# Chromatographic Separation of Selenium and Arsenic: A Potential 72Se/72As Generator

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## Summary

An anion exchange method was developed to separate selenium and arsenic for potential utility in a $^{72}$Se/$^{72}$As generator. The separation of the daughter $^{72}$As from the $^{72}$Se parent is based on the relative acid-base behavior of the two oxo-anions in their highest oxidation states. At pH 1.5, selenate is retained on strongly basic anion exchange resin as $HSeO_4^-$ and $SeO_4^{2-}$, while neutral arsenic acid, $H_3AsO_4$, is eluted.

## Keywords

As-72; Se-72; radionuclide generator; ion exchange

## 1. Introduction

Positron emission tomography (PET) has become a routine nuclear medicine imaging technique with the approval of 2-deoxy-($^{18}$F)-fluoroglucose ($^{18}$F-FDG). PET imaging allows for better resolution nuclear medicine images than single photon emission computed tomography (SPECT) and allows for quantitation. Several positron emitting radionuclides are currently available but most are short-lived (e.g., $t_{1/2}$ = 110 min for $^{18}$F, $t_{1/2}$ = 20 min for $^{11}$C, $t_{1/2}$ = 12.7 h for $^{64}$Cu) [1]. Longer-lived positron emitting radionuclides would allow the imaging of biological processes that require longer localization times such as radiolabeled antibodies. Zirconium-89 is being developed as a longer-lived, metallic positron emitter ($t_{1/2}$ = 3.27 d), and enhanced chelate development is currently underway [2]. Iodine-124 is a long-lived, non-metallic, positron emitter ($t_{1/2}$ = 4.176 d) being developed for molecular imaging applications, but minimizing radioiodine impurities and

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maximizing [124]I production yield during either proton or deuteron irradiation remains a trade-off [3–6]. An alternative non-metallic, positron emitting radionuclide is [72]As ($t_{1/2} = 26.0$ h, 88%, $\beta^+$, 2.49 MeV endpoint energy, 12% EC), which is available from the decay of [72]Se ($t_{1/2} = 8.4$ d, EC, 46 keV $\gamma$ (58% abundant)) and makes this parent-daughter pair suitable for development of a [72]Se/[72]As generator [7]. Compared to most PET radionuclides, the longer half-life of [72]As allows for attachment to antibodies and proteins for quantitative imaging of biochemical and physiological processes and receptor mapping [8–12]. The chemistry of arsenic is diverse (oxidation states of 0, +3 and +5) allowing for the development of potentially valuable PET radiopharmaceuticals. Direct labeling of the monoclonal antibody Bavituximab with [72]As for tumor-specific PET imaging has been reported [13].

Selenium-72, the parent of [72]As, can be produced from charged particle reactions on Ge, Br, or As targets [14]. The most common production routes involve $\alpha$ or [3]He irradiation of [70]Ge, [72]Ge, or [nat]Ge, generally as the oxide (e.g., [70]Ge($\alpha$,2n)[72]Se or [70]Ge([3]He,n)[72]Se), or a proton induced nuclear reaction on stable arsenic ([75]As(p,5n)[72]Se) [15–18]. Cross-section data suggest that these are the best production methods [15,19,20]. Alternatively natural NaBr targets irradiated with higher energy protons (72–92 MeV) generate Se radioisotopes ([72]Se, [73]Se, ^{75}Se) with fewer by-products than higher energy reactions [7,21,22]. The higher beam currents available (average of 101.4 $\mu$A) at the Los Alamos National Laboratory Isotope Production Facility (LANL-IPF) make this a viable route although radioselenium impurities are generated [22]. Any of these routes will produce [72]Se for generator development, with the [72]Se generally separated from its target as Se(0) [15,17] or Se(IV) [16,18,20].

Development of a column generator for the separation of [72]As from [72]Se, similar to the Mo-99 alumina column generator for [99m]Tc (e.g., Ultra-TechneKow®) or the Sr-82 stannic oxide column for [82]Rb (e.g., Cardiogen-82®) would be advantageous. Selenium-72/Arsenic-72 ([72]Se/[72]As) radionuclide generators have been proposed in the literature [14], however complex separation/isolation methods are required. Separation of the daughter from the parent can occur via repeated distillation of "grown-in" [72]AsCl$_3$ from carrier added [72]Se stock solutions [16,21]; electroplating of [72]Se as Cu$_2$Se on Cu backings [23], solvent extraction [22] or solid phase extraction of [72]Se [17,24]. However, with the exception of the solid phase extraction method, the reported methods require laborious and/or significant handling. The solid phase approach as reported in the literature is limited to very small dimensions (500 $\mu$L cartridge volume, few $\mu$Ci of [72]Se activity) involving reagents such as HF, requires Se in its elemental state under an inert atmosphere to prevent oxidation, and scale up to clinically useful 10 mCi batches may not be feasible [17,22,24].

Extraction chromatographic methods, including batch distribution ratio methods and column chromatography, have provided simple, robust methods for the separation of parent/daughter radionuclide pairs [25–27] including the development of a higher activity ^{137}Cs/[137m]Ba generator system for industrial and environmental applications [27]. Using several radioisotopes of arsenic and selenium, an anion exchange method was developed for the separation of As from Se with each element in its highest oxidation state, namely as selenate

(Se(VI)$O_4^{2-}$) and arsenic acid (H$_3$As(V)O$_4$). A low activity generator was prepared and evaluated.

## 2. Experimental

*Caution!* Selenium-75 ($^{75}$Se), $^{72}$Se, $^{72,73,74,76}$As, and $^{22}$Na are radioactive and all work involving these radionuclides were carried out in approved laboratories following the appropriate radiation safety procedures. Selenium-75 ($t_{1/2}$ = 119.78 d) used as a Se tracer in column and batch studies was produced at the University of Missouri Research Reactor (MURR) via the $^{74}$Se (n, $\gamma$) $^{75}$Se reaction of an encapsulated $^{74}$Se (as SeO$_2$; 98.85%) target, which was dissolved in 1 M HNO$_3$. Arsenic-76 ($t_{1/2}$ = 1.0942 d) used as an As tracer in batch studies was produced at the MURR via the $^{75}$As (n, $\gamma$)$^{76}$As reaction of an encapsulated $^{75}$As (as $^{nat}$As$_2$O$_3$) target, which was dissolved in 1 M NaHCO$_3$. A mixture of radioactive selenium ($^{72,75}$Se), arsenic ($^{72,73,74}$As), and sodium ($^{22}$Na) in 0.1 M HCl and ~3 M NaCl (from processing) [22], produced by the irradiation of natural NaBr targets with 72–92 MeV protons at the Los Alamos National Laboratory – Isotope Production Facility (LANL-IPF), was evaluated for producing a $^{72}$Se/$^{72}$As generator. On receipt, the sample contained 19.34 μCi of $^{22}$Na, 2.8 μCi of $^{72}$As (with $^{72}$Se in equilibrium), 62.51 μCi of $^{73}$As, 56.26 μCi of $^{74}$As, and 35.24 μCi of $^{75}$Se as determined by gamma spectroscopy (HPGe). Table 1 lists the radionuclides produced at LANL during irradiation of the NaBr target, their half-lives in days, the initial activities measured at LANL, and the activities measured at the University of Missouri after processing and receipt.

Table 2 lists the various radionuclides, their nuclear properties, and their specific activities as used in these studies.

### 2.1 Materials

Reagents and solvents were purchased from Alfa Aesar (Ward Hill, MA), Fisher Scientific (Pittsburg, PA), Mallinckrodt (St. Louis, MO), and Sigma-Aldrich (St. Louis, MO) and used as received. Dowex anion exchange resins were obtained from Bio-Rad Corporation (Hercules, CA) and Sigma-Aldrich (St. Louis, MO). Alumina basic form (Sigma Aldrich), alumina neutral form (Sigma-Aldrich), alumina acidic form (Fisher Scientific), silica gel 60 (Brinkman, Des Plaines, IL), magnesium oxide (Mallinkrodt), Florisil (Fisher Scientific), and zirconium oxide (Alfa Aesar) were evaluated. All water used was purified on-site (deionized water from a Millipore system to >18MΩcm).

### 2.2 Instrumental analysis

Ion pairing HPLC analysis was performed using a Perkin-Elmer Model 410 LC pump with controller and Turbochrome software equipped with a Rainin Dynamax UV-1 variable UV-visible detector. An analytical reversed phase C18 column (Alltech Hypersil, 120 Å, 0.46 cm × 25 cm × 5 μm) with an isocratic mobile phase of 20:80 acetonitrile: phosphate buffer (60 mM, pH 7.2, 20 mM tetrabutylammonium hydroxide as the ion pair agent) at a flow rate of 1 mL/min and UV detection at 210 nm was utilized for HPLC. Retention times for selenite, arsenate and selenate were 3.1, 3.4 and 3.9 minutes, respectively.

Radiochemical assays for samples containing solely ^{75}Se were determined by gamma counting using a Harshaw integral line NaI(Tl) solid scintillation well detector with a Canberra HV power supply and Ortec preamp.

Radiochemical assays for [22]Na, [72, 73,74]As, and ^{75}Se were performed by γ-ray spectroscopy using a Canberra Model GC2018S HPGe detector system. The detector diameter was 60.5 mm, detector length was 30.5 mm, and the distance from the window was 5 mm. The detector's specified FWHM at 1.33 MeV was 1.8 keV. Spectral analyses were performed by a Canberra Model 9600 multichannel analyzer.4

### 2.3 Determination of optimum chromatographic media

For the development of a [72]Se/[72]As generator, various support media were evaluated to elute arsenate while retaining selenate. Silica gel, acidic alumina, neutral alumina, basic alumina, Florisil, zirconium oxide, magnesium oxide, Dowex 1-X8 and Dowex 1-X2 were tested for their ability to retain selenate while allowing arsenate to elute under neutral to slightly acidic conditions (Table 3). Columns were separately loaded with non-radioactive sodium selenate and sodium arsenate dibasic heptahydrate dissolved in water, and were then evaluated for ion retention by elution with DI $H_2O$ and either dilute HCl (<1 M) or dilute $HNO_3$ (<1 M). The collected elution fractions were evaluated for selenate and arsenate using ion pairing reversed phase HPLC analysis described above.

### 2.4 Optimization of elution parameters

To optimize the elution parameters on Dowex anion exchange resin, sorption studies were performed via the batch method for selenite, selenate, and arsenate. Prior to use, samples were verified to contain purely selenate, selenite, or arsenate by comparison to standards using HPLC. All experiments were performed at room temperature (20°C). The removal of selenium and arsenic from varying pH (−1 to 7) solutions was measured by mixing 50 mg of AG 1-X8 resin with 1.5 mL of a [75]Se- or [76]As-spiked solution. The selenium tracer was 117 kBq (3.16 μCi)/sample and the arsenic tracer was 146 kBq (3.95 μCi)/sample. The liquid-solid system was mixed by vortexing for 2 minutes and immediately centrifuged for 2 minutes at 7500 rpm. Two 500-microliter aliquots of the supernatant ($A_s$) were transferred into clean counting vials. Additionally, a 500-microliter aliquot of the original solution ($A_0$) was transferred to a clean counting vial to determine the original activity. The distribution ratio, $K_d$, is calculated as follows:

$$K_d = \frac{(A_0 - A_s)}{A_s} * \frac{V}{m}$$

$A_0$ is the original selenium or arsenic activity of the aqueous solution. $A_s$ is the selenium or arsenic activity remaining in solution following contact with the resin. ($A_0 − A_s$) is the amount of activity adsorbed by the resin. The volume (V) of the aqueous solution is measured in milliliters and the mass (m) is measured in grams resulting in the distribution ratio having units of mL/g. A larger distribution ratio correlates to a more effective removal of selenate, selenite, or arsenate from the aqueous solution.

### 2.5 Oxidation of Se(IV) to Se(VI)

An oxidation method was developed to treat the samples to ensure that selenium was present as Se(VI). The optimum method was as follows: samples of ^{75}Se were treated using 1 mL of 30% $H_2O_2$ and refluxed for 1 hour. Each sample was quantitatively loaded onto a 1 mL bed volume of Dowex AG1-X8 (100–200 mesh) column and washed with DI $H_2O$ (4 × 1 mL), 0.03 N $HNO_3$ (4 × 1 mL), and 0.6 N $HNO_3$ (4 × 1 mL) with all fractions collected and counted using the NaI(Tl) well detector. Selenous acid ($H_2SeO_3$) eluted with 0.3 N $HNO_3$ and selenic acid ($H_2SeO_4$) eluted with 0.6 N $HNO_3$, allowing determination of Se oxidation state present.

The LANL-IPF mixture (222 kBq; 6 μCi/sample) was evaluated by this technique to determine the Se oxidation state. Treated and untreated control samples were eluted from Dowex columns in the manner described above and the fractions were counted on the HPGe detector, and evaluated for [22]Na, [73]As, and ^{75}Se content (Table 1).

### 2.6 Application of method to Se/As generator column

A large, preparative column (2.2 cm i.d. × 12.5 cm) packed with a 48 mL bed volume of Dowex 1-X2 (50–100 mesh) resin was conditioned with 3 M HCl followed by DI $H_2O$ (~10 bed volumes). Approximately 11.1 MBq (300 μCi) of the LANL-IPF sample was neutralized, oxidized and diluted (total volume = 300 mL), and then added to this pre-concentrating column followed by DI $H_2O$ washes (2 × 50 mL). Arsenate was eluted from the column with 0.03 N $HNO_3$ (6 × 50 mL) followed by DI $H_2O$ (2 × 50 mL). Selenate was then eluted from the column with 0.6 N $HNO_3$ (6 × 50 mL) followed by DI $H_2O$ (2 × 50 mL). The 2nd and 3rd fractions of the 0.6 N $HNO_3$ elution (containing the majority of the selenate) were combined, the volume slowly reduced to 0.1 mL by evaporation in a hot water bath under air flow, and then neutralized (1 M $NaHCO_3$), oxidized (30% $H_2O_2$), and diluted with DI $H_2O$ to 32 mL. This sample was loaded onto a generator Dowex 1-X2 column (50–100 mesh; 4 mL wet resin volume; 0.7 cm i.d. × 10 cm; conditioned as above), washed with DI $H_2O$ (10 mL), and then eluted after 1, 3, and 8 days with 0.03 N $HNO_3$ (5 × 5 mL) and DI $H_2O$ (2 × 10 mL). The fractions were counted on the HPGe to analyze for [72,73,74]As and ^{75}Se content.

### 2.7 Effect of radiation on Dowex resin

With the anticipated, large radiation dose to the resin expected from a [72]Se/[72]As generator, the Dowex 1-X8 resin was exposed to 2880 Gy (288 MRad) over 2 weeks using a ^{60}Co source. The elution profile of two columns (0.6 cm i.d. × 4.2 cm; 1.2 mL wet volume), one containing the exposed resin and one the control unexposed resin, were prepared as described above to evaluate any difference in breakthrough of [75]Se. The same activity of [75]Se (1850 Bq (50 nCi); 0.24 ng; 0.206 Ci/mg) was applied to each column, with ^{75}Se used as radiotracer for [72]Se for easier counting.

## 3. Results and discussion

Several methods have been reported to separate [72]As from [72]Se, none are straight-forward and most require extractions or harsh chemical treatments (e.g., HF) to generate [72]As in the

+3 oxidation state, which is readily oxidized at the tracer level to the +5 oxidation state. To this end, an anion exchange method has been developed that takes advantage of the difference in acid strength between selenic acid ($H_2SeO_4$) and arsenic acid ($H_3AsO_4$) with the $^{72}As$ in its +5 oxidation state.

### 3.1 Determination of optimum chromatographic media

Several chromatographic materials were evaluated for their ability to retain selenate and elute arsenate, with Dowex anion exchange resins (1X-8 and 1X-2) determined to be the best suited (Table 2). At pH = 1.5 the arsenate ion is nearly fully protonated and is eluted from the anion exchange resin while the selenate ion retains a negative charge and is not readily displaced. None of the hydrous metal oxides evaluated gave a suitable separation as they retained arsenate and eluted selenate or both species were similarly retained.

### 3.2 Optimization of elution parameters

To determine the best pH for selenate retention and arsenate elution, equilibrium distribution ratios were acquired as a function of pH. The retention of selenate, selenite, and arsenate was impeded by the high ionic strength of the pH −1 and 0 solutions. Since selenic acid is a strong acid, selenate was readily retained by the resin and quickly reached its maximum $K_d$. Maximum $K_d$ is a function of the minimal detectable activity of the detector and the activity of the spike; the maximum $K_d$ in this experiment is 3570 mL/g. Since selenous and arsenic acids are weaker acids, selenite ($pK_{a1} = 2.62$) and arsenate ($pK_{a1} = 2.19$) are weakly retained until reaching a solution pH where the anionic species is the primary form. The several orders of magnitude difference in the retention of selenite and selenate highlights the necessity of an oxidation step to prevent loss of selenium from the column. Additionally, the results indicate eluting at pH 2 allows the parent, selenate, to be retained on the column while the arsenate daughter is removed (Figure 1).

### 3.3 Oxidation of Se(IV) to Se(VI)

The first $pK_a$ value of selenous acid ($H_2SeO_3$) is close to that of arsenic acid ($H_3AsO_4$) (2.62 and 2.19, respectively), which would make their separation very difficult as shown in Figure 1. Thus, a method was developed to ensure that all of the selenium was present as selenic acid to afford a separation from the weak arsenic acid. An ion pairing reversed phase HPLC method was used to determine the speciation of selenate, selenite and arsenate present and to optimize the conditions for the oxidation of Se(IV) to Se(VI). Refluxing 30% $H_2O_2$ was found to completely convert selenite to selenate. Variations in pH had no effect on the conversion or elution.

The LANL-IPF sample, which contained radioisotopes of Se, As and Na, and had been oxidized with hydrogen peroxide/HCl to remove bromide isotopes as $Br_2$ prior to shipping to the University of Missouri Research Reactor (MURR), required additional treatment with refluxing 30% $H_2O_2$ to convert all of the selenium to selenate. Either the original oxidation was insufficient to convert the selenium to Se(VI) or over time the selenium originally converted was reduced to a range of lower oxidation states (Figure 2). The sample contained principally $^{22}Na$, $^{73}As$, $^{74}As$ and $^{75}Se$ as observed by HPGe gamma spectroscopy with $^{75}Se$ in the greatest abundance. At low levels of $^{72}Se$ activity the $^{72}Se$ gamma signal is not readily

quantitated from the high Compton background at 45 keV. Chromatographic separation of the non-oxidized sample showed selenium in at least three different forms: an uncharged or cationic species eluting with water, selenite eluting with 0.03 N $HNO_3$ and selenate eluting with 0.6 N $HNO_3$. The arsenic present eluted with water as an uncharged or cationic species and the sodium also eluted with water.

Treatment with refluxing 30% $H_2O_2$ oxidized the arsenic to arsenate, and the selenium to selenate. No lower oxidation state As or Se species were observed (Figure 3).

### 3.4 Application of method to Se/As generator column

To examine the practical application of the separation method to a generator preparation, a portion of the neutralized, oxidized, diluted LANL-IPF sample was pre-concentrated to reduce the volume and remove Na and As radioisotopes produced during irradiation with 72–92 MeV protons. A portion of the neutralized, oxidized, diluted LANL-IPF sample was directly added to a preparative column; the eluent resulting from the addition yielded 481 kBq (13 μCi) of [22]Na and trace amounts of [74]As. No selenium was detected during the addition. Elution with 0.03 N $HNO_3$ removed approximately 24 μCi of [74]As and 41 μCi of [73]As and a much smaller amount of [72]As. Again no Se was detected. Elution with 0.6 N $HNO_3$ yielded over 1.61 MBq (43.5 μCi) of ^{75}Se and trace quantities of [72,73,74]As. From the concentrated fractions, more than 97 % of the ^{75}Se was transferred to the 4 mL (bed volume) generator column. No radioisotopes were detected in the resulting eluent from addition of the sample to the generator column. Results from elution with 0.03 N $HNO_3$ are illustrated in Figure 4. After the first elution, which removed the residual arsenic isotopes, [72]As was the only arsenic radioisotope eluted. The activity of the LANL sample was measured by HPGe to contain 35.2 μCi/mL of ^{75}Se and 2.8 μCi/mL of [72]As (and also [72]Se in equilibrium) on 08/23/2010. These activities were decay corrected to the expected activities during the generator column experiment, 26.4 μCi/mL of ^{75}Se and 0.045 μCi of [72]As ([72]Se). Note the significantly higher activity for ^{75}Se over [72]Se at this time. The final elution of the generator column yielded 50 nCi of [72]As and 1.5 nCi of [75]Se. The expected [72]Se breakthrough was determined from the [75]Se:[72]Se ratio present in the sample at this time. Only 0.0026 nCi of [72]Se would have co-eluted with the [72]As, which represented 0.005% of the [72]Se activity on the column.

### 3.5 Effect of radiation on Dowex resin

Additionally, a test was performed to assess possible changes in selenium breakthrough upon exposure of the Dowex anion exchange resin to gamma radiation. After irradiation with 2880 Gy (288 MRads) cumulative dose over two weeks using a ^{60}Co source, the resin was darker in color and did not wet as well as the untreated resin. The column prepared from the irradiated resin appeared to have a slightly higher flow rate than the column prepared from the original resin (visually observed; not measured). Breakthrough of Se(VI) occurred on day 16 (elution 12) for the ^{60}Co irradiated resin, and on day 23 (elution 16) for the untreated resin. The column capacity to retain Se(VI) was only marginally affected by the strong extended gamma radiation conditions. The complete breakthrough curves are shown in Figure 5.

## 4. Conclusions

The development of new drugs utilizing longer-lived radionuclides will allow for the continued growth and increased application of PET imaging for quantitation, especially radioimmunoimaging. Arsenic-72, a long-lived, non-metallic positron emitter, is best produced in high specific activity from the decay of its $^{72}$Se parent. A $^{72}$Se/$^{72}$As generator system was developed and will greatly increase the availability and application of $^{72}$As. Dowex 1-X2 and 1-X8 resins retained selenate while allowing no-carrier-added $^{72}$As arsenate to be eluted.

Distribution coefficient studies showed selenite has over an order of magnitude weaker retention on Dowex 1-X8 than selenate, which would lead to undesired $^{72}$Se breakthrough and product loss. A simple oxidation method was developed to ensure selenium is completely in the selenate form.

A $^{72}$Se/$^{72}$As generator, prepared from an irradiated NaBr target, demonstrated the production and elution of $^{72}$As from the decay of $^{72}$Se. It was further demonstrated that Dowex 1-X8 held up relatively well to radiation damage, which should allow for elutions to be performed over a longer time period without $^{72}$Se breakthrough due to radiolysis.

Future work will include preparation and evaluation of a $^{72}$Se/$^{72}$As generator containing higher activities similar to those used in a clinical setting. This would allow determination of radiation resistance of the polystyrene-divinylbenzene copolymer backbone of Dowex AG 1-X8 or 1-X2 resins or whether other resin backbones are needed. A $^{72}$Se/$^{72}$As generator would make $^{72}$As available as a tracer for medical, toxicological and environmental studies.

## Acknowledgments

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**Highlights**

- Ion exchange separation for $^{72}$Se and $^{72}$As developed.

- Oxidation method to Se(VI) for column loading developed.

- $H_3AsO_4$ eluted at pH 1.5 while $SeO_4^{2-}/HSeO_4^-$ retained.

- $K_d$ values reported for As(V), Se(IV) and Se(VI).

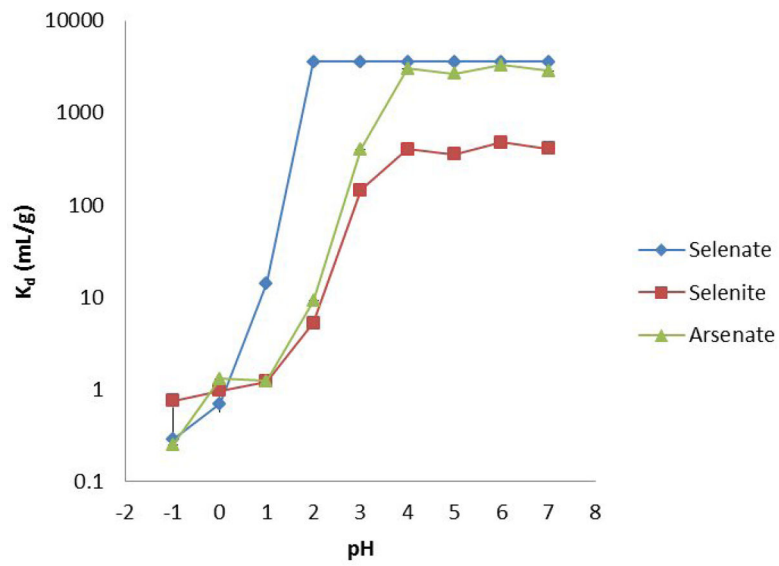- Resin gamma irradiation has minimal effects on Se breakthrough.

**Figure 1.**
Distribution coefficient for selenate, selenite, and arsenate as a function of pH (n = 6; the error bars are included).
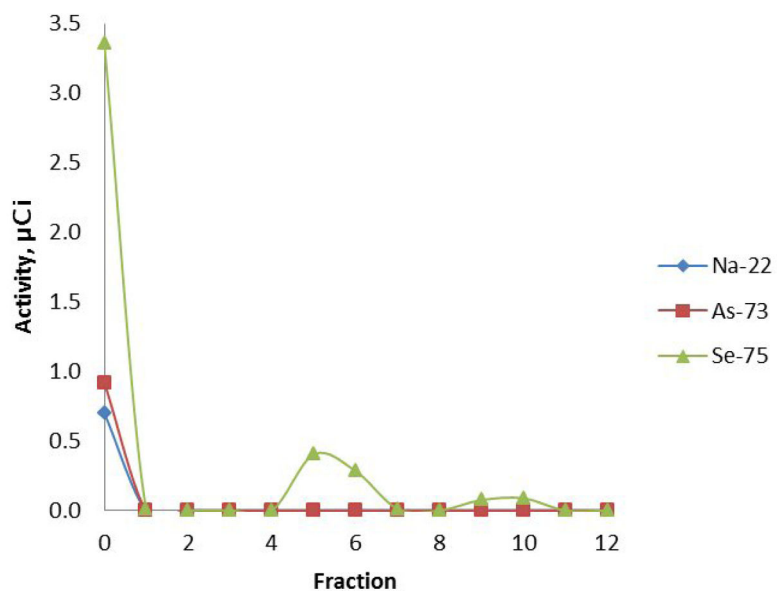
**Figure 2.**
Un-oxidized aliquot of the LANL-IPF sample evaluated on Dowex 1X-8 (100–200 mesh). Eluent of fractions: 0 = sample addition; 1–4 = water; 5–8 = 0.03 M $HNO_3$ (pH 1.5); 9–12 = 0.6 M $HNO_3$ (pH 0.2). Selenium is present in three different chemical forms: Se(0) in fraction 0; selenite in fractions 5–6; selenate in fractions 9–11.
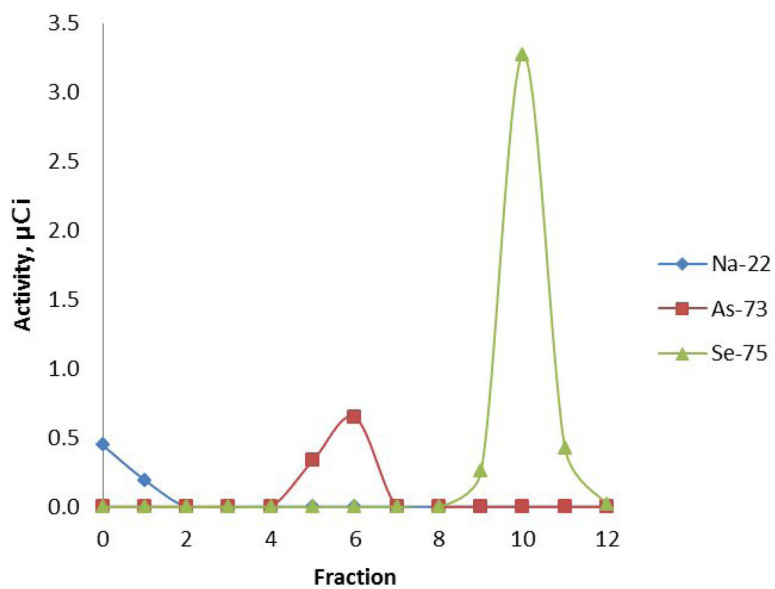
**Figure 3.**
Oxidation of LANL-IPF sample with 30% $H_2O_2$ evaluated on Dowex 1X-8 (100–200 mesh). Eluent of fractions: 0 = sample addition; 1–4 = water; 5–8 = 0.03 M $HNO_3$ (pH 1.5); 9–12 = 0.6 M $HNO_3$ (pH 0.2). Arsenate elutes in fractions 5 and 6, while selenate elutes in fractions 9–11.
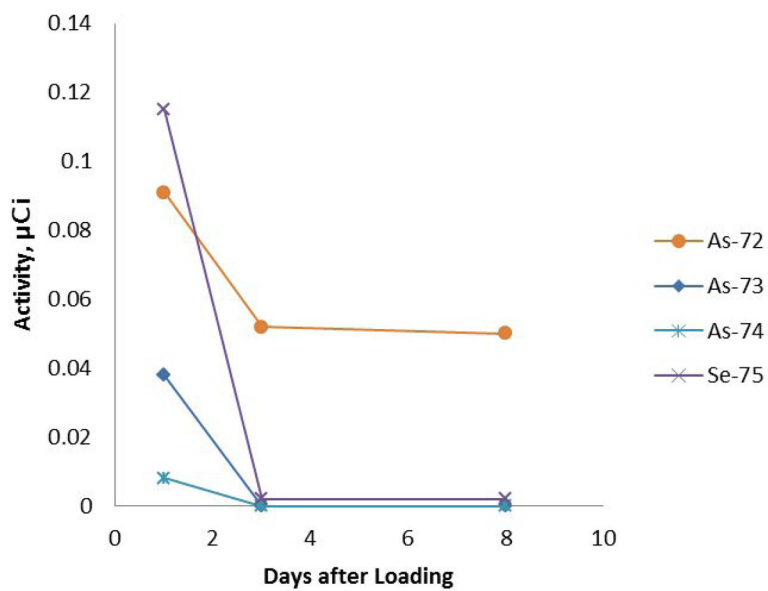
**Figure 4.**
Elution profile of generator column. Each elution included 0.03 M $HNO_3$($4 \times 5$ mL) followed by a water rinse ($2 \times 5$ mL).
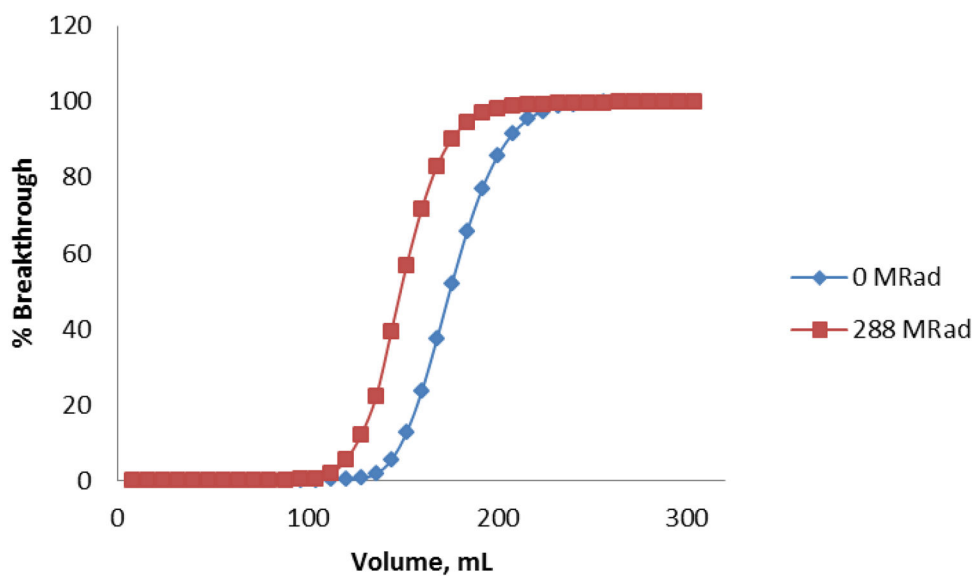
**Figure 5.**
Se(VI) breakthrough comparison of irradiated and non-irradiated Dowex 1×8 resins. The columns were eluted with 0.03 M $HNO_3$ (4 × 1 mL) followed with a water rinse (4 × 1 mL).

**Table 1**

Activities of sample from LANL.

| Nuclide | Literature[28] $t_{1/2}$ (days) | Measured Initial Activity at LANL on 07/24/2011[22] | Measured Activity at MURR on 08/23/2011 |
|---|---|---|---|
| [22]Na | 950.5 | Not determined | 19.34 µCi/mL |
| [71]As | 2.72 | 0.19 ± 0.01 Ci | Decayed |
| [72]As | 1.08 | Not determined | 2.8 µCi/mL |
| [73]As | 80.3 | [a] Not determined | 62.51 µCi/mL |
| [74]As | 17.77 | 0.046 ±0.001 Ci | 56.26 µCi/mL |
| [72]Se | 8.4 | 0.03 ± 0.01 Ci | [b] Not determined |
| [75]Se | 119.78 | 0.050 ± 0.002 Ci | 35.24 µCi/mL |
| [76]Br | 0.68 | 8.1 ± 0.7 Ci | [c] Not present |
| [77]Br | 2.38 | 4.2 ± 0.6 Ci | [c] Not present |
| [82]Br | 1.47 | 0.27 ± 0.06 Ci | [c] Not present |

[a] Accurate measurement of the low energy (56 keV) was impossible with the high activities present.

[b] Accurate measurement of the low activity, low energy (46 keV) gamma was not possible.

[c] Br isotopes had been removed prior to our receipt.

**Table 2**

Evaluated sodium, arsenic, and selenium isotopes and their associated gammas [28].

| Isotope | Half-Life | Gamma Emissions (Abundances) | Specific Activity |
|---------|-----------|------------------------------|-------------------|
| $^{22}$Na | 2.6027 a | 511 (180%), 1274.5 keV (99.9%) | 1.65 MBq/mg ($4.5 \times 10^{-5}$ Ci/mg)[*] |
| $^{72}$As | 26.0 h | 511 (176%), 834.0 keV (81.0%) | nca (6.18 GBq/mg; 1670 Ci/mg) |
| $^{73}$As | 80.30 d | 53.4 keV (10.3%) | nca (825 MBq/mg; 22.3 Ci/mg) |
| $^{74}$As | 17.77 d | 511 (58%), 595.8 keV (59.0%) | nca (3.67 GBq/mg; 99.3 Ci/mg) |
| $^{76}$As | 26.3 h | 559.1 keV (45.0%) | 5.74 MBq/mg (0.155 mCi/mg) at EOB[*] |
| $^{72}$Se | 8.4 d | 45.89 keV (57.2%) | nca (7.99 GBq/mg; 216 Ci/mg) |
| $^{75}$Se | 119.78 d | 121 (17%), 136.0 keV (58.3%) 264 (59%), 279 (25%), 400 (11%) | nca in LANL sample 9.36 MBq/mg (0.253 Ci/mg)[*] |

nca = no carrier added; specific activities calculated.

[*] Specific activities determined from target mass irradiated.

**Table 3**

Column and eluent conditions evaluated.

| Matrix (volume) | Eluent | Arsenate | Selenate |
|---|---|---|---|
| Silica (1.5 mL) | Water or 0.12 M HCl | Elutes rapidly | Elutes rapidly |
| Acidic Alumina (1.5 mL) | Water, neutral saline, 0.12 M or 0.24 M HCl | Did not elute | Did not elute |
| Neutral Alumina (1.5 mL) | Water or normal saline | Did not elute | Elutes |
| Basic Alumina (1.5 mL) | Water, normal saline, 0.12 M or 0.24 M HCl | Did not elute | Elutes |
| Florisil (1.5 mL) | Water, normal saline, 0.012 M or 0.12 M HCl | Elutes | Elutes |
| Zirconium Oxide[*] (0.5 mL) | 0.001 M, 0.01 M or 0.1 M $HNO_3$ | Elutes | Elutes |
| Magnesium Oxide[*] (0.5 mL) | 0.012 M HCl | Elutes | Elutes |
| Dowex 1-X2 or X8 (1.5 mL) | Water or normal saline | Did not elute | Did not elute |
|  | 0.012 M or 0.12 M HCl | Elutes | Did not elute |
|  | 1.2 M HCl | Elutes | Begins to elute very slowly |

[*] Fine powders; smaller volumes for reasonable flow rates.