4H-Silicon Carbide as Field Deployable Sensor for Trace Actinide Detection

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Introduction and Motivation

- Analysis of in-field samples have provided valuable information
  - Scale of incident
  - Critical path to containment/treatment
- Exporting samples to be characterized in labs can be time consuming.
  - Loss of critical time that can be used for containment
    - Mobility of actinides in environment (air, soil, ground water, etc…)
- On site, rapid sample analysis can mitigate the need for exporting of samples to lab and can be done in-field.

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Introduction and Motivation

- Characterization is possible due to common decay modes of actinides.
- The high-noise environment associated with in-field sampling poses a great application for rad-hard, wide band-gap semiconductors such as 4H-Silicon Carbide (SiC).
- 4H-SiC is a wide band gap semiconductor that possesses great characteristics for in-field sample analysis
  - High breakdown voltage
  - Low leakage current
  - High radiation tolerance
  - Variable temperature operation
- 4H-SiC Schottky Barrier Diodes (SBD) alpha spectroscopy coupled with differential pulse voltammetry provides small form factor for rapid sample analysis.

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Goals and Objectives

Goals:

- Characterize 4H-SiC SBD devices for applications as a field deployable sensors.
- Investigate efficacy of on-site sampling coupling DPV with alpha spectroscopy.

Objectives:

- Demonstrate rapid characterization of trace electrodeposited actinide samples with DPV and alpha spectroscopy

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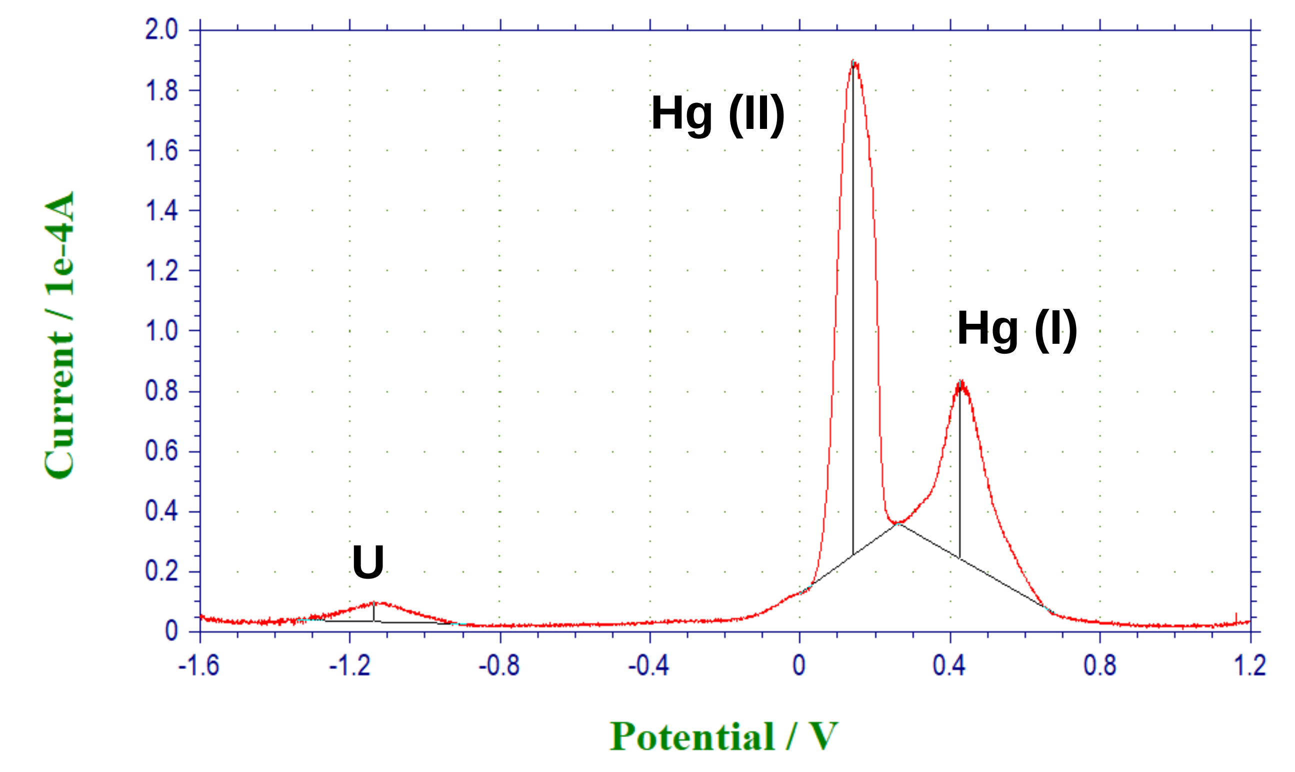
Methods and Materials – Proposed Process

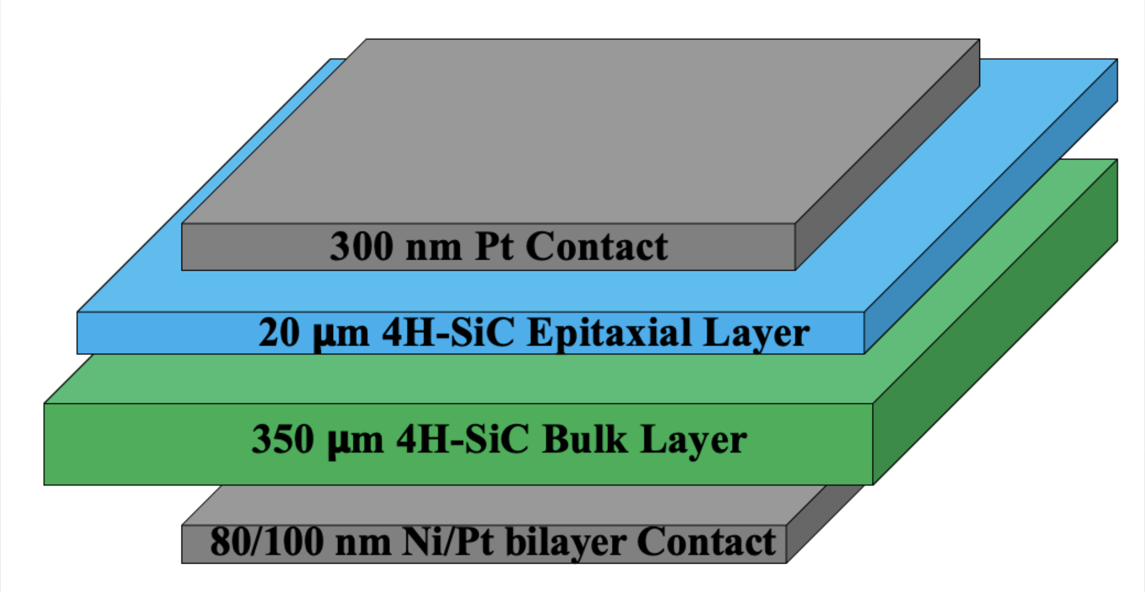
This study focuses primarily on liquid samples.

1. Sample is collected then run through DPV analysis.
2. Post-DPV, if any special nuclear material (SNM) is identified, sample is prepared for electrodeposition.
   a.) DPV solution is very similar to electrodeposition solution to provide ease of transition into electrodeposition step
3. Sample is electrodeposited onto the surface of the 4H-SiC SBD contact.
4. Alpha spectroscopy is collected.
5. Device is then electrochemically stripped cleaned via physical abrasion to be prepared for another sample.



Methods and Materials – 4H-SiC SBD

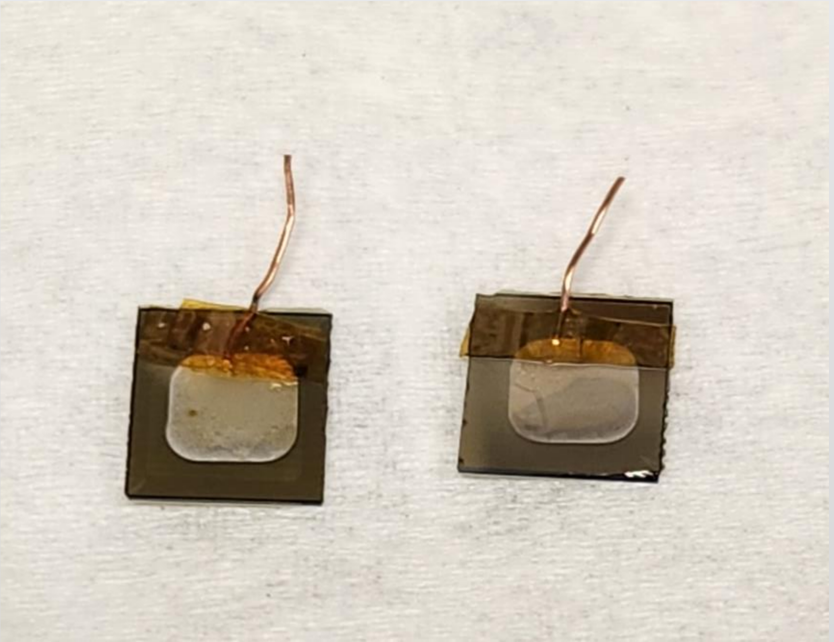
- 4H-SiC Schottky Barrier Diodes
  - 300 nm Pt Schottky Contact
  - 20 µm - 2E14 Nitrogen-doped epitaxial layer
  - 350 µm – Bulk/substrate layer
  - 80/100 nm Ni/Pt bilayer ohmic contact.
- Contact area is 4x4 mm to provide large surface for electrodeposition.
- Pt Contact is highly-corrosive resistant to prevent any damage from acids associated with electrodeposition and electrochemical stripping process.



Methods and Materials – Electrodeposition

2 main steps occur in the electrodeposition process

- A thin layer of mercury is deposited onto the contact metal
  - The layer of mercury serves multiple purposes
    - Prevents hydrogen evolution reactions (HER) during the electrodeposition process
    - Bubbles limit electrodeposition and causes spikes in DPV analysis
- The deposition of actinides is deposited onto the layer of mercury.
  - In real application, isotopes of interest would be Plutonium (Pu) and Uranium (U).
    - ^{241}Am is used in place of Plutonium (NIST – 4322-d)
    - A NIST standard containing ^{234}U, ^{235}U, and ^{238}U is also used in the electrodeposition (NIST – 4321-d)



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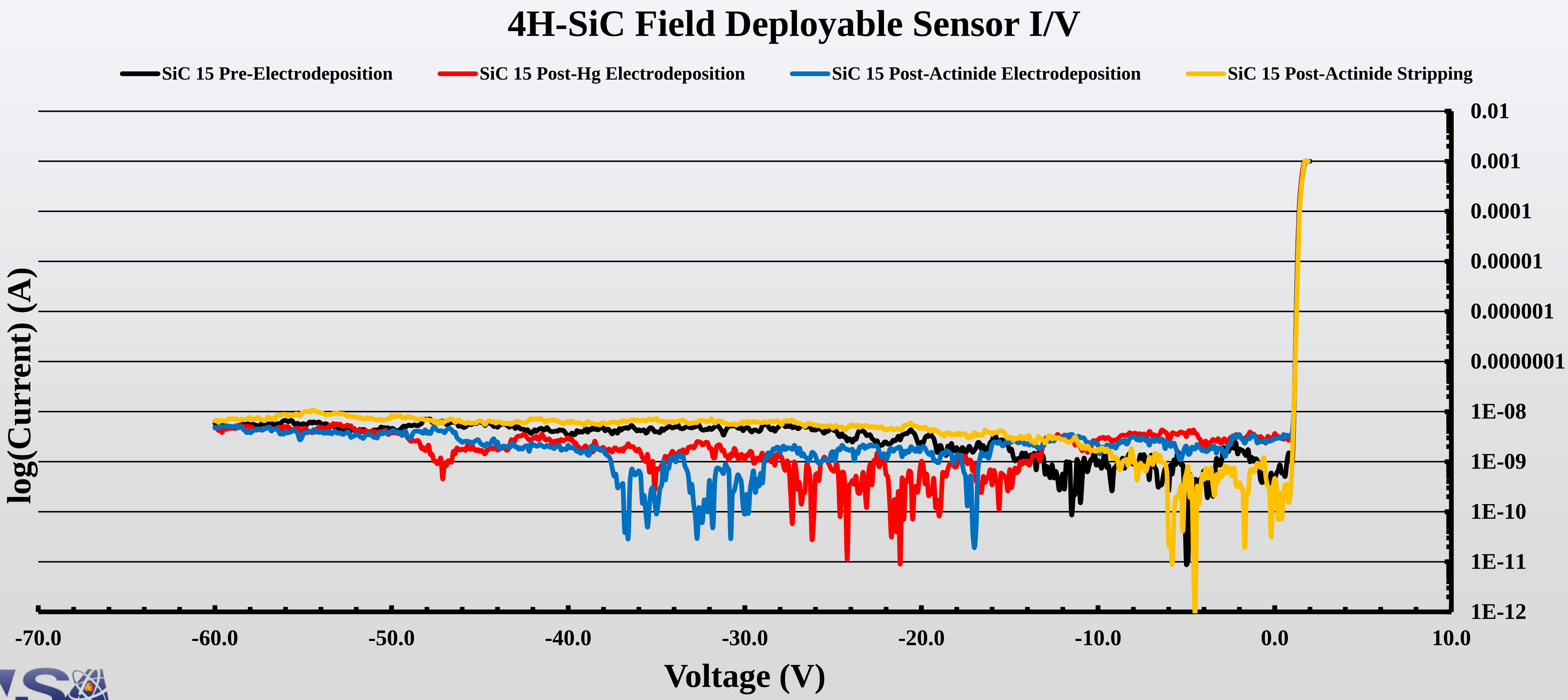
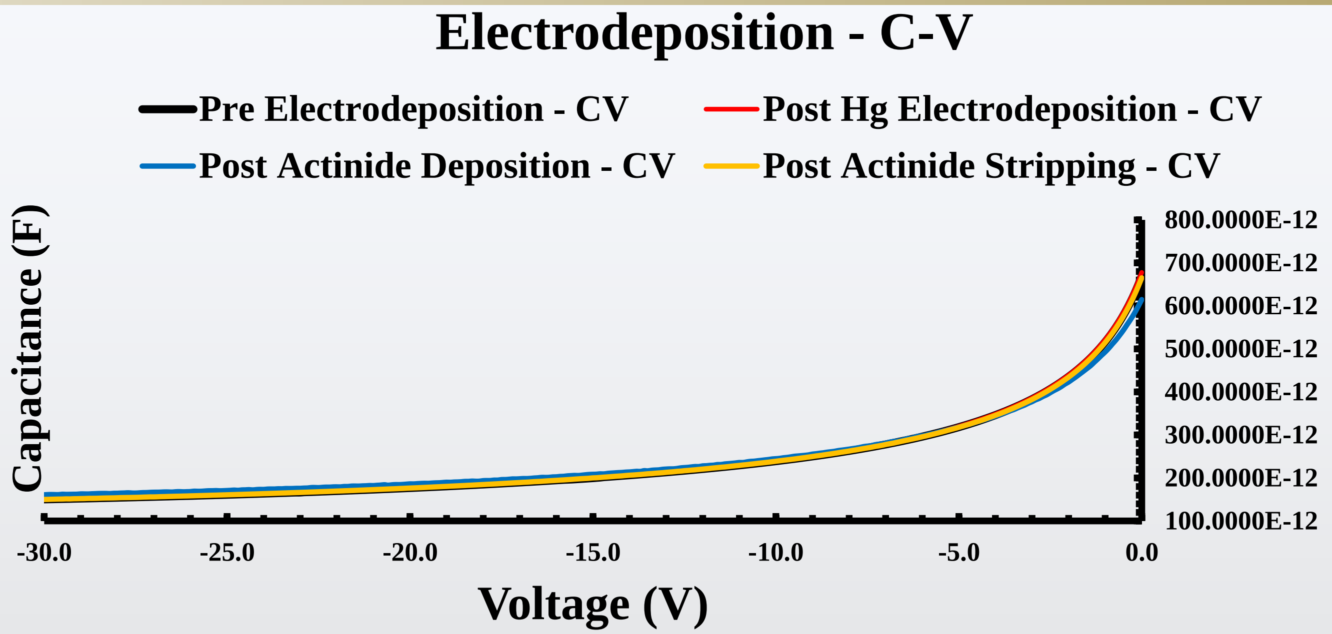
Technical Work and Results – I/V and C-V

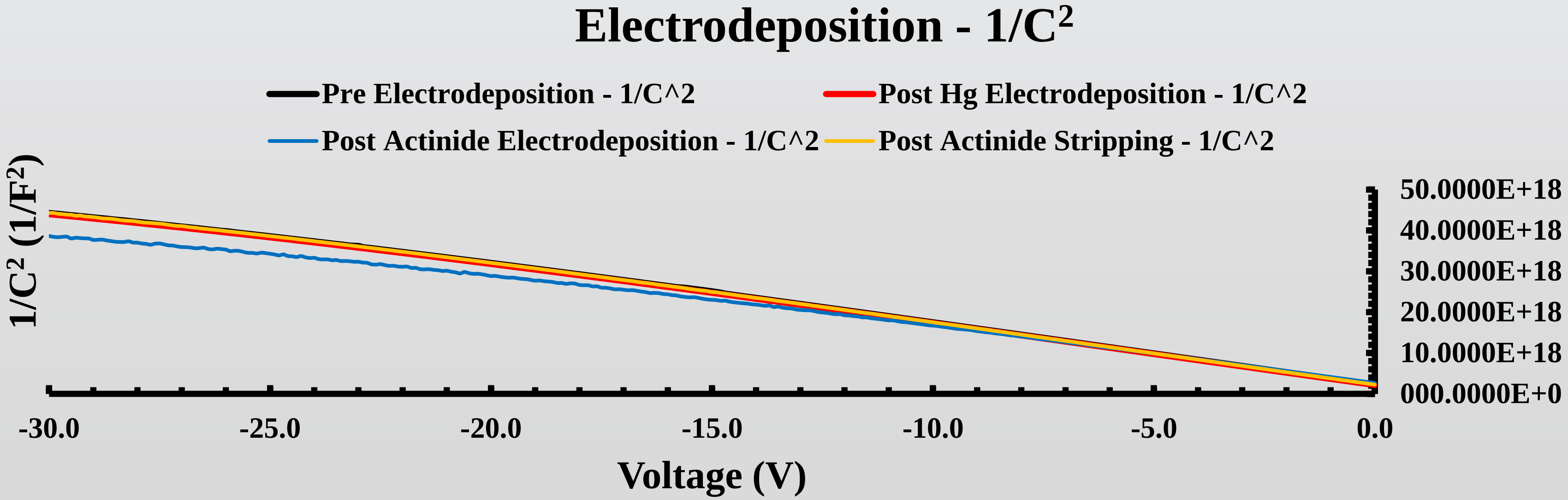
- I/V curves collected at each stage of the electrodeposition process to ensure repeatable measurements
- No major changes in leakage current or turn-on voltage at any stage in the electrodeposition process



Technical Work and Results – I/V and C-V

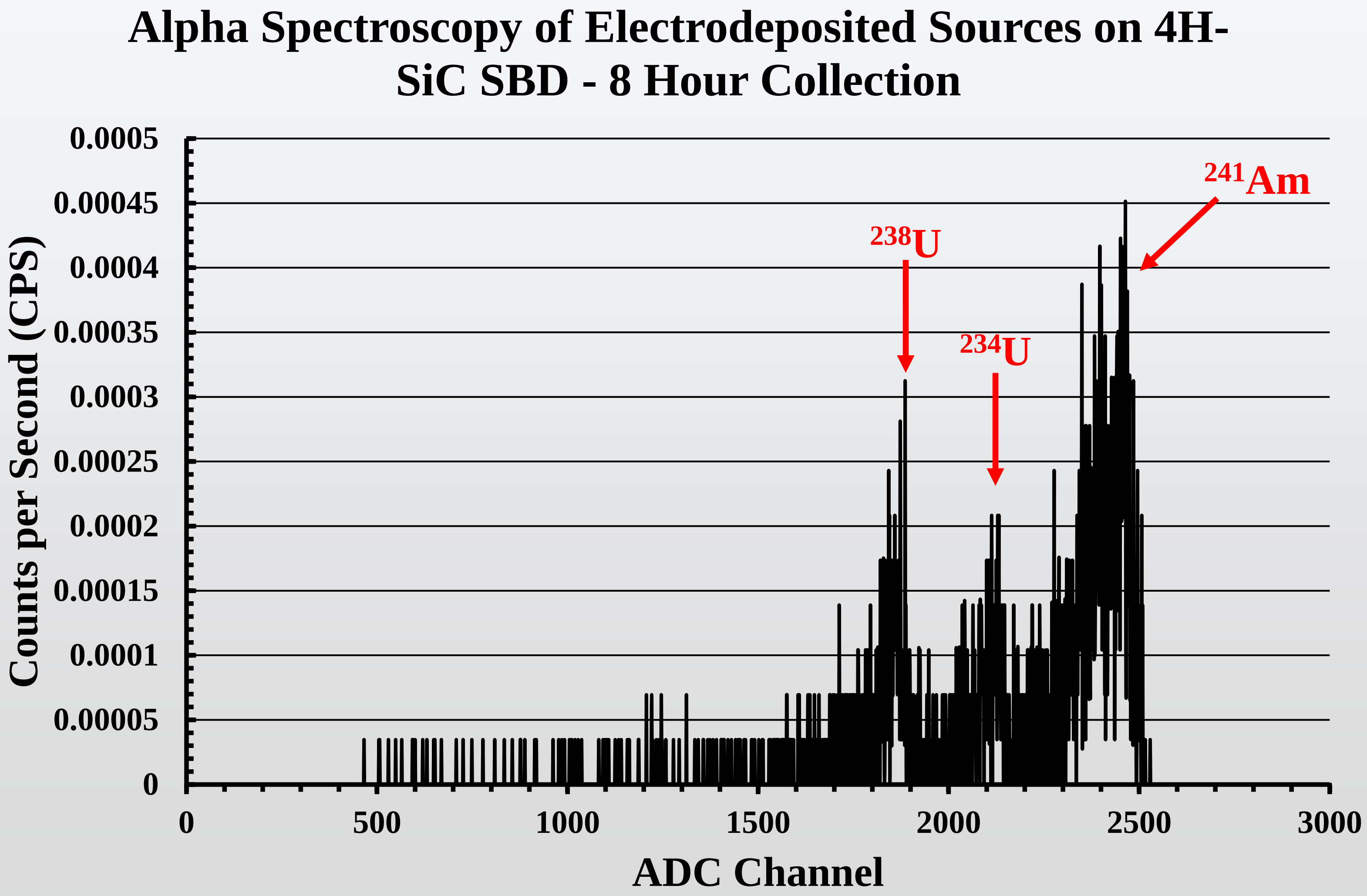
- C-V analysis is also conducted at each stage to confirm no changes in:
  - Depletion Region
  - Resolution
- $1/C^2$ is also extracted to look for any change in dopant profile or built-in voltage.





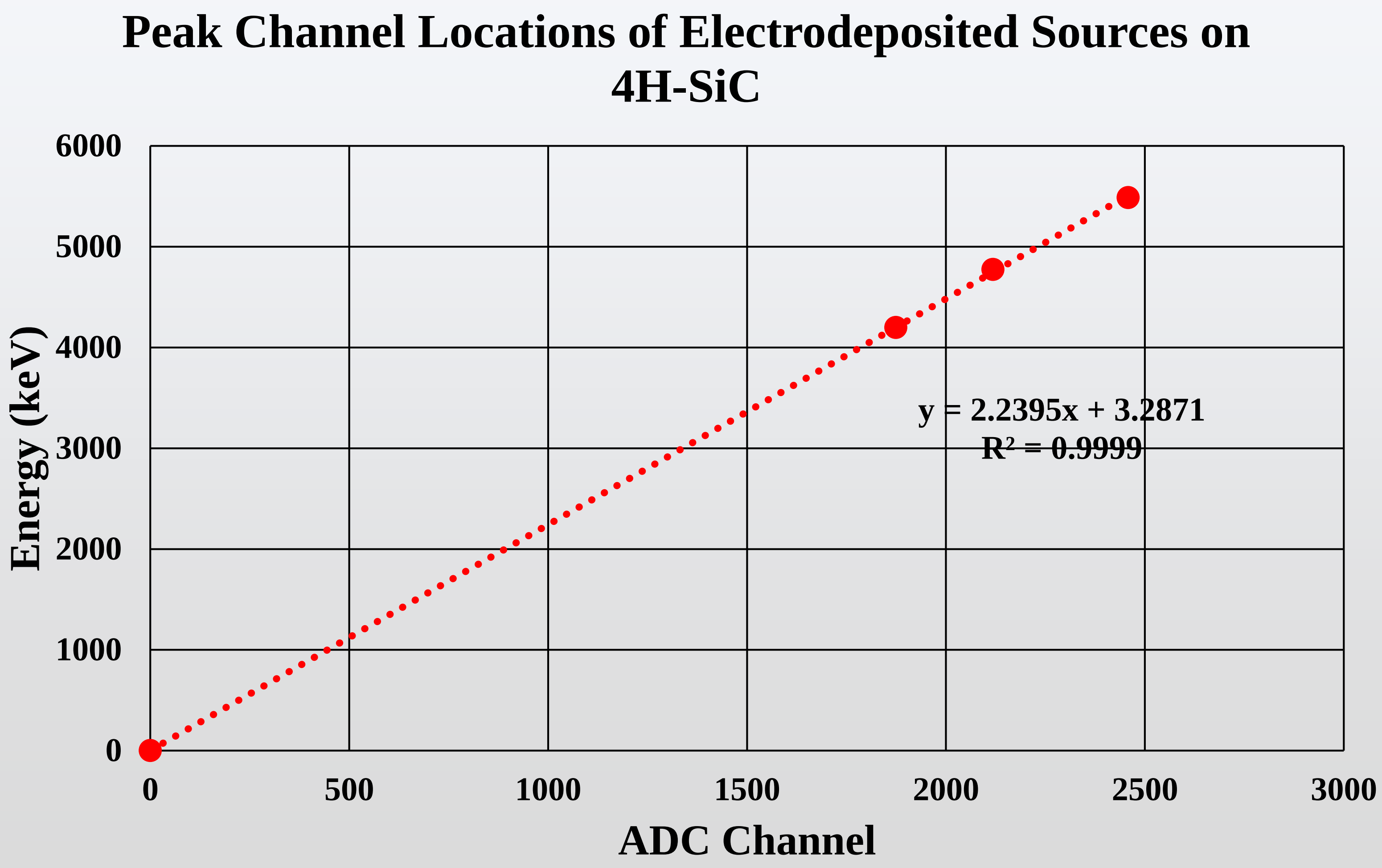
Technical Work and Results – Alpha Spectroscopy

- After 8 hours of collection, we can resolve most actinides present in solution.
  - ^{235}U is present in NIST – 4321d in extremely small quantities and does not appear in alpha spectrum due to low specific activity (1.535E-8 Ci/g)
- Alpha spectroscopy provides a general idea of the isotopes present in the solution

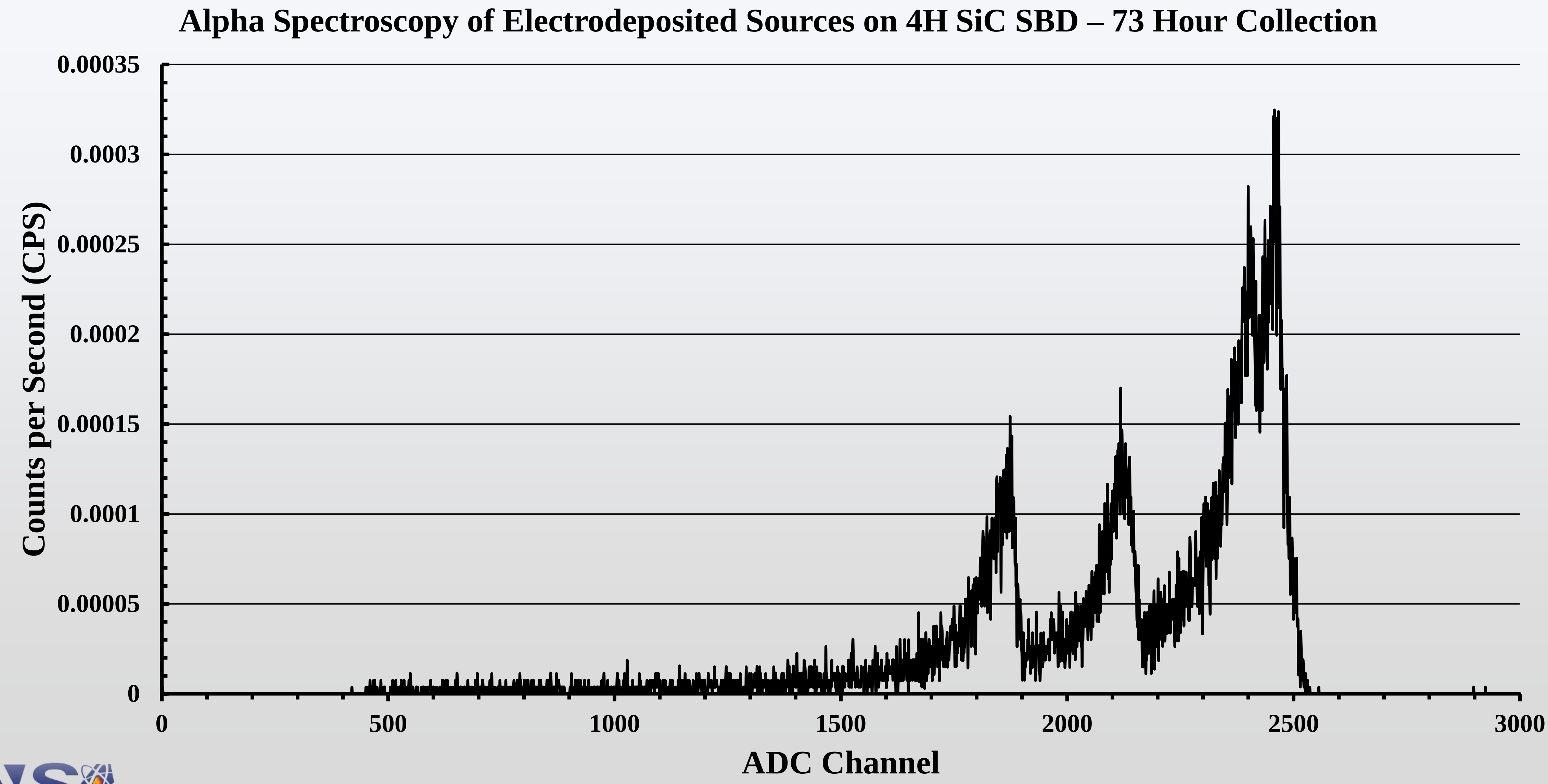


Technical Work and Results – Alpha Spectroscopy

- Peak channels are extracted from alpha spectrum to generate an energy calibration curve for this detector under these conditions (Electrodeposited actinides and Hg)
- Linear behavior suggest reliable spectrum is collected and device operation is as expected.
- Extended alpha spectrum was collected to confirm results and extract isotope ratios.



Technical Work and Results – Alpha Spectroscopy



Technical Work and Results – Isotopic Ratios

- Peaks were separated and integrated accordingly to their respective isotopes then normalized to each isotopes specific activity to provide atomic % of isotopes collected in spectroscopy.
- **Shows agreement with NIST standard reported values.**

Isotopes	Specific Activities (Ci/g)	Peak Integration	Activity (Bq) - 2Pi	Activity (Bq) - 4Pi	Convert to Ci (Ci)	Normalized to Specific Activity (Mass in g)	Number of Atoms (Uranium)	Number of Atoms (Americium)	Isotopic Ratio by Total Mass	Atomic Ratio of Uranium
^{238}U	3.36E-07	3389	0.01275038	0.02550076	6.8921E-13	2.05E-06	5.19E+15		9.9994E-01	9.99939E-01
^{234}U	6.30E-03	3835	0.014428359	0.028856717	7.79911E-13	1.24E-10	3.18E+11		6.03E-05	6.13824E-05
^{241}Am	3.4	10653	0.04007961	0.08015922	2.16647E-12	6.37196E-13		1591290920	3.10623546E-07	

Isotopes	Atomic % - NIST (%) Normalized without ^{235}U	Atomic % - Alpha Spectroscopy (%)
^{238}U	99.994715	99.99386
^{234}U	0.005285	0.006138

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Future Work

- Confirm isotopic ratios extracted from alpha spectroscopy with inductively coupled plasma mass spectrometry (ICP-MS) results.
- Perform multiple cycles on 4H-SiC SBD to understand total device lifetime in this setup.
  - Current device has undergone 2 cycles successfully.
  - Adjustments to design to prolong device lifetime
- Compare analysis time to other forms of in-field sampling analysis.

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Conclusion

- 4H-SiC SBD were electrodeposited with actinides electrodeposited from a DPV analyzed solution.
- 4H-SiC SBD I/V and C-V was monitored at each step of the electrodeposition process to ensure stability of device through out the analysis.
- Alpha spectroscopy was collected from the 4H-SiC SBD to gain isotope information about electrodeposited actinides.
    - Peaks clearly distinguished at 8 hours
- Alpha spectroscopy collection was extended to confirm isotopic information and extract more information from samples.
- Isotope ratio data extracted shows agreement with NIST standard 4321d. Data to be further validated by ICP-MS results.

ACKNOWLEDGEMENTS

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