Integrating Microfluidic Technology into Bioassay Programs for Enhanced Efficiency and Safety – 25469

Sarah Lu *, Tiffany Sanders *, Allison Wende *, April Cardon * Annelise Cardon *, Matthew Sanborn * Jeremy Inglis *, Robert Steiner *, Stephen LaMont *
* Los Alamos National Laboratory

ABSTRACT

The nuclear industry, with over 90 active reactors in the U.S., requires strict safety measures for managing radioactive materials. Traditional bioassay methods for monitoring worker exposure are labor-intensive and time-consuming, leading to delays and significant infrastructure needs. This study investigates microfluidic technology as a solution to these challenges, enabling integrated separation and recovery on a microscale. By reducing sample volume, reagent use, and minimizing waste, this approach has the potential to accelerate analytical processes and improve temporal accuracy, enhancing overall responsiveness. Our research specifically examines the potential for microfluidic systems to optimize the PuTIMS process, addressing sources of plutonium (Pu) loss and matrix interference caused by elements from stainless steel planchets, such as chromium (Cr), manganese (Mn), cobalt (Co), nickel (Ni), copper (Cu), and molybdenum (Mo). The results demonstrate that microfluidic systems have the potential improve recovery rates and detection limits for radionuclides, offering substantial benefits for the nuclear industry, including enhanced data resolution, reduced operational costs, and improved safety protocols. Integrating this technology into bioassay programs ultimately sets a new standard for modern monitoring practices, improving both worker safety and regulatory compliance in the nuclear sector.

INTRODUCTION

Our research explores the use of microfluidic technology to enhance *in vitro* bioassay measurement programs used to monitor nuclear workers for occupational intakes of radionuclides, a critical area for the safe handling of radioactive materials.[1] Traditional bioassay methods are labor-intensive and involve multi-step pre-treatment and processing [2] to decompose the high organic matter content of the sample which is necessary for subsequent separation.[3] A common practice is to conduct evaporation or co-precipitation to concentrate analytes from the urine followed by acid digestion and purification using single ion-exchange columns.[3] These steps are time, infrastructure, and reagent-intensive leading to delays in data availability. To improve analytical efficiency and sustainability, direct analysis of urine for radionuclides with little or no pre-processing has been proposed, e.g. direct analysis of urine by inductively coupled plasma mass spectrometry.[4] These less-labor intensive and timelier techniques are attractive as the nuclear industry continues to grow,[5] as there is an increasing demand on bioassay programs, which in turn requires a more skilled workforce and robust infrastructure to support these complex processes. With limited laboratory space and the time required to train personnel, it is crucial to develop technologies that can accommodate this growing demand while optimizing space and resource utilization.

An example of the increasing need for bioassay monitoring is the Los Alamos National Laboratory (LANL), the plutonium *in vitro* urine bioassay program. This program has been established for several decades and currently monitors thousands of workers exposed to potential radionuclide intake.[6] The LANL bioassay program currently monitors workers using two analytical procedures, alpha spectrometry and thermal ionization mass spectrometry (TIMS).[7] First, alpha spectrometry is used to measure the ^{238}Pu and $^{239+240}$Pu Content. However, due to the closely spaced alpha energies of ^{239}Pu and ^{240}Pu, alpha spectrometry cannot distinguish between these isotopes.[8] To address this limitation, the samples are further analyzed using thermal ionization mass spectrometry, which can resolve the ^{239}Pu and ^{240}Pu isotopes independently and has the required sensitivity to detect a Pu intake equivalent to a 20 mRem, 50 yr CEDE. The PuTIMS analysis represents the cornerstone of the LANL bioassay program, offering highly sensitive isotope data

for dose assessment. The TIMS isotope data can also be used as a forensic signature of the possible source of the contamination in the event of worker exposure.

Despite the effectiveness of these methods, several challenges persist, particularly in the PuTIMS chemistry process. Two recurring issues are the variable chemistry yields and inconsistent Pu ionization in the TIMS instrument, which can compromise the consistency and reliability of dose assessments for ^{239}Pu and ^{240}Pu. These inconsistencies are thought to stem from matrix interference during the purification stages, which can lead to either plutonium losses during rinsing and the retention of contaminants, or poor Pu ionization in the TIMS source. While the exact causes are not fully understood, it is suspected that these interferences in the early phases of TIMS preparation significantly contribute to the problem.

Microfluidics offers the potential to simplify bioassay sample preparation in radiochemistry. We chose to focus initial efforts on the TIMS sample preparation chemistry, given its more limited matrix and evaluate the replacement of more traditional gravity-feed anion exchange chemistry with a microfluidic technique. Building on the success of this method, there is further potential for improvement by incorporating advanced technologies like microfluidic systems. These systems could accelerate the PuTIMS process by automating steps and reducing sample volumes, leading to faster screening and more timely results. Additionally, the process can be improved by reducing variability in yields through introducing supplementary purification loops to minimize matrix interference and plutonium losses. This integration would expedite the PuTIMS process, further streamlining the overall workflow.

Current LANL Bioassay Chemistry Procedure

Our work focuses on optimizing the plutonium (Pu) purification step following leaching from a stainless-steel planchet after alpha spectrometry, which is a key part of the plutonium urine bioassay process. Alpha spectrometry is used in multiple Department of Energy (DOE) employee bioassay surveillance programs to measure isotopic concentrations of Americium (Am), Plutonium (Pu), and Uranium (U).[9]

In this process, urine samples are spiked with ^{242}Pu as an internal yield tracer prior to any chemical treatment.[7] Sample preparation begins with an alkaline earth phosphate precipitation followed by an anion exchange.[7] This process separates the Pu, Am, and U from the majority of the sample matrix by exploiting the limited solubility of actinide phosphates under alkaline conditions. . The sample is then wet ashed with nitric acid and dissolved in 8 M HNO_3 for ion-exchange. The plutonium is separated by sorption to an anion-exchange resin (BioRad AG 1x4), rinsing with additional 8 M HNO_3, and it is elution with 0.36 M HCl and a concentrated HI-HCl solution. The eluted plutonium is evaporated to dryness, ashed with a H_2O_2–HNO_3 mixture, and then electrodeposited from a bi-sulfate buffer onto a 12.7 mm stainless-steel planchet.[9, 10] After the electroplating process, the planchet is analyzed using alpha spectrometry to measure the isotopic concentrations of $^{239+240}$Pu, and ^{238}Pu. The tracer's alpha-particle emissions recorded in a calibrated energy spectrum provide data to calculate chemical recovery, based on the ratio of tracer counts to its activity in becquerels (Bq), ensuring accurate results.

To independently measure ^{239}Pu and ^{240}Pu and decrease detection limits, thermal ionization mass spectrometry (TIMS) is employed. After alpha spectrometry analysis the plutonium is extracted from the planchets by soaking them in 8 M HNO_3.[7] The solution is passed through an anion-exchange column (BioRad AG MP-1), rinsed with additional 8 M HNO_3, plutonium is eluted with conc. HBr, then evaporated. The sample is digested in HNO_3 and $HClO_4$ to digest any organic residue from the ion-exchange process, then digested again with conc. HCl. Finally, the purified plutonium is co-electrodeposited with platinum onto a rhenium filament, which is over-plated with platinum to create the surface ionization filament used for plutonium measurement by TIMS.[11] Two recurring issues are the variable chemistry yields and irregular Pu ionization in the TIMS instrument, which can compromise the consistency and reliability of detection limits and ultimately dose assessments for ^{239}Pu and ^{240}Pu. These inconsistencies are thought to stem from matrix interference during the purification stages, which can lead to either plutonium losses during rinsing and the retention of contaminants, or poor Pu ionization in the TIMS source. While

the exact causes are not fully understood, it is suspected that these interferences from the stainless steel planchet and insufficient purification during TIMS preparation chemistry significantly contribute to the problem.

EXPERIMENTAL OUTLINE

The primary goal of this study was to identify potential areas of Pu loss and matrix interference effecting the PuTIMS chemistry and then to identify areas in which microfluidics could improve the chemical process. To accomplish this, a series of experimental steps were designed to identify the presence of elements that may interfere with TIMS during the leaching of alpha spectroscopy planchets. The focus was on examining the behavior of elements commonly associated with stainless steel, including chromium (Cr), manganese (Mn), cobalt (Co), nickel (Ni), copper (Cu), molybdenum (Mo) and cerium (Ce) because elevated levels of cerium are associated with low ionization of plutonium in the TIMS process. These elements were tracked across various fractions during the column sample traversal after leaching plutonium from the alpha spectroscopy planchets. The primary goal is to identify potential steps where Pu is lost, and the presence of other elements from the leaching of alpha spectroscopy planchets, where the chemical yields of range between 77.4% to 83.5% for quality control planchets and 79% to 86.9% for blank planchets. Inductively Coupled Plasma Mass Spectrometry (ICP-MS) was then used to screen for these elements at different stages of the leaching and purification process. After initial alpha spectrometry the sample planchets for the quality control, urine blank and process blank (Table 1) samples were transferred to 50 mL tubes, 2 mL of 8M HNO_3 was added to each tube, and the tube rack was placed on an orbital shaker and leached overnight. Each sample was then vortexed and ~ 5 mg of $NaNO_2$ was added to each tube. MilliQ water was added to columns (Bio- Spin) to begin flow, 0.5 mL of AG MP-1(BioRad), 100-200 mesh resin was then added to each column. The column was then rinsed twice with 1.5 mL of 0.12 M HCl and then rinsed twice with 1.5 mL of 8M HNO_3. The leached solution from leaching the stainless steel planchets were loaded onto the column.

Table 1. Sample planchet identification in urine bioassay.

Sample planchet	
Blank	The blank planchet, processed alongside samples, is used to monitor for contamination and loss of analytes throughout the sample preparation procedure.
Quality Control	The quality control planchet, spiked with a known quantity of Pu to ensure the accuracy and consistency of the bioassay results to be tested alongside customer samples verifies that the assay is performed within established parameters and detecting any potential issues with the process batch
Urine Blank	Blank urine samples act as a matrix matched control specimen during bioassay.

The sample planchets were then rinsed twice in a 50 mL tube with 1.5 mL of 8M HNO_3 and the solutions added to the column. The rinse from these planchets was then collected after it had passed through the columns (Figure 1) and labelled as ‘Load rinse’ (Table 2).

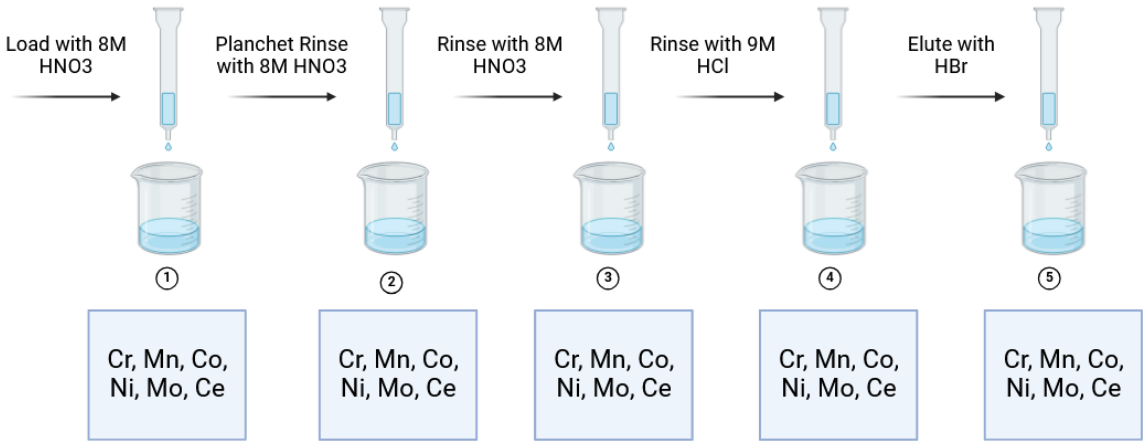


Figure.1 Rinse steps in PuTIMS single column procedure.

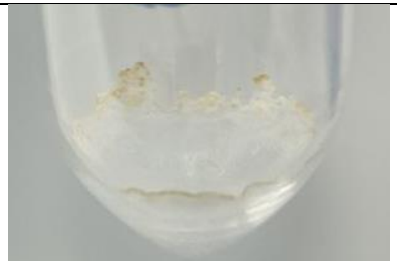
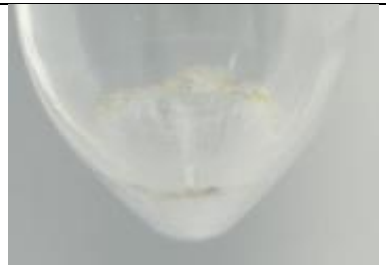
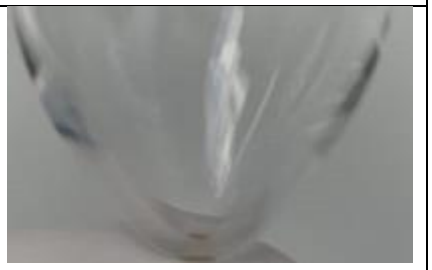
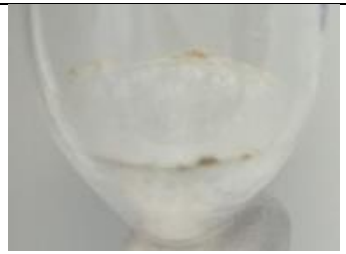
The columns were then rinsed four times with 1.5 mL of 8 M HNO_3 and the solutions from the planchets collected and labelled as 'First Rinse'. The columns were then rinsed twice with 1.5 mL of 9 M HCl, the rinse was collected and labelled as 'Second Rinse'. Plutonium was eluted with four rinses of 0.75 mL of conc. HBrHBr, this elution was collected and labelled as 'Elution Step'. This process was repeated for each blank, quality control and urine blank.

Table 2. Rinse steps in PuTIMS single column procedure.

Step	**Reagent(s)**	**Purpose**
Load Rinse	HNO_3	Dissolve plutonium and associated contaminants for further analysis.
First Rinse	HNO_3	Rinse to remove residual contaminants that were not removed during the initial loading.
Second Rinse	9M HCl	Extract additional impurities that may interact with plutonium or are detrimental to determination by TIMS.
Elution Step	HBr	Purified Pu fraction

Each collection was then dried down overnight in a quartz crucible and spiked with 2 pg of ^{244}Pu, then dried down and redissolved in 2% Nitric Acid 0.005 M HF to prepare it for screening by ICPMS. After the collections were dried down residue was noted in the quart crucibles (Table 3). Most residue was observed after the load rinse and initial rinse, whilst the second rinse and elution step demonstrated significantly less visible residue after the drying down process.

Table 3. Residue in quart crucible after dry down process.

	Step			
	Load Rinse	**First Rinse**	**Second Rinse**	**Elution Step**
Urine Blank				
Blank				
Quality Control				

Upon completing the recovery steps and resuspending the samples in 2% nitric acid, the samples were analyzed using ICP-MS. The results are reported as the $\log_{10}$ of acid-blank subtracted isotope counts per second (cps) for each element. The instrument sensitivity was approximately 2.0 $x10^6$ cps ^{238}U / ppb for all analytical sessions. The primary objective is to screen the concentrations of chromium (Cr), manganese (Mn), cobalt (Co), Nickel (Ni), copper (Cu), and molybdenum (Mo) to evaluate the effectiveness of the purification process. This screening will guide the design of additional rinsing and purification columns or loops to be integrated into the microfluidic system. By pinpointing areas where further purification is required, the results will inform the strategic placement of these additional steps, thereby improving sample purity prior to Thermal Ionization spectrometry (TIMS) and enhancing overall process efficiency.

Reagents and Materials

All acids (HNO_3, HCl, HF, HBr) used were OptimaTM grade (Fisher ChemicalTM, Fisher scientific, USA). For the radiochemical separation of Pu AG 1-x4 (100-200 mesh, Chloride form, BioRad, USA) ion exchange resin was used. The resins were filled in 10 mL Poly-Prep® chromatography columns (BioRad, USA). All reagents used ($NaNO_2$) were of analytical grade. Ultrapure water (resistivity 18.2 MΩ at 25 °C) was used.

RESULTS AND DISCUSSION

The experimental analysis of the eluent from the stainless steel planchets used in the PuTIMS process revealed substantial levels of matrix elements, including chromium (Cr), manganese (Mn), cobalt (Co), nickel (Ni), copper (Cu), , and molybdenum (Mo), which are primary constituents of stainless steel. Cerium (Ce) was also detected in the rinses and elution. These elements were consistently detected across all fractions during the rinsing and purification steps, which included the blank, urine blank, and quality control planchets. To enhance data visualization, counts per second (CPS) were recorded and log-transformed to a base-10 scale, allowing all rinse fractions and the elution step to be displayed on a single graph for each element. This transformation helped to highlight the significant presence of matrix elements and their potential interference with plutonium recovery. The presence of these matrix elements is noteworthy, as their interactions with plutonium (Pu) can lead to complexation, which may reduce plutonium recovery and affect the accuracy of the TIMS analysis. Histograms of the log-transformed counts revealed that decontamination varied among the elements, with certain metals such as chromium (the presence of iron is inferred from the presence of chromium and other stainless steel derived elements) contributing substantially to the matrix interference. The blank planchet typically shows the lowest levels of all elements in each rinse and elution fraction. In comparison, the urine blanks have slightly higher levels and exhibit smaller discrepancies between the two across fractions. In general, the quality control planchets show higher levels of elements in the rinse and elution fractions, suggesting that the plated plutonium interacts with the stainless steel. These findings underscore the importance of optimization of the PuTIMS chemistry purification steps to minimize potential plutonium loss or contamination.

Chromium (Cr) and Manganese (Mn): Chromium, a key component of stainless steel, and manganese were detected in all rinses and the elution step (Figure 2 & 3), with log-transformed counts showing consistent levels throughout the purification. Both metals, as transition elements with partially filled d-orbitals, form stable complexes with plutonium due to their redox flexibility. Chromium can exist in multiple oxidation states (e.g., Cr(III), Cr(VI)), and manganese in Mn(II), Mn(IV), and Mn(VII), allowing them to readily interact with plutonium and form complexes. This process is further stabilized by electrostatic interactions due to the high charge density of both iron/chromium and plutonium, facilitating complex formation. These interactions, particularly under oxidizing conditions during the load and rinse steps, may reduce the available plutonium for analysis or introduce matrix interference, complicating TIMS measurements. While counts decrease by an order of magnitude after the load rinse, log-transformed data suggest that additional rinsing or purification steps are needed to mitigate chromium- and manganese-induced interference. These interactions underline the need for optimized purification strategies to minimize

matrix interference, ensuring more efficient plutonium separation and recovery for subsequent TIMS analysis.

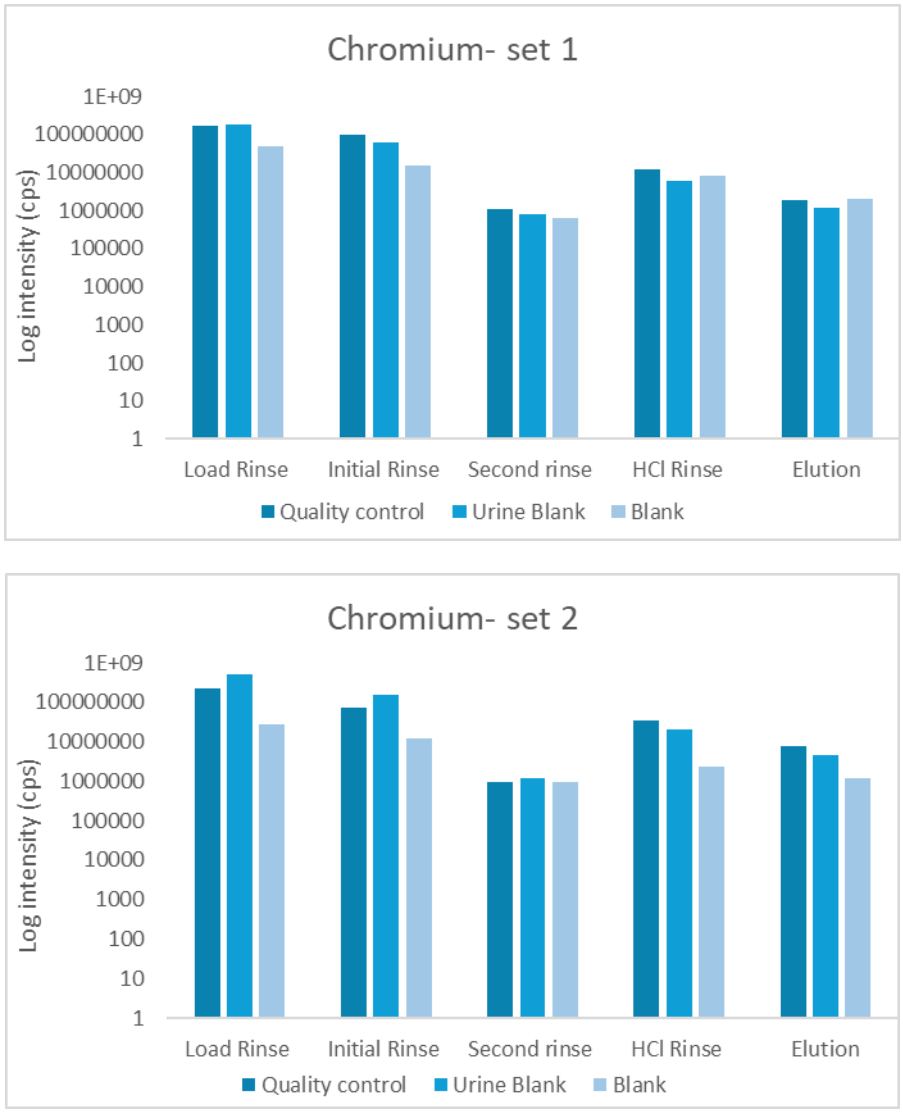


Figure 2. Histograms showing the distribution of chromium in the rinses and elution steps. The data is presented as the log10 of isotope counts per second (cps), with acid-blank values subtracted.

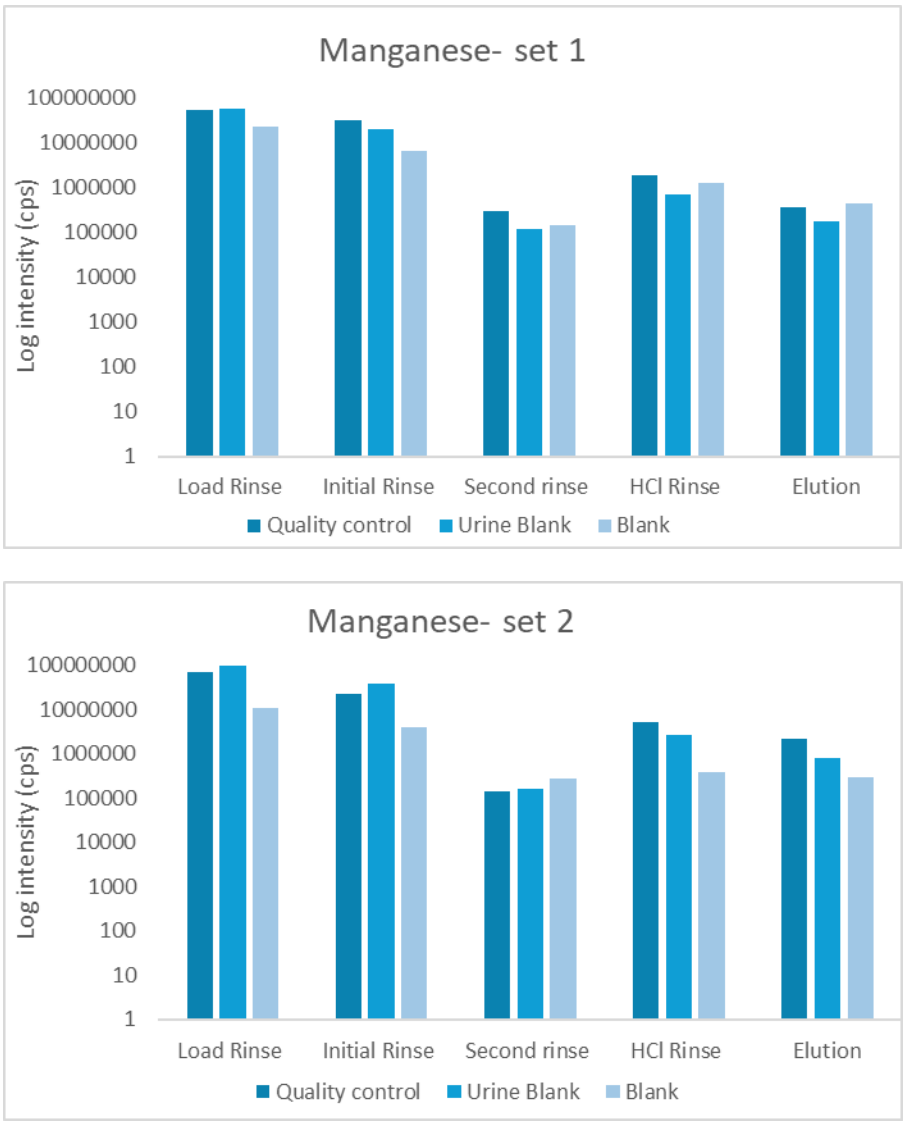


Figure 3. Histograms showing the distribution of Manganese in the rinses and elution steps. The data is presented as the log10 of isotope counts per second (cps), with acid-blank values subtracted.

Nickel (Ni) and Cobalt (Co): Nickel and cobalt, common components of stainless steel, were detected in the rinse fractions (Figure 4 & 5), with both levels decreasing across successive rinses, and the most significant reduction occurring after the first and second rinses. Both metals, as transition elements with partially filled d-orbitals, can complex with plutonium, particularly under acidic conditions like those in the hydrochloric acid rinse. These interactions, driven by d-orbital coordination, may hinder plutonium separation, causing potential loss or interference during analysis. Cobalt, like nickel, can form similar complexes with plutonium, potentially complicating the purification process. To reduce this interference, additional purification steps could ensure higher plutonium yields and more accurate analysis.

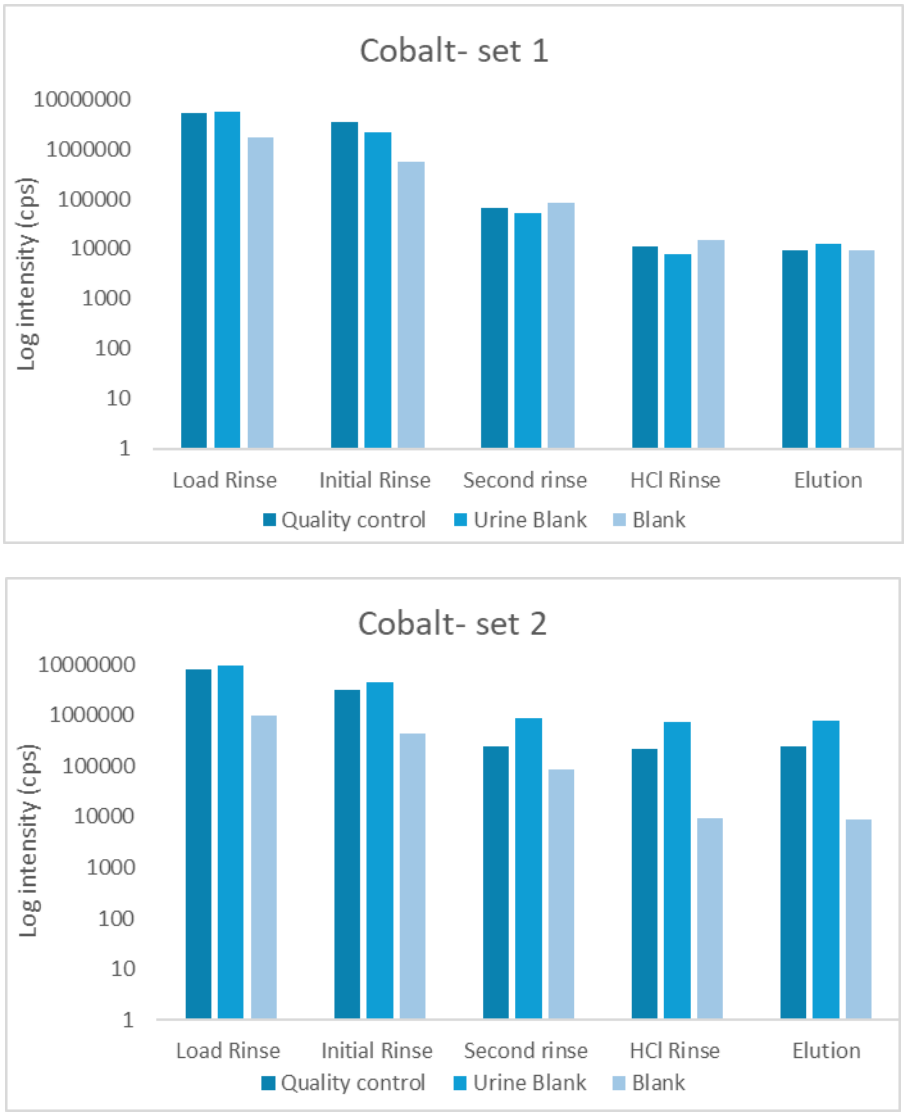


Figure 4. Histograms showing the distribution of Cobalt in the rinses and elution steps. The data is presented as the log10 of isotope counts per second (cps), with acid-blank values subtracted.

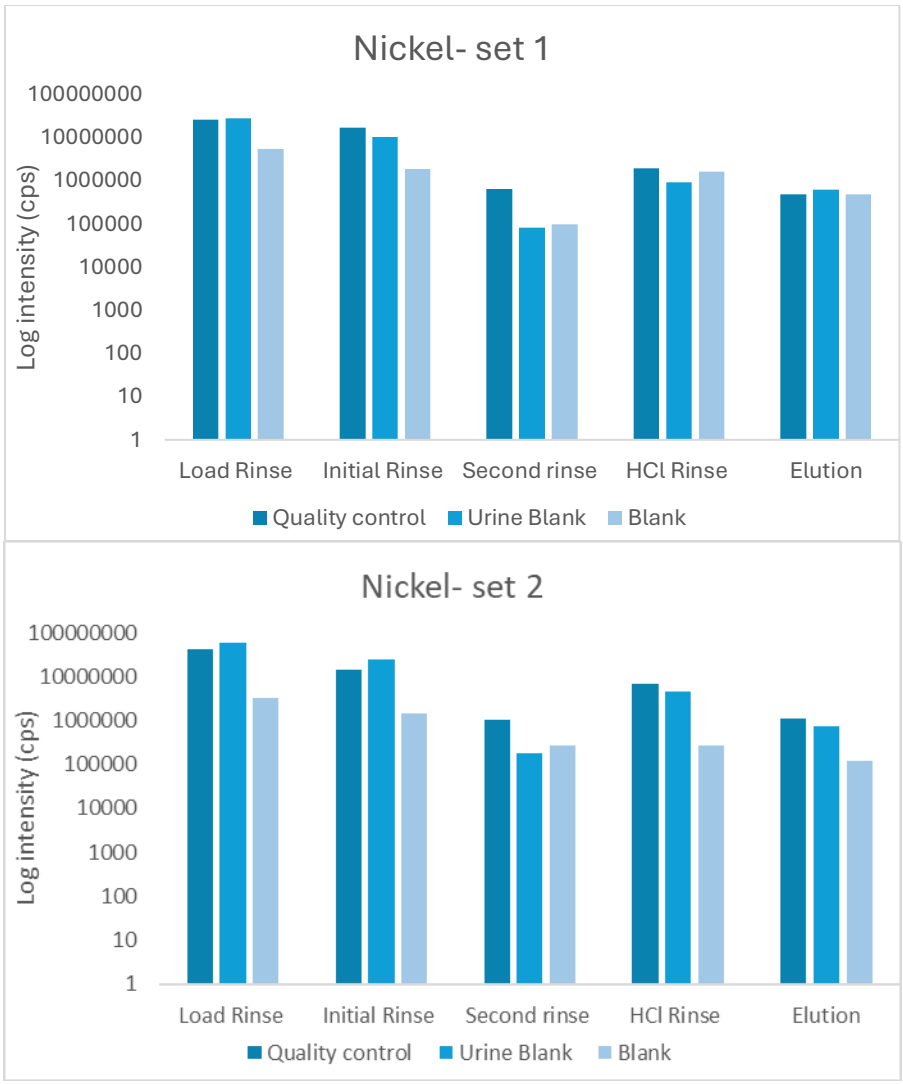


Figure 5. Histograms showing the distribution of Nickel in the rinses and elution steps. The data is presented as the log10 of isotope counts per second (cps), with acid-blank values subtracted.

Copper (Cu) and Molybdenum (Mo): Copper and molybdenum are additives elements to stainless steel were detected in smaller quantities (Figure 6 & 7) than chromium or manganese. Overall cps for these elements indicates that while their impact on plutonium recovery was less significant than other elements, copper, and molybdenum still contribute to matrix interference, potentially distorting chemical yields or causing plutonium loss. To minimize this, further purification steps, such as additional rinsing or selective ion-exchange methods, are recommended to reduce these elements and improve plutonium recovery for more accurate analysis.

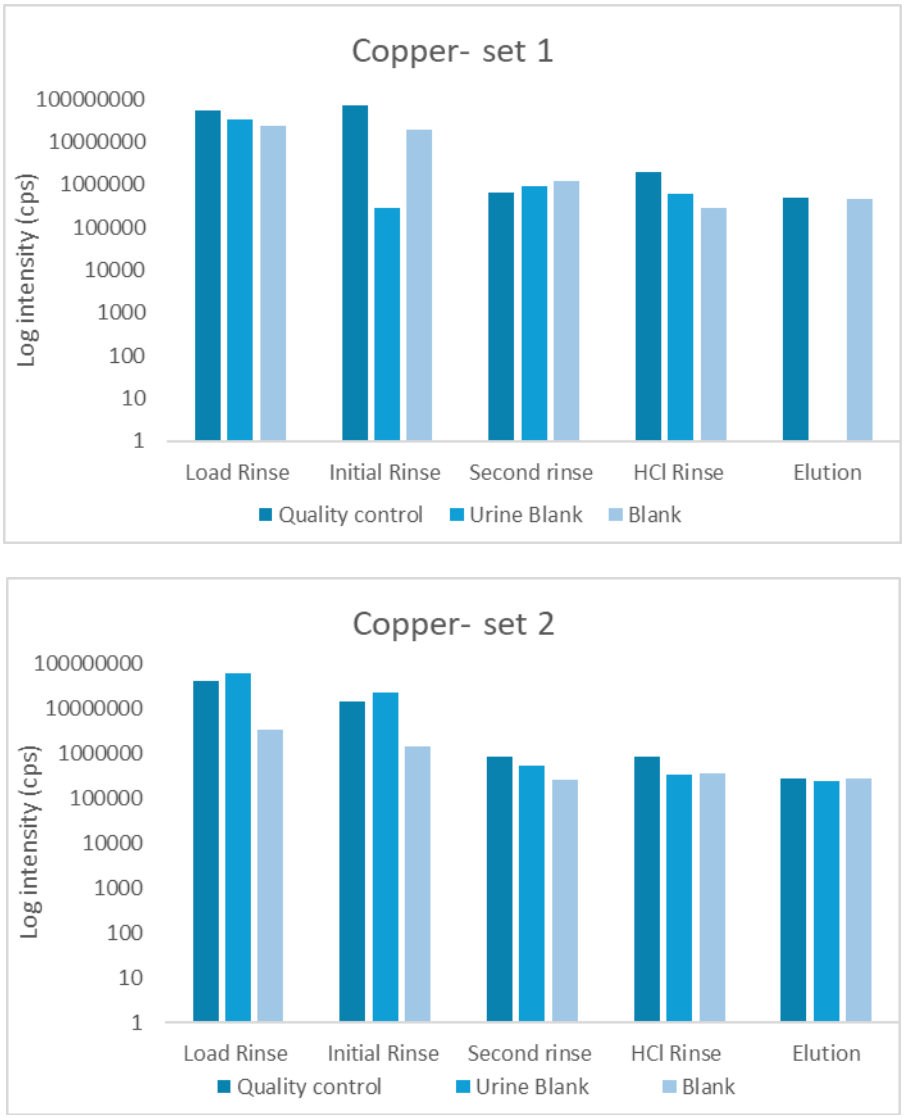


Figure 6. Histograms showing the distribution of Copper in the rinses and elution steps. The data is presented as the log10 of isotope counts per second (cps), with acid-blank values subtracted.

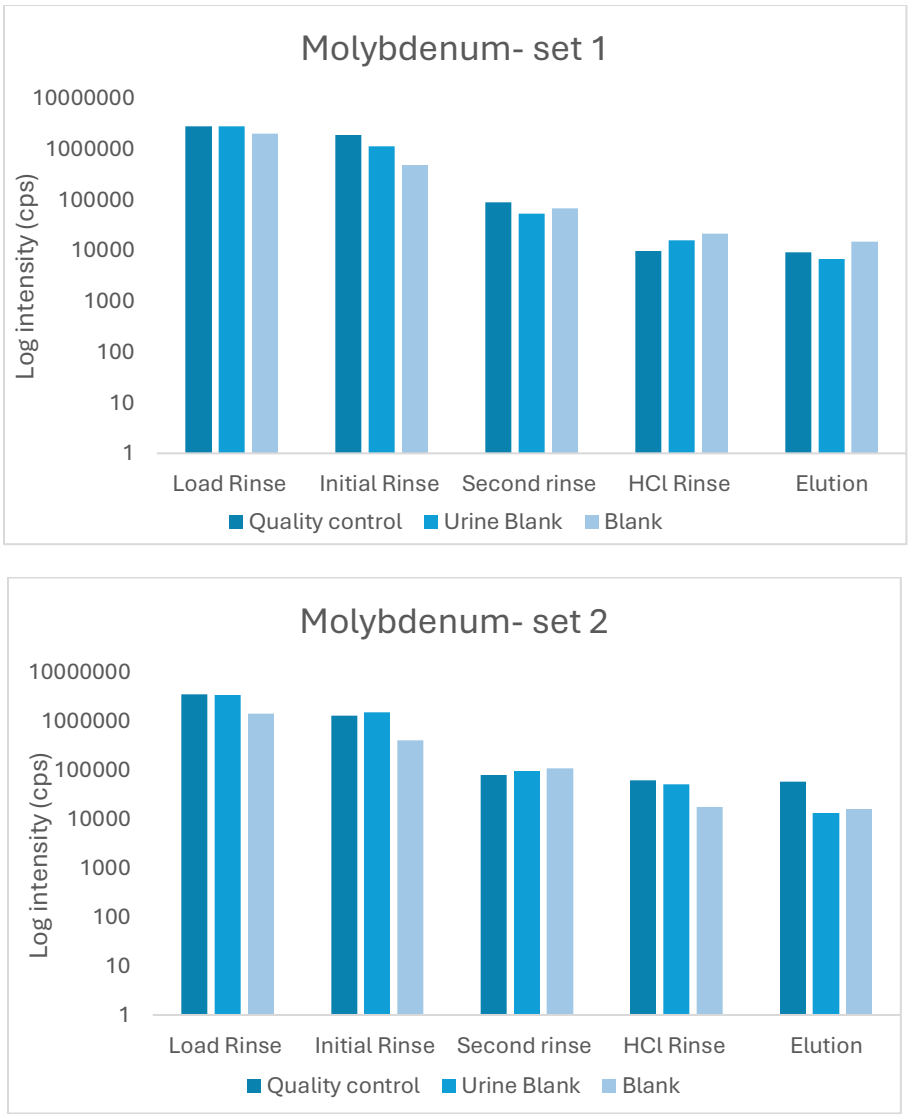


Figure 7. Histograms showing the distribution of Molybdenum in the rinses and elution steps. The data is presented as the log10 of isotope counts per second (cps), with acid-blank values subtracted.

Plutonium Losses and Matrix Interference: The leaching and purification procedure, as demonstrated by the residue observed in the quartz crucibles (Table 3), showed a decrease in visible residue following the first and second rinse steps. Which correlated with overall cps reduction between these steps for all elements. The log-transformed counts indicated that contaminants were more effectively removed during these stages, suggesting that the rinsing process was successful in reducing the overall matrix load. However, the continued presence of stainless steel-derived elements, such as chromium, manganese, nickel, and the inferred presence of iron, during these rinses indicates that complexation between plutonium and these matrix elements may still be occurring. These metal ions, due to their ability to form stable complexes with plutonium, can bind to plutonium and reduce its availability for recovery, leading to potential plutonium losses during purification. Cerium (Figure 8) is often associated with reduced ionization of plutonium in TIMS analysis, and its presence in all elution and rinse steps, without consistent reduction between rinse steps, indicates that cerium may be interfering with the ionization process. The data suggests that while the rinsing process helps reduce some matrix interference, further optimization of the purification steps is required to fully minimize plutonium losses. Specifically, additional rinsing or selective purification methods targeting the removal of stainless steel derived elements would help to prevent complexation with

plutonium. This could enhance the efficiency of plutonium recovery and improve the accuracy of subsequent analysis by reducing matrix interference and ensuring a purer final sample.

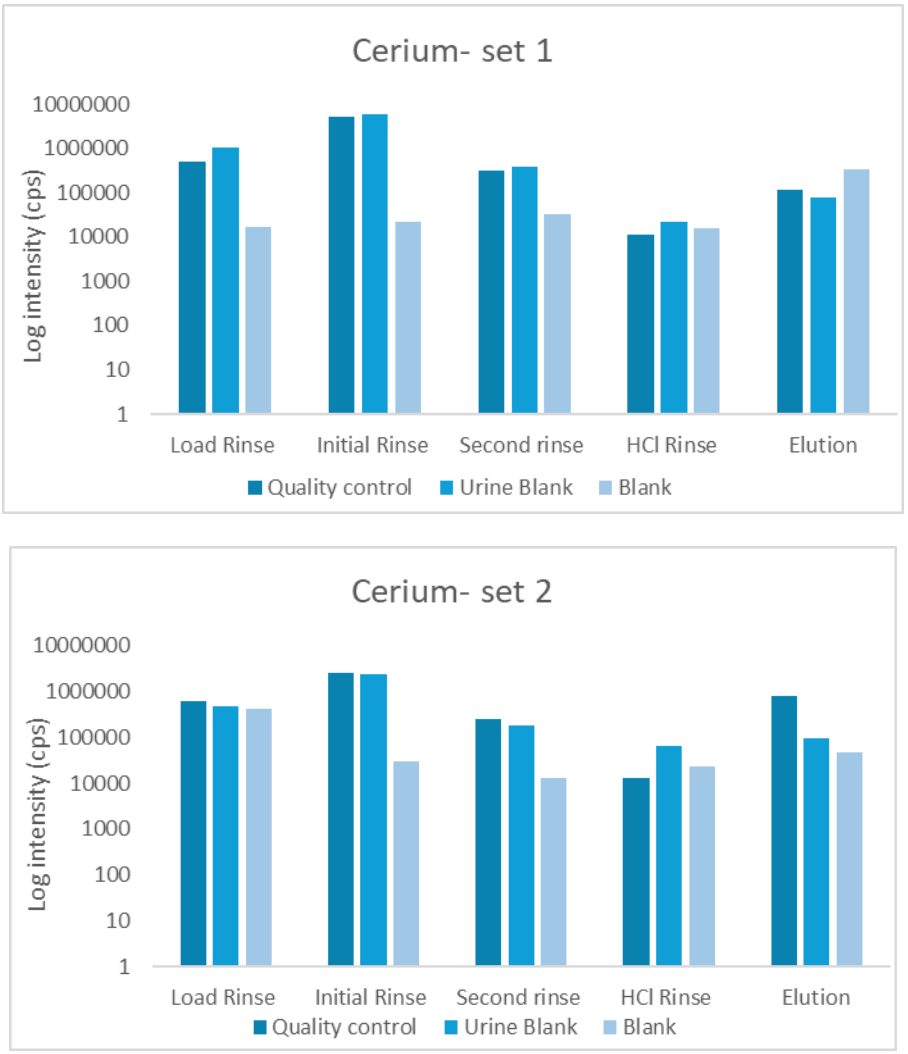


Figure 8. Histograms showing the distribution of Cerium in the rinses and elution steps. The data is presented as the log10 of isotope counts per second (cps), with acid-blank values subtracted.

Impact on Sample Purity and TIMS Analysis: ICP-MS screening revealed that matrix elements, particularly iron (inferred by presence of chromium) could significantly interfere with the subsequent TIMS analysis by altering the chemical speciation of plutonium. These transition metals have the potential to form stable complexes with plutonium, especially under the conditions of the purification process. The log-transformed data further suggests that if these matrix elements are not adequately removed, they may distort the isotopic distribution or mass spectra of plutonium, leading to inaccuracies in TIMS analysis. The presence of these elements can cause shifts in the plutonium isotopic ratios or affect the mass spectra, thus compromising the accuracy and reliability of the plutonium measurements.

To mitigate this interference, additional purification steps are essential. Strategies such as supplementary purification loops, which can target specific contaminants, or the use of microfluidic systems for more precise separation, could help further reduce matrix interference. These advanced purification methods would enhance plutonium recovery by selectively removing problematic matrix elements, ensuring a higher purity sample for TIMS analysis. This approach would improve the accuracy of the plutonium isotopic ratios, thereby yielding more reliable results and minimizing the impact of matrix-induced errors.

FUTHER WORK & MICROFLUIDIC R&D

The initial experiments and screenings have identified specific areas where the bioassay process can be optimized using microfluidic technology. These findings highlight opportunities to improve key parameters such as reducing sample volume, minimizing reagent usage, and enhancing detection capabilities. We anticipate that these changes will significantly improve the timeliness, sensitivity, and overall efficiency of analyses, while also enhancing worker safety and ensuring stronger regulatory compliance. These improvements hold promise for optimizing purification steps, leading to higher yields, more reliable results, and faster bioassay processes. In particular, the presence of elements from stainless steel planchets especially iron, presents challenges by complexing with plutonium. This leads to both plutonium loss and matrix interference during the leaching and purification process. The data underscores the importance of refining the purification process to minimize these interferences and improve plutonium recovery. We propose the inclusion of additional purification and rinsing steps (Figure 9) to address these challenges. Integrating microfluidic systems into the PuTIMS process here would improve analytical capability without drastically increasing reagent consumption.

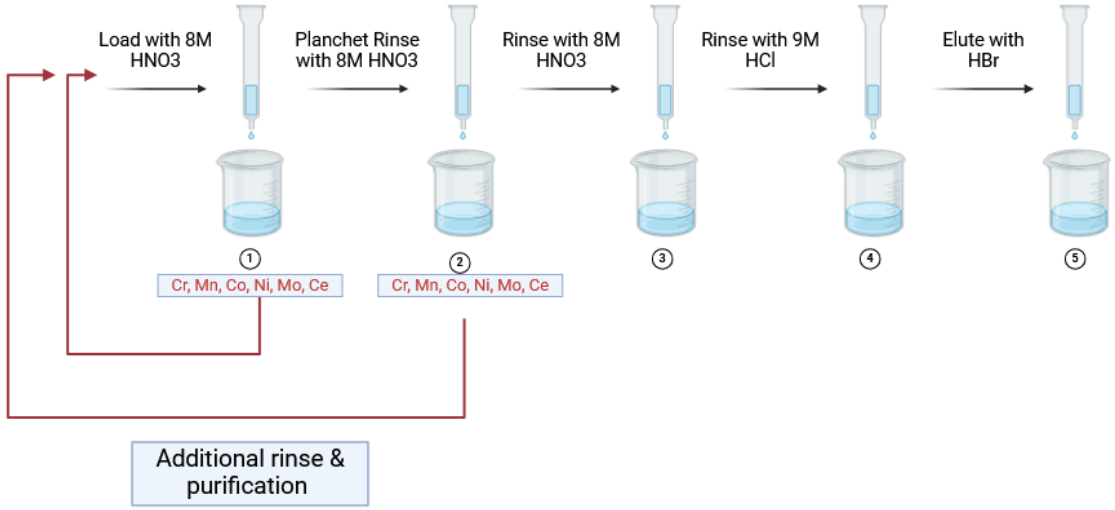


Figure 9. Suggested placement of additional microfluidic processes.

The ongoing research and development of bioassay methods at LANL holds broad applicability for advancing practices across industries, particularly those relying on stringent safety and compliance measures. In sectors like nuclear energy, where global nuclear fuel cycle activities are expanding, the industry requires stringent safety measures to protect workers and ensure compliance with regulatory standards. By incorporating microfluidic technology into bioassay programs, we can improve data resolution, reduce operational costs, and enhance worker safety. This technology allows for more frequent sampling, increasing turnover and better supporting the As Low As Reasonably Achievable (ALARA) principle, which helps minimize the risk of missing rapidly excreted isotopes, such as uranium, while enabling more precise exposure assessments. To achieve these goals, our initial work focused on screening the PuTIMS chemistry process to identify areas where microfluidic systems can optimize the PuTIMS chemistry, we aim to improve yields, minimize purification interferences, and expedite bioassay processes. The resulting enhancements will deliver high-quality, reliable results more quickly, setting new benchmarks for bioassay practices not only in the nuclear sector but also in other industries that require robust, accurate, and efficient safety assessments.

In the long term, the adoption of microfluidic technology in bioassay programs will enable faster, more efficient, and higher sensitivity measurements that contribute to worker exposure monitoring. It will support the nuclear sector's ongoing commitment to worker safety and regulatory compliance while promoting more cost-effective and sustainable laboratory practices. As this technology evolves, we foresee its integration

driving further innovations, improving both operational workflows and the long-term viability of monitoring radioactive materials in a safe and environmentally responsible manner. Ultimately, microfluidic systems will become an important method for bioassay monitoring, enhance the nuclear industry's ability to protect workers, manage potential occupational exposures more efficiently, and meet measurement quality requirements.

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