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A Simple Method for the Separation of ^{111}In from Silver by Thermochromatography

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A dry thermochromatographic process for the quantitative separation of ^{111}In from α -particles bombarded with silver targets at reduced pressure is proposed. The separation is performed in a quartz apparatus using gas phases containing different mixtures of HCl- and steam.

More than 98% of the ^{111}In can be released from a 5.7 g silver target within 0.5 h thereby losing 0.6% of the silver. The ^{111}In adsorbed on a quartz surface can easily be dissolved by a 0.05 M HCl solution.

1. Introduction

Indium-111 is an important radionuclide for nuclear medicine. It is usually produced by irradiation of cadmium with protons or deuterons, or by irradiation of silver targets with α -particles or ^3H -ions. In the latter case impurities of the undesirable $^{114\text{m}}\text{In}$ are completely excluded, but the yield of ^{111}In from the nuclear reactions is lower.

For the separation of the ^{111}In from the target material coprecipitation with hydroxide⁽¹⁾ followed by isopropyl ether extraction,⁽²⁾ anion exchange methods⁽³⁾ and extraction methods^(4,5) have been used. A disadvantage of all these methods is their multistep form.

Westgard *et al.*,⁽⁶⁾ Rudstam *et al.*,⁽⁷⁾ and Eichler *et al.*,^(8,9) have investigated the thermochromatographic behaviour of metal chlorides. It was shown that the adsorption temperature (T_a) of traces of indium chlorides on a quartz surface and the condensation temperature of silver chloride are significantly different. This idea was used by Shevelev *et al.*⁽¹⁰⁾ for separating ^{111}In from molten AgCl.

We propose here a dry thermochromatographic process for the separation of ^{111}In from irradiated silver at reduced pressure and in gas phases containing different mixtures from HCl and steam.

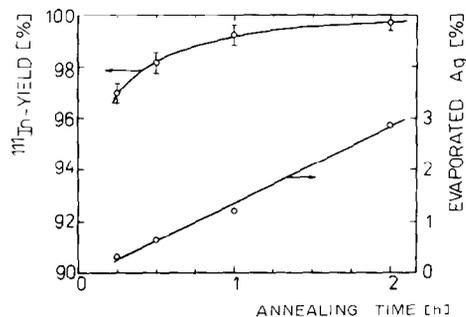


Fig. 1. Dependence of the ^{111}In -extraction and of silver evaporation on time.

2. Experimental

The target (silver, 2 mm thick) was bombarded by α -particles (27 MeV, 100 μAh) in the external beam of the Rossendorf U-120 cyclotron. Irradiated silver samples (0.1-6 g) were inserted into a special quartz furnace (Fig. 1).

After evacuation to an initial pressure of 2.6×10^{-3} Pa the gas mixtures for the chlorination were supplied via the needle valve. The gas mixtures were generated using HCl-solutions of different concentrations (7.2 M, 9.0 M, 10.3 M) (1) to obtain different gas compositions according to the ratios of the partial pressures $p_{\text{HCl}}/p_{\text{H}_2\text{O}}$.

The experiments were performed at total pressures ranging from 0.3 to 13.3 Pa within 15-120 min. The sample temperature was kept constant at (1310 ± 20) K.

The ^{111}In -release from the irradiated silver as well as the indium distribution along the thermochromatographic column were measured by means of a high resolution Ge(Li)-spectrometer.

3. Results and Discussion

Radioactive indium can be separated from silver quantitatively under reduced pressure and defined chlorination conditions (Table 1). More than 98% of the ^{111}In was released from a 5.7 g silver target within 0.5 h (Fig. 1), during this time only 0.6% of the silver was lost. The ^{111}In was adsorbed on a quartz surface and could be easily dissolved in a 0.05 M HCl solution.

The thermochromatogram of ^{111}In (Fig. 2) demonstrates two peaks at the temperatures $T_a = (385 \pm 10)$ K and $T_a = (475 \pm 10)$ K. These peaks do not coincide with the visible precipitation zone of AgCl. A third small ^{111}In -peak was found at $T_a = (655 \pm 20)$ K which always appears in experiments shorter than 1 h.

The relations between the conditions during the thermochromatography and the ^{111}In -fractions adsorbed at the regions 1, 2 and 3 (Fig. 2) (Table 2) permit the identification of the indium compounds as the tri-, mono- and oxychlorides respectively.

Table 1. The release of ^{111}In from 0.3 g irradiated silver samples at: $T = 1315$ K, experimental time: 1 h, ratio of gas composition: $p_{\text{HCl}}/\text{H}_2\text{O} = 1/10$.

Total pressure of the gas mixtures [Pa]	0.40	1.33	4.00	6.67	13.3
Fractional release [%]	99.85 \pm 0.08	99.91 \pm 0.09	99.98 \pm 0.02	99.91 \pm 0.08	99.89 \pm 0.11

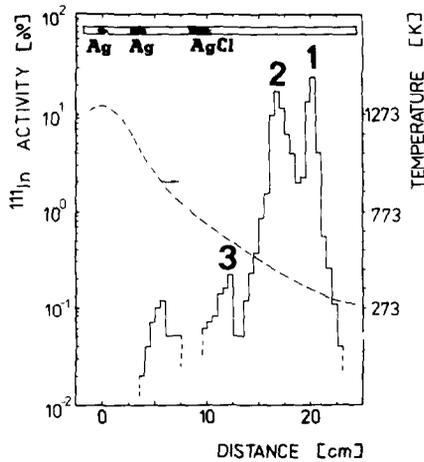


Fig. 2. Thermo-chromatogram of the indium chlorides, evaporated into the gas phase from the irradiated silver experimental time: 1 h, $p_{\text{HCl}}/p_{\text{H}_2\text{O}} = 0.1$, total pressure: 6.67 Pa.

The results of our investigations were used to develop a simple technology for the high temperature separation of ^{111}In from silver (Fig. 3).

Figure 3 shows a diagram of the remote control led process. Chips of silver cut from the target surface (thickness: 100–150 μm) are inserted in a quartz test tube (2) which is sealed and evacuated to 10^{-2} Pa. The HCl steam mixture is supplied via a needle valve adjusting to a total pressure of about 1.3 Pa in the device (4). The generator for the chlorinating gas mixture is a solution of 7.2 M HCl. The furnace (1) with a power of 500 W is switched on and the silver is brought to a temperature of (1310 ± 20) K for 0.5 h, the furnace is then switched off and the gas mixture supply valve is closed. After cooling the apparatus to 320 K the test tube is filled with a solution of 0.05 M HCl via a system of

Table 2. Thermo-chromatographic distribution at different conditions ($T = 1315$ K).

$T_{\text{fraction 1}} = 385$ K,
 $T_{\text{fraction 2}} = 475$ K,
 $T_{\text{fraction 3}} = 655$ K

Total pressure [Pa]	$p_{\text{HCl}}/p_{\text{H}_2\text{O}}$	Time [h]	% of ^{111}In fraction		
			3	2	1
0.67	10/1	0.5	0.97	65.8	32.8
0.67	10/1	1.0	0.80	53.3	45.3
6.67	10/1	0.5	0.68	31.3	66.9
6.67	10/1	1.0	0.35	27.5	71.6
6.67*	1/10	1.0	1.85	52.9	45.1
6.67	1/10	2.0	0.10	8.7	90.6

* Figure 2.

valves and tubes. This solution can be drawn through a quartz filter (3) into the ^{111}In container (5) 3–5 min later. The ^{111}In solution may be concentrated by ion-exchange chromatography and the final solution, in 0.05 M HCl, sterilized by filtration through a millipore filter. The obtained ^{111}In preparations thus contains less than 1 nM Ag in 20 mL solution.

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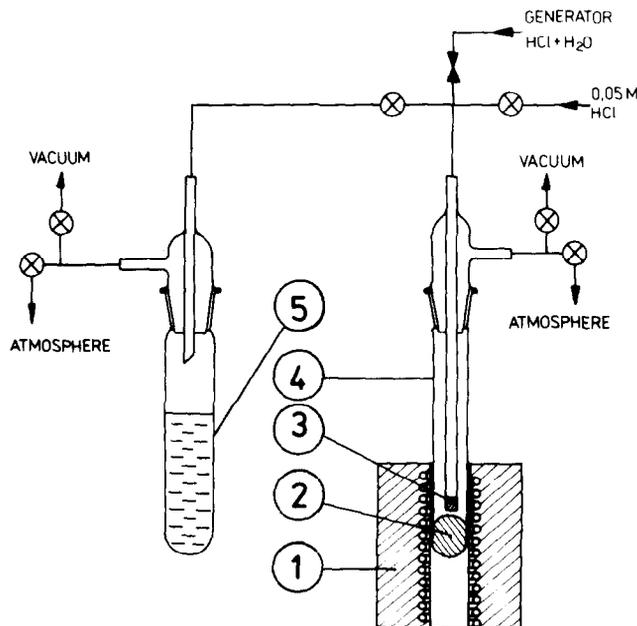


Fig. 3. Remotely controlled version of the method for the high temperature extraction of ^{111}In from silver. 1—electric furnace, 2—silver sample, 3—quartz filter, 4—quartz device for ^{111}In -release, 5— ^{111}In -container.

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