ANL/CSE-13/34

Design of Column Separation Processes for Recovery of Molybdenum from Dissolved High-Density LEU Target—Further Studies

Chemical Science and Engineering Division

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Design of Column Separation Processes for Recovery of Molybdenum from Dissolved High-Density LEU Target—Further Studies

by
Dominique C. Stepinski, Elizabeth O. Krahn, Pei-Lun Chung, George F. Vandegrift
Chemical Science and Engineering Division, Argonne National Laboratory

prepared for
U.S. Department of Energy, National Nuclear Security Administration,
Office of Defense Nuclear Nonproliferation

March 28, 2012

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DESIGN OF COLUMN SEPARATION PROCESSES FOR RECOVERY OF MOLYBDENUM FROM DISSOLVED HIGH-DENSITY LEU TARGET

1 INTRODUCTION

The Global Threat Reduction Initiative (GTRI) Conversion Program develops technology necessary to convert civilian facilities that use high enriched uranium (HEU) to low enriched uranium (LEU) fuels and targets. The conversion of conventional HEU dispersion targets to LEU for the production of ^{99}Mo production requires approximately five times the uranium in a target to maintain the ^{99}Mo yield per target. Under GTRI, Argonne National Laboratory (Argonne) is developing two frontend options for the current processes to allow the use of LEU-foil targets. In both processes, the aim is to produce a product after the frontend that will be compatible with current purification operations and that will, with the same number of targets irradiated, provide the same or a higher yield of ^{99}Mo at the end of processing. The goal of the frontend process is to deliver a product solution that is of the same or higher Mo purity than the current solution and is of equal or better compatibility with current purification process. In the first frontend option, the LEU foil (contained in a thin [10–15 µm] Ni fission-recoil barrier) is removed from the annular target and dissolved in nitric acid. In the dissolution, the uranium, nickel, and all fission and activation products are dissolved; the solution will be ~7 mM Mo and ~450 g-U/L, and the nitric-acid concentration after dissolution will be ~1 M. The resultant solution will then be run through a column to recover Mo from other target components; the Mo will be stripped from the column with a sodium hydroxide solution that will be compatible with the current purification processes used for dispersion targets. The volume of dissolved-foil solution is assumed to be 500 mL, and the pressure drop, ΔP, constraint of 0.8 atm is based on the use of vacuum/gravity fluid transfer system. Therefore pressure loss is limited to <1 atm; hence a value of 0.8 was chosen.

In the previous report, isotherm parameters, column, intraparticle diffusivity, and column designs were determined for the Sachtopore, S80, sorbent using VERSE simulation program (Stepinski et al. 2011). It was shown that column designs with a loading velocity of 5 cm/min were most efficient in terms of mass of sorbent versus loading time. Therefore, column designs at 5 cm/min were tested experimentally and were shown to load 98% of Mo. Recovery of Mo with 0.1-M NaOH was achieved by counter-current stripping at 3 and 5 cm/min. Mo recovery ranged between 88% and 105% ± 5%, with an average of 95.5% ± 5%.

A full-scale design was chosen based on target total Mo-recovery-process time of ~1 h. The design employs a 2.5 × 8 cm column loaded at 5 cm/min. In this process (Table 1), 500 mL of solution is loaded in 20.4 min. Subsequently, the column is washed with 10 bed volumes (BV) (393 mL) of 1-M HNO_3 for 8 min at 10 cm/min. This step is followed by a water wash, using 196 mL of water loaded for 4 min at 10 cm/min. Finally, Mo is stripped from the column using 16 BV (629 mL) of 0.1-M NaOH at 5 cm/min for 25.6 min. This design was tested experimentally using 500 mL of 450 g-U/L uranyl nitrate solution, 1 M H^+, 7 mM stable Mo, and tracer Mo-99. In this run, 96% ± 5% of Mo was loaded on the column and 90% ± 5% was recovered. The total process time is 58 min.

TABLE 1 Process Design for Recovery of Mo from LEU Target Solution Using a 2.5 × 8 cm S80 Column

Step	Volume (mL)	$t_{loading}$ (min)	Flow Rate (mL/min)	u_s (cm/min)
Load	500	20.4	24.5	5
Wash, 1-M HNO_3	393	8.00	49.1	10
H_2O wash	196	4.00	49.1	10
Strip, 0.1-M NaOH	628	25.6	24.5	5

In this report, we would like to describe the testing of the above column design with irradiated target solution to monitor partitioning of Mo and tracer level fission products. The details of this work are described in Section 2. In addition, because S80 sorbent is no longer available on the market, S40 and S110 were evaluated for recovery of Mo from the LEU target solution. A description of new column designs and results of verification experiments are also presented in Section 2.

2 RESULTS AND DISCUSSION

2.1 S80 COLUMN DESIGN VERIFICATION WITH IRRADIATED TARGET SOLUTION

A 4-g depleted uranium (DU) target encapsulated in aluminum casing was irradiated at the 20-MeV Argonne Linear Particle Accelerator (LINAC). Irradiation was done using a tungston photoconverter to generate high-energy X-ray that generated photofissions in the target. The target was irradiated for 5.5 h at 18 MeV and 200 microA. The calculated ^{99}Mo yield, after a 14-h cooling time, was 0.5 mCi. After a 15-h cooling time, the aluminum casing was removed and 4 g of 125-µm-thick DU foils were dissolved in 12 mL of 10 M HNO_3. The dissolution took ~3 h and was maintained at ~100°C using a water bath. The dissolution flask was equipped with a dry ice/acetone filled condenser to minimize loss of water and nitric acid. The resulting 10 mL of dissolved target solution was added to 500 mL of UNH solution containing 450 g-U/L uranyl nitrate, 1-M H^+, and 7-mM Mo.

The irradiated target solution was loaded on a 2.5 × 8 cm column containing 51 g of S80 sorbent at 24.5 mL/min for 20.4 min, at 60°C. Details of loading times, volumes, and flow rates are in Table 2. The loading was followed by 1 M-HNO_3 and H_2O washes. Mo was stripped from the column with 0.1-M NaOH at 5 cm/min at 25°C. The strip was divided into four sections containing 4, 6, 6, and 14 BV of strip solution.

The counting results indicate that 98.3% of Mo was loaded on the column. Subsequently, 85.0% of Mo was in strip fraction 2, and 10.0% of Mo was found in strip fraction 3. Strip fraction 4 contained 0.2% of Mo. Therefore, stripping with 16 BV of 0.1 NaOH is sufficient to recover Mo from the column. About 1.5% Mo remained in the sorbent after stripping with 30 BV

TABLE 2 Experimental Conditions during Verification of Column Design with Irradiated Target Solution

Step	$t_{loading}$ (min)	Volume (mL)	Flow Rate (mL/min)	u_s (cm/min)	Solution Volume (BV)
Load	20.4	500	24.5	5	
Wash, 1-M HNO_3	8.0	392.7	49.1	10	10
1st H_2O wash	4.0	196.3	49.1	10	5
Strip fraction 1, 0.1-M NaOH	6.4	157.8	24.5	5	4
Strip fraction 2, 0.1-M NaOH	9.6	235.6	24.5	5	6
Strip fraction 3, 0.1-M NaOH	9.6	235.6	24.5	5	6
Strip fraction 4, 0.1-M NaOH	22.4	549.8	24.5	5	14
2nd H_2O wash	1.6	78.5	49.1	10	2

of 0.1-M NaOH. The nitric acid wash, water washes, and strip fraction 1 each contained <0.1% Mo. Mo partitioning between process streams is summarized in Table 3.

Partitioning of actinides and fission products between process streams was determined using gamma counting (Table 4). It was found that majority of radionuclides, including uranium, iodine, alkaline (Ba, Sr), transition metals (Cd, Rh, and Ru), and lanthanides (Ce, Nd) partition into the eluent and nitric acid wash streams. The fission products found in the Mo product stream are Te (~18% in the strip fractions 2 and 3) and I (~8% from fission + growing-in from Te), as well as <3% of Ru, Rh, Sb, and Zr. Te (~50%), Sb (>90%), and Zr (~80%, most likely as a precipitate) remained on the sorbent. Since 14% of Te was in strip fraction 4, which is not projected for the actual process conditions, ~60% of Te is expected to remain on the sorbent.

The activity levels of Ag-112, I-131, I-132, La-140, and Nb-97 in the product stream were determined at t = 26 h (Table 5). Their activities were not corrected to t = 0 because they originate as fission products and are growing in from their parent radionuclides: Pd-112, Te-131m, Te-132, Ba-140, and Zr-97, respectively. Their activities are presented as µCi/mCi Mo-99 at t = 26 h.

During the actual production of Mo-99, ~99% of I is expected to be removed during the dissolution step. Therefore, I activity in the product stream can be expected to be 100 times lower.

2.2 SORBENT SELECTION

Two sorbents, S40 and S110, were considered for new column designs for recovery of Mo from dissolved LEU target solution. The VERSE simulation program was used to determine MTZ (mass transfer zone) for S40 and S110 at 3 and 5 cm/min, when Mo concentration in the

TABLE 3 Partitioning of Mo between Process Streams during Column Design Verification Experiments Using a Tracer Irradiated Target Solution

Stream	Volume (mL)	Mo-99 (µCi)	Mo-99 (%)
Eluent	500	7.67	1.7
Strip, fraction 1	157.8	0.14	0.03
Strip, fraction 2	235.6	368.5	85.0
Strip, fraction 3	235.6	43.3	10.0
Strip, fraction 4	549.8	0.86	0.20
Mo Loaded			98.3
Total Mo in strip fractions 1–4			95.2

TABLE 4 Partitioning of Isotopes between Process Streams during Column Design Verification with Tracer Irradiated Target Solution

`	Half-life (days)	Eluent	1 M HNO_3 Wash	1st H_2O Wash	Strip fraction 1	Strip fraction 2	Strip fraction 3	Strip fraction 4	2nd H_2O Wash	Sorbent	Mass Balance
Ba-140	12.8	95	7			0.2					103
Cd-115	2.2	93	6								99
Ce-141	32.3	95	6							0.2	101
Ce-143	1.4	94	7	0.0		0.1			0.02		101
I-133	0.9	80	7	0.1	0.2	8	1.0	0.3	0.04		96
I-135	0.3	78	7	0.1	0.3	7	1.1	0.8	0.1		94
Mo-99	2.7	1	0.1	0.1	0.0	83	10	0.2		1.5	97
Nd-147	11.1	86	12		0.0		0.1				98
Pm-151	1.2	96	7		0.1						102
Rh-105	1.5	91	6	0.1	0.2	2			0.1		100
Ru-103	39.4	91	7			2	0.2	0.2		0.9	101
Sb-127	3.9	43	1			3	0.4			39	86
Sr-91	0.4	94	6	0.6	0.0						100
Te-132	3.2	3	0.3	0.0	0.0	14	4	14	3	48	85
U-237	6.8	89	6	0.0						0.0	96
Y-93	0.5	100	7			0.1	0.2	1.4	0.1		108
Zr-95	64.4		0.1			2	0.0			80	82

TABLE 5 Radionuclides Growing from Parents in the Product Steam at *t* = 26 h

Isotope	Half-life (days)	Parent	Half-life (days)	Total Activity, (µCi) at t = 26 h	% 1-σ	µCi/mCi Mo-99
Ag-112	0.13	Pd-112	0.88	7.52E-01	1.9	10
I-131	8.02	Te-131m	1.25	2.92E+00	0.7	12
I-132	0.10	Te-132	3.25	2.56E+01	0.1	199
La-140	1.68	Ba-140	12.79	7.05E-02	19.6	0.35
Nb-97	0.05	Zr-97	0.70	1.46E+00	0.5	55

TABLE 6 VERSE Simulation Results for Length of $MTZ_{0.1\%Cf}$ and Column Volume Calculations for S40 and S110 Sorbents

Sorbent	u_s (cm/min)	$MTZ_{0.1\%Cf}$ (cm)	CV_{min} (mL)
S40	3	2.06	*17.1*
	5	3.47	17.4
S110	3	5.23	43.6
	5	8.45	42.2

bulk reached 0.1% of the feed concentration. Both sorbents have the same titania composition as S80 and a 60-Å pore size. For both sorbent isotherms, sorbent and mass transfer parameters were assumed to be the same as those for S80 sorbent. Based on the MTZ length for S40 and S110, column volumes were calculated for both sorbents (Table 6). The results indicate that for recovery of Mo from dissolved LEU target solutions, columns based on S110 would be ~2.5 times larger than those based on S40 sorbent. Hence S40 was chosen for recovery of Mo from LEU targets.

2.3 COLUMN DESIGN AND VERIFICATION

Column designs based on S80 sorbent were calculated for 99% Mo capture. However, with a temperature variation to 55°C from 60°C and a change in viscosity from 1.40 cp to 1.52 cp, the intraparticle diffusivity was observed to decrease to 7.80×10^{-6} cm^2/min, resulting in Mo capture decreasing to 95%. Therefore, a small variation in process conditions can lead to a Mo recovery that is outside of the target recovery. To increase the margin for process conditions and variation, and to repeatedly achieve 99+% Mo recovery, the Mo loading goal was increased from 99% to 99.9%.

2.3.1 Isotherm Parameters

When sorption rates are higher than the controlling mass transfer rate, local equilibrium can be achieved between the solid and pore phases. Under such conditions, the solid phase concentrations are related to pore-phase concentrations by an equilibrium isotherm. The Langmuir model is tested for the sorption of Mo in this study:

$$q_i = \frac{a_i C_{p,i}}{1 + \sum_{j=1}^{N} b_j C_{p,j}} \tag{1}$$

where q_i is the amount of species i sorbed on the column packing and equilibrated with the concentration in mobile phase, $C_{p,i}$. In this study, both q_i (meq/L) and $C_{p,i}$ (mol/L) of the Mo isotherm is described on the basis of volume, assuming a 1.3-g/mL dry packing density of TiO_2 sorbent. Langmuir isotherm parameters for S40 and S110 were based on experimental data obtained for S80 sorbent and were fitted using Origen 8.5.1 (Figure 1).

The data from S80 batch equilibrium experiments were used to estimate the effective Mo isotherm parameters for TiO_2 sorbents. All batch equilibrium data were obtained for samples equilibrated for 24 h at 60°C. The calculated Langmuir isotherm model parameter a = 289.4, and b = 0.36 mM^{-1}.

2.3.2 Estimation of Other Mass Transfer Parameters of Mo

The axial dispersion coefficient, E_b, was estimated using the Chung and Wen (1968) correlation. The mass transfer coefficient, k_f, was estimated using the Wilson and Geankoplis (1966) correlation. Brownian diffusivity (D_∞) values of $HMoO_4^-$ in aqueous solution at 25°C used in the simulations were reported to be 8.3 cm^2/min (Marcus 1997). The Stokes-Einstein equation was then used to correct the D_∞ for solution viscosity and temperature effects. At 60°C, the viscosity of 450 g-U/L $UO_2(NO_3)_2$ and 1-M HNO_3 was calculated to be 1.40 cp, using a correlation listed in HW-57386 (Krigens 1968). Therefore, at 60°C, $D_{\infty,Mo}$ in 450 g-U/L $UO_2(NO_3)_2$ and 1-M HNO_3 solution was estimated to be 5.85×10^{-4} cm^2/min.

2.3.3 Estimation of the Mo Intraparticle Diffusivity Values (Dp) for S40 from Breakthrough Curves

A breakthrough experiment using 450 g-U/L, 6.5-mM Mo, and 1-M H^+, was run to estimate D_p from breakthrough curve (Figure 2). The breakthrough curve was obtained by loading 150 m L of feed solution onto a 6.6 × 35 mm S40 column at 3 cm/min at 60°C.

The intraparticle diffusivity was estimated by comparing the experimental Mo breakthrough curves with VERSE simulations. In 450 g-U/L $UO_2(NO_3)_2$ and 1-M HNO_3

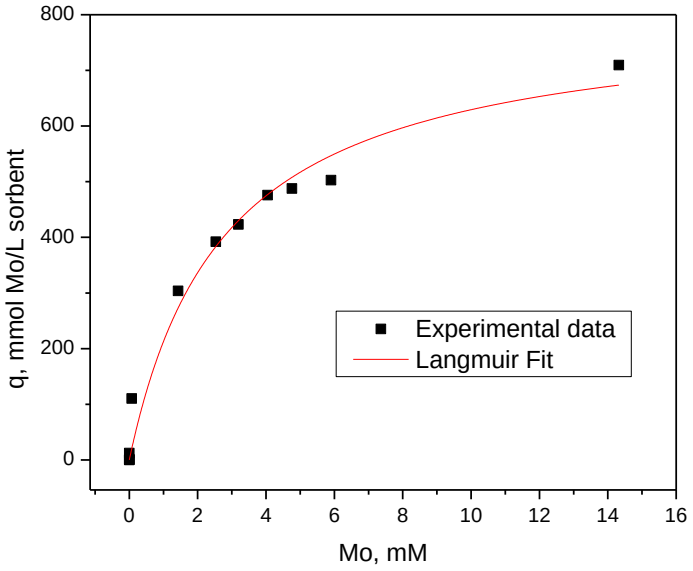

FIGURE 1 Langmuir Isotherm Data from Batch Tests and Langmuir Isotherm Model Fit for Uptake of Mo on S80 Sorbent

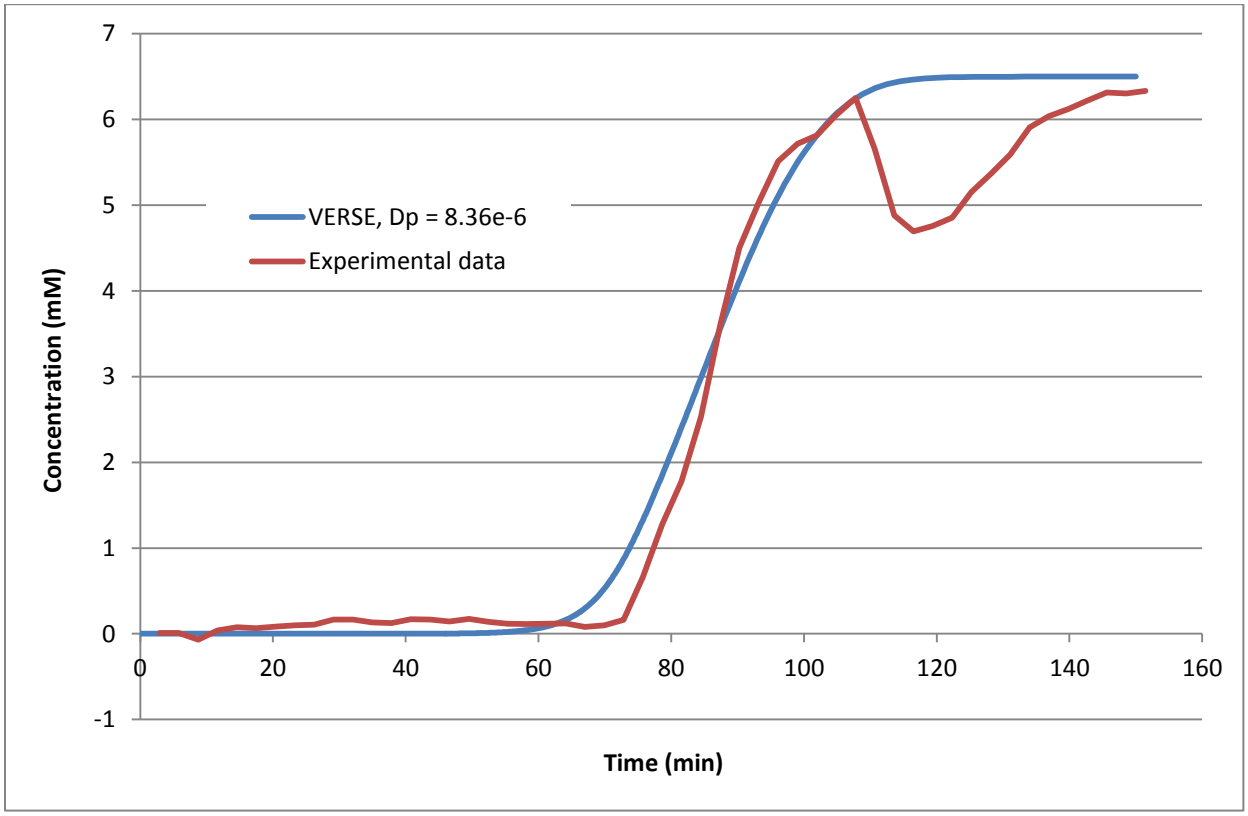

FIGURE 2 Estimation of Dp for S40 Sorbent from Breakthrough Experiment

solution, the intraparticle diffusivity of Mo on S40, assuming $D_\infty/D_p = 70$, is estimated to be $D_p = 8.36 \times 10\text{-}\ cm^2/min$. Intraparticle diffusion of Mo is assumed to be pore diffusion. As shown in a recent paper of Chung et al. (2010), in a linear isotherm range, surface diffusion effects cannot be distinguished from pore diffusion effects, and therefore an effective pore diffusion coefficient is sufficient to take into account of pore diffusion, surface diffusion, or parallel pore diffusion.

The agreement between experimental Mo breakthrough curves and model prediction indicates that (a) the adsorption of Mo in 450 g-U/L $UO_2(NO_3)_2$ and 1-M HNO_3 can be described by the effective Langmuir isotherm; and (b) the numerical parameters and the intrinsic model parameters (including void fractions, isotherm, and mass transfer parameters) are sufficiently accurate to predict the Mo breakthrough curves in 450 g-U/L $UO_2(NO_3)_2$ and 1-M HNO_3 solution for S40 sorbent.

2.3.4 Column Designs for Mo Recovery from 450 g-U/L $UO_2(NO_3)_2$ and 1-M HNO_3, 7-Mm Mo

Column designs were calculated for loading 500 mL of LEU target solution in 20 min to limit the total column process time to 1 h. Therefore, for loading the LEU target solution at 25 mL/min inner column diameters were chosen to be 5, 3.5, and 2.5 cm. These diameters reflect dimensions of commonly manufactured columns. The VERSE simulations were carried out to estimate the MTZ at resulting linear velocities for 20 min loadings (Table 7). For the 2.5 × 9 cm column design, ΔP exceed the 0.8-atm pressure drop limit. The 5 × 2.5 and 3.5 × 5 cm sizes are viable column designs.

The 3.5 × 5 cm column design was tested experimentally at 60°C using 450 g-U/L solution with 7-mM Mo, 1-M H^+, and tracer levels of Mo-99. Loading velocity and column length were kept constant at 2.60 cm/min and 5 cm, respectively, while the column's inner diameter (ID) and the solution volume were reduced to 1 cm and 40.8 mL, respectively. Mo was stripped from the column with 0.1-M NaOH at 3 cm/min at 25°C. The load, wash, and strip fractions were collected as specified in Table 8. Then, 99.4% ± 3% of Mo was loaded on the

TABLE 7 Column Designs for Recovery of Mo from 500 mL of LEU Target Solution in 20 min

Column ID (cm)	Velocity (cm/min)	$MTZ_{0.1\%}$ (cm)	Column Length (cm)	Column Volume (mL)	Sorbent Weight (g)	ΔP (atm)
5	1.27	1.19	2.50	49	64	0.09
3.5	2.60	2.41	5.00	48	63	0.35
2.5	5.09	4.70	9.00	87	113	1.24

TABLE 8 Experimental Details of Verification of Column Design for Recovery of Mo from LEU Target Solution

Step	$t_{loading}$ (min)	Volume (mL)	Flow Rate (mL/min)	u_s (cm/min)	Solution Volume (BV)
Load	20	40.8	2.0	2.60	
Wash, 1-M HNO_3	8.1	31.4	3.9	5	8
1st H_2O wash	3.0	11.8	3.9	5	3
Strip fraction 1, 0.1-M NaOH	6.5	15.7	2.4	3	4
Strip fraction 2, 0.1-M NaOH	9.8	23.6	2.4	3	6
Strip fraction 3, 0.1-M NaOH	9.8	23.6	2.4	3	6
Strip fraction 4, 0.1-M NaOH	22.9	55.0	2.4	3	14

column and 99.9% ± 3% was recovered in strip fractions 2 and 3. Mo-99 activity in the nitric acid and water wash fractions and strip fraction 1 and 4 were below detectable levels.

Based on these results, the Mo recovery process was designed using a 3.5 × 5 cm column with S40 sorbent (Table 9). In this process, 500 mL of solution is loaded at 60°C in 20 min at 25 mL/min. The loading is followed by 1-M HNO_3 and H_2O washes at 48.1 mL/min at 25°C. Mo is stripped at 25°C with 770 mL of 0.1-M NaOH at a flow rate of 28.9 mL/min for 26.7 min. The total process time is 58 min. The calculated pressure drop for the loading, washing, and strip are 0.35, 0.43, and 0.26 atm, respectively.

TABLE 9 Process for Recovery of Mo from LEU Target Solution Using a 3.5 × 5 cm Column and S40 Sorbent

Step	$t_{loading}$ (min)	Volume (mL)	Flow Rate (mL/min)	u_s (cm/min)	Solution Volume (BV)
Load	20	500	25.0	2.60	
Wash, 1-M HNO_3	8	385	48.1	5	8
Wash, H_2O	3	144	48.1	5	3
Strip, 0.1-M NaOH	26.7	770	28.9	3	16

3 CONCLUSIONS

Partitioning of actinides, lanthanides, and fission products between different process streams was determined, using a representative titania-based column. The majority of activity from radionuclides, including iodine, actinides, lanthanides, and alkaline and transition metal ions reports in the eluent and wash fractions (waste). Fission products found in the Mo product stream are transition metals: Te (~30%) and <3% of Ru, Rh, Sb, and Zr. About 8% of fission I was found in the Mo product stream. However, the activity of iodine in the product stream is expected to be ~100 times lower when I is removed during the target dissolution step.

New column recovery process was developed based on S40 sorbent. This process uses a 3.5 × 5 cm column with 63 g of sorbent. The loading time is 20 min and the column recovery process is designed to be complete in 1 h. This process design was verified with solution containing 450 g-U/L, 1-M H^+, 7-mM Mo, and tracer levels of Mo-99. The experimental results indicate that 99.4% ± 3% of Mo was loaded on the column and 99.9% ± 3% was recovered in strip fractions 2 and 3.

4 EXPERIMENT

Titania sorbents (S80, S110, and S40 [80, 110 and 40 μm, respectively, 60 Å pore size]) were purchased from ZirChrom Separations, Inc. (Anoka, Minnesota).

Uranyl nitrate, $UO_2(NO_3)_2$, solutions were prepared by dissolving a known amount of uranium metal in 8-M HNO_3. The resulting solution was titrated in presence of oxalate and H^+ was adjusted with concentrated HNO_3. Molybdenum was added as 0.3-M Na_2MoO_4, pH 0 solution.

Mo-99 was obtained from a commercial Tc-99m generator by stripping with 1-M NH_4OH, evaporating to dryness and dissolving in nitric acid.

Isotherm data were obtained by equilibrating 10 ± 1 mg of sorbent with 1 mL of 450 g-U/L containing an appropriate amount of Mo for 24 h at 60°C.

The amount of Mo-99 activity in the aqueous samples was determined using a NaI(Tl), HPGe well detector, or NaI(Tl) flow-through detector. Mo-99 was quantified by measurement of its 739-keV γ-ray. The activity of Mo-99 in each sample was corrected for decay. Activities in the irradiated target solution and process streams were determined using HPGe planar detector.

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Chemical Science and Engineering Division
Argonne National Laboratory
9700 South Cass Avenue, Bldg. 205
Argonne, IL 60439-4837

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