

SEPARATION OF IODINE-131 RADIONUCLIDE BY ADSORPTION-CHROMATOGRAPHIC METHOD.

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ABSTRACT

In order to obtain pure radionuclides of ^{131}I , the adsorption-chromatographic separation of the iodine-131 radionuclide from the tellurium target material was studied. In the experiments, we used aluminum oxide impregnated with silver nitrate as a sorbent. The conditions for the sorption of the ^{131}I radionuclide on a modified sorbent in an alkaline medium and its desorption by solutions of Na_2S and $\text{Na}_2\text{S}_2\text{O}_3$ were studied.

KEYWORDS: *Iodine-131, Tellurium dioxide, carrier free, half-life, isotop, separation, radionuclide, irradiating, thermal neutron, nuclear reactor, nuclear reaction, aluminum oxide, silver nitrate, Peristaltic pump, chromatographic column, sorption, desorption, adsorption-chromatography.*

INTRODUCTION

The history of the use of radionuclides iodine-131 in the diagnosis and treatment of thyroid disease begins with the 40-ies of the last century. Such a long period of experience in the application of iodine-131 in the nuclear medicine have shown that it is by far one of the most effective methods in this field for the diagnosis and therapy of thyroid benign and malignant diseases. There is an ever expanding list of radiopharmaceuticals labeled with ^{131}I , which are currently being used or have the potential to be used for the therapy and diagnosis of [1].

In addition to the treatment of thyroid cancer, radionuclide ^{131}I is used in the synthesis of labeled compounds for diagnosis and therapy. One of them is Metaiodobenzylguanidine ($\text{MIBG-}^{131}\text{I}$) for functional imaging of tumors - pheochromocytoma and paraganglioma and for the treatment of chromaffin cell tumors (neuroblastoma, pheochromocytoma, and paraganglioma) [2–6].

^{131}I is a β and γ -emitting radionuclide with a half-life of $T_{1/2} = 8,1$ days, with a main γ -line of 364 KeV (81%) and a β -particle with a maximum energy of 0,61 MeV, an average energy of 0,192 MeV and the average the range of beta particles in soft tissue is 0,4 mm. Radionuclide iodine-131 can be produced in a nuclear reactor by irradiation of compounds of uranium-235 in a yield of 3.076% [7]. However, the extraction of iodine-131 from uranium fission products requires expensive and laborious processing in a complex installation Radionuclide iodine-131 can be produced in a nuclear reactor by irradiation of compounds of uranium-235 in a yield of 3.076% [7]. However, the excretion of iodine-131 from uranium fission products requires expensive and laborious processing in a complex installation.

Another practical method of producing iodine-131 is irradiation tellurium dioxide by thermal neutrons of the reactor. Figure 1. In this case, it is necessary to separate the iodine-131 radionuclide from the tellurium target material. There are several ways to separate the ^{131}I radionuclide, such as: dry distillation, wet distillation, extraction, ion exchange, etc. In the world, the production of the ^{131}I radionuclide is mainly carried out by separating ^{131}I from tellurium mainly by dry distillation, including at the State Enterprise "Radiopreparat" INP AN RUz.

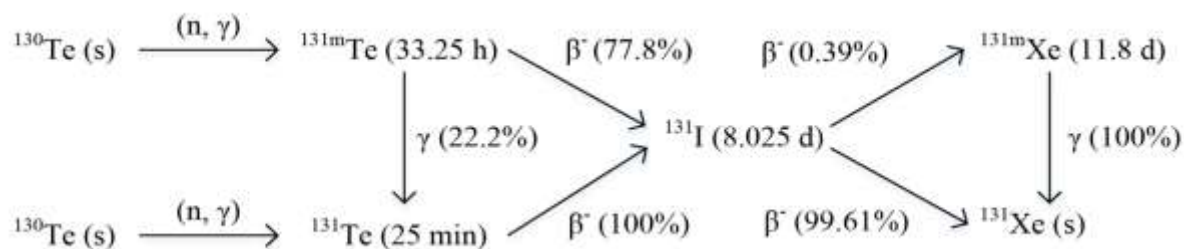


Fig. 1. Nuclear transformation of ^{131}I .

The disadvantage of this technique is that during the thermal distillation of iodine-131 at a temperature of 700-750 ° C, together with iodine-131 vapors, tellurium vapors sublime and the resulting product of sodium iodide with iodine-131 contains tellurium impurities in the form of sodium tellurite Na_2TeO_3 . In addition, during dry distillation, there is a possibility of contamination of workers and nearby hot chambers with radionuclide of iodine-131.

The present work is devoted to the study of the preparation of the iodine-131 radionuclide, the radiochemical separation of the iodine-131 radionuclide from the tellurium target material, by the adsorption chromatographic method. Thus, the problem of developing a simple, convenient and effective method of obtaining the radionuclide iodine-131 still remains relevant

EXPERIMENTAL

As the irradiated target material was used the tellurium dioxide enriched with tellurium-130 (enrichment degree -99.9%).

Irradiation of samples was conducted in quartz ampoules placed in its turn in block containers of aluminum in vertical channels Research Reactor WWR –SM (INP AS RUz). The irradiation time of the samples was 100-140 hours. The irradiated target was dissolved in a hot solution of 2,5 M NaOH at the rate of 7,0 ml/g of the tellurium dioxide.

The reagents: sodium hydroxide, hydrochloric acid, silver nitrate, ascorbic acid, used in the experiments were of chemically pure grade, as a sorbent in the process of adsorption chromatography, was used aluminum oxide for chromatography with a size of 74-150 μm . In the course of the experiments, for the stir the phases was used in a static conditions was used the universal apparatus shaking liquids the ABU-1.

The contacting of the phases carried out in glass vials stoppered, for 10 min. Quantity and quality majoring of radionuclides carried out on gamma spectrometer devices ASPECT SU – 03P with semiconductor Ge (Li) detector. Identification of radionuclides carried out with their gamma-lines.

Impregnate of silver nitrate on aluminum oxide.

Oxide of aluminum for chromatography 74-150 with a size of 74-150 μm washed deionization water and the fine fraction of the sorbent was discarded by decantation. The washed part of the aluminum oxide was treated with 3,0 M HCl solution, shaking on the ABU-1 apparatus for 1 hour. The resulting suspension was allowed to settle, and then washed with deionized water to pH -2, then dried in a drying oven. To the dried alumina (10 g.) was impregnated with silver by adding thereto 6,0 mL of 4,0 M silver nitrate solution. The resulting mixture is dried again. Thereafter, to the mixture is added 10 mL of 2,0 M ascorbic acid solution heated to 50 ° C and the mixture was heated for 15 minutes, stirring occasionally. After that, the supernatant

liquid was removed, washed with cold deionized water until nitrate ions were completely removed and dried in a drying oven at 200 ° C for 4 h.

Investigation of the adsorption and desorption of the ^{131}I radionuclide on the aluminum oxide impregnated with silver nitrate

In 10 glass stoppered vials each added 1,0 ml of a solution of an irradiated sample with an activity of ^{131}I 10 mCi. Then, a NaOH solution or deionized water was added to each tube, bringing their volume to 3.0 ml and the NaOH concentration to 0.1; 0.25; 0.75; 1.0; 1.25; 1.5; 2.0; 2.5; 3.0; 3.5 M respectively. To each tube was added 200 mg of silver impregnated aluminum oxide and shaken for 10 min. After that, the contents of each tube were filtered on 10 columns with a filter, and the filtrates of each sample were measured on a gamma spectrometer. Columns with aluminum oxides adsorbed with ^{131}I were washed with NaOH solutions of the corresponding concentration of the sorption process, deionized water to a neutral medium, and desorbed with solutions of thiosulfate and sodium sulfide of various concentrations, 10 ml each separately.

The process of separating radionuclide ^{131}I

The process of adsorption chromatography of a solution of an irradiated tellurium sample with iodine-131 was performed on a special chromatographic column made of glass, shown in Figure-2 with dimensions Ø-0.5 mm, h-50 mm, containing the sorbent alumina impregnated with silver nitrate. The solution of the irradiated sample was fed to the chromatographic column using a peristaltic pump. The effluent solution (eluate) from the chromatographic column passed through the opening of the protective lead casing with the detector unit of the dosimetric device to control the radioactive background of the eluate.

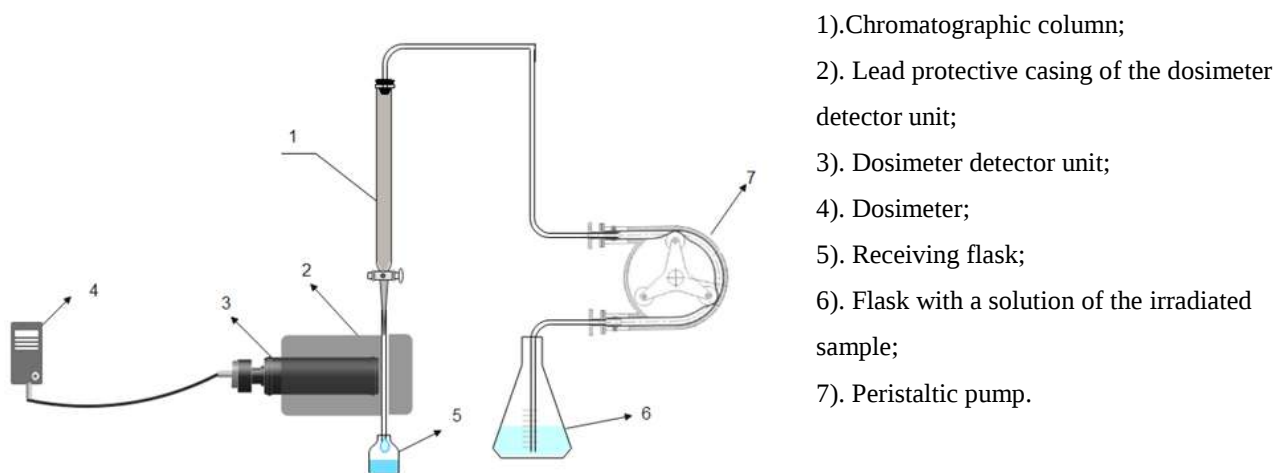


Figure 2 Chromatographic column.

RESULTS AND DISCUSSION

The study of the adsorption of the ^{131}I radionuclide on aluminum oxide coated with silver nitrate was carried out at various concentrations of NaOH. The results of these experiments are presented in table-1.

Sorption of ^{131}I radionuclide on aluminum oxide impregnated with silver nitrate.

Table 1.

C_{NaOH} , mol/L	The amount of injected ^{131}I , mCi	The amount of unsorbed ^{131}I in solution, mCi	Sorption of ^{131}I to the sorbent, %
0,1	10	0,17	98,3
0,25		0,11	98,9
0,75		0,14	98,6
1,0		0,09	99,1
1,25		0,16	98,4
1,5		0,17	98,3
2,0		0,16	98,7
2,5		0,06	99,4
3,0		0,15	98,5
3,5		0,18	98,2

It follows from the results that the adsorption of ^{131}I - ions on the adsorbent of aluminum oxide impregnated with silver nitrate from NaOH solutions, with a concentration of 0,1 to 3,5 mol/l, is more than 98%.

To select the optimal concentration of NaOH, to separate iodine-131 from tellurium, the volume of the formed solution of the irradiated sample and the fluidity of the solution in the chromatographic column were taken into account.

Taking this into account, the optimal concentration for the separation of ^{131}I from tellurium on a chromatographic column with aluminum oxide impregnated with silver nitrate, the concentration of a NaOH solution was 2,5 mol/L.

The study of the desorption of the ^{131}I radionuclide was carried out sorbed on alumina impregnated with silver nitrate at various levels of sodium thiosulfate and sodium sulfide.

The results of these experiments are presented in Tables 2 and 3.

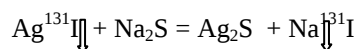
Desorption of radionuclide of ^{131}I from the adsorbent of aluminum oxide by sodium sulfide solutions.

Table 2.

C_{Na_2S} , mol/L	Sorbed amount of ^{131}I , mCi	Desorbed amount of ^{131}I , mCi	Desorption of ^{131}I from sorbent, %
0,1	9,95	2,4	24,0
0,25		3,25	32,5
0,5		6,28	62,8
0,75		8,96	89,6
1,0		9,65	96,5
1,5		9,66	96,6

From the results of the table it can be seen that the concentration of Na_2S for quantitative desorption of the ^{131}I radionuclide from the adsorbent of aluminum oxide is a Na_2S solution with a concentration of 1,0 mol/l.

The desorption process proceeds due to the exchange of the adsorbed $^{131}\text{I}^-$ ion for the S^{2-} ion, due to the difference in the values of the solubility products AgI and Ag_2S , which are $1,5 \cdot 10^{-16}$ and $7,2 \cdot 10^{-50}$, respectively, where the reaction proceeds according to the following scheme:



Desorption of radionuclide of ^{131}I from the adsorbent of aluminum oxide by sodium thiosulfate solutions.

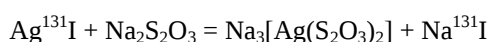
Table

3

$\text{C}_{\text{Na}_2\text{S}_2\text{O}_3}$, mol/l	Sorbed amount of ^{131}I , mCi	Desorbed amount of ^{131}I , mCi	Desorption of ^{131}I from sorbent, %
0,1	9,95	1,79	18,0
0,25		2,2	22,5
0,5		4,25	42,8
0,75		6,9	69,6
1,0		9,5	95,5
1,5		9,6	96,6

The presented results of the table show that for the completeness of desorption of the ^{131}I radionuclide from the adsorbent of aluminum oxide by the $\text{Na}_2\text{S}_2\text{O}_3$ solution, the concentration is 1,0 mol / l.

In this case, the process of desorption of $^{131}\text{I}^-$ ions proceeds due to the formation of a complex of silver with the thiosulfate anion.



Adsorption chromatographic separation of sodium iodide-131

The irradiated sample of tellurium dioxide in the form of tablets weighing 1,0 g, transferred to a round-bottom flask with a flat bottom, containing 7,5 ml of 2,5 M NaOH solution, stirring occasionally until complete dissolution and heated to a boil.

After dissolution and cooling of the solution, an aliquot selected for measuring the amount of radionuclide ^{131}I and it was 27,3 mCi/mL with a total activity 204,75 mCi. Then the solution was passed through a chromatographic column using a peristaltic pump.

Then, for purification from traces of tellurium, the column was washed with 15,0 ml of 2,5 M NaOH solution and deionized water to achievement neutral medium and eluted with 1,0 M sodium sulfide solution. Figure-3 shows the elution profile of iodine-131 with 1,0 M sodium sulfide.

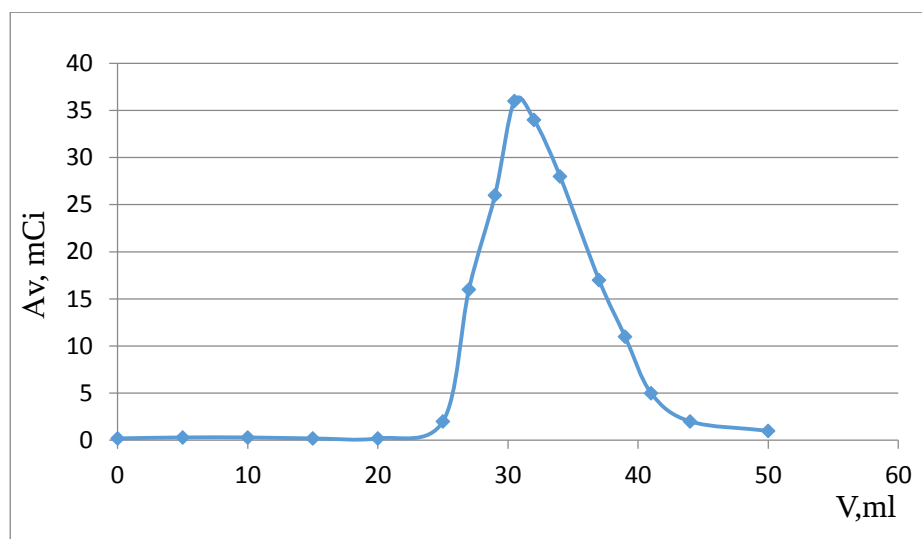


Fig. 3. The profile of elution of sodium iodide-131 with 1,0 M Na₂S solution

As a result, by the adsorption chromatographic method of separating iodine-131 from the tellurium target material, 195,7 mCi of sodium iodide with iodine-131 was obtained with a radiochemical yield of 95,6%.

CONCLUSION

Thus, the studies carried out to study the conditions for the separation of the ¹³¹I radionuclide from the tellurium target material showed that the maximum sorption of ¹³¹I ions on the sorbent of aluminum oxide impregnated with silver nitrate was observed both in weakly alkaline and strongly alkaline media. Quantitative desorption of ¹³¹I from the sorbent was observed with solutions of Na₂S and Na₂S₂O₃ with a concentration of 1,0 mol/l.

Based on the results of the work, it can be concluded that to obtain the iodine-131 radionuclide, carrier free in the form of sodium iodide-131, free from tellurium traces, by separation from the target material of tellurium, may be used the adsorption chromatography method using the sorbent of aluminum oxide impregnated with silver nitrate.

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