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 distillation 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(α,n)-, $^{71}$Ga(α,3n)-, $^{71}$Ga($^3$He,2n)-reactions. More interestingly, however, is its availability as the daughter isotope of $^{72}$Se ($T_{1/2} = 8.5$ d). $^{72}$Se itself can be produced via direct processes such as $^{70}$Ge(α,2n)- and $^{72}$Ge($^3$He,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$^6$ adsorbed, while $^{72}$As was eluted 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 hydrazinium hydrochlorid and its filtration with $^{72}$As remaining in solution. Prior to the subsequent separation cycle, Se must be oxidised using $\text{H}_2\text{O}_2$.  

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Production and isolation of $^{72}\text{Se}$

$^{72}\text{Se}$ was produced via the $^{70}\text{Ge}(^{3}\text{He},3\text{n})^{72}\text{Se}$-reaction (FZ Juelich, Germany). To isolate $^{72}\text{Se}$ 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 $^{72}\text{Se}$ (and generated $^{72}\text{As}$) is quantitatively remaining.

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

The HCl solution containing $^{72}\text{Se}$ 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 $^{72}\text{Se}$ compounds and $^{72}\text{As}[\text{AsCl}_3]$ [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 $^{72}\text{Se}$ fraction inside the tube (1) is raised from 50 to 140°C. The $^{72}\text{As}$ 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 $^{72}\text{As}$ is nearly quantitatively desorbed from the cartridge in > 90% yields using < 5 ml of H$_2$O or NaOH and it can be used immediately for labelling reactions.

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

References

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Fig. 1

Scetch of the $^{72}\text{Se}/^{72}\text{As}$ 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-$^{72}\text{Se}$ fraction;
8-upper end of the inner tube;
9-adsorber;
10-electric resistance oven

Fig. 2

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