Research Square

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Electrodeposition of Highly Enriched Uranium Thin Films for Advanced Fission Detectors

Brian W. Miller
brian.miller@inl.gov
Idaho National Laboratory
Brian K. Harris
Idaho National Laboratory
Michael A. Reichenberger
Idaho National Laboratory
R. Duane Ball
Idaho National Laboratory
Harry W. Rollins
Idaho National Laboratory
Dani G. Ottaway
Idaho National Laboratory
Delaney Stine
Idaho National Laboratory
Andrew W. Holland
Idaho State University

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Abstract

Real-time, in-core neutron flux monitoring is essential for characterizing irradiation conditions in research reactors. Micro-Pocket Fission Detectors (MPFDs) operate as an ionization chamber in which a thin fissile coating electrodeposited onto a platinum substrate generates measurable electrical pulses upon neutron-induced fission, enabling continuous, real-time flux monitoring. Optimal sensor performance requires a deposited uranium mass of 0.3 to 1.0 μg, sufficient to produce a measurable fission rate while remaining thin enough to allow fission fragments to escape into the gas cavity. This article describes the electrodeposition of 93% Highly Enriched Uranium (HEU) onto platinum (Pt) substrates to achieve this target mass, along with a standardized protocol for quantifying the deposited mass using alpha-particle spectrometry. The efficiency of thin-film formation by cyclic voltammetry (CV) was compared to bulk electrolysis (BE) to identify the optimal deposition method. ICP-MS independently validated the radiometric mass calculations. Optimizing both the fabrication and quantification techniques facilitates the development of reliable, high-resolution sensors suitable for deployment in diverse research reactor environments.

1. Introduction

Time-dependent and spatially dependent neutron flux measurements are critical for evaluating the performance of new materials and fuels in research reactors. Traditional passive sensors, such as the uranium-aluminum fission wires currently used to characterize fission rates at the Advanced Test Reactor Critical Facility (ATR-C) [12], solely provide integral fluence data upon completion of an experiment, while in-situ sensors provide real-time data capturing the evolution of physical phenomena over time. Micro-Pocket Fission Detectors (MPFDs) enable high-speed, real-time neutron flux measurements in high-radiation environments within a variety of research reactors [1, 3]. The deployment of these sensors require the fabrication of ultra-thin neutron-sensitive layers to achieve high sensitivity while minimizing disruption to the local neutron flux [1, 3]. Specific experimental constraints dictate the deposited mass of fissile material remain sufficiently low to avoid signal attenuation effects degrading measurement accuracy [4]. Previous research has developed deposition methods for natural and low-enriched uranium using cyclic voltammetry (CV) [4, 5]. This project builds upon those efforts by characterizing the deposition of 93% Highly Enriched Uranium (HEU) on platinum (Pt) substrates.

The MPFD operates as an ionization chamber [9, 10]. A thin fissile coating, in this case electrodeposited HEU, is applied to a platinum electrode housed within a small gas-filled cavity. When the sensor is placed inside a nuclear reactor, the high neutron flux causes fission events within the ^{235}U atoms of the coating. Each fission produces two high-energy fission fragments that travel in opposite directions, carrying roughly 160–170 MeV of kinetic energy. Due to the planar geometry of the deposited film, one fragment is emitted into the substrate and absorbed, while the other escapes the thin uranium film and enters the gas cavity, where it ionizes the fill gas, creating ion pairs along its path. An applied electric field collects the resulting free electrons and ions, generating a discrete electrical pulse for each fission event. The pulse rate is directly proportional to the neutron flux, enabling real-time, continuous monitoring throughout a reactor experiment. This capability is particularly valuable for capturing transient flux profiles, power ramps, and spatial flux variations across the reactor core, data entirely unavailable from passive dosimetry methods.

The thickness of the fissile coating is the critical fabrication parameter governing sensor performance [9, 10]. If the coating is too thin, insufficient ^{235}U atoms are present to produce a measurable fission rate. If the coating is too thick, fission fragments from deeper within the film lose energy before escaping into the gas cavity and are never detected, degrading the signal. The penetration depth of fission fragments in solid materials is typically less than 1 μm, while fission fragments can travel several centimeters in gases [9]. The optimal mass range for these sensor geometries is 0.3 to 1.0 μg of deposited uranium [4, 7]. Achieving this target reliably and reproducibly across multiple sensors is a prerequisite for deploying multi-node sensor arrays capable of capturing spatially resolved flux profiles within the reactor core [9, 10].

The primary objective of this study is to optimize and demonstrate the electrodeposition chemical process for depositing 93% HEU onto Pt substrates within the target mass range, validated using multiple mass-quantification methods: alpha-particle spectrometry, which allows for the correlation of measured activity with the actual mass of the deposited material [6], and ICP-MS, which provides independent confirmation of the radiometric mass calculations. This study compares CV versus bulk electrolysis (BE) to determine the optimal method for producing consistent, sub-microgram fissile layers for use in the fabrication of optimized sensors to enable high-fidelity flux monitoring in research reactors.

93% HEU was selected for this study for consistency with the existing flux-monitoring practice at ATR-C, which has used HEU-based flux monitors since its startup [12]. ATR-C's existing monitors employ HEU in a larger wire geometry to measure very low neutron flux; by contrast, the MPFD application addressed here requires a very small deposited mass to measure comparatively high neutron flux, making the high ^{235}U enrichment of HEU advantageous: the elevated ^{235}U content provides sufficient neutron-capture sensitivity per microgram of deposited uranium despite the small total mass. Ongoing regulatory efforts to transition research reactor fuels and materials toward High-Assay Low-Enriched Uranium (HALEU) motivate future work bridging the deposition and quantification methods developed here from HEU to HALEU source material.

2. Materials and Methods

This section describes the solution preparation and electrodeposition of 93% HEU onto Pt substrates. Electrodeposition utilized both CV and BE to deposit the fissile material, allowing for a comparison of the effectiveness of each technique.

2.1. Substrate Fabrication

The experimental substrates consisted of 250 μm diameter Pt electrodes deposited via deposition onto polished alumina wafers with a diameter of 3.175 mm and thickness of 500 μm. Each substrate consisted of a 0.005 μm titanium adhesion layer followed by a 0.05 μm platinum electrode layer deposited onto the alumina wafer surface. These substrates were fabricated using metal lift-off techniques to ensure surface uniformity and precise geometric definitions required for the MPFD design [4, 5]. Platinum was selected as the working electrode material due to its corrosion resistance during the electrolytic process and its low neutron absorption cross-section [4].

2.2. Solution Preparation

The electrolytic solution was prepared using 93% HEU to meet the sensitivity requirements for research reactor experiments. This method is similar to previous studies [4, 5] in employing a slightly buffered uranyl nitrate, $UO_2(NO_3)_2$. Instead of preparing a fresh electrodeposition solution for each individual deposition run, a primary stock solution of $UO_2(NO_3)_2$ and a separate working solution of buffered ammonium nitrate (NH_4NO_3) were used for all electrodeposition runs [11]. The electrodeposition process is highly sensitive to electrolyte pH, which strongly influences uranium deposition yield. At pH values below 2.0, the deposited uranium layer can redissolve into the electrolyte, while at pH values above 4.0, uranium tends to form hydroxide complexes of varying stoichiometry that inhibit deposition [11]. For these reasons, the working solution pH was carefully controlled to 3.4 throughout all deposition runs.

2.3. Electrodeposition Protocols

Two distinct electrodeposition techniques were evaluated to determine the optimal method for achieving the target fissile mass of approximately 1.0 µg. A total of 13 samples were prepared using CV and eight samples were prepared using BE.

Cyclic Voltammetry

The first set of samples was prepared using the cyclic potential sweep method. The potential of the working electrode was swept between − 0.62 V and − 0.64 V relative to an Ag/AgCl reference electrode at a scan rate of 0.01 V/s. This method was previously identified as effective for producing uniform, thin layers of uranium [4, 5]. The number of sweep cycles varied from 10 to 100 to assess both the increase in deposited uranium mass per cycle and the maximum quantity of material that could be deposited before the method lost effectiveness.

Bulk Electrolysis

The second set of samples was prepared using a BE approach at an applied potential of − 0.325 V relative to the Ag/AgCl reference electrode using a CHI620E potentiostat. Analysis of the raw potentiostatic data revealed the experiment was conducted under constant-potential (potentiostatic) rather than constant-current conditions, with the current decaying naturally over the deposition period. Samples in the BE 2 series were terminated at fixed time intervals (60, 120, 300, and 600 seconds), while samples in the BE 3 series were terminated automatically when the current declined to 20% of its initial value (End Current Ratio = 20%), resulting in actual deposition durations of 53, 62, 63, and 85 seconds for BE 3A through BE 3D, respectively. Sample BE 2E was similarly terminated at an End Current Ratio of 10%, yielding an actual duration of 120 seconds. Initial current densities ranged from approximately 66 to 115 mA/cm² across all samples, declining to between 4.5 and 17.1 mA/cm² at termination, with average current densities ranging from 8.5 to 25.0 mA/cm

2.4. Alpha-Particle Spectrometry and Analysis

^{234}U is not the isotope used for real-time flux monitoring in the final sensor; rather, its comparatively high specific activity (approximately 231 MBq/g, versus approximately 80 kBq/g for ^{235}U) makes it a practical non-destructive alpha-spectrometric indicator for assessing the relative quantity of uranium deposited on the MPFD substrate, independent of its role in neutron detection. ^{234}U enriches quantitatively and proportionally

with ^{235}U throughout the electrodeposition process, allowing its measured activity to serve as a reliable proxy for total deposited uranium mass. While ^{235}U activity can also be measured directly, its long half-life results in a much lower specific activity than ^{234}U, making it less suitable than ^{234}U for a rapid, non-destructive post-fabrication mass verification method.

Following deposition, the samples were rinsed with isopropyl alcohol and air-dried. The quantity of electrodeposited uranium was determined using an ORTEC Octête Plus high-resolution alpha-particle spectrometer manufactured by AMETEK ORTEC [8] equipped with passivated implanted planar silicon (PIPS) detectors. The spectrometer was calibrated for energy and efficiency using a standard radionuclide source (Analytics, Atlanta, GA; Source ID: 64001-121), consisting of a 24.2 mm diameter × 0.75 mm thick stainless steel disk onto which a mixture of alpha-emitting radionuclides was electrodeposited. The calibration source maintains traceability to the National Institute of Standards and Technology (NIST) through Measurement Assurance Programs as described in USNRC Regulatory Guide 4.15, Rev. 1. Source activities were certified on June 10, 2002, with a total activity of 381.6 dpm and an expanded uncertainty of 11.5 dpm (k = 3). The isotopic composition and associated parameters of the calibration source are summarized in Table 4.

Table 1
Analytics Standard Radionuclide Source (ID: 64001-121), Calibration Source Properties

Isotope	Activity (dpm)	Half-Life	Expanded Uncertainty, dpm (k = 3)	Energy Range (keV)
U-238	93.1	4.468×10^9 y	3.2	3900–4290
U-234	91.8	2.455×10^5 y	3.2	4560–4880
Pu-239	97.4	2.41×10^4 y	3.4	4900–5240
Am-241	94.4	4.322×10^2 y	3.3	5260–5620
Note: U-235 impurity reported as < 4 dpm. Active area diameter: 24.2 mm. Calibration date: June 10, 2002. Total activity: 381.6 dpm; expanded uncertainty: 11.5 dpm (k = 3).				

Due to the varying activity of the deposited material, count times were extended to minimize total peak uncertainty where possible rather than adhering to a strict percentage threshold. The net activity in picocuries (pCi) was converted to total uranium mass using the specific activity of ^{234}U (6.23×10^{-3} Ci/g) [2] and the ^{234}U weight fraction of 1.005% for the 93% HEU source material, verified via ICP-MS intensity analysis with concurrent natural uranium isotopic standards run on the same instrument to characterize measurement bias (Section 3.4), yielding a conversion factor of 1.597×10^{-2} µg total U per pCi ^{234}U. Equivalently, the specific activity of pure ^{234}U can be expressed as 6,230 pCi per µg ^{234}U, which is the form used later (Section 3.4) to convert ICP-MS-derived ^{234}U mass back into an activity value for direct comparison with the alpha spectrometry measurements.

2.5. Inductively Coupled Plasma Mass Spectrometry

To independently validate the radiometric mass calculations, a subset of samples was analyzed using inductively coupled plasma mass spectrometry (ICP-MS). Prior to analysis, total specimen mass was measured and recorded for each disc. Analyses were performed using a Thermo Scientific iCAP TQ ICP-MS operated in Kinetic Energy Discrimination (KED) mode. The instrument was calibrated using certified uranium

and platinum metal standards from NIST-traceable sources, using six increasing concentrations to establish the calibration curve; the uranium standards were prepared from depleted U stock, with isotopic abundances for ^{234}U, ^{235}U, and ^{238}U established using a separate natural uranium reference source. Data acquisition and processing were performed using Qtegra software (version 2.18.5492.610) in isotopic calibration mode.

3. Results

The electrodeposition experiments produced quantitative data on mass deposition kinetics, method efficiency, and isotopic characterization of the resulting HEU layers. Alpha-particle spectrometry and ICP-MS were used to independently verify the deposited fissile mass.

Table 2
Cyclic Voltammetry Radiometric Results. All uncertainty values are 1 total propagated uncertainty (TPU) in pCi as reported by AlphaVision v6.0.

σ						
Sample ID	**Cycles**	**^{234}U Activity (pCi)**	**^{235}U Activity (pCi)**	**^{238}U Activity (pCi)**	**Meas. Unc. 1σ TPU (pCi)**	**Det. Eff. (%)**
9A	100	7.35×10^{-1}	—	—	1.60×10^{-2}	32.97
9B	100	7.28×10^{-1}	—	—	1.65×10^{-2}	32.53
9C	100	7.14×10^{-1}	—	—	1.50×10^{-2}	34.27
9E	100	6.40×10^{-1}	—	—	1.43×10^{-2}	26.70
10A	100	1.31	—	—	2.71×10^{-2}	26.60
10B	100	1.83	—	—	3.72×10^{-2}	26.21
10C	100	7.27×10^{-1}	—	—	1.54×10^{-2}	33.93
10 Cycles	10	2.93×10^{-1}	3.10×10^{-2}	—	3.36×10^{-2}	32.97
20 Cycles	20	6.48×10^{-1}	5.42×10^{-2}	3.30×10^{-2}	2.82×10^{-2}	32.53
30 Cycles	30	8.69×10^{-1}	1.03×10^{-1}	5.70×10^{-2}	3.70×10^{-2}	34.27
50 Cycles	50	9.76×10^{-1}	1.33×10^{-1}	9.85×10^{-2}	3.29×10^{-2}	31.56
100 Cycles	100	1.23	1.15×10^{-1}	6.41×10^{-2}	4.05×10^{-2}	26.70

Table 3

Bulk Electrolysis Radiometric Results. All uncertainty values are 1 total propagated uncertainty (TPU) in pCi as reported by AlphaVision v6.0.

σ							
Sample ID	**Method**	**Dep. Time (s)**	**^{234}U Activity (pCi)**	**^{235}U Activity (pCi)**	**^{238}U Activity (pCi)**	**Meas. Unc. 1σ TPU (pCi)**	**Det. Eff. (%)**
BE 2B	Bulk	60	1.721	1.48×10^{-1}	3.32×10^{-2}	4.77×10^{-2}	32.97
BE 2C	Bulk	300	1.340	1.13×10^{-1}	6.68×10^{-2}	3.64×10^{-2}	32.53
BE 2D	Bulk	600	1.055	1.25×10^{-1}	6.21×10^{-2}	3.82×10^{-2}	34.27
BE 2E	Bulk	120	2.394	1.36×10^{-1}	6.18×10^{-2}	5.34×10^{-2}	31.56
BE 3A	Bulk	53	1.344	1.19×10^{-1}	6.15×10^{-2}	4.12×10^{-2}	26.70
BE 3B	Bulk	62	1.050	9.07×10^{-2}	5.32×10^{-2}	3.72×10^{-2}	26.60
BE 3C	Bulk	63	1.149	9.62×10^{-2}	5.24×10^{-2}	3.87×10^{-2}	26.21
BE 3D	Bulk	85	1.255	8.83×10^{-2}	4.81×10^{-2}	3.89×10^{-2}	27.51

Table 3

a. Bulk Electrolysis Current Density Summary. Current density calculated using Pt electrode surface area of 4.91 × 10− 4 cm2 (250 µm diameter).

Sample ID	**Dep. Time (s)**	**End Current Ratio (%)**	**Initial CD (mA/cm^2)**	**Final CD (mA/cm^2)**	**Avg CD (mA/cm^2)**	**Total Charge (C)**
BE 2B	60	0	115.4	17.1	25.0	7.37×10^{-4}
BE 2C	300	0	90.2	7.4	12.6	1.85×10^{-3}
BE 2D	600	0	77.6	4.5	8.5	2.50×10^{-3}
BE 2E	120	10	68.4	11.1	15.3	9.04×10^{-4}
BE 3A	53	20	66.4	13.2	18.0	4.69×10^{-4}
BE 3B	62	20	66.0	13.2	17.5	5.32×10^{-4}
BE 3C	63	20	65.8	13.1	17.9	5.53×10^{-4}
BE 3D	85	20	66.2	13.2	16.9	7.06×10^{-4}

3.1. Deposition Kinetics and Saturation Thresholds

The correlation between the number of potential sweep cycles and mass accumulation was analyzed to determine the operational limits of the CV method. Experimental data from the 10-cycle to 100-cycle trials

revealed a linear increase in uranium deposition during the initial stages. The sample prepared with 10 cycles yielded a net ^{234}U activity of 0.293 ± 0.034 pCi. Increasing the cycle count to 100 raised the net activity to 1.226 ± 0.040 pCi.

A similar slowing in the rate of deposited activity was observed in the BE samples at extended deposition times. Samples prepared via constant-potential BE yielded net ^{234}U activities ranging from 1.05 to 2.39 pCi across deposition times of 60 to 600 seconds. The decline in activity at longer BE durations likely reflects a combination of mass transport limitations as the uranium concentration near the electrode surface becomes locally depleted, and the potential for partial re-oxidation or re-dissolution of the deposited layer under the prevailing solution chemistry.

3.2. Comparative Efficiency of Deposition Methods

A comparison of the two electrodeposition methods demonstrated BE achieved greater mass deposition rates than CV. Samples prepared via BE for 120 seconds (e.g., Sample BE 2E) achieved activities up to 2.394 ± 0.053 pCi. The CV method required significantly longer total processing times to approach a nominal value of 1.3 pCi. The optimized BE protocol used for the BE 3 series produced an average activity of 1.20 ± 0.15 pCi. These data confirm maintaining controlled solution pH and applying a constant potential enhanced the reproducibility of the fabrication process compared to prior trials.

3.3. Characterization of HEU Layers

Alpha-particle spectrometry analysis confirmed the successful deposition of 93% HEU across the BE 2 and BE 3 sample series. Using the corrected specific activity conversion factor of 1.597×10^{-2} µg total U per pCi ^{234}U, alpha-derived total uranium masses ranged from 0.0168 to 0.0382 µg for the BE2 series and 0.0168 to 0.0215 µg for the BE 3 series. Independent validation of these mass values by ICP-MS is discussed in Section 3.4. The energy spectra for these samples displayed distinct peaks for ^{234}U and ^{235}U without significant low-energy tailing, indicating the films were sufficiently thin to minimize self-absorption while providing the signal intensity required for reactor flux monitoring [1].

3.4. ICP-MS Validation

ICP-MS analysis was conducted on six electrodeposited samples (BE 2B, BE 2C, BE 2D, BE 3B, BE 3C, and BE 3D) to independently verify the radiometric mass calculations and confirm the isotopic signature of the deposited HEU material. The isotopic composition of the 93% HEU source material, averaged across two independent measurement sets and verified via ICP-MS signal intensity analysis, is U-234: 1.005%, U-235: 93.421%, and U-238: 5.574%, yielding a theoretical $^{238}U/^{235}U$ ratio of 0.0597. To characterize instrumental measurement bias, five natural uranium check standards were analyzed using the same instrument run and calibration as the HEU samples. The measured natural uranium composition averaged U-234: 0.00505%, U-235: 0.679%, and U-238: 99.316%, compared to accepted natural abundance values of U-234: 0.005%, U-235: 0.720%, and U-238: 99.274%. This corresponds to a $^{238}U/^{235}U$ ratio of approximately 146:1 in the natural standards, compared to the accepted natural ratio of approximately 138:1, indicating an approximately 6% measurement bias affecting the ^{235}U determination under these instrument conditions. The ICP-MS measured average isotopic composition across the six HEU samples was U-234: 1.037%, U-235: 93.143%, U-238: 5.820%,

in reasonable agreement with the source material composition given the characterized measurement bias ($^{238}U/^{235}U$ ratio within 4.7% of theoretical based on average composition, ranging from 1.6% to 13.6% across individual samples), indicating the isotopic enrichment of the deposited material was largely preserved throughout the electrodeposition process.

ICP-MS derived total uranium masses, calculated using each sample's actual measured dilution mass rather than an assumed fixed volume, ranged from 0.2289 to 0.3559 µg across the six samples (average 0.2849 µg). Using the corrected specific activity conversion factor of 1.597×10^{-2} µg total U per pCi ^{234}U (equivalently, 6,230 pCi per µg pure ^{234}U), the corresponding ^{234}U activities were calculated from the ICP-MS masses and compared directly to the alpha spectrometry measurements. For example, for sample BE 2B: the ICP-MS ^{234}U mass yields a calculated activity of 2.314 pCi, compared to the alpha spectrometry measurement of 1.721 pCi, giving an alpha spectrometry to mass spectrometry (AS/MS) ratio of 74.3%. The AS/MS ^{234}U activity ratios ranged from 56.6% to 79.2% across all six samples, with an average recovery of 69.4%. This systematic underestimation of activity by alpha spectrometry relative to ICP-MS is more consistent with a calibration effect (e.g., energy or efficiency calibration, or detector-source geometry) than with self-attenuation of alpha particles within the electrodeposited film. A calibration effect would be expected to produce a systematic offset across samples without distorting peak shape, consistent with the alpha energy spectra showing no significant low-energy tailing (Section 3.3), whereas self-attenuation is expected to scale with film thickness and mass; the weak observed correlation between deposited mass and AS/MS recovery ($r \approx -0.27$) does not support a mass-dependent attenuation effect. Further investigation, including alpha spectrometer calibration verification and a larger sample set, would be needed to confirm calibration as the source of this discrepancy.

The ^{234}U isotope demonstrated the most consistent agreement between the two techniques, supporting its use as the primary activity indicator for radiometric mass quantification. A second dissolution pass of the original discs may help quantify the degree of residual uranium retention and further characterize the attenuation behavior. The complete isotopic data and AS/MS comparisons are summarized in Table 5.

Table 4
ICP-MS Uranium Isotopic Data for Selected BE Samples. Masses calculated using actual measured per-sample dilution mass, not a fixed 10 mL assumption.

Sample ID	^{234}U Conc. (ppb)	^{235}U Conc. (ppb)	^{238}U Conc. (ppb)	Total U Mass (µg)	ICPMS ^{234}U (pCi)	Alpha Spec ^{234}U (pCi)	AS/MS Ratio (%)	$^{238}U/^{235}U$ Meas.	$^{238}U/^{235}U$ Theor.
BE 2B	0.03716	3.3209	0.2251	0.3559	2.314	1.721	74.3	0.0678	0.0597
BE 2C	0.03127	2.8414	0.1774	0.3073	1.947	1.314	67.5	0.0624	0.0597
BE 2D	0.03010	2.7170	0.1647	0.2948	1.874	1.060	56.6	0.0606	0.0597
BE 3B	0.02840	2.5444	0.1570	0.2750	1.768	1.050	59.4	0.0617	0.0597
BE 3C	0.02333	2.0987	0.1280	0.2289	1.452	1.150	79.2	0.0610	0.0597
BE 3D	0.02555	2.2691	0.1394	0.2473	1.591	1.260	79.2	0.0614	0.0597

Average recovered mass 0.2849 µg, range 0.2289–0.3559 µg. AS/MS ratios unaffected by the dissolution-mass correction, since both alpha spec and ICP-MS-derived activities scale by the same factor.

4. Discussion

The experimental data identified the operational boundaries for uranium electrodeposition on planar electrodes, highlighted the physical limitations of the CV method, and confirmed that the fabricated sensors meet the mass and isotopic specifications required for high-fidelity reactor flux monitoring. Planned reactor irradiation of the fabricated MPFD sensors to confirm operational performance under neutron flux was not completed as part of this study due to scheduling delays, and remains recommended for future work.

4.1. Evaluation of Mass Deposition Limits

Experimental results revealed a diminishing rate of uranium mass deposition using CV on a planar electrode as cycle count increased. Data from the CV trials showed a linear increase in alpha activity up to 100 cycles, with the deposition rate slowing as the cycle count increased, consistent with the progressive decrease in cathodic current observed in cyclic voltammograms reported in prior work [4, 5]. This behavior is consistent with deposition becoming increasingly competitive with a concurrent oxidative stripping process at the electrode surface, as discussed in Section 3.1. BE effectively deposited thicker layers, reaching a substantially higher activity plateau than CV achieved over the range tested, indicating that BE reaches a greater deposited mass before encountering a comparable limitation.

The optimal fissile mass for these sensor geometries ranged between 0.3 µg and 1.0 µg. This mass range provided sufficient signal intensity for real-time monitoring while maintaining a sufficiently thin film to prevent excessive alpha self-absorption [4]. ICP-MS-confirmed total masses (Section 3.4), corrected for actual per-sample dissolution volume, fall within or near this target range (0.229–0.356 µg); the lower alpha-spectrometry-derived masses (Section 3.3) reflect the combined effect of the volume correction and the unresolved systematic AS/MS discrepancy discussed in Section 3.4, rather than indicating the target mass was not achieved.

4.2. Methodological Improvements and Reproducibility

Comparative analysis of the two deposition techniques indicated BE offered distinct advantages over CV for the production of higher-mass samples. CV provided precise control for depositing extremely thin layers [4]. The BE method demonstrated a higher deposition rate and superior time-efficiency for reaching the 1.0 µg target. ICP-MS-confirmed masses, corrected for actual dissolution volume (Section 3.4), indicate BE depositions approached the lower end of the 0.3–1.0 µg target range (up to 0.356 µg for BE 2B), though consistent achievement of the full target range across all samples was not demonstrated in this study. The stability observed in the BE 3 sample series (Samples BE 3A through BE 3D) was attributed to strict control of the electrolytic solution chemistry. Precision pipetting maintained the pH at 3.4, consistent with prior protocols identifying controlled electrolyte pH as critical for preventing the premature hydrolysis of uranium and re-dissolution of the deposit in acidic conditions, both recognized sources of inconsistency in earlier electrodeposition protocols [6, 11]. These procedural refinements enabled reliable fabrication of multiple

sensors with nearly identical fissile masses, a critical requirement for deploying multi-node sensor arrays in reactor experiments.

5. Conclusion

This investigation confirms BE provides a more efficient mechanism than CV for depositing disc-shaped fissile masses approaching the 0.3 to 1.0 μg target required for MPFD operation in research reactors, with ICP-MS-confirmed masses (corrected for actual per-sample dissolution volume) reaching up to 0.356 μg. Cycle-dependence data up to 100 cycles define the operational range of the CV process characterized in this study, with a diminishing deposition rate at higher cycle counts attributed to a reduction in electrochemical effectiveness as the uranium layer builds up on the electrode surface. BE utilizing a constant potential improves on the result under the same limitation, achieving higher deposited masses with superior reliability. Optimal electrolytic solution chemistry requires maintaining pH at 3.4 to prevent uranium hydrolysis and re-dissolution, consistent with prior protocols [6, 11].

Spectral analysis of the 93% HEU samples demonstrated the fabrication process preserved the required isotopic signature. Higher-activity samples yielded well-resolved alpha energy peaks for ^{234}U and ^{235}U. The distinct peak resolution confirmed the electrodeposited films remained sufficiently thin to minimize alpha self-absorption, ensuring accurate radiometric mass quantification [4].

ICP-MS provided independent validation of the electrodeposition methodology. The measured $^{238}U/^{235}U$ isotopic ratios agreed with the theoretical value to within 4.7% on average (ranging from 1.6% to 13.6% across individual samples), indicating the isotopic signature of the 93% HEU was largely preserved throughout the deposition process. Alpha spectrometry recovered an average of 69.4% of the ^{234}U activity measured by ICP-MS; the alpha spectra showed no significant low-energy tailing and the correlation with deposited mass was weak, so this discrepancy does not appear to be explained by film self-attenuation and is more consistent with a calibration effect. A larger sample set, along with alpha spectrometer calibration verification, would be needed to fully quantify and confirm the source of this discrepancy.

The fabricated sensors meet the operational criteria defined by computational simulations for real-time in-core measurements [7]. Further investigation, including coulometric analysis of the current-time data and post-deposition surface microscopy, would help distinguish between mass transport limitations and partial re-dissolution of the deposit as contributing mechanisms to the observed activity decline at extended deposition times, and is recommended for future work.

Declarations

Author Contributions

Brian W. Miller: Lead author, original draft preparation, review and editing, data analysis, investigation, and validation. Brian K. Harris: Methodology, investigation, performed electrolysis of samples, data curation, and review and editing. Michael A. Reichenberger: Supervision, conceptualization, methodology, validation, resources, and review and editing. R. Duane Ball: Investigation, ICP-MS analysis, data curation and analysis, and review and editing. Harry W. Rollins: Investigation, performed electrolysis of samples, and resources. Dani

G. Ottaway, Delaney Stine: Reviewing and editing. Andrew W. Holland: Formal analysis, resources, validation, writing – review & editing.

Declaration of Competing Interest

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During the preparation of this manuscript, the authors used Claude (Anthropic) to assist with editing, organizing, and formatting the text and figures, including verification of internal data consistency across tables and text. All content was reviewed, verified, and approved by the authors. The authors take full responsibility for the accuracy and integrity of the published work. Generative AI was not used to create or alter any research data.

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Figures

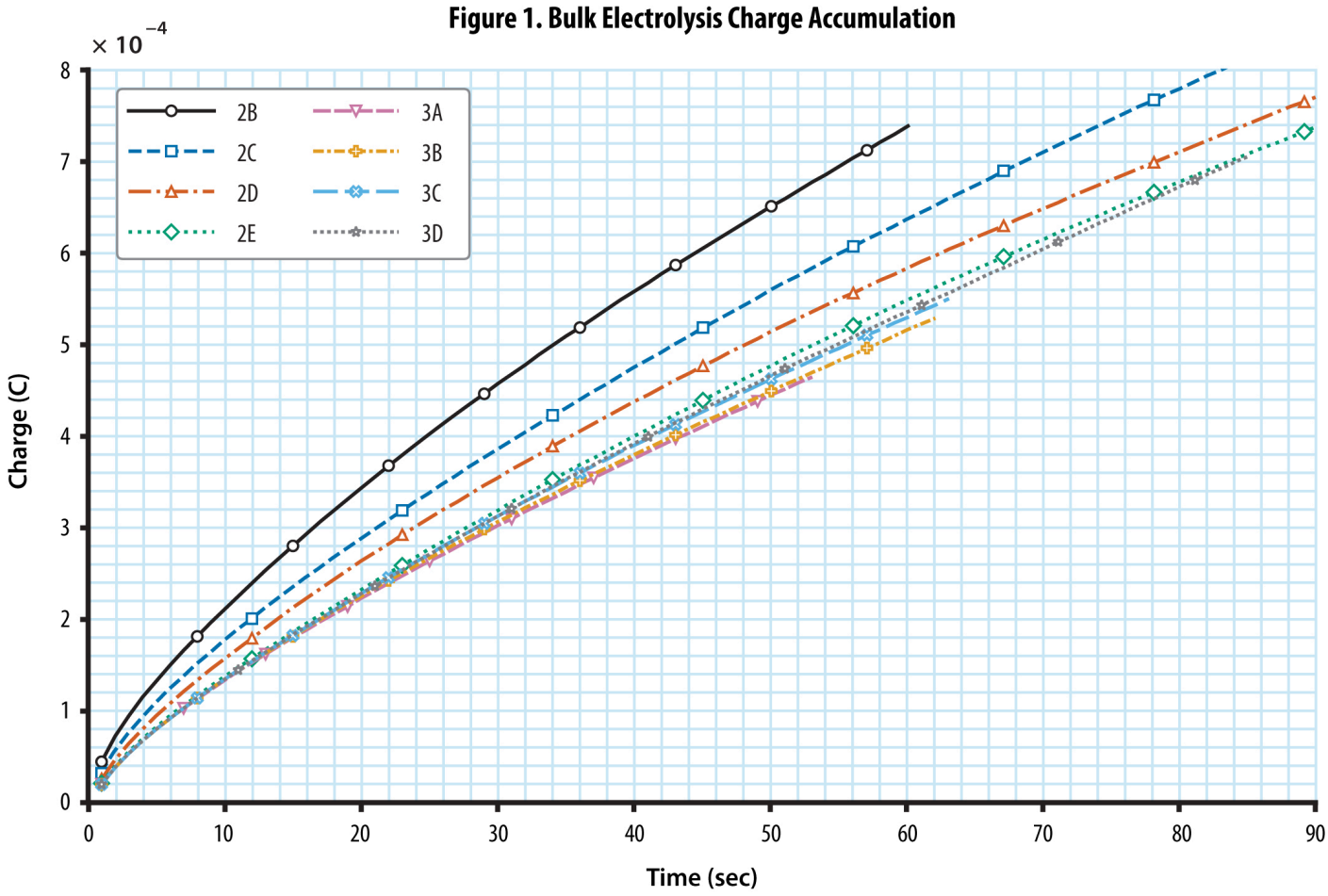


Figure 1

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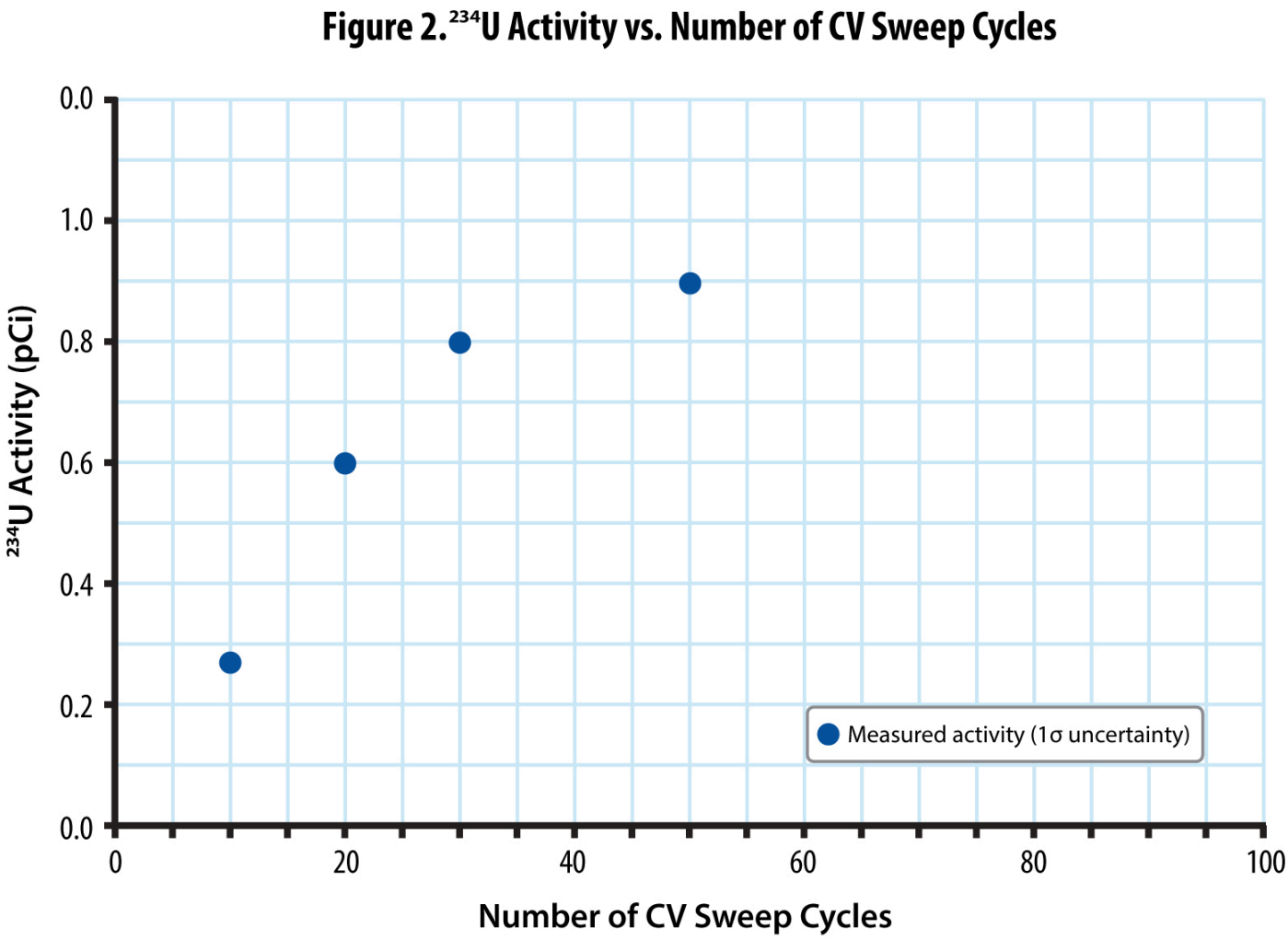


Figure 2

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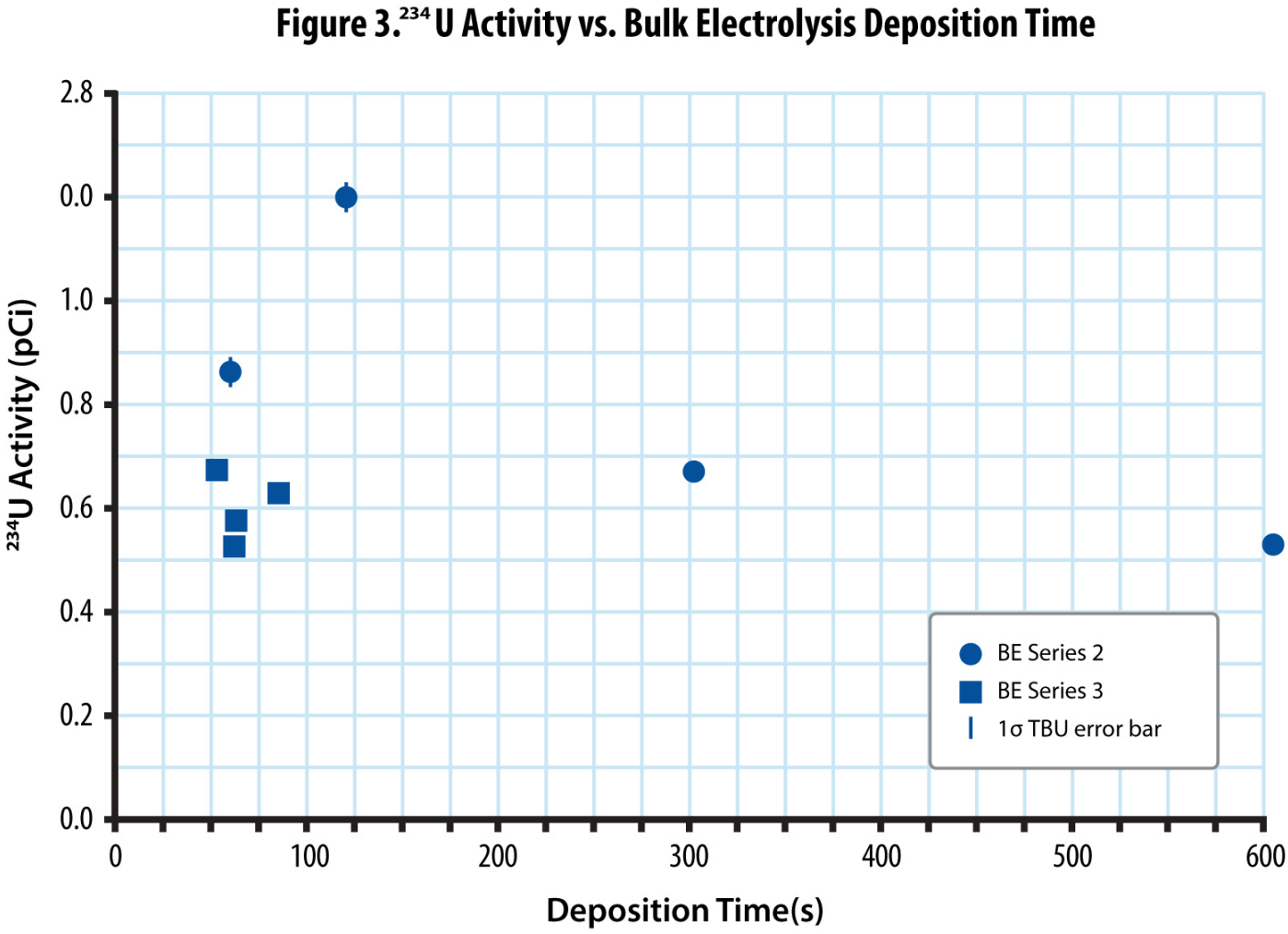


Figure 3

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Figure 4. Representative Alpha-particle Energy Spectrum for Bulk Electrolysis Sample BE2B (60 s deposition time)

Detector Efficiency: 32.97 ± 1.21% (2σ); ^{234}U Activity: 1.721 ± 0.048 pCi (1σ TPU)

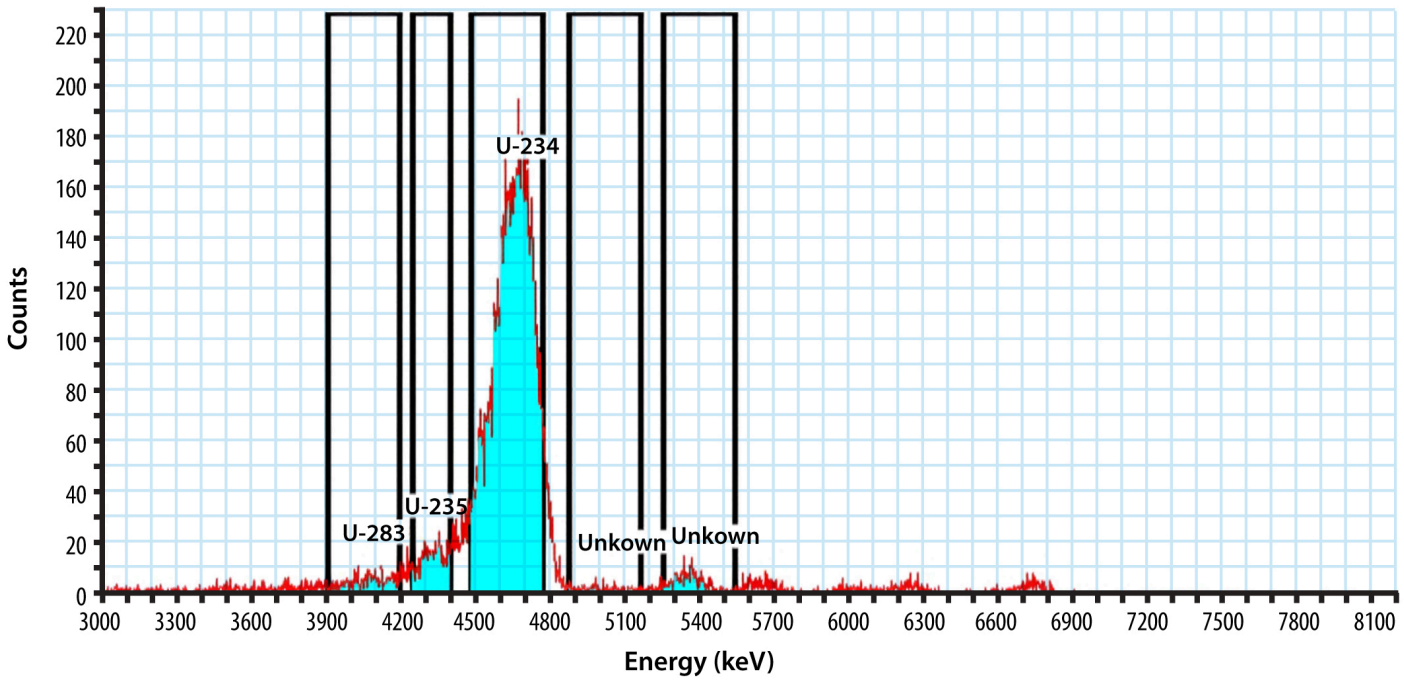


Figure 4

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