

Development a recovery method of ^{131}I from the ^{235}U fission products

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ABSTRACT

^{131}I is a iodine radioisotope widely used in nuclear medicine that can be used either for diagnostic or for treatment due to its physical decay by β - and its high emission of rays- γ . It is produced at IPEN through the irradiation of TeO_2 targets in the IEA-R1 nuclear reactor. There is also the possibility produced it by the fission of ^{235}U . The aim of this work is to develop a recovery method of ^{131}I in the production process of ^{99}Mo through the route of acid dissolution of ^{235}U targets, with the quality to be used in Nuclear Medicine. ^{131}I finds itself in two stages of the process, either in the gaseous produced in the acid dissolution of metallic U targets and the smallest part in solution. In this work was studied the recovery of ^{131}I in these two stages. Several materials were used for the capture and recovery of ^{131}I at the two phases of the process. Anionic cartridges, Ag cartridges, anion exchange resin, activated charcoal columns and AgI precipitation were tested. Solutions with ^{131}I in 0.1 mol.L^{-1} NaOH were percolated through the materials and the eluted solutions were analyzed in a dose calibrator. Among all the tests that were executed, at first, the anion exchange resin and AgI precipitation have showed the best retention result (100%). The results of elution have varied according to the material, the activated charcoal presented a elution yield between 70% and 82%. At first, it is possible to conclude that anion exchange resin and AgI precipitation show better results for ^{131}I retention and the column and activated charcoal have a great potential for the elution of ^{131}I in the right chemical state.

1. INTRODUCTION

Nuclear Medicine is a medical specialty that uses open sources of radionuclides for diagnostic and therapy, being differentiated from other imaging techniques such as MRI and CT that have, the detection ability of metabolism, anatomy and physiology of the organ or system studied [1].

Iodine radioisotopes are commonly used in nuclear medicine in both diagnosis and therapy for several diseases and can be used in a simple molecule or labeling with organic compounds[2].

The iodine isotopes used in medicine are ^{123}I , ^{124}I , ^{125}I and ^{131}I [3]. The ^{123}I is used for thyroid study and diagnostic through the SPECT technique, the ^{124}I is used for PET techniques, the ^{125}I technique can be used in the radioimmunoassay and currently is used in the form of iodine seeds in the brachytherapy technique and the ^{131}I that is used both as diagnostic and treatment of diseases of the thyroid and other studies [1].

^{131}I is mainly obtained in nuclear reactors as a byproduct of the fission of ^{235}U or by irradiating compounds of ^{130}Te , but is also possible to obtain the radionuclide in cyclotron through the $^{130}\text{Te}(d,n)^{131}\text{I}$ reaction[4].

^{235}U fission is the base of the production method for this work. This process produces about 300 different elements coming directly from irradiation of ^{235}U or from his radioactive decay, such as: ^{89}Sr , ^{90}Sr , ^{90}Y , ^{99}Mo , ^{131}I , ^{132}Te , ^{133}Xe , ^{140}Ba . One of the advantages of using this method of production of ^{131}I is the high specific activity obtained with a high radioactive concentration that facilitates the production of labeled compounds with ^{131}I and allows their widespread use inside the producer country. On the other hand this route has disadvantages regarding the cost, investments in infrastructure, management of radioactive waste and the complexity of separation methods from the other elements obtained in fission of ^{235}U , specially due to the chemical interference of other fission products.

The separation of ^{131}I from ^{235}U can be performed by solvent extraction and initially CCl_4 was used in acid solutions, mainly in HCl or HNO_3 . Other techniques were also performed, such as dissolution of ^{235}U in HNO_3 followed by distillation of the acid solution. After cooling the solution was percolated in a Zeolite resin 13X to retain the iodide ion I^- , that subsequently is eluted from the column with water[5]. One system with 2 consecutive columns was used to separate the element in ionic form, one column containing Ag_2O and another with NiCO_3 [6].

^{131}I is produced at IPEN through the irradiation of TeO_2 targets in the IEA-R1 nuclear reactor, where these targets are pressed and irradiated inside Al capsules with 7 cm height and 2 cm diameter. After the irradiation, the ^{131}I is separated by dry distillation, where the targets are put inside an oven, heated at 760°C for 2 hours and the ^{131}I volatile is carried by an O_2 gas stream. This gas runs through 3 traps: the first, containing H_2SO_4 to retain Te, the second containing 0.1 mol.L^{-1} NaOH at low temperature to retain ^{131}I in the form of iodine, and the last, containing 0.1 mol.L^{-1} NaOH at room temperature to retain any ^{131}I that was not retained in the second trap.

2. OBJECTIVE

The aim of this work is to develop a recovery method of ^{131}I in the production process of ^{99}Mo through the route of acid dissolution of ^{235}U low enrichment targets (LEU) with the required quality to be used in Nuclear Medicine.

^{131}I finds itself in two stages of the process, either in the gaseous produced in the acid dissolution of metallic U targets and the smallest part in solution. This work is going to study the recovery of ^{131}I in solution.

3. MATERIALS AND METHODS

3.1. Samples

^{131}I samples were obtained directly from IPEN's production. The first step was to check the activity of samples through a dose calibrator and further dilution in $0.1 \text{ mol}\cdot\text{L}^{-1}$ NaOH if necessary for fractionation and use in each stage of the study.

3.2. Separation of ^{131}I by precipitation of AgI

Precipitation studies of ^{131}I were performed in the form of silver iodide (AgI) and a possible recovery after the dissolution of the precipitate.

The precipitate was obtained using I⁻, the solution was acidified with $1 \text{ mol}\cdot\text{L}^{-1}$ HNO₃, 10 % AgNO₃ (in $0.1 \text{ mol}\cdot\text{L}^{-1}$ HNO₃) and $1 \text{ mol}\cdot\text{L}^{-1}$ HCl, forming AgI and AgCl. After the formation of the precipitate, it was filtered through a Millipore filter and washed with $1 \text{ mol}\cdot\text{L}^{-1}$ HCl and $1 \text{ mol}\cdot\text{L}^{-1}$ HNO₃.

The precipitation was performed at room temperature, in order to optimize the precipitation process, and before filtration the heating of the precipitate for 5 minutes at a temperature of approx. 75°C was tested. The solution used for dissolving the precipitate was $1 \text{ mol}\cdot\text{L}^{-1}$ NH₄OH and 5% Na₂S₂O₃ at room temperature or warmed.

For the tests with ^{131}I the same conditions for the formation of the precipitate were used, however ^{131}I was added to the solution with an activity of 4.70 MBq (127 mCi). The solution containing the precipitate was heated and after cooling was passed through two Millipore filters interconnected. This filter was washed with $1 \text{ mol}\cdot\text{L}^{-1}$ HNO₃. The dissolution of the precipitate was performed using 3 ml of $1 \text{ mol}\cdot\text{L}^{-1}$ NH₄OH, and the solutions were measured in a dose calibrator.

In another experiment with ^{131}I , 10% AgNO₃ was used (in $0.1 \text{ mol}\cdot\text{L}^{-1}$ HNO₃) and 10 mg of iodide carrier, for the formation of the AgI precipitate. The pH of the solution was adjusted to 2 using $1 \text{ mol}\cdot\text{L}^{-1}$ HNO₃. The dissolution was performed using warm 5% Na₂S₂O₃.

3.3. Separation of ^{131}I by adsorption

3.3.1. Activated charcoal columns

Studies have been performed using liquid chromatography columns Luer-Lock 1.0 cm x 20 cm (Sigma-Aldrich) containing activated carbon previously macerated.

The procedure was as follows: a solution containing known activity of ^{131}I was percolated through the column and fractions were collected. The column was washed with $1 \text{ mol}\cdot\text{L}^{-1}$ HCl and finally the ^{131}I elution was studied using NaOH solutions. The column was washed with H₂O before the addition of NaOH. Was used a dose calibrator for evaluated the recovery.

The variables studied were: pH of the solution of ^{131}I (1, 2, 3, 6, 8 and 13), column size (1.5 and 3.0 cm), temperature (room temperature and boiling) and concentration of NaOH solution (0.1 and $0.4 \text{ mol}\cdot\text{L}^{-1}$).

Some experiments were performed using the acid solution obtained after the acid dissolution of uranium metal non-irradiated targets.

3.3.2. Cartridges

The studies were carried out using the following commercial cartridges: Sep-Pak QMA Light (anion exchange adsorber) from Waters and Ag 1.0cc (cation exchange containing Ag adsorber) from Dionex.

The anionic Sep-Pak QMA light cartridge was preconditioned with 20 mL of H₂O and a solution containing ¹³¹I with activity was percolated through the column at both pH 1 and 13. The elution of ¹³¹I was tested with 0.1 mol L⁻¹ NaOH and 4 mol L⁻¹ NaOH. All steps were quantified in dose calibrator.

The Ag cartridge was preconditioned with 20 mL of H₂O. At principle a solution obtained after the dissolution of uranium metal containing ¹³¹I was percolated through the column. The pH of the solution was changed to 2 and 4 using a solution of 0.1 mol L⁻¹ NaOH. All steps were quantified using a HPGe detector due to the low ¹³¹I activity.

3.3.3. Anion exchange resin

Experiments were performed using anion exchange resin AG 1X8 resin (100-200 mesh) assembled inside a liquid chromatography Luer-Lock (1.0 cm x 20 cm) column.

The resin was previously activated with 0.1 mol L⁻¹ NaOH and 0.1 mol L⁻¹ HCl solutions. After that the resin was conditioned with 0.1 mol L⁻¹ NaOH. The elution of ¹³¹I was tested using solutions containing 5 mL of 0.1 mol L⁻¹ NaOH, 5 mL of 0.5 mol L⁻¹ sodium carbonate and 10 mL of 1 mol L⁻¹ NaOH containing 100 µL of H₂O₂. All steps were quantified in dose calibrator.

4. RESULTS

4.1. Separation of ¹³¹I by precipitation of AgI

The experiments with non radioactive iodide solutions showed that the precipitate could be dissolved with warm 1 mol L⁻¹ NH₄OH solution.

In the experiments containing ¹³¹I, 100% of ¹³¹I remained in the precipitate but there was no dissolution of the precipitate with 1 mol L⁻¹ NH₄OH solution. The dissolution was achieved with 5% Na₂S₂O₃ solution yielding 80% of dissolution. The dissolution reached 95% when the solution of 5% Na₂S₂O₃ was heated at 80°C. In the test using only AgNO₃ the same good results were achieved.

4.2. Separation of ¹³¹I by adsorption

4.2.1. Activated charcoal columns

The results of the experiments with the charcoal columns are shown in Table 1.

Table 1: Retention and recovery of ^{131}I in columns of activated charcoal

Column size (cm)	pH	^{131}I Retention (%)	Eluent concentration NaOH (mol L ⁻¹)	^{131}I Elution (%)
3.0	2	81	0.1	42
3.0	2	82	0.1 *	81
3.0 (after acid dissolution)	2	66	**	**
3.0	13	54	0,1	83
1.5	1	52	0,1	53
1.5	2	49	0,4	67
1.5	3	35	0,1 *	59
1.5	6	54	0,4 *	80
1.5	8	68	0,1 *	71

* Boiling NaOH

** no recovery

The results show that the best results were achieved using 3 cm of charcoal, pH 2 and using boiling NaOH solution as elutant for ^{131}I .

4.2.2. Cartridges

Table 2 shows the results using commercial cartridges.

Table 2: Retention and recovery of ^{131}I in commercial cartridges

	pH	^{131}I Retention (%)	^{131}I Elution (%)
Ag (after acid dissolution)	2	91	11
Ag (after acid dissolution)	4	83	0
QMA	13	86	82
QMA	13	97	81
QMA	13	88	90

The best results were achieved using QMA columns loading the ^{131}I solution at pH 13. There was practically no elution of ^{131}I when Ag cartridges were employed.

4.2.3. Anion exchange resin

The anion resin AG 1X8 showed 100% of retention of ^{131}I , but it was not possible to elute ^{131}I from the resin with neither solution employed.

5. CONCLUSIONS

The precipitation of AgI is a very good method for trapping ^{131}I that can be eluted with a warm 5% $\text{Na}_2\text{S}_2\text{O}_3$ solution. Further experiments must be performed in order to separate ^{131}I from Ag.

So far the most effective method for the recovery of ^{131}I in alkaline solution is the use of commercial QMA cartridges.

The anionic resin AG 1X8 is very effective to trap ^{131}I but its elution proved to be very difficult.

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