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Studies of Sorption Properties of Impregnated Porous Scintillator Based on *p*-Terphenyl

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For dosimetry of β -radionuclides in surface and potable water, an impregnated porous scintillator based on *p*-terphenyl is proposed, which is a matrix with through pores with a sorbent selectively absorbing radionuclides on the pore surface. Synthetic hydroxyapatite is used as sorbent for strontium-90. High measurement sensitivity is ensured by concentration of the emitter from large volume of water and realization of 4π -geometry of radiation detection.

Keywords: porous scintillator; sorption; radionuclides; hydroxyapatite

INTRODUCTION

To improve modern environmental conditions and prevent ecologically dependent diseases, the problem of deactivation of environmental objects is of primary importance ^[1]. Especially important problem is determination of radionuclide ^{90}Sr in natural, technical and potable water, including the lower limit of the concentration range. The most efficient method of radiation monitoring is still the scintillation method.. Difficulties consist in that effects of self-absorption of short-range β -particles in the bulk samples should be avoided. To determine elements with sufficient sensitivity and required accuracy, preliminary concentration of elements traces is necessary ^[2].

The present work was aimed at the development and use in radiation monitoring of new sorption materials and methods of their application for selective determination of radionuclide ^{90}Sr in the environmental waters.

To do this, we have proposed a new material that is a matrix with through pores, on the surface of which a selective sorbent is fixed.

The material structure allows penetration of the analyzed liquid through it. The determined element is accumulated in the scintillator volume. Thus, 4π -geometry is ensured for radiation detection, and simultaneously the radiochemical problem is solved of radionuclide concentration.

EXPERIMENTAL

As selective complex-forming agent, we used synthetic hydroxylapatite (SHA). Studies of its properties were carried out by means of static sorption from solutions with Sr(II) content $10\text{ }\mu\text{g/ml}$. Weighted sorbent was shaken with 100 ml of the studied solution; pH values were varied from 5 to 12, sorbent sample weight - from 0.01 to 1.0 g. To determine the degree of strontium extraction, its concentration in the solutions under study was evaluated by an inductively coupled plasma spectrometer Trace Scan Advantage (Thermo Jarrell Ash).

The impregnated porous scintillator in the form of cylindrical tablets of 15 mm diameter and 3-5 mm height was prepared by pressing of the uniform mixture of granules of activated *p*-terphenyl, ammonium hydrocarbonate and SHA taken in ratio 4:1:1 by mass. Granules of the said substances had linear dimensions of 0.5, 0.1 and 0.05 mm, respectively. Pressing was carried out in a steel press-form under pressure of 20 GPa for 5 minutes at room temperature. Then, to ensure decomposition of ammonium hydrocarbonate, the pressed tablets were put into a quartz ampoule and heated on water bath for 2 hours.

Structure studies of the sample surface were carried out using a MBS-9 optical microscope. The light output of the tablets was measured

using a $^{90}\text{Y}+^{90}\text{Sr}$ beta-source with limiting energy 2.2 MeV. Diphenyl-butadiene-doped *p*-terphenyl crystals were used as reference.

RESULTS AND DISCUSSION

In accordance with the purpose of the present work, the material obtained ensured simultaneous concentrating and direct measuring of the radionuclide ^{90}Sr content.

Preliminary studies showed that the optimum matrix for concentration of radionuclides and measurement of their activities was *p*-terphenyl doped with diphenylbutadiene, ensuring high light output and short decay time.

To extract strontium (II) from the aqueous medium, we have synthesized a selective sorbent - hydroxylapatite. It has high sorption capacity and mechanical strength, is colorless, does not exhibit luminescence, is stable with respect to temperature and radiation.

Apatite belongs to the group of inorganic anionites with three-dimensional crystal lattice. Its chemical composition is $\text{Ca}_{10}(\text{PO}_4)_6(\text{F},\text{OH})_2$ [3]. Selectivity of hydroxylapatite of composition $\text{Ca}_{10}(\text{PO}_4)_6(\text{OH})_2$ to strontium (II) ions is due to isomorphic substitution of Ca^{2+} ions by Sr^{2+} ions in the crystal lattice [4].

To optimize the experimental conditions, we have studied the degree of strontium sorption as function of pH of the analyzed solution, which was measured in static conditions (Fig. 1a). The rising branch of the $R(\text{pH})$ plot is due to active sorbent centers being blocked by H^+ ions at low pH, and the falling branch - to formation of hydroxocomplexes of strontium and lowering of equilibrium concentration of ions Sr^{2+} . Thus, optimum region for carrying out sorption is the region with pH of aqueous phase from 6 to 11, where sorption degree is 99%.

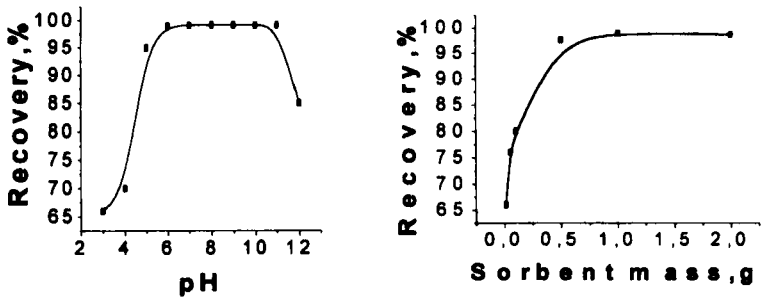


Fig.1. Degree of sorption as function of pH (a) and degree of sorption as function of sorbent mass (b)

From dependence of sorption degree on sorbent mass (Fig.1b), sorbent capacity for Sr^{2+} was calculated, which was found to be 1.09 mmol/g.

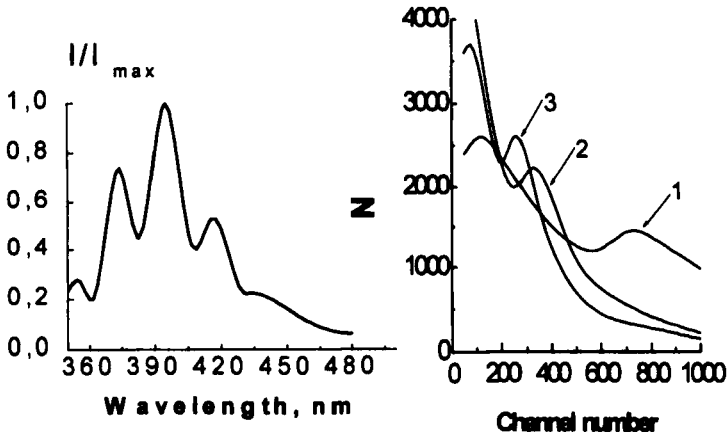


Fig.2. Luminescence spectrum of porous scintillator (a) and energy spectrum of porous scintillator (b)

The main condition for the free flow of the analyzed solution through the scintillator is the presence of through pores. The determined radionuclides can accumulate in the scintillator volume and to ensure 4π -geometry of radiation detection, resulting in high accuracy and sensitivity of the measurements. An easy and reliable way to create pores was introduction of gas-forming substance into the tablets with its subsequent thermal decomposition. We used ammonium hydrocarbonate (decomposition temperature 36-60°C).

At the next stage, the complex-forming sorbent SHA was introduced into the tablet, ensuring sorption completeness and mechanical strength.

Fig.2a shows luminescence spectrum of the prepared material comprising, together with *p*-terphenyl, ammonium hydrocarbonate and SHA. These two substances were neither luminescent, nor quenchers.

Fig.2b shows energy spectrum of point beta-source $^{90}\text{Y}+^{90}\text{Sr}$. Curve 1 corresponds to the reference *p*-terphenyl crystal, 2 - to porous scintillator without complex-forming agent, and 3 - to porous scintillator with SHA.

Light yield of the tables, though 1.5-2 times lower as compared with the reference, still could be considered as satisfactory.

CONCLUSIONS

The proposed method of detection using porous scintillators allows to detect 3.7×10^{-13} g/l of strontium-90, which corresponds to its maximum acceptable concentration in the potable water.

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