*Int. J. Adv. Res. Sci. Technol. Volume 12, Issue 01, 2023, pp.897-899*

IJARST

*International Journal of Advanced Research in Science and Technology*

journal homepage: www.ijarst.com

**ISSN 2319 – 1783 (Print)**

**ISSN 2320 – 1126 (Online)**

# Recovery of Ytterbium Enriched by Ytterbium-176 from Spent Irradiated Samples of Ytterbium-176.

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**ARTICLEINFO**

***Article history:***

| | |
|---|---|
| Received | 09 Jan 2023 |
| Accepted | 17 Jan 2023 |
| Available online | 31 Jan 2023 |

**Keywords:**
Ytterbium-176,
enrichment degree,
stable isotope,
radionuclide,
recovery,
extraction chromatography,
solid extractant,
chromatographic column,
sorption,
desorption.

PAPER-QR CODE

**ABSTRACT**

To date, one of the promising directions in the development of modern oncology is radionuclide therapy using targeted radiopharmaceuticals, that is, targeted effects on tumor tissues. For the synthesis of such targeted radiopharmaceuticals is suitable the radionuclide $^{177}$Lu with high specific activity. Currently, in world practice, the lutetium-177 radionuclide with a high specific activity is obtained by irradiating ytterbium targets enriched (with an enrichment degree >96%) in the stable isotope of ytterbium-176. Due to the high cost of highly enriched stable isotopes, their availability is not always possible. In this regard, the importance of developing a technology for the regeneration of $^{176}$Yb from spent irradiated targets with a high technological yield is promising. This work is devoted to the development of a technology for the regeneration of grams amounts of ytterbium enriched in the $^{176}$Yb isotope in the form of chloride from spent irradiated $^{176}$Yb samples with a high radiochemical yield, while ensuring high chemical purity.

Citation: Usarov Z.O., Abdukayumov A.M., Rikhsiev A.Z., Akhmedov J.A., Sherov O.O. Recovery of Ytterbium Enriched by Ytterbium-176 from Spent Irradiated Samples of Ytterbium-176. *Int. J. Adv. Res. Sci. Technol.. Volume 12, Issue 01, 2023, pp.897-899.*

## INTRODUCTION

To date, one of the promising directions in the development of modern oncology is radionuclide therapy using targeted radiopharmaceuticals, that is, targeted effects on tumor tissues. For the synthesis of such targeted radiopharmaceuticals, the $^{177}$Lu radionuclide with high specific activity, which has optimal nuclear physical characteristics with an average half-life ($T_{1/2}$ = 6,7 days), is suitable; acceptable energy of β-particles (maximum 0,5 MeV), which allows the destruction of small tumors and metastases without affecting healthy tissues, and soft accompanying γ-radiation with sufficient energy for imaging.

Currently, in world practice, the lutetium-177 radionuclide with a high specific activity close to theoretical is obtained by irradiating ytterbium targets enriched (with an enrichment degree >96%) in the stable isotope of ytterbium-176. Typically, the cost of highly enriched stable isotopes is high. In addition, the availability of highly enriched stable isotopes is not always possible. In this regard, when obtaining the radionuclide of $^{177}$Lu carrier free by irradiation of enriched with $^{176}$Yb, it is very important to develop a technology for the recovery of $^{176}$Yb from spent irradiated targets, in which the loss of the raw material-target $^{176}$Yb was minimal.

According to the known method [1] for the regeneration of ytterbium enriched in ytterbium-176, which consists in collecting a fraction of the solutions of Yb/$\alpha$-HIBA, enriched by $^{176}$Yb, and the solution is acidified to pH-1. The acidified solution is then loaded onto a column packed with Eichrom DGA resin. In this case, the maximum loaded weight of $^{176}$Yb should not exceed 50% of the total resin capacity. The column is then washed with 1,0 M $HNO_3$ and $^{176}$Yb is eluted with 0,1 M HCI.

At the next stage, $^{176}$Yb is quantitatively precipitated with a 10-fold amount of oxalic ($C_2H_2O_4$) acid in the form of ytterbium oxalate [$^{176}Yb_2(C_2H_2O_4)_3$], the precipitate is separated from the supernatant solution, washed with a saturated solution of oxalic acid, and calcined in a quartz tube at 700 $^0$C. According to [2], the regeneration of ytterbium enriched by ytterbium-176 is carried out after 3-4 cycles of separation of the $^{177}$Lu fraction on a chromatographic column, the accumulated Yb-76 was concentrated on a column with a cation exchange resin AG 50W-X12. And in this work, the regenerated column was designed for a loaded target of no more than 50% of the total resin capacity. The pH of the regenerating target solution was adjusted to approximately pH=1 with 65% $HNO_3$. After concentrating of the $^{176}$Yb on the column, the resin was washed with 0,2 M $HNO_3$ solution. Then $^{176}$Yb was eluted with 4 M solution of the $HNO_3$ and evaporated under $N_2$. The resulting regenerated $^{176}$Yb solution, about 60 ml, was evaporated to dryness.

In both cases, the regenerated amount of ytterbium is calculated in milligram quantities.

In addition, during the regeneration of $^{176}$Yb after purification, the solution with $^{176}$Yb is released by precipitation in the form of oxalate, which will lead to the loss of $^{176}$Yb, and to purify $^{176}$Yb from oxalate, the precipitate is calcined and $^{176}$Yb is obtained in the form of oxide $^{176}Yb_2O_3$, which requires additional the process of dissolution in concentrated hydrochloric acid after irradiation and evaporation to a dry residue, and this complicates the regeneration method and will lead to a decrease in technological yield.

In this work, a procedure is presented for the recovery of some grams quantities of ytterbium enriched in the yttebium-176 isotope in the form of chloride with a high radiochemical yield while maintaining high chemical purity.

## Experimental

### Materials and equipment

Irradiated ytterbium enriched in ytterbium-176 (99,8%) used to obtain the substance "Lutetium chloride ($^{177}LuCI_3$) with $^{177}$Lu, without carrier" was used as a regenerated material.

**Reagents:** All reagents and laboratory reagents used in the work were of the highest purity (unless otherwise indicated).

Radiochemical procedures for isolation, purification and concentration of enriched ytterbium-176 were carried out by extraction chromatography. As an extractant, di-2-ethylhexylorthophosphoric acid (D2EHPA) was used in the experiments, and organic solvents, o-xylene, benzene, were used as a diluent for extractants.

Commercial D2EHPA was subjected to preliminary purification from the monofraction by the method described in [3]. Fluoroplast-4 (FT-4) powder with a particle size of ≥38 μm was used as a liquid extractant carrier in the case of extraction-chromatographic experiments. Solid extractant D2EHPA/FT-4 for extraction chromatography was prepared according to the procedure [3].

The extraction-chromatographic column had dimensions d=1.8 mm and H=550 mm.

The control over the process of sorption and desorption during extraction chromatography was carried out by a DRG device connected to a recorder, and the DRG detector was connected to the eluate line at the outlet of the chromatographic column. Quantitative measurements of ytterbium-176 were carried out by measuring the activities of the ytterbium-175 radionuclide on an ASPEKT SU-03P gamma spectrometric device with a semiconductor Ge(Li) detector.

## RESULTS AND DISCUSSIONS

### Regeneration of ytterbium enriched with ytterbium-176

For the regeneration of ytterbium enriched by ytterbium-176, accumulated from 6 cycles [4], spent solutions of ytterbium-176: an acetic acid solution of ytterbium-176 obtained from amalgam, and solutions of the fraction with $^{176}$Yb-α-HIBA after the isolation of the $^{177}$Lu fraction [5], containing a total of 6 g of $^{176}$Yb, combined and acidified with 6,0 M HCI solution to a pH value of 2,5.

After that, the resulting solution is passed through a chromatographic column at a rate of 10 ml/min. Control of the sorption process is carried out by detection of the $^{175}$Yb radionuclide. The chromatographic column is filled with a solid extractant consisting of D2EHPA/FT-4 in a ratio of 1:2, prepared by impregnation of the liquid extractant (cation exchanger) D2EHPA on fluoroplast-4 with particle sizes ≥ 35 μm swollen in ethyl alcohol. To wash away the residues of $NH_4^+$, $K^+$, α-HIBA- and $Cit^{3-}$ ions and possible divalent cations, such as $Zn^{2+}$, $Cu^{2+}$, the column is washed at the same rate, sequentially with three free column volumes of 0,01; 0,5; 1,0 M HCI solutions, respectively.

Enriched $^{176}$Yb is then eluted from the chromatographic column with 6,0 M HCl solution by changing the flow direction of the eluent solution from bottom to top (ascending) at a flow rate of 2,0 ml/min. The resulting solution of $^{176}YbCI_3$ in 6,0 M HCI is passed through an anion-exchange column to remove possible traces of $Fe^{3+}$ and the solution is evaporated on a rotary evaporator to dryness in rotation mode and a vacuum of 10 Pa at a temperature of 80 $^0$C.

The result is 5,85 g of $^{176}$Yb in the form of chloride with a chemical yield of more than 97%.

Table 1 shows the results of the sorption of acidified waste solutions of ytterbium-176 with a concentration of 4,5 mg/ml to different pH values on a chromatographic column filled with a solid extractant D2EHPA/FT-4.

**Table 1. Dependence of sorption of waste solutions of ytterbium-176 at different acidity, pH.**

| Acidification of spent solutions $^{176}$Yb, pH | Radioactive concentration of unsorbed $^{175}$Yb, (mCi/ml) | Total activity of unsorbed $^{175}$Yb, mCi | Sorption of $^{176}$Yb on the chromatographic column, % |
|---|---|---|---|
| pH -1,0 | 0,04 | 45,9 | 85,8 |
| pH -1,3 | 0,029 | 39,15 | 87,9 |
| pH -1,5 | 0,0192 | 25,92 | 92,0 |
| pH -2,0 | 0,0092 | 12,32 | 96,2 |
| pH -2,5 | 0,0036 | 4,86 | 98,5 |
| pH -3,0 | 0,094 | 12,7 | 96,08 |
| pH -3,5 | 0,03 | 40,5 | 87,5 |

Based on the results of the experiment given in the table, it can be seen that the optimal value of acidification of spent ytterbium solutions for sorption on a chromatographic column is pH -2,5, which is 98,5%. Whereas with acidification of the pH value of 3,5, the percentage of sorption of ytterbium decreases to 87%, and when the solution is acidified to pH -1,0 - 1,3, the percentage of sorption also decreases to 85,0%.

Table 2 shows the percentage of sorption of ytterbium-176 from a combined solution of spent solutions of ytterbium-176 on a chromatographic column with a solid extractant D2EHPA/FT-4 with a change in the flow rate of the transmission of the solution (sorption), and in table 3 the effect of speed and the mode of elution of $^{176}$Yb from the chromatographic column with 6,0 M HCI solution.

**Table 2. Influence of the solution flow rate on the sorption of ytterbium-176.**

| The flow rate of the solution during the sorption of $^{176}$Yb pH -2.5; ml/min. | Sorption time, min | Sorption of $^{176}$Yb on the chromatographic column, % |
|---|---|---|
| 2,0 | 675 | 99,0 ± 0,5 |
| 4,0 | 337,5 | 99,0 ± 0,5 |
| 6,0 | 225 | 98,5 ± 0,5 |
| 8,0 | 168,75 | 98,5 ± 0,5 |
| 10,0 | 135 | 98,5 ± 0,5 |
| 12,0 | 112 | 96,8 ± 0,5 |

**Table-3. Influence of the rate and mode of elution (desorption) of ytterbium-176.**

| Elution rate $^{176}$Yb; ml/min. | Downward Elution Time/Volume, min/mL | Time/Volume of ascending elution, min/ml |
|---|---|---|
| 1,0 | 100/100 | 80/80 |
| 2,0 | 150/300 | 50/100 |
| 4,0 | 115/460 | 40/160 |
| 6,0 | 100/600 | 35/210 |

The results of the series of experiments performed showed that during the sorption of $^{176}$Yb from spent solutions of ytterbium-176, acidified with a 6,0 M HCI solution to a pH value of 2.5, on a chromatographic column filled with a solid extractant, consisting of D2EHPA/FT-4 with particle sizes ≥ 35 μm, the most optimal solution flow rate was 10 ml/min. In order to achieve maximum elution of $^{176}$Yb from the chromatographic column, with minimum time spent, the effects of the rate and mode of $^{176}$Yb elution were investigated. The results showed that with the downward elution, the time and volume of the eluted solution is greater than in the upward elution mode.

This is apparently due to the fact that during the sorption of ytterbium the column is saturated from top to bottom, while during elution from bottom to top, i.e. ascending method, the elution distance decreases. Most importantly, it promotes uniform movement of the solution through the column, bypassing the formation of gaps in the column, thereby reducing the elution time and the amount of eluent. Sorption and elution were monitored by detection with the $^{175}$Yb radionuclide.

## CONCLUSION

The sorption of spent ytterbium solutions from irradiated samples of ytterbium enriched by stable isotope Yb-176 with different acidity of solutions onto a chromatographic column filled with a solid extractant D2EHPA/FT-4 was studied. The conditions for sorption of spent ytterbium solutions from irradiated samples of ytterbium-176 and the regime of desorption of ytterbium enriched by ytterbium-176 are established. Thus, from the obtained results of the work, it can be concluded that the technology for the regeneration of spent ytterbium solutions from irradiated samples of ytterbium-176 is suitable.

This technology for the regeneration of ytterbium from spent irradiated samples enriched with ytterbium-176 (solutions) has been introduced into production, and the resulting high-quality product is used as a raw material in the production of the substance "Lutetium chloride ($^{177}$LuCI$_3$) with $^{177}$Lu, without carrier" by the State Enterprise "Radiopreparat".

## REFERENCES

1. TECHNISCHE UNIVERSITÄT MÜNCHEN // Lehrstuhl für adiochemie// Production of noncarrier added (n.c.a.) $^{177}$Lu for radiopharmaceutical applications//Christoph Barkhausen Vollständiger Abdruck der von der Fakultät für Chemie der Technischen Universität München zur Erlangung des akademischen Grades eines, Doktors der Naturwissenschaften (Dr. rer. nat.), genehmigten Dissertation.
2. Moreno, Josue. 2009.
3. Report of the Institute of Atomic Energy named after I.V. Kurchatov "A new effective method for obtaining pure di-2-ethylhexylphosphoric acid (D2EHPA)" - Moscow, 1975, No. 12/13.- 58 p.
4. Abdukayumov A.M., Rikhsiev A.Z., Usarov Z.O., Akhmedov Zh.A., Sherov O.O. // Method for producing lutetium-177 radionuclide, without carrier in the form of chloride. Patent No. FAP UZ 01893, G21 G4 08; 2006.01; Appl. 20200337; Pub. 04/29/2022. Bull. No.4
5. Usarov Z.O., Rustamov N.M., Nishanov Sh.Zh., Abdukayumov A.M., Rikhsiev A.Z., Abdukayumov M. // Method for producing lutetium-177 radionuclide without carrier in the form of chloride. UZ Patent No. IAP 05930, G21 G4 08; 2006/01;G21 G100; 2006.01; Appl. 20140232; Published on December 31, 2015; Bull.12.