Further Developments on a $^{72}Se/^{72}As$ Isotope Generator based on Distillation

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Introduction: Recently a generator system based on the distillation of nca $^{72}AsCl_3$ was developed [1,2]. This generator was now optimized in terms of distillation temperature and time, HCl-flow, radiochemical yield of nca arsenic and retention of the selenic generator charge.
Isotope: To simulate the behaviour of ^{72}Se, ^{75}Se was used, which was produced via (n,γ)-reaction at the nuclear research reactor at the HMI Berlin. To simulate the behaviour of nca ^{72}As, ^{77}As was used, which was produced via (n,γ)-reaction on natural germanium at the TRIGA reactor of the Institute of University of Mainz, giving ^{77}Ge which decays to ^{77}As with a half-life of 11.3 h. The ^{72}Se itself was produced at the Forschungszentrum Juelich via (^{3}He,3n)-reaction on natural germanium at a beam current of 5 µA for 12 h, giving a yield of 5 mCi.
Experimental: For a detailed description of the experimental setup and used materials, see [1,2]. In this work, various salts (Fig.3) and different HCl-flow-rates were investigated (Fig.2). To determine the influence of the HCl-flow-rates, the active coal-cartridge was substituted by a 100 ml glass-syringe.
Results and Discussion:

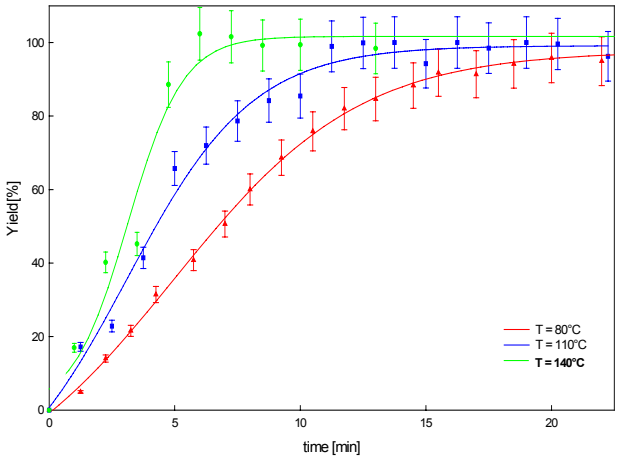

Fig. 1: Distillation kinetics for $^{77}AsCl_3$ at different maximum temperatures applied to the position of ^{72}Se (40 ml/min HCl flow; KCl)

The separation of As/Se is based on the formation of volatile $AsCl_3$ at different temperatures, beginning with 80°C in the presence of alkali halides and other salts (Figs. 1, 3).
The ^{72}As yield increases strongly at a temperature of about 100°C. At a temperature of 100°C, 50 % yield after 6 minutes and 99.9 % yield after 20 minutes were observed. However, a complete oxidation of Se with aqua regia is recommended prior to subsequent generator runs. The Se-retention is 99.9 % after 1 hour running of the generator system.
The effect of HCl flow rate is illustrated in Fig. 2. A triplication of HCl flow rate is followed by an approximate triplication in yield at t=10 min. With a flow rate of 20 ml/min the yield of 100 % can not be achieved.

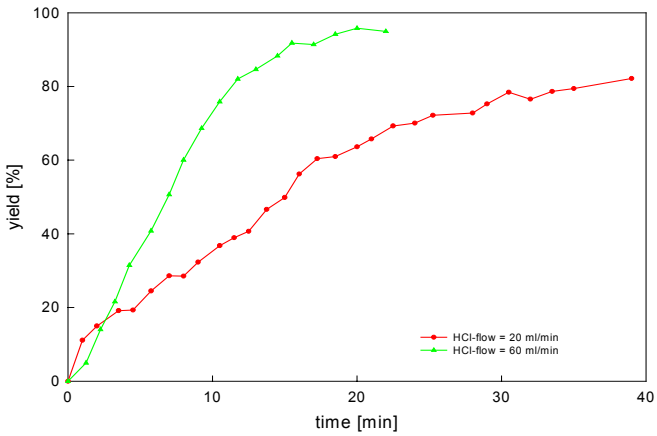

Fig. 2: Distillation kinetics of nca $^{77}AsCl_3$ at different HCl-flow-rates, T=100°C; KCl

The salts indicated in Fig. 3 were added to the ^{72}Se-fraction. Obviously, KCl seems to provide the optimum results.

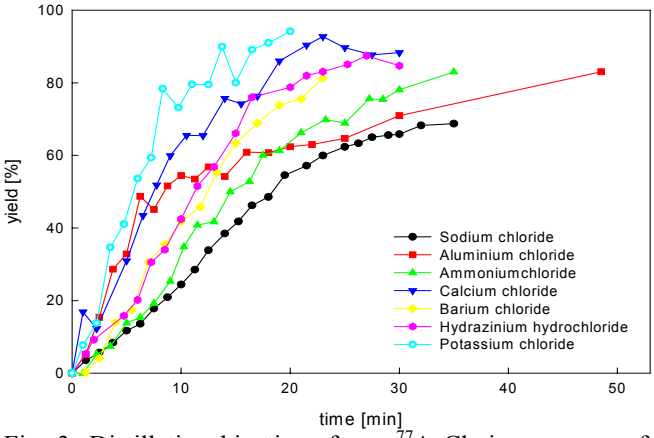

Fig. 3: Distillation kinetics of nca $^{77}AsCl_3$ in presence of various salts

Conclusion: Seven salts have been tested and the originally used KCl was found to be the optimum. It could be shown that high temperatures (T=140°C) give best results in terms of ^{77}As volatilization. It is possible to obtain 90 % yield of radioarsenic in about 5 minutes. During this time less than 0.075 % of selenic are released. A disadvantage of the generator is its redox-instability. The filling has to be transferred to aqua regia and refluxed for 1 hour to completely reoxidize the selenium before each separation. Otherwise the selenium released with the arsenic fraction reaches 20 %.

References:
[1] Novgorodov A.F. et al., Annual Report 2000, Institute for Nuclear Chemistry, University of Mainz
[2] Novgorodov A.F. et al., J. Labelled Comp. Radiopharm., **44** S1, 2001