

A Laboratory Setup for Increasing the Technological Yield of ^{123}I from a ^{124}Xe Target under Proton Bombardment

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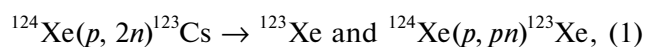
Abstract—One of the methods for obtaining ^{123}I is the bombardment of gaseous ^{124}Xe with protons, in which nuclear reactions of production and decay of ^{123}Xe and ^{123}I isotopes occur. After irradiation, the gas phase is condensed from the target into a special “decay container,” in which the target isotope ^{123}I is produced and accumulated during ^{123}Xe decay. The amount of ^{123}I produced in the target and deposited on its walls during the irradiation is comparable to the amount of ^{123}I obtained in the decay container. A laboratory setup has been created and a process technology for extracting ^{123}I from the walls of the target has been developed to increase the total yield of ^{123}I . Organic solvents (acetone and diethyl ether) are used for this purpose. The proportion of the ^{123}I extracted by washing off from the walls of the aluminum target is at least 84%. The loss during subsequent vacuum distillation of solvents does not exceed 5%. After vacuum distillation, the extracted ^{123}I is dissolved in NaOH. At this stage, the efficiency of ^{123}I washing-off with a 0.01 M NaOH solution is at least 95%. Nevertheless, even taking into account these losses, the proposed method makes it possible to additionally extract the ^{123}I radionuclide from the target in an amount equal to or greater than the activity of the ^{123}I produced using the existing technology.

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1. INTRODUCTION

Radioactive isotope ^{123}I is widely used for diagnostics of many diseases. There are several methods today for producing ^{123}I radionuclide for medical applications using accelerators. Extraction of ^{123}I accumulated in ^{124}Xe gas targets under irradiation is mainly carried out by washing off with water. Depending on the target material, weak solutions of NaOH, NaCl, and NaHSO₃ are also used in addition to water, and extraction is followed by concentration and purification by applying various radiochemical methods [1–3].

The Kurchatov Institute National Research Center has developed and implemented a method for obtaining ^{123}I radionuclide [4, 5], in which the ^{123}I radioisotope is produced by proton bombardment of ^{124}Xe isotope according to the reactions



The ^{124}Xe and ^{123}Xe gases are cryogenically condensed after irradiation into a “decay container,” where they are kept for the decay of ^{123}Xe and the

accumulation of ^{123}I , after which ^{124}Xe is recondensed into a storage and reuse tank. The efficiency of different technologies for collecting ^{123}I after irradiation was analyzed in [4, 5]. The best (in terms of the minimum losses) is the technology in which ^{123}I is extracted both from the target (immediately after irradiation) and from the decay container (at the moment when the ^{123}I activity in it reaches a maximum).

The method currently being implemented by the Kurchatov Institute National Research Center does not allow the extraction of ^{123}I produced in the target during irradiation. Extraction is carried out only from the decay container, which makes it possible to obtain high-purity ^{123}I radionuclide; however, a significant part of it is lost. Although the use of the method in which ^{123}I is extracted only from the decay container leads to large ^{123}I losses, this method has been selected for the study since it is easier and faster to implement. The use of this method with frequent short exposures has made it possible to meet the initial needs of obtaining ^{123}I in an amount of ~1 Ci and to move on to a higher-efficiency extraction technique based on this method.

Table 1. Ratio of the activity of ^{123}I decaying inside the target after 6-h-long irradiation to the maximum activity of ^{123}I in the decay container

Δt , h	0	1	2	3	4	5	6	7
q , rel. units	1.65	1.57	1.49	1.41	1.34	1.27	1.21	1.15

2. KINETICS OF ^{123}I ACCUMULATION

Let us estimate the level of unused ^{123}I remaining inside the target after standard 6-h-long irradiation and ^{123}Xe condensation into the decay container.

The relative amounts of ^{123}Xe and ^{123}I isotopes that are produced and decay during and after the irradiation are represented by simple kinetic equations using only the ^{123}Xe and ^{123}I decay constants.

2.1. ^{123}Xe and ^{123}I in the Target

The change in the amounts of ^{123}Xe and ^{123}I isotopes during irradiation is

$$n_{\text{Xe}}(t) = \frac{V}{\lambda_{\text{Xe}}} (1 - e^{-\lambda_{\text{Xe}} t}), \quad (3)$$

$$N_{\text{I}}(t) = V \left[\frac{1}{\lambda_{\text{I}}} - \frac{1}{\lambda_{\text{I}} - \lambda_{\text{Xe}}} e^{-\lambda_{\text{Xe}} t} + \frac{\lambda_{\text{Xe}}}{\lambda_{\text{I}} (\lambda_{\text{I}} - \lambda_{\text{Xe}})} \times e^{-\lambda_{\text{I}} t} \right], \quad (4)$$

where $\lambda_{\text{Xe}} = 0.333 \text{ h}^{-1}$ and $\lambda_{\text{I}} = 0.052 \text{ h}^{-1}$ are the decay constants of ^{123}Xe and ^{123}I , $n_{\text{Xe}}(t)$ and $N_{\text{I}}(t)$ are the amounts of ^{123}Xe and ^{123}I in the target during irradiation, $V [\text{h}^{-1}]$ is the cumulative rate of the nuclear reaction, and $t [\text{h}]$ is the irradiation time.

Within 6 h after the irradiation and condensation of the gas phase into the decay container, the accumulated ^{123}I remains on the walls of the target body and is subject to decay.

2.2. ^{123}Xe and ^{123}I in the Decay Container

Provided that all the ^{123}Xe , the potential source of the next portion of ^{123}I , is completely transferred to the decay container, the change in their amounts is

$$n_{2\text{Xe}}(t) = n_{\text{Xe}}(6) e^{-\lambda_{\text{Xe}} t}, \quad (5)$$

$$N_{2\text{I}}(t) = n_{\text{Xe}}(6) \frac{\lambda_{\text{Xe}}}{\lambda_{\text{I}} - \lambda_{\text{Xe}}} (e^{-\lambda_{\text{Xe}} t} - e^{-\lambda_{\text{I}} t}), \quad (6)$$

where $n_{2\text{Xe}}(t)$ and $N_{2\text{I}}(t)$ are the amounts of ^{123}Xe and ^{123}I isotopes in the decay container during decay.

Figure 1 shows graphs of the change in the ^{123}Xe and ^{123}I activities in relative units. The ^{123}I activity accumulated in the target body for 6 h, which is $A_{1\text{I}}(6) = \lambda_{\text{I}} N_{\text{I}}(6)$, is referred to unity. After the end of irradiation, the ^{123}I activity begins to decrease due to

decay (Fig. 1a). The iodine activity $A_{2\text{I}}(t) = \lambda_{\text{I}} N_{2\text{I}}(t)$ in the decay container reaches its maximum $\max(A_{2\text{I}}) = 0.606$ rel. units after the exposure of 6.2–7.0 h (Fig. 1b). This time is determined only by the ^{123}Xe and ^{123}I decay constants, is independent of the irradiation parameters, and is the most advantageous for the ^{123}I extraction from the decay container. The amount of ^{123}I accumulated in the target by the end of the 6-h-long irradiation is 1.65 times larger than the maximum amount of ^{123}I extracted from the decay container. The collection of ^{123}I from the decay container at the time of its maximum activity has already been worked out well, but the technology for extracting the ^{123}I remaining in the target has not yet been developed. Table 1 presents the values of ratio q of the ^{123}I activity inside the target to the maximum activity in the decay container as a function of the extraction time:

$$q(\Delta t) = A_{1\text{I}}(\Delta t) / \max(A_{2\text{I}}),$$

where Δt is the time from the end of irradiation to the moment of ^{123}I extraction from the target.

The data presented in Table 1 allow one to estimate the achieved technological yield of ^{123}I upon timely extraction of ^{123}I from the target body. In order to reduce the time from the end of irradiation to the extraction of ^{123}I from the target, it is necessary that the technology, design, and computer control of this process be developed.

3. SETUP DESIGN

Judging by the available literature, no significant attention has been paid to the search for alternative solvent types for the extraction of the ^{123}I remaining in the target. The ^{123}I isotope was obtained by proton bombardment of xenon enriched in the ^{124}Xe isotope to 20 and 40% [6]. The target bodies made of fused silica and cobalt alloy (Havar) were washed sequentially with 0.1 M NaOH and 0.1 M HCl solutions. A work is known in which highly enriched ^{124}Xe (99.9%) was used for irradiation and the target body was made of stainless steel. The target was washed with a weak water solution of NaI and NaHSO_3 , and $75 \pm 5\%$ of iodine was extracted [7]. The KIPROS system in which the target is made of aluminum with an internal nickel coating has become widespread [8]. The target is washed with water, and iodine is then concentrated by ion exchange chromatography and subsequent dissolution in 0.02M NaOH.

The method that we developed is based on the use of organic solvents that provide high solubility of iodine and do not interact with the material of the target walls. The use of organic solvents offers the following advantages:

1. an increase in the ^{123}I extraction efficiency;
2. the absence of contact between the inner surface of the target and water;

3. the applicability of simple methods for purifying the extracted ^{123}I of radionuclide impurities using the vacuum distillation methods.

Acetone and diethyl ether were selected for the experiments since they are low-toxicity (Class 3) and high-volatility solvents and are completely removed during vacuum distillation. The use of “wet” chemistry in the ^{123}I extraction procedure has required complications in the target design.

Figure 2 shows the diagram of the laboratory setup for ^{123}I radionuclide production by bombarding ^{124}Xe with protons, collecting gaseous products into a decay container, and extracting ^{123}I from the target walls using organic solvents. The setup consists of two units: a gas-vacuum unit (the metal tubes of this unit are indicated with bold lines in the diagram) and an extraction unit (polymer tubes of the extraction unit are indicated with thin lines). Both units are connected to the target. A 3D-model and a photo of the target are shown in Fig. 3.

The target body is made of AD1 grade aluminum. The working chamber is a cylinder 15 mm in diameter and 50 mm in length. The chamber volume is 8.8 cm^3 . The entrance membrane made of aluminum with a thickness of $200\text{ }\mu\text{m}$ is sealed with Viton gaskets.

4. DEVICES AND REAGENTS

The sample activity was measured at successive technological stages of processing by detecting γ rays from ^{123}I with energy $E_\gamma = 159\text{ keV}$ using a GEM 35P4 γ -ray spectrometer from ORTEC (United States) with an HPGe detector. The weight balance during operations was controlled gravimetrically using Mettler PM6100 analytical scales; the gas pressure was monitored by AIR-20/M2-DA sensors from ELEMER (Moscow, Zelenograd, Russia). Acetone (ultrapure), diethyl ether (analytically pure), sodium hydroxide (ultrapure), and deionized water were used in the study. After working out the main technological operations, we proceeded to work with samples of ^{124}Xe isotope enriched to 99.9% by AO PO Electrochemical Plant (Zelenogorsk, Krasnoyarsk krai, Russia) and ^{123}I radionuclide produced at the U-150 cyclotron of the Kurchatov Institute National Research Center.

5. TECHNOLOGICAL OPERATIONS

5.1. Filling the Target with ^{124}Xe

The preparation and the target filling were carried out as follows (Fig. 2). Before the ^{124}Xe was fed into the working communication chamber, measuring trap 4 and target 6 were evacuated using adsorption trap 2 to a residual pressure of 1 Pa. For ^{124}Xe gas puffing into the target to a required pressure, xenon from the vessel 3 was condensed into the measuring trap 4 with a volume of 1 cm^3 . The measuring trap was then slowly

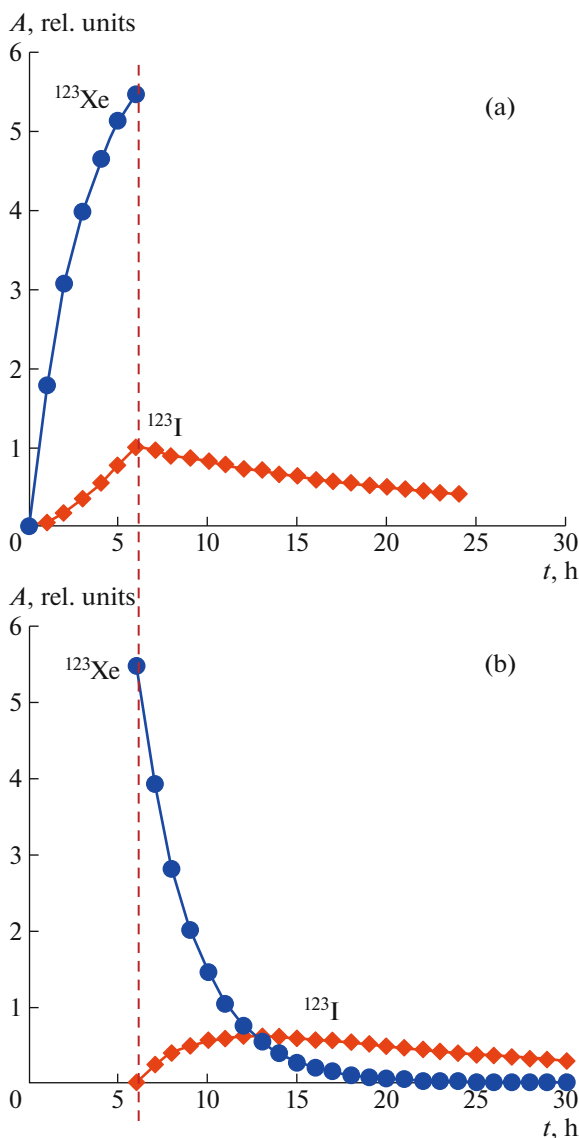


Fig. 1. ^{123}Xe and ^{123}I relative activities A normalized to the ^{123}I activity generated in the target during 6-h-long irradiation (a) in the target and (b) in the decay container.

warmed up, and ^{124}Xe was fed into the target to a pressure of 300 kPa. The residual ^{124}Xe was condensed back into vessel 3.

5.2. Cyclotron Irradiation

The target with valve $v6$ was disconnected from the setup and was transferred for irradiation to the U-150 cyclotron. The standard irradiation for testing the technology for extracting ^{123}I from the target using organic solvents was carried out with 30-MeV protons at a current of $1\text{ }\mu\text{A}$ for 10 min. After the irradiation, the target waited for 24 h, the ^{123}I activity was mea-

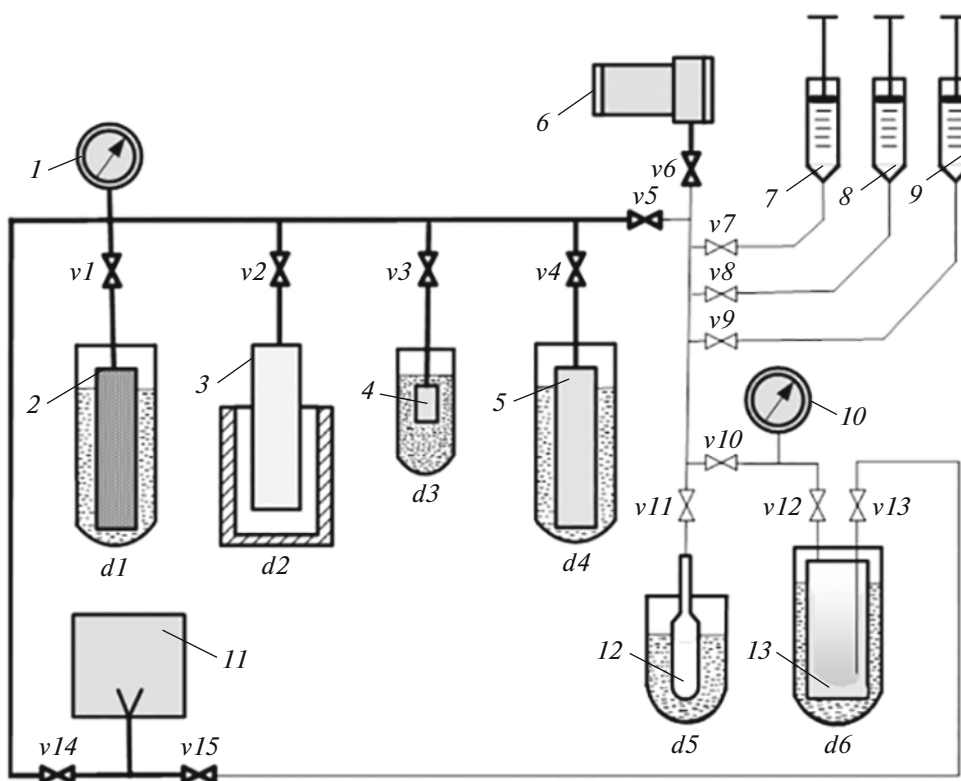


Fig. 2. Diagram of the laboratory setup: (1, 10) AIR-20/M2-DA pressure sensors, (2) adsorption trap with activated carbon, (3) container with the source ^{124}Xe , (4) measuring (auxiliary) trap for ^{124}Xe , (5) trap for collecting irradiated ^{124}Xe and ^{123}Xe , (6) target, (7) syringe dispenser with acetone, (8) syringe dispenser with diethyl ether, (9) syringe dispenser with 0.01 M NaOH, (11) backing vacuum pump, (12) fused silica flask for collecting washed-off samples, (13) pumped trap, (d1–d6) Dewar vessels, and (v1–v15) valves.

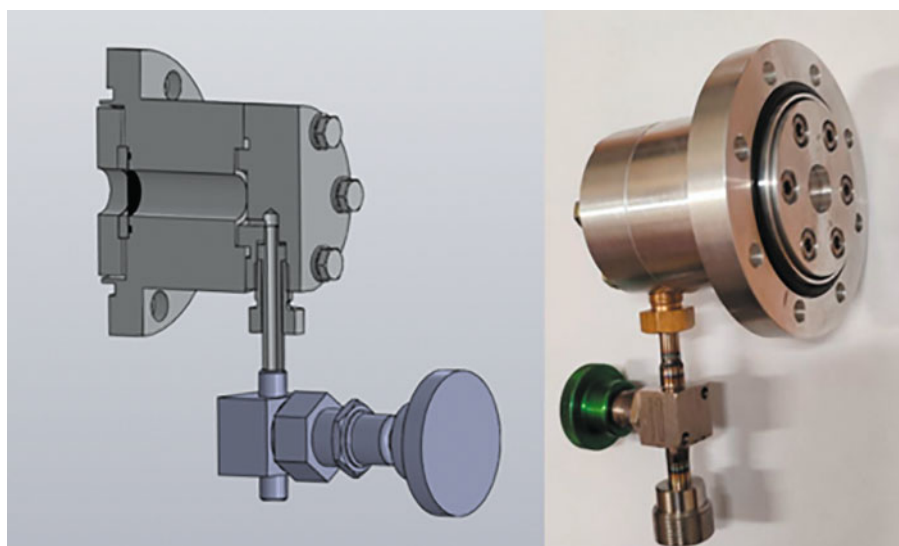


Fig. 3. Design of the target 6 in Fig. 2.

sured by the γ -ray spectrometer, and the target was then connected to the setup.

5.3. Removal of Irradiated ^{124}Xe

The setup was evacuated, and ^{124}Xe condensed from the target into receiving trap 5 to a residual pressure of 1 Pa. Next, the target with valve $v6$ was disconnected from the setup and the remaining ^{123}I activity was measured in it.

5.4. Filling the Target with Solvent and Draining the Eluate

The target was connected to the setup to perform ^{123}I extraction. For this purpose, with the help of the backing vacuum pump 11, the pipelines limited by valves $v5$ – $v11$ and the fused silica flask 12 were evacuated. Valve $v11$ on the fused silica flask and valve $v10$ were then blocked. Valve $v6$ on the target was then opened, and the target was filled with acetone, diethyl ether, or a mixture of them from syringe dispensers 7 and 8 via valves $v7$ and $v8$. After the solvent filled the target, valves $v6$, $v7$, and $v8$ were blocked and the pipelines were evacuated, while solvent residues were captured in pumped trap 13, which was cooled with liquid nitrogen. To carry out the extraction, the solvent was kept in the target for 15 min. The solvent was then drained from the target into the evacuated fused-silica receiving flask 12, which was cooled with liquid nitrogen. To do this, valves $v11$, and $v6$ were sequentially opened. After the solvent with the extracted ^{123}I radionuclide was drained, valves $v6$ and $v11$ were closed and the target with valve $v6$ and the receiving fused-silica flask with valve $v11$ were disconnected from the setup in order to measure the ^{123}I activity in them.

5.5. Vacuum Distillation of Solvent

After measuring the activity, the target and the fused silica flask were connected to the setup and the solvent was vacuum-distilled from the fused silica flask at room temperature. The solvent was captured in pumped trap 13, which was cooled with liquid nitrogen. The duration of vacuum distillation of the solvent was 20–30 min. The distillation was carried out to a residual pressure of 5 Pa in the flask. After the vacuum distillation was completed, the receiving fused-silica flask 12 together with valve $v11$ was disconnected from the setup to measure the ^{123}I activity. After measuring the activity, the fused silica flask was connected to the setup and the pipelines were evacuated.

5.6. Washing ^{123}I off with 0.01 M NaOH Solution

By sequentially opening valves $v11$ and $v9$, 0.01 M NaOH solution with a volume of 5 cm³ was fed into the flask to wash off the ^{123}I radionuclide from the

walls of the flask. Valves $v11$ and $v9$ were then blocked, and the pipelines were washed with a small amount of acetone from syringe dispenser 7 and evacuated through pumped trap 13. The receiving fused-silica flask 12 was disconnected from the setup, and 0.01 M of NaOH solution was manually sampled using a syringe with a capillary tube. After the sampling, the activity in the fused silica flask and the activity of the solution in the syringe were measured to estimate the washing-off efficiency.

5. RESULTS AND DISCUSSION

Experiments on the extraction of accumulated ^{123}I from the target were carried out using acetone, diethyl ether, and a mixture of these solvents with a composition of 50 : 50 vol %.

The target was preliminarily evacuated before filling with solvents. When the valve was opened, the solvents were drawn into it due to the pressure drop. The results of the model experiments have shown that the degree of filling depends on both the solvent type and the filling conditions. The temperatures of the target device, pipelines, and solvent during filling were the same. The filling capacity of the inner volume of the target with acetone was at least 95% under the vacuum condition in the target and a pressure of 1 ata over the liquid acetone. In the case of filling with diethyl ether, the filling efficiency decreased to ~85%; at the same time, it was necessary to create an excess pressure of approximately 1 ata above the liquid ether during filling. Apparently, this was caused by the formation of a vapor plug in the target due to the high elasticity of diethyl ether vapor. The solvent-filled target device was kept at room temperature for 15 min.

The proportion of the solvent extracted from the target changed depending on the conditions of the process. When the solvent was drained into the evacuated fused-silica flask 12 having room temperature, it was possible to collect 85% of ether and 90% of acetone. When the flask was cooled with liquid nitrogen, the collection efficiency approached 100%. Cooling the receiving fused-silica flask 12 was also necessary to make the distillation process smoother and eliminate boiling of the solvent at the initial stage.

The next stage, i.e., the vacuum distillation of solvents from the flush condensed at a temperature of -196°C , was carried out when the trap was heated in an air atmosphere at room temperature. For diethyl ether, the duration of vacuum distillation under the same conditions was reduced by approximately one-third relative to the acetone; it was ~20 min for diethyl ether and ~30 min for acetone.

The obtained ^{123}I radionuclide was washed off from the fused silica flask after vacuum distillation of organic solvents with a 0.01 M NaOH solution. As a result of this operation, the washing-off efficiency, which was determined by the ratio of the ^{123}I activity in

Table 2. Efficiency of ^{123}I extraction from the target

Solvent composition	Efficiency of ^{123}I extraction from the target by washing off, %	^{123}I loss during solvent distillation, %	Total ^{123}I extraction efficiency, %
Acetone 100%	84	17	70 ± 5
Diethyl ether 100%	40	3	39 ± 5
Acetone + diethyl ether 50 : 50 vol %	84	5	80 ± 5

the flask before and after washing-off, was at least 95%.

The efficiency of ^{123}I extraction from the target and the level of losses during solvent distillation were evaluated by measuring the ^{123}I activities in the target and the flask before and after the operations. The measurement results are presented in Table 2.

Comparison of the results presented in Table 2 shows that the efficiency of ^{123}I extraction with acetone is twice as high as the efficiency of ^{123}I extraction with diethyl ether. At the same time, the ^{123}I loss during vacuum distillation of acetone is more than five times higher than the loss during distillation of diethyl ether. These results show the possibility of further optimizing the procedure by changing the composition of the solvent mixture. This has been demonstrated by using a mixture of acetone and diethyl ether with a composition 50 : 50 vol % as an extractant, which provided the highest percentage of ^{123}I extraction.

7. CONCLUSIONS

The laboratory setup to work out the technological process of ^{123}I extraction from the target by washing off with organic solvents has been developed, manufactured, tested, and used. It has been demonstrated that the developed method allows extraction of at least 80% of the ^{123}I remaining on the inner walls of the target during the ^{124}Xe irradiation.

We believe that the developed design of the laboratory setup and technology allow computer control of the process, which will help to reduce the time from the end of irradiation to the ^{123}I extraction from the target and to additionally extract the ^{123}I radionuclide in an amount equal to or greater than the accumulated ^{123}I activity by using the existing technology without increasing the cyclotron irradiation time.

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CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

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