Production of ^{77}As for potential endoradiotherapeutic application

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For the development of new arsenic based radiopharmaceuticals a reliable logistic supply with radioactive arsenic isotopes is needed. ^{77}As ($T_{1/2}$ = 38.8 h, β^- = 100 %) is a cheap alternative to conventionally used positron emitting isotopes. Due to its low γ-dose and 100 % emission of electrons, it might be also a valuable asset to endoradiotherapy (ERT).

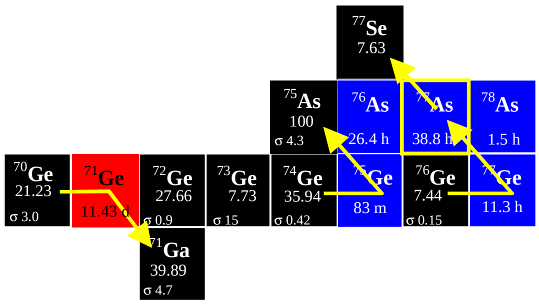

Fig. 1: Production of arsenic isotopes. Extract from the chart of nuclides

^{77}As can easily be produced in nca state at nuclear reactors via (n,γ) reaction on $^{nat}GeO_2$ (see fig. 1). At the TRIGA reactor Mainz, about 2 MBq of ^{77}As are yielded after an irradiation of 100 mg $^{nat}GeO_2$ for 6 h, which is the daily irradiation length maximum. Thus we are interested in the production of ^{77}As at high flux reactors and have investigated the following systems during irradiation mathematically:

System I:

$$^{76}Ge \xrightarrow{(n,\gamma)} {}^{77}Ge \xrightarrow{\beta^-;T_{1/2}\ =\ 11.3h} {}^{77}As$$

$$\xrightarrow{\beta^-;T_{1/2}\ =\ 38.8h} {}^{77}Se$$

general:

$$A \xrightarrow{(n,\gamma)} B \xrightarrow{\lambda_B} C \xrightarrow{\lambda_C} D$$

System II:

$$^{74}Ge \xrightarrow{(n,\gamma)} {}^{75}Ge \xrightarrow{\beta^-;T_{1/2}=83\text{ min}} {}^{75}As$$

System III:

$$^{70}Ge \xrightarrow{(n,\gamma)} {}^{71}Ge \xrightarrow{\varepsilon;T_{1/2}=11.43\text{ d}} {}^{71}Ga$$

general:

$$A \xrightarrow{(n,\gamma)} B \xrightarrow{\lambda_B} C$$

Differential equation system I:

$$A: \frac{dN_A}{dt} = 0$$

$$B: \frac{dN_B}{dt} = -N_B \cdot \lambda_B + \phi \cdot \sigma \cdot N_A$$

$$C: \frac{dN_C}{dt} = -N_C \cdot \lambda_C + \lambda_B \cdot N_B$$

$$D: \frac{dN_D}{dt} = N_C \cdot \lambda_C$$

Solution of the differential equation system I:

$$A: \quad N_A = N_A \qquad A_A = 0$$

$$B: \quad N_B = \frac{N_A \cdot \phi \cdot \sigma}{\lambda_B} \cdot \left(1 - e^{-\lambda_B \cdot t}\right)$$

$$A_B = \lambda_B \cdot N_B = N_A \cdot \phi \cdot \sigma \cdot \left(1 - e^{-\lambda_B \cdot t}\right)$$

$$C: \quad N_C = \frac{N_A \cdot \phi \cdot \sigma}{\lambda_C \cdot (\lambda_B - \lambda_C)} \cdot \left[\lambda_B - e^{-\lambda_C \cdot t} \cdot \lambda_B + \left(e^{-\lambda_B \cdot t} - 1\right) \cdot \lambda_C\right]$$

$$A_C = \lambda_C \cdot N_C = \frac{N_A \cdot \phi \cdot \sigma}{\lambda_B - \lambda_C} \cdot \left[\lambda_B - e^{-\lambda_C \cdot t} \cdot \lambda_B + \left(e^{-\lambda_B \cdot t} - 1\right) \cdot \lambda_C\right]$$

$$D: \quad N_D = \frac{N_A \cdot \phi \cdot \sigma \cdot e^{-(\lambda_B + \lambda_C) \cdot t}}{\lambda_B \cdot \lambda_C (\lambda_C - \lambda_B)} \cdot \left[\begin{matrix} -\lambda_B^2 \cdot e^{\lambda_B \cdot t} + \lambda_C^2 \cdot e^{\lambda_C \cdot t} - e^{(\lambda_B + \lambda_C) \cdot t} \cdot (\lambda_B - \lambda_C) \\ \cdot \left(-\lambda_C - \lambda_B + \lambda_B \cdot \lambda_C \cdot t\right) \end{matrix}\right]$$

$$A_D = 0$$

Differential equation system II / III:

$$A: \frac{dN_A}{dt} = 0$$

$$B: \frac{dN_B}{dt} = -N_B \cdot \lambda_B + \phi \cdot \sigma \cdot N_A$$

$$C: \frac{dN_C}{dt} = N_B \cdot \lambda_B$$

Solution of the differential equation system II / III:

$$A: \quad N_A = N_A \qquad A_A = 0$$

$$B: \quad N_B = \frac{N_A \cdot \phi \cdot \sigma}{\lambda_B} \cdot \left(1 - e^{-\lambda_B \cdot t}\right)$$

$$A_B = \lambda_B \cdot N_B = N_A \cdot \phi \cdot \sigma \cdot \left(1 - e^{-\lambda_B \cdot t}\right)$$

$$C: \quad N_C = \frac{N_A \cdot \phi \cdot \sigma}{\lambda_B} \cdot \left[e^{-\lambda_B \cdot t} + \lambda_B \cdot t - 1\right]$$

$$A_C = 0$$

The calculations show that ^{77}As can be produced in GBq yields at high flux reactors. The use of highly enriched $^{76}GeO_2$ targets would increase the specific activity of ^{77}As significantly and increase the absolute achievable activity by a factor of up to 13 by decreasing the by-products ^{71}Ge and ^{75}Ge.

Reactor	ϕ [$n \cdot cm^{-2} \cdot s^{-1}$]	^{77}Ge [MBq]	^{77}Ge [atoms]	^{77}As [MBq]	^{77}As [atoms]	^{77}Se [atoms]	^{75}Ge [GBq]	^{75}As [atoms]	^{71}Ge [GBq]	^{71}Ga [atoms]
TRIGA	$4.0 \cdot 10^{12}$	25.7	$1.51 \cdot 10^{12}$	21.15	$4.32 \cdot 10^{12}$	$5.27 \cdot 10^{12}$	0.348	$1.48 \cdot 10^{14}$	0.39	$8.8 \cdot 10^{13}$
BER II	$1.0 \cdot 10^{14}$	642	$3.7 \cdot 10^{13}$	536	$1.08 \cdot 10^{14}$	$1.3 \cdot 10^{14}$	8.69	$3.69 \cdot 10^{15}$	9.76	$2.2 \cdot 10^{15}$
FRM II	$4.0 \cdot 10^{14}$	2570	$1.51 \cdot 10^{14}$	2150	$4.32 \cdot 10^{14}$	$5.27 \cdot 10^{14}$	34.8	$1.48 \cdot 10^{16}$	39	$8.8 \cdot 10^{15}$
Dimitrovgrad, SM	$2 \cdot 10^{15}$	12800	$7.54 \cdot 10^{14}$	10700	$2.16 \cdot 10^{15}$	$2.64 \cdot 10^{15}$	174	$7.38 \cdot 10^{16}$	195	$4.4 \cdot 10^{16}$

Tab. 1: Calculated activities and numbers of produced atoms after 120 h of irradiation of 100 mg $^{nat}GeO_2$ at various reactors