linear dependence of the optical absorption on the dose within wide limits (10^3–10^6 Gy) and a weak dependence on temperature and humidity [11].

In some cases, to obtain RFDs the reaction of an acid-base dye with an acid is employed. As a rule, in these systems the films change their color as a result of formation of the acid form of the dye, having a spectrum of actinic absorption that differs from that of the neutral form. At the same time other spectral characteristics, for instance, the luminescence spectrum, can undergo changes. In this respect RFDs based on poly-

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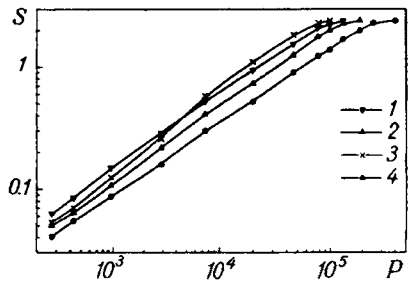

Fig. 1. Optical density of the coloring vs. absorbed dose for layers with different content of sodium tungstate: 1) 70 wt.%; 2) 74; 3) 86; 4) 60.

vinyl butyral (PVB) containing fluorescein and acid chloride are of interest [12]. Upon irradiation the acid form of fluorescein is produced and colorless or weakly colored PVB films become green at λ_{max} = 450 nm and display yellow fluorescence at λ_{max} = 470 nm. The range of the doses measured is 20–700 Gy. An analysis of the literature [7-13] concerned with RFDs reveals that they possess various aftereffects (spontaneous decolorization, i.e., "fading," sensitivity to the ultraviolet spectral region). Therefore, only a few RFDs find wide application in radiation chemistry and radiation technology and, as a rule, none of the dosimetric systems meets all the requirements imposed on a dosimeter since in the general case the type of radiation and its energy exert an influence on the yield of radiolysis products. Thus, RFDs based on PVB containing *para*-rhodanyline cyanide and *para*-nitrobenzoic acid can be used for dosimetry of 10 meV protons at doses up to $4 \cdot 10^5$ Gy and for 1 meV electrons within the dose range up to $7.5 \cdot 10^4$ Gy [13]. In connection with the aforesaid, it remains urgent to search for and develop dosimetric systems with a diverse complex of properties for various fields of application.

In investigating radiative chemical transformations in solid solutions of polymer compositions containing added heavy metals, we established that W-containing complexes of carboxymethyl cellulose exposed to α- or γ-radiation undergo optical changes in their visible spectral region. As is known [1], measurements of the absorption of a dose of heavy charged particles by chemical methods involves a number of difficulties attributable to the fact that the dosimetric systems for which the radiation yields under the action of heavy charged particles have been determined are limited in number. In this connection it seems worthwhile to investigate the dosimetric properties of a material based on W-containing complexes of carboxymethyl cellulose as a function of its composition.

Samples of the material were prepared by pouring an aqueous solution of a composition containing highly substituted carboxymethyl cellulose, sodium tungstate, and a plasticizer onto a baryta paper base followed by drying. The thickness of the film layer was 100 μm. The samples were irradiated by an α-particle unit (FAKT-1, ^{210}Po isotope). The absorbed dose was calculated by the formula $P = 1.6 \cdot 10^{-10} IE/R$ (Gy/sec) [15]. The optical density of the colored portions of the material was measured by a reflecting densitometer, model DO-1, at a wavelength of 620 nm.

As a result of this investigation, we determined the preparation conditions and the constitution of the polymer composition that provide the optimum radiative chemical yield of recovery of the tungsten complexes. Since the content of the tungsten salt is sufficiently high to prevent salting out of the composition, a type and content of plasticizer are chosen that exert no influence on the radiative chemical yield of the system and allow production of materials with good physical and mechanical properties.

The absorption band of the colored product formed upon irradiation has a maximum at 650 nm, which allows visual evaluation of the dose. Figure 1 shows the change in the optical density of coloring as a function of the absorbed dose for different compositions of the material. With a content of 60–86 wt.% tungsten salt in the composition a linear dependence of the optical density of coloring (S) on the dose is observed within the range of $4 \cdot 10^2$–$5.1 \cdot 10^5$ Gy, which testifies to the possibility of using the material as a dosimeter in the indicated dose range. A decrease in the tungsten concentration in the composition to less than 60 wt.% causes a marked decrease in the threshold sensitivity of the layer (to $7 \cdot 10^4$ Gy). As the experiment revealed, the humid-

TABLE 1. Dependence of the Optical Density of the Coloring on the Dose for Different Kinds of Radiation

Kind of radiation, dose, Gy	Optical density
α-particles, LET = 30.0	
10^3	0.12
$5 \cdot 10^3$	0.35
10^4	0.55
10^5	2.4
^{60}Co γ-rays, LET = 0.2	
10^3	0.11
$5 \cdot 10^3$	0.36
10^4	0.56
10^5	2.38

ity (investigations were conducted at 90–95% relative humidity of the air) and the irradiation temperature (10–60°C) do not exert a substantial influence on the optical density of the layer coloring.

The sensitivity of the reaction was evaluated by the relation $\rho = \Delta S/\Delta P$, where ΔS is the change in the coloring density for the dose ΔP absorbed by the material. The quantity ρ is proportional to the radiative chemical yield and is 0.3–0.4 for the compositions studied, which is an order of magnitude higher than the sensitivity of systems based on cellulose diacetate containing 1,4-diaminoanthraquinone [14] and is approximately equal to the sensitivity of PVB-based films containing leucocyanide and *para*-nitrobenzoic acid [13]. This allows evaluation of the absorbed dose in real time not only by measuring S but also by comparing the visually observed change in the coloring to a calibrated scale that represents a sequence of fields of different density of coloring corresponding to the coloring of the sensitive layer at different prescribed radiation doses. These systems are advantageous due to their simplicity and small size.

As is known, heavy charged particles are characterized by higher values of the linear energy transfer (LET, keV/μm) than γ-rays and high-speed electrons. This circumstance is responsible for a change in the yields of radiolytic transformations in many systems. Therefore, in conducting dosimetric measurements of fluxes of heavy charged particles using dosimetric systems, it is necessary either to know the LET or to use systems in which the transformation yield is independent of the LET.

Table 1 provides data on the change in S of the material exposed to radiation of different kinds (α-particles and ^{60}Co γ-rays) up to the same dose. It is evident that S of the material is determined by practically just the absorbed dose and is independent of the LET level (at each point five parallel measurements are made and average values are taken). This testifies to the possibility of using the material for dosimetry of ionizing radiation with an unknown energy spectrum. Such materials can be used not only for dose determination but also for simple monitoring of the spatial distribution of the absorbed energy, which is of importance for technical dosimetry, particularly in using radiation in complex multicomponent equipment where radiation is absorbed by different elements and existing dosimetric methods turn out to be complicated and time-consuming as applied to this problem.

Thus, the possibility of using W-containing complexes of carboxymethyl cellulose as radiochromic dosimeters within the dose range of $4 \cdot 10^2$–$5.1 \cdot 10^5$ Gy is shown.

NOTATION

I, radiation intensity, $cm^{-2} \cdot sec^{-1}$; E, energy of the irradiating particles, MeV; R, mean free path of the particles in the layer, $g \cdot cm^{-2}$; S, optical density of the coloring; P, absorbed dose, Gy; ρ, sensitivity of the reaction; λ, wavelength, μm.

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