A no-carrier-added $^{72}Se/^{72}As$ isotope generator

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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 and $E_{\beta+mean}$ = 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 days 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 cromatographic columns, with ^{72}Se as Se^o 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

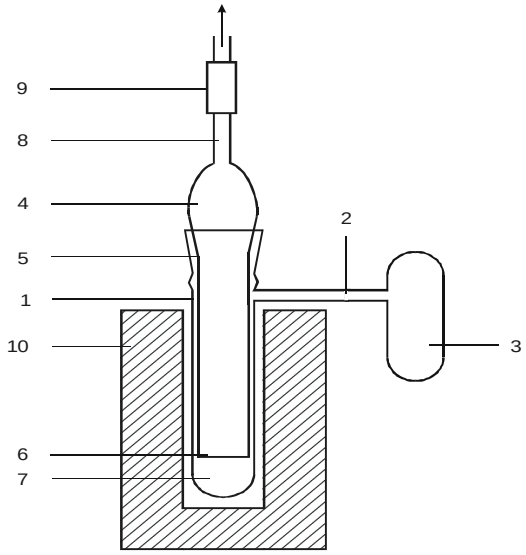
^{72}Se was produced via the $^{nat}Ge(^3He,3n)$ ^{72}Se-reaction (FZ Juelich, Germany).

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 with no-carrier-added ^{72}Se (and generated ^{72}As) 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 pumped 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 – 110°C), > 99.7% of ^{72}Se are still present at position (7) and are ready for the next separation cycle without further treatment.



1-outer quart or glas tube; 2-inlet of HCl; 3-HCl vessel; 4-inner quart or glas tube; 5-ground joint; 6-open lower ende of the inner tube; 7-^{72}Se fraction; 8-upper end of the inner tube; 9-adsorber; 10-electric resistance oven

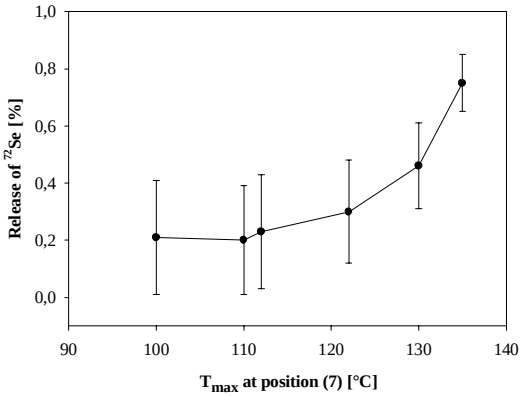


Fig. 2: Release of ^{72}Se depending on the temperature (separation parameters: 1 g KCl, 1 ml conc. HCl, HCl stream of 20 ml/min, t = 10 min)

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3 S.H. A-Kouraishi, G.G.J. Boswell, Int. J. Appl. Rad. Isot. 29 (1978) 607
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Fig. 1: Scetch of the $^{72}Se/^{72}As$ generator apparatus