A NO-CARRIER-ADDED $^{72}Se/^{72}As$ ISOTOPE GENERATOR

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Summary

A no-carrier-added $^{72}Se/^{72}As$ generator has been developed for the isolation of ^{72}As, relevant for eventual application in the syntheses of ^{72}As-labelled radiopharmaceuticals. Avoiding the addition of Se carrier and using a thermochromatographic destillation process, no-carrier-added ^{72}As is nearly quantitatively released within 10 min. ^{72}Se remains almost quantitatively (> 99.7%) in solution when a temperature of the separation process of 100°C is applied.

Introduction

Arsenic-72 is a positron emitting isotope with properties which are promising for eventual application in ^{72}As-labelled radiopharmaceuticals. It has a positron emission rate of 88% and positron energies of $E_{\beta+max}$ = 2.5 MeV; $E_{\beta+mean}$ = 1.0 MeV [1]. Although the positron emission decay is accompanied by photons of 834 keV (79.5%), 630 keV (7.9%), 1464.1 keV (1.1%), and others (< 0.5%), the long physical half-life of 26 hours might turn ^{72}As into the PET isotope of choice for biochemical / physiological processes with longer biological half-lives. It can be directly produced at medium-energy cyclotrons via the $^{72}Ge(p,n)$- or $^{72}Ge(d,2n)$-, $^{69}Ga(\alpha,n)$-, $^{71}Ga(\alpha,3n)$-, $^{71}Ga(^3He,2n)$-reactions. More interestingly, however, is its availability as the daughter isotope of ^{72}Se ($T_{½}$ = 8.5 d). ^{72}Se itself can be produced via direct processes such as $^{70}Ge(\alpha,2n)$- and $^{72}Ge(^3He,3n)$- or via proton induced spallation reactions on RbBr [2]. It was the aim of this work to develop a $^{72}Se/^{72}As$ generator relevant for the routine separation of ^{72}As.

Chemical approaches applied until now were based on chromatographic columns, with ^{72}Se as Se^0 adsorbed, while ^{72}As was eluated in rather large volumes of 15 ml [3]. Due to the amount of Se carrier, the separation yields are less than 70%. Another $^{72}Se/^{72}As$ generator was described in [4]. The separation of ^{72}As is achieved under addition of selenic acid carrier in each cycle, followed by reduction to metallic Se using hydrazonium hydrochlorid and its filtration with ^{72}As remaining in solution. Prior to the subsequent separation cycle, Se must be oxidised using H_2O_2.

Production and isolation of ^{72}Se

^{72}Se was produced via the $^{nat}Ge(^{3}He,3n)^{72}Se$-reaction (FZ Juelich, Germany). To isolate ^{72}Se the irradiated Germanium targets are dissolved in HCl/HNO_3 (2:1). After dissolution and distillation of HNO_3, HCl is added. $GeCl_4$ is removed from the solution via distillation while no-carrier-added ^{72}Se (and generated ^{72}As) is quantitatively remaining.

Cyclic separation of no-carrier-added ^{72}As from no-carrier-added ^{72}Se

The HCl solution containing ^{72}Se is transferred to a quartz or glass tube system as shown in Fig. 1, which is inserted vertically into an electric resistance oven (10). 1 g of KCl and 1 ml of conc. HCl are added under formation of non-volatile ^{72}Se compounds and $^{72}As[AsCl_3]$ [5]. Hydrochloric acid (3) is passed through the inlet (2) into the apparatus with a stream of 20 ml/min. The temperature at position (7) of the ^{72}Se fraction inside the tube (1) is raised from 50 to 140°C. The ^{72}As is immediately volatilised as $AsCl_3$ and transported with the stream of hydrochloric acid through tube (4). It is not adsorbed on the inner tube even at its outlet (8), but on a cartridge (9) containing an adequate material (such as charcoal for example). The whole process takes about 10 min. No-carrier-added ^{72}As is nearly quantitatively desorbed from the cartridge in > 90% yields using < 5 ml of H_2O or NaOH and it can be used immediately for labelling reactions.

The no-carrier-added ^{72}Se almost quantitatively remains in solution. Depending on the temperature of the separation process applied (100 – 140°C), > 99.7% of ^{72}Se are still present at position (7) and are ready for the next separation cycle without further treatment.

References

1 Browne E., Firestone R.B.: Table of Radioactive Isotopes (Ed. Shirley V.S.), John Whiley & Sons, 1986
2 Phillips D.R., United States Patent, Nr.: 5,371,372, Dez. 6, (1994); WO 93/04768, 18.03.1993
3 S.H. A-Kouraishi, G.G.J. Boswell, Int. J. Appl. Rad. Isot. 29 (1978) 607
4 Phillips D.R., United States Patent, Nr.: 5,371,372, Dez. 6, (1994); WO 93/04768, 18.03.1993
5 Gmelin, Handbuch der anorganischen Chemie 8 Auflage, System-Nummer 17, Arsen, Verlag Chemie Weinheim, 383, (1952)

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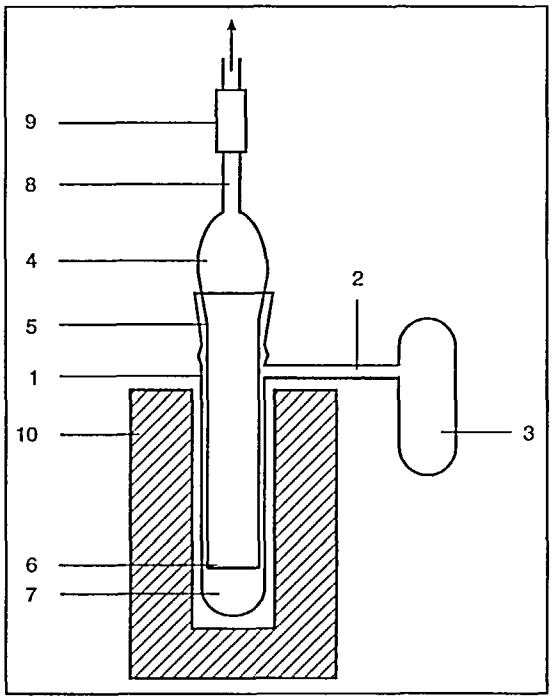

Fig. 1

Scetch of the $^{72}Se/^{72}As$ generator apparatus
1-outer quartz or glas tube;
2-inlet of HCl;
3-HCl vessel;
4-inner quartz or glass tube;
5-ground joint;
6-open lower end of the inner tube;
7-^{72}Se fraction;
8-upper end of the inner tube;
9-adsorber;
10-electric resistance oven

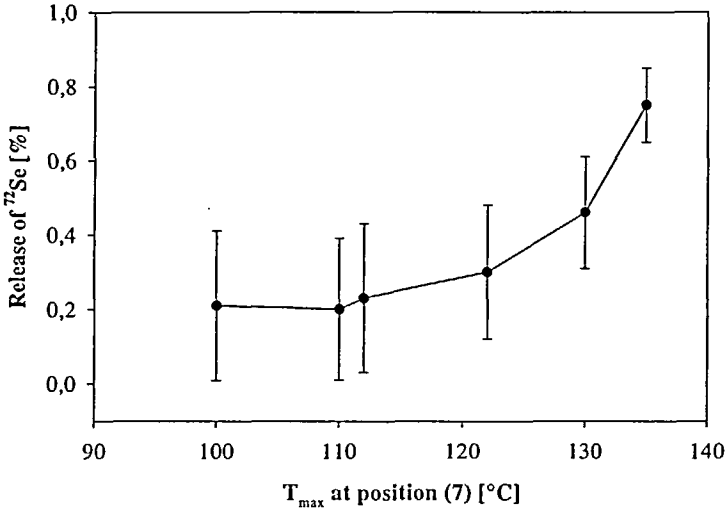

Fig. 2

Release of ^{72}Se depending on the temperature (separation parameters: 1 g KCl, 1 ml conc. HCl, HCl stream of 20 ml/min, t = 10 min)