

Diamonds are a Reactor's Best Friend: Novel Sensor to Investigate High-Valence Actinides in Molten Salts

Dr. Hannah Patenaude

12 August 2026

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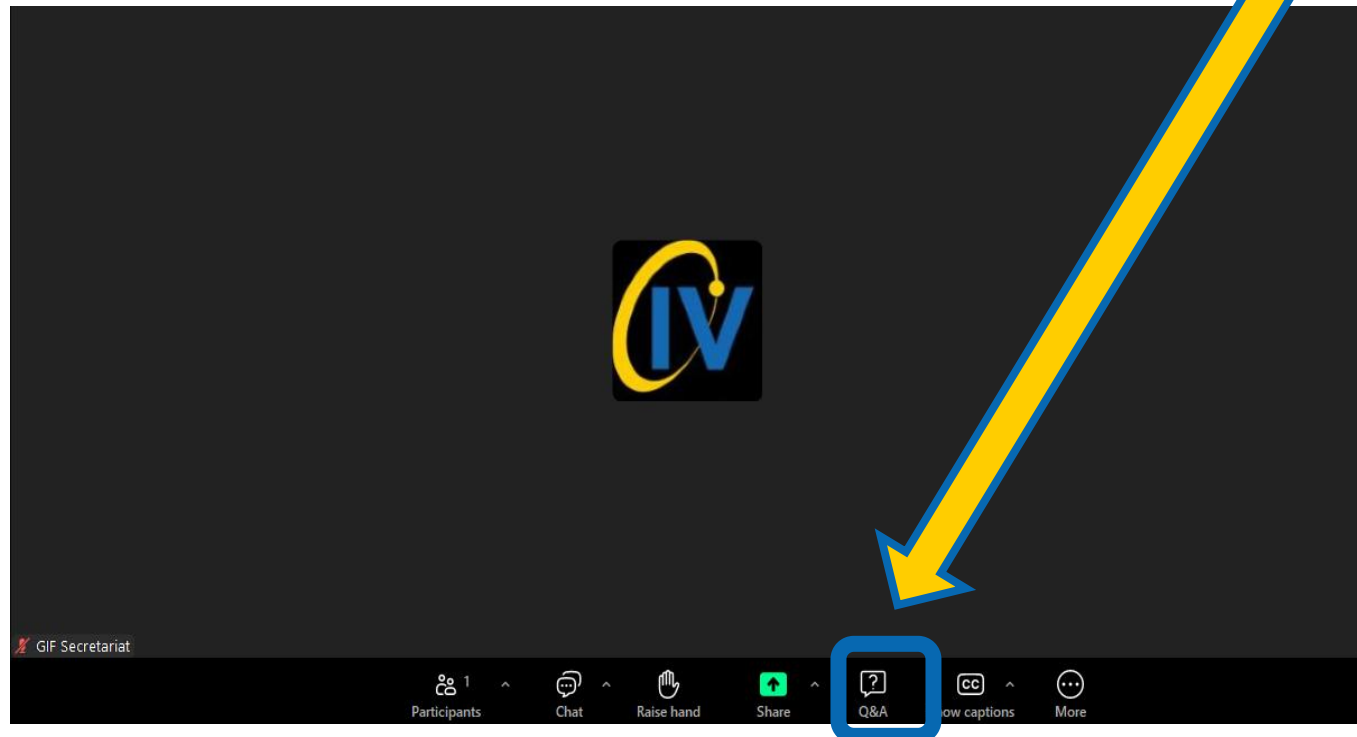
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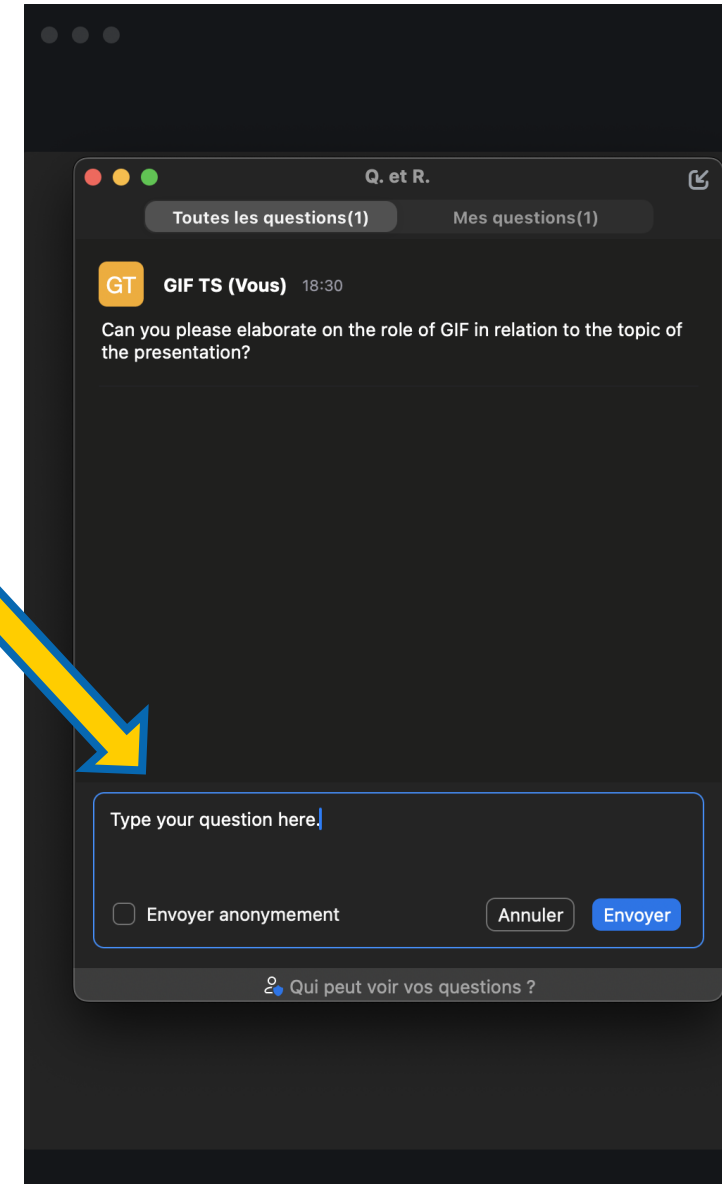
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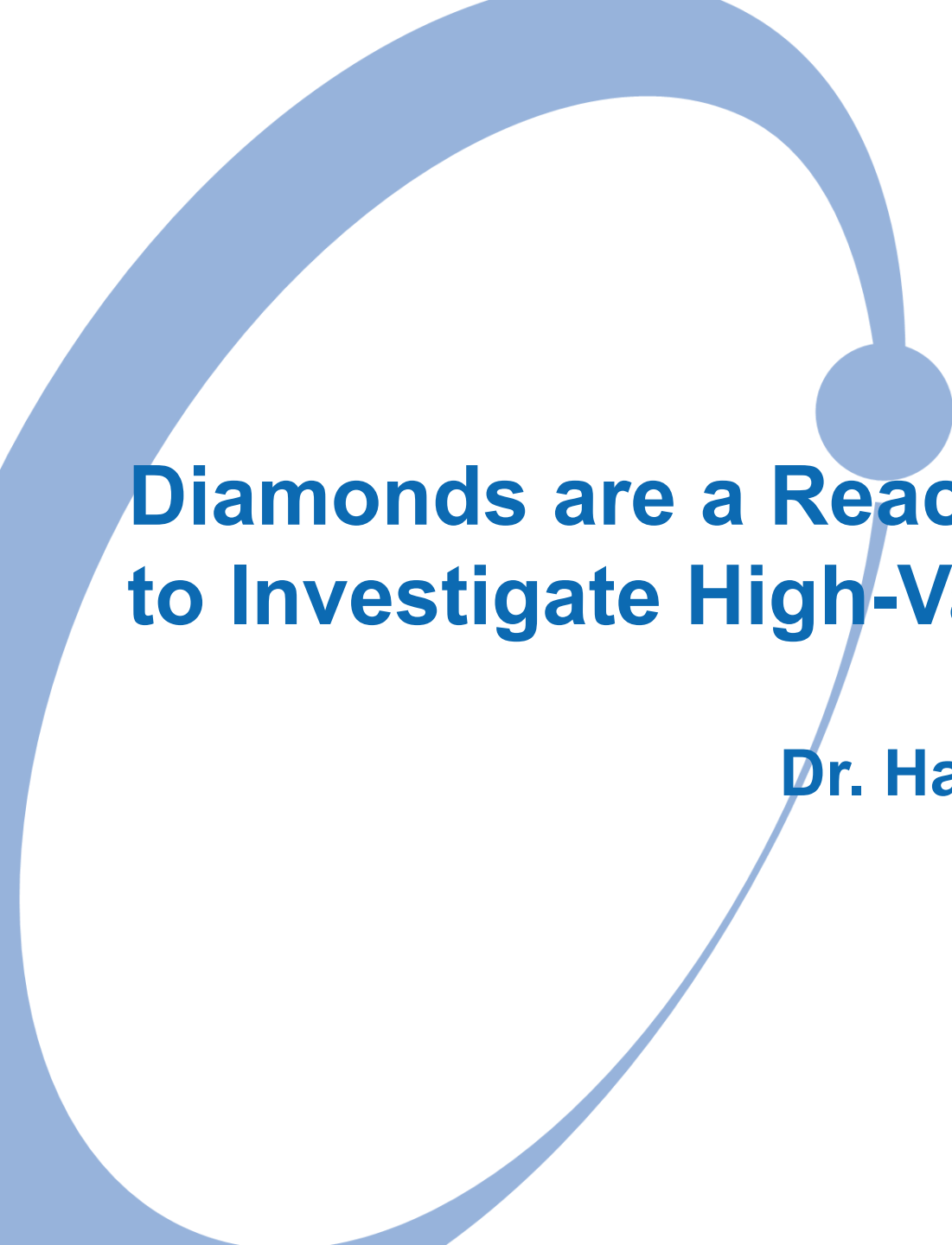
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you to type
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question.





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Meet the Presenter

- **Dr. Hannah Patenaude** is a Director's Postdoctoral Research Fellow in the Inorganic, Isotope, and Actinide Chemistry group at the Los Alamos National Laboratory.
- As a recent graduate of the University of Nevada, Las Vegas Radiochemistry Program, Hannah is experienced in actinide chemistry, specifically electrochemical processes, thermophysical properties measurements, and materials science techniques with an emphasis on molten salt systems to support the safe, secure deployment of Molten Salt Reactors.
- Outside of the laboratory, Hannah is involved in public policy and studies the rhetoric and public perceptions of nuclear technology. She is active in organizations like the American Nuclear Society, serving on the Executive Committee of the Fuel Cycle & Waste Management Division and as the Vice Chair for the Trinity Section. Hannah spends her free time with her dog, Clementine—named after a LANL Manhattan Project era nuclear reactor—who provided support in Hannah's 2025 Pitch Your Gen IV Research video.









**Fukushima Daiichi
Nuclear Power Plant**
Japan 2011

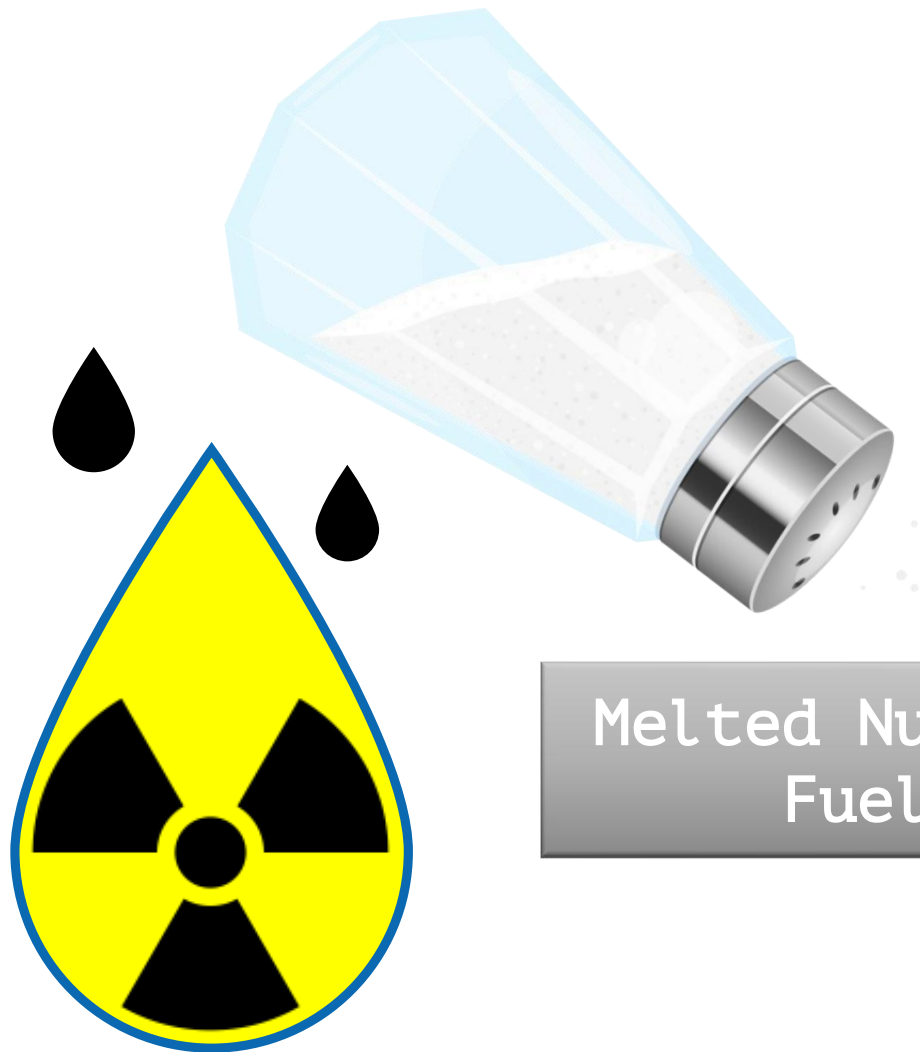


**Chernobyl
Nuclear Power Plant**
Ukraine SSR 1986

Nuclear Energy ? Nuclear Meltdown



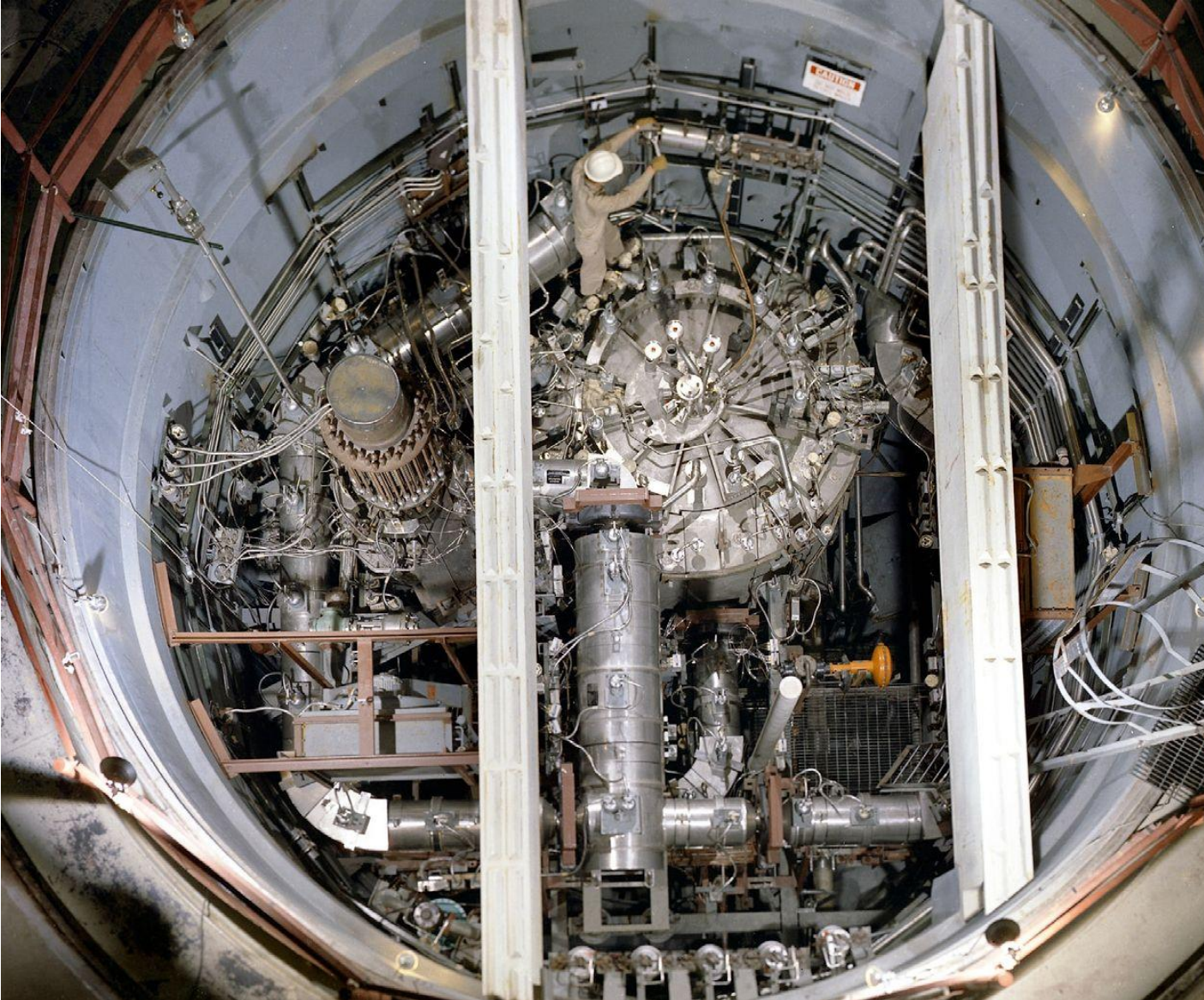
www.grs.de



Melted Nuclear
Fuel



Molten Salt Technology

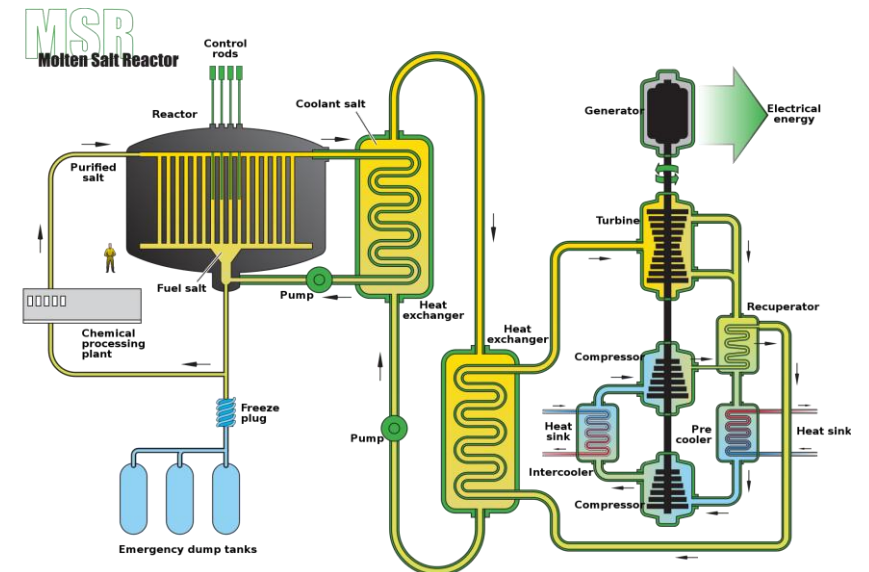


Molten Salt Reactor Experiment, Oak Ridge National Laboratory (1965-1969)

Rosenthal, ORNL/TM-2009/181, 2009.



Molten Salt Chemistry Laboratory, LANL



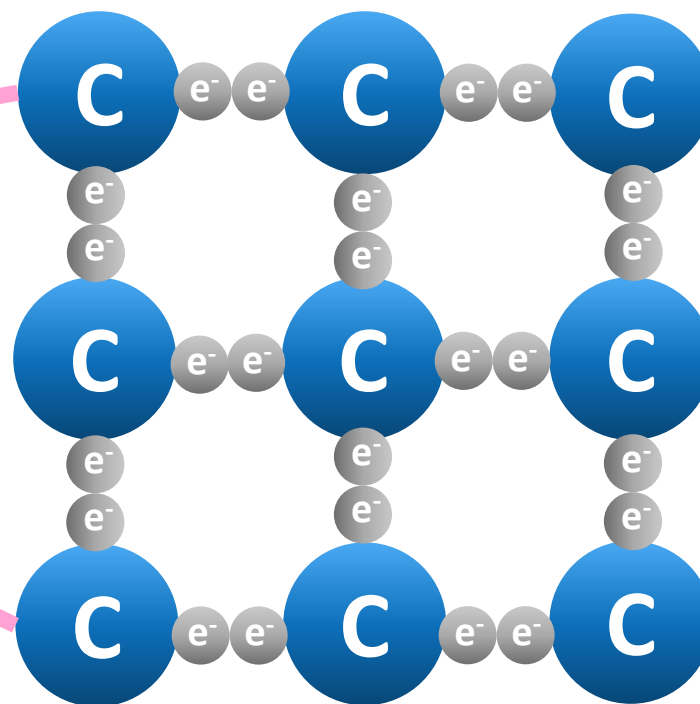
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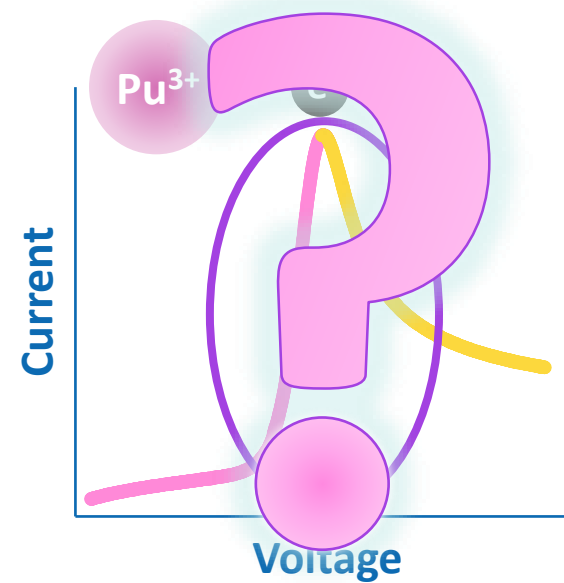
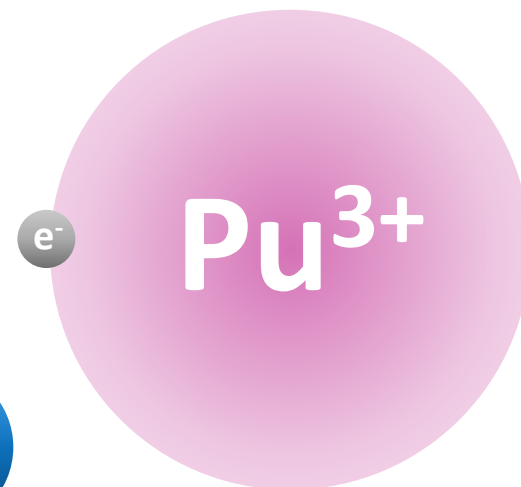
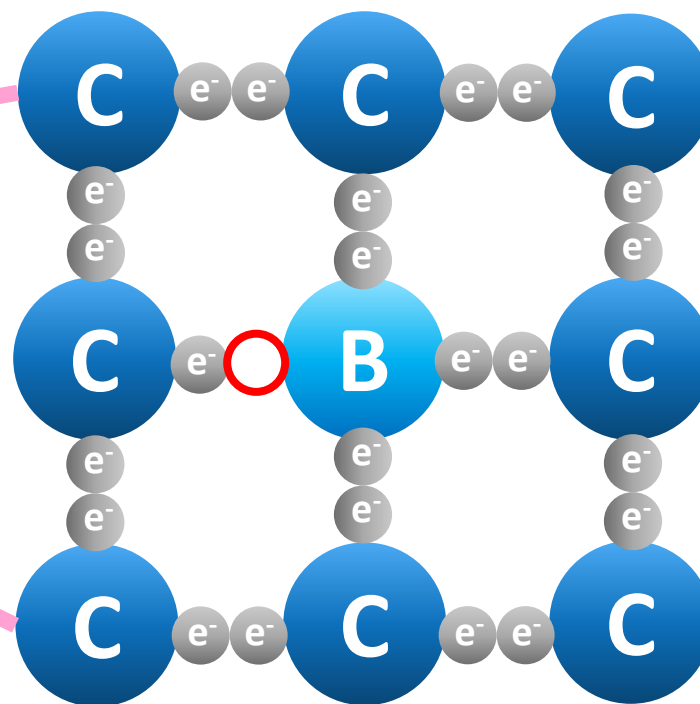
Molten Salt Reactor

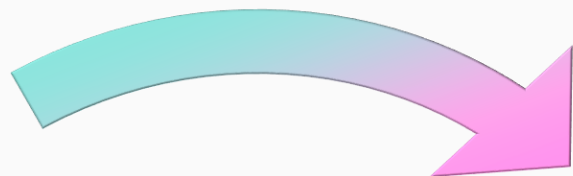


How to track Pu?

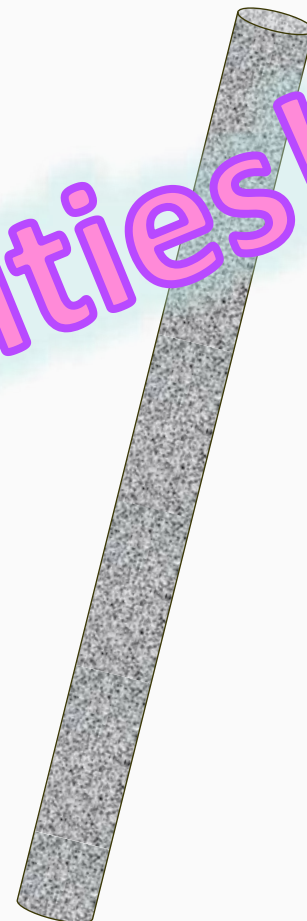


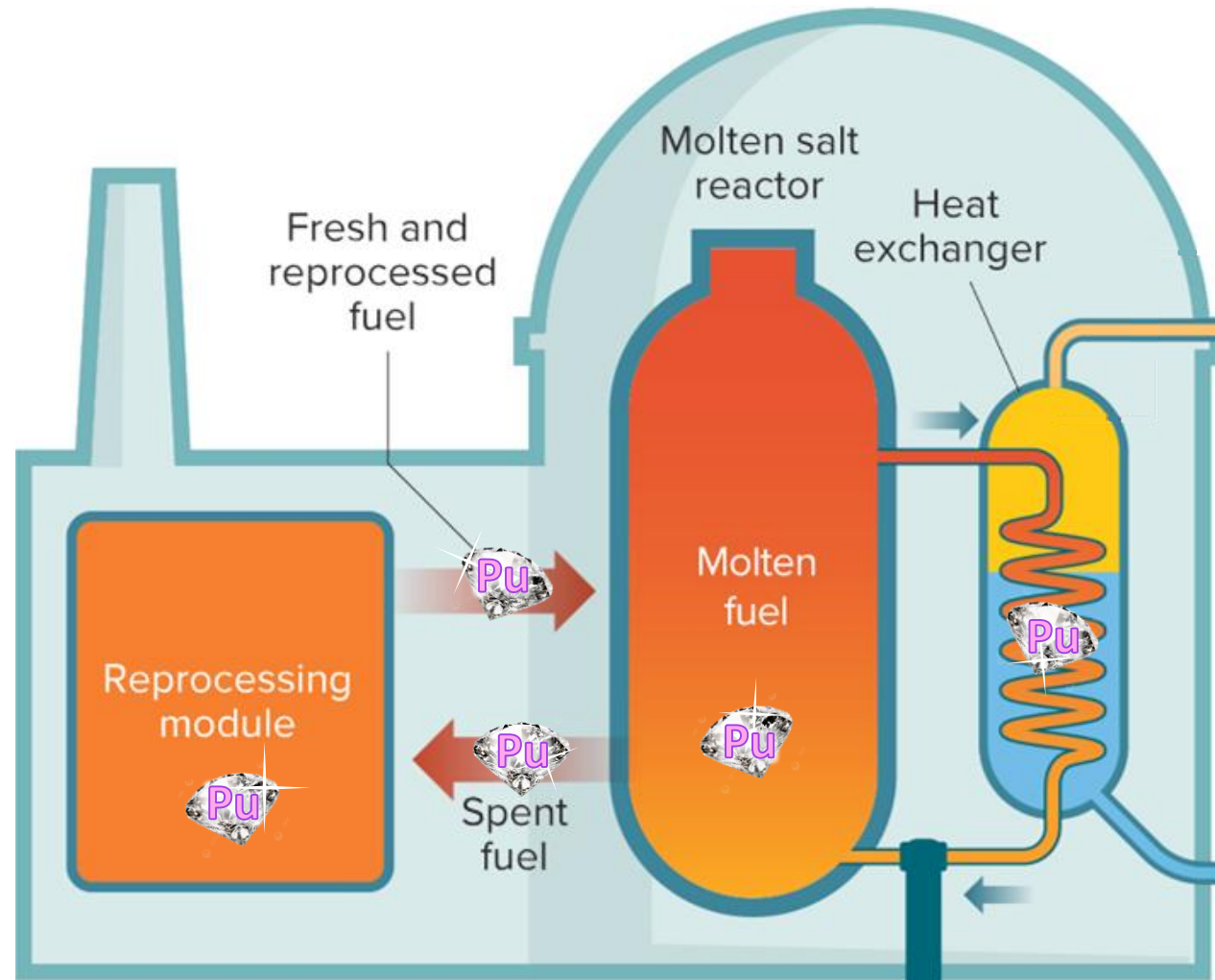


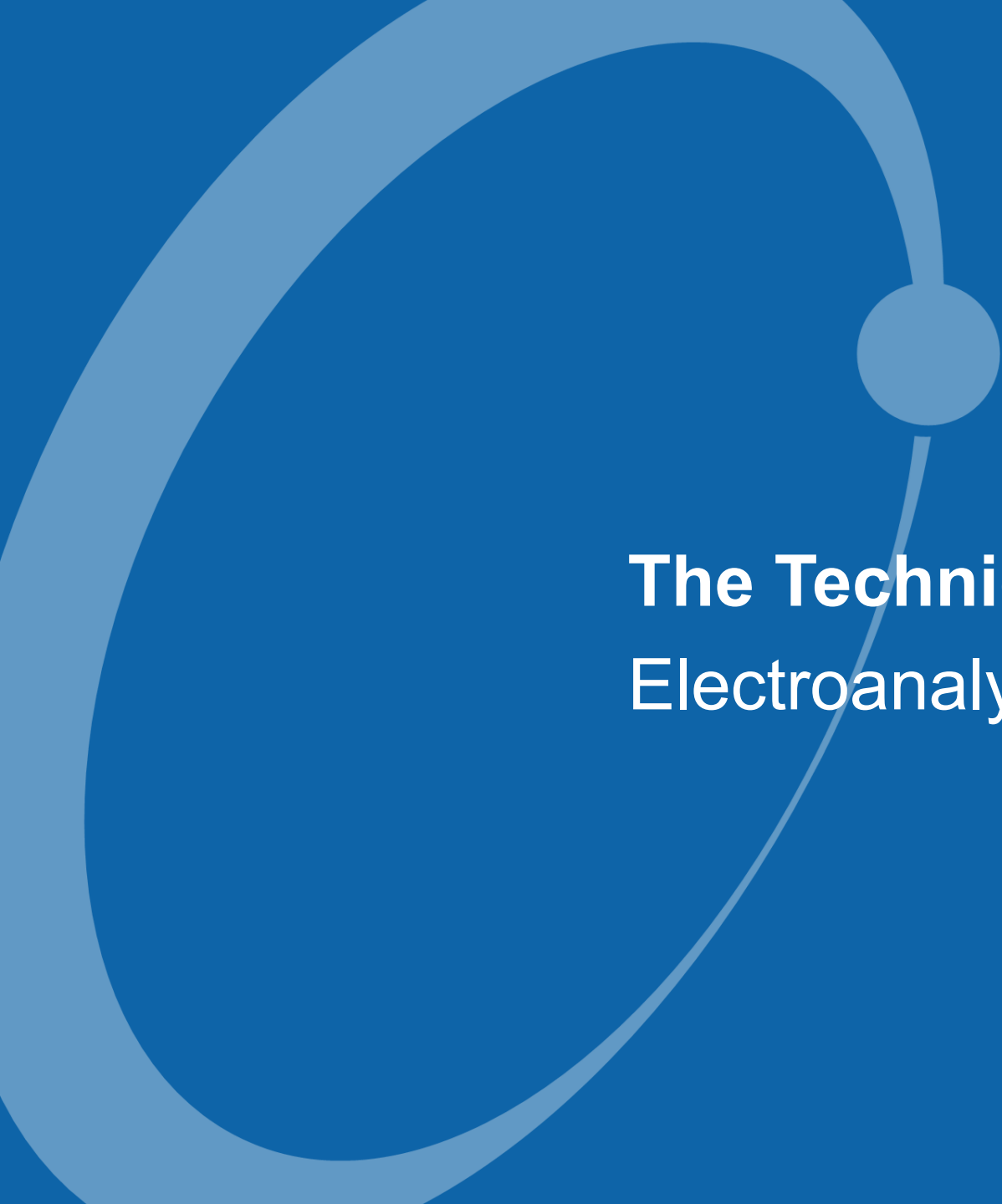




Endless Possibilities!

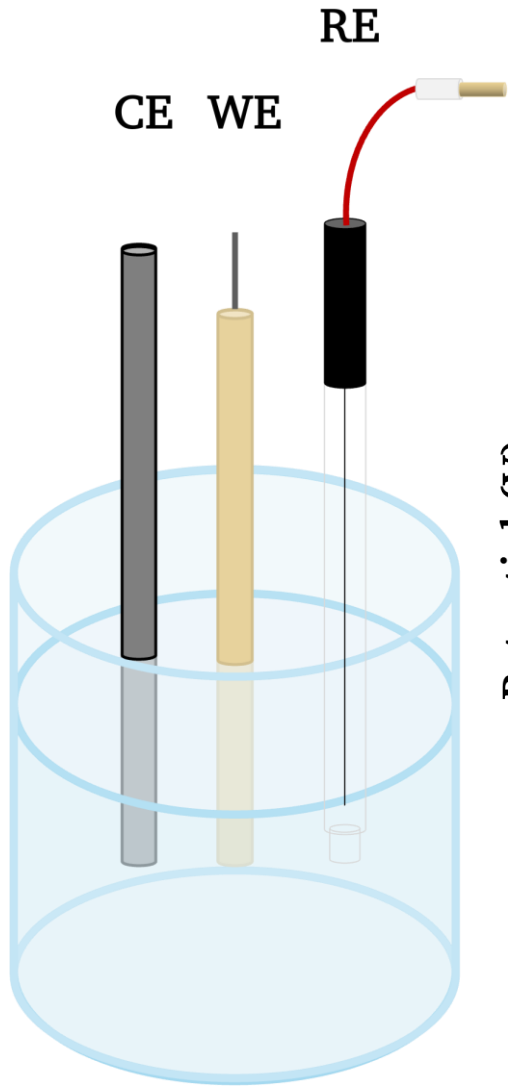




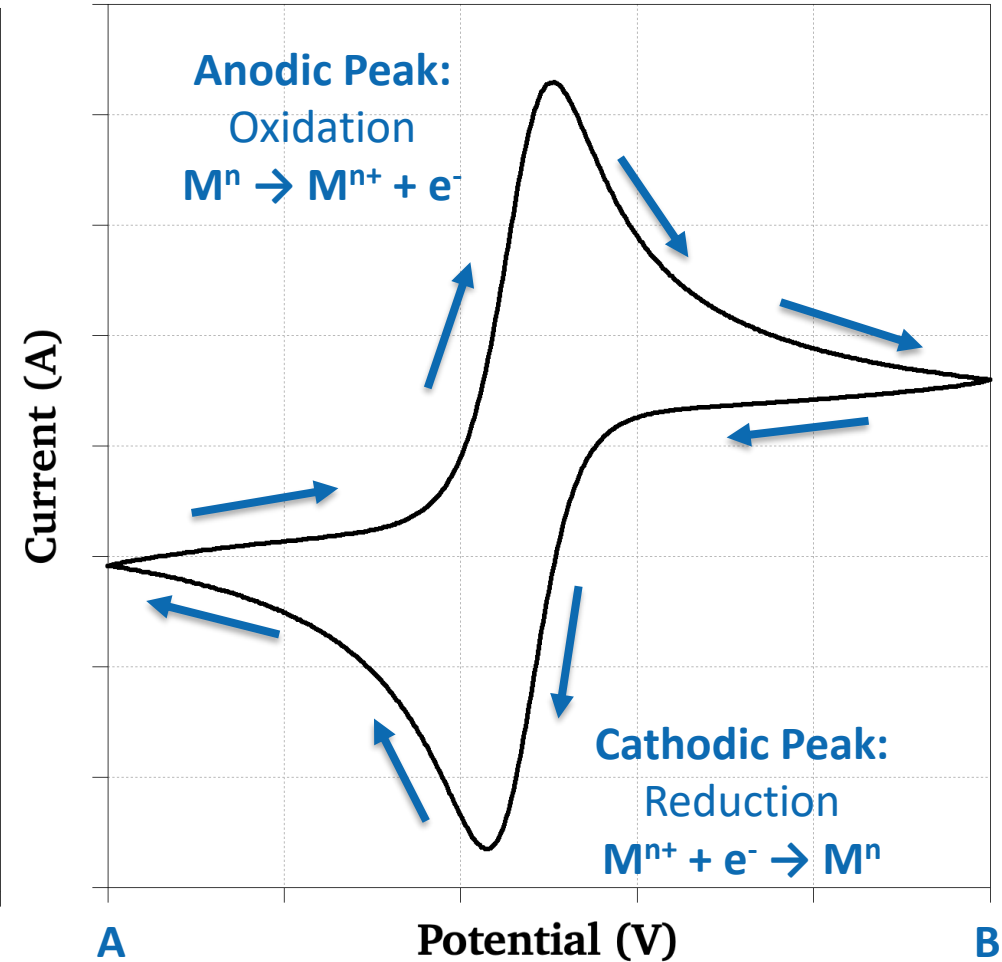
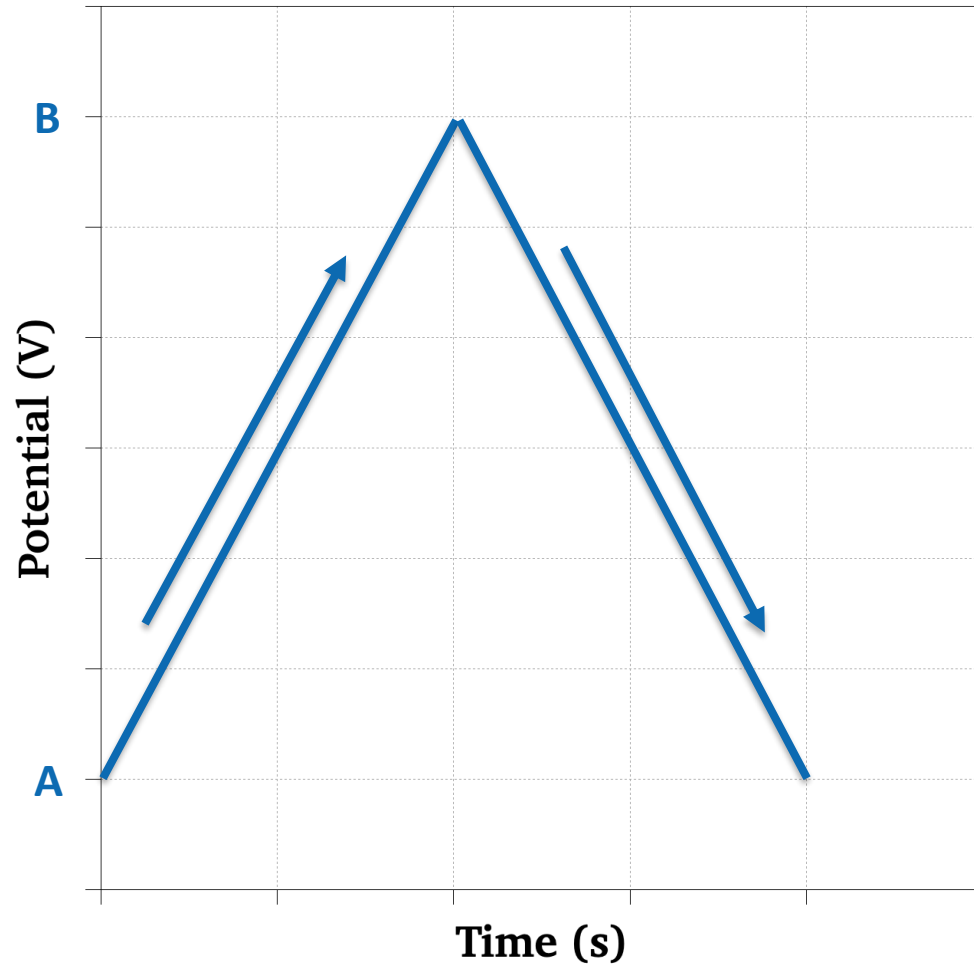


The Technique: Electroanalysis

Electroanalytical Chemistry



Cyclic Voltammetry



Electroanalytical Chemistry

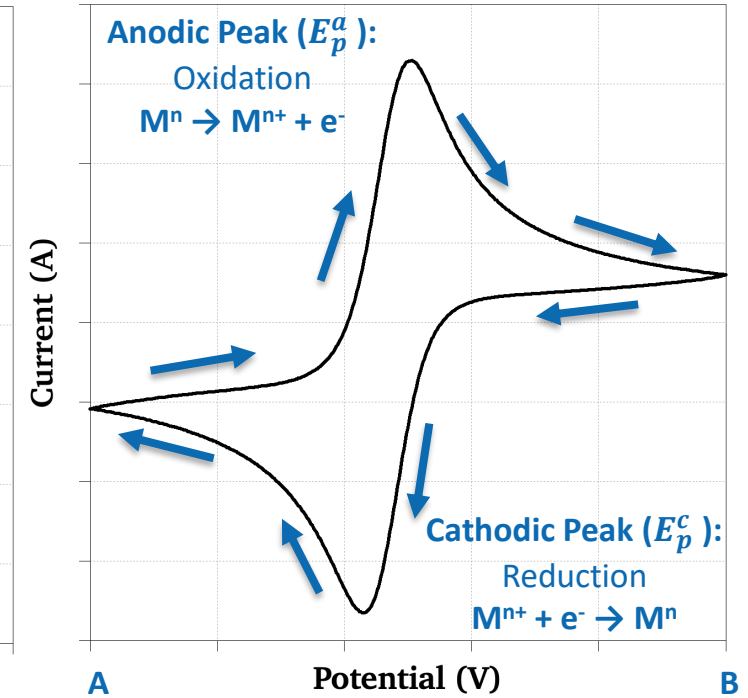
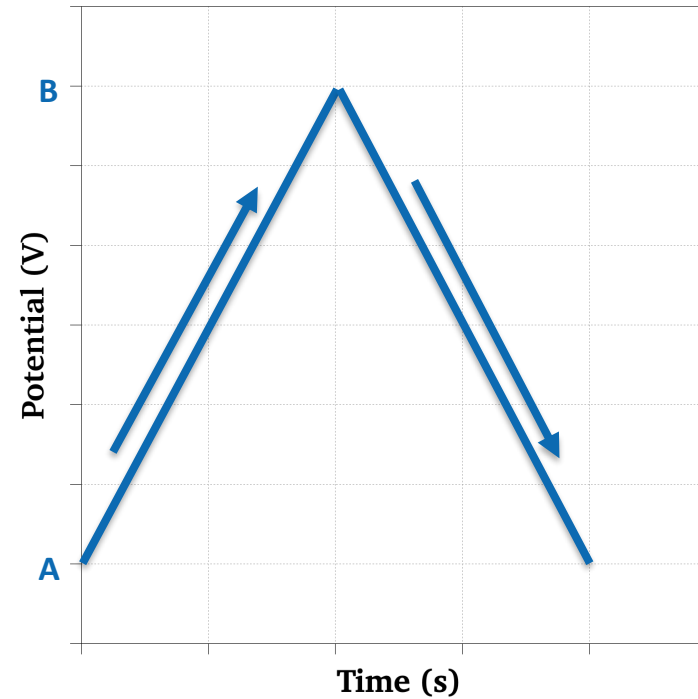
Cyclic Voltammetry

Potential & Thermodynamics:

$$E = E^0 + \frac{RT}{nF} \ln \frac{[Ox.]}{[Red.]}$$

$$E^0 = \frac{E_p^a + E_p^c}{2} \quad \Delta E_p = E_p^a - E_p^c$$

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Electroanalytical Chemistry

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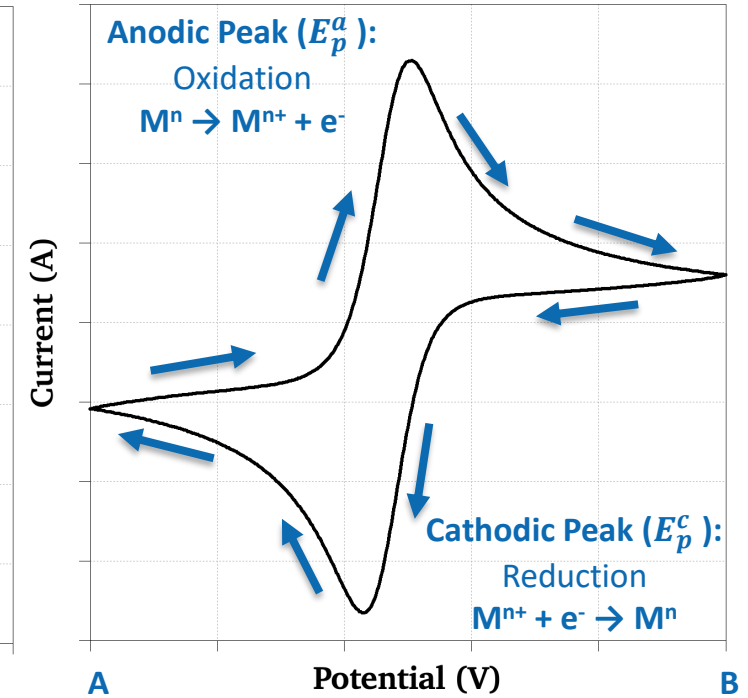
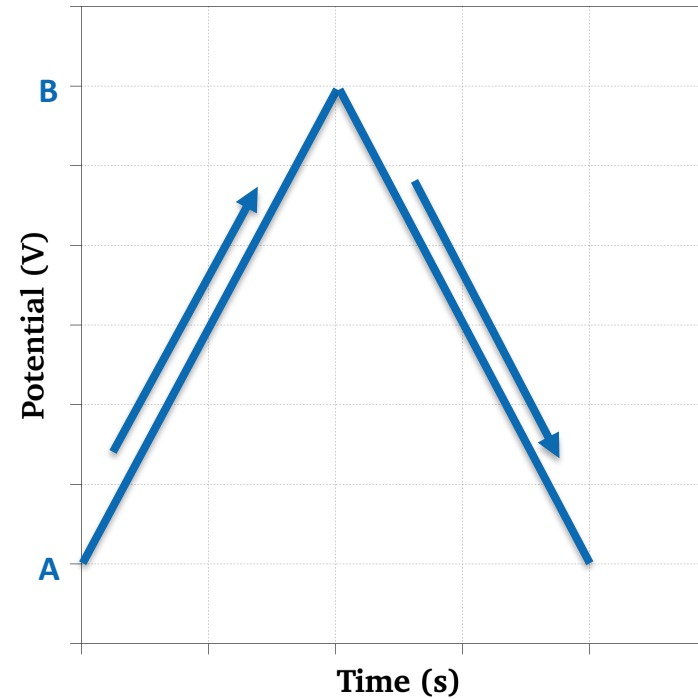
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Analyte Diffusion:

$$i_p^{a/c} = 0.496nFAC \sqrt{\frac{D_{R/o}(\alpha n_\alpha)Fv}{RT}}$$

$$i_p^{a/c} = 0.4463nFaC \sqrt{\frac{nFvD_{R/o}}{RT}}$$

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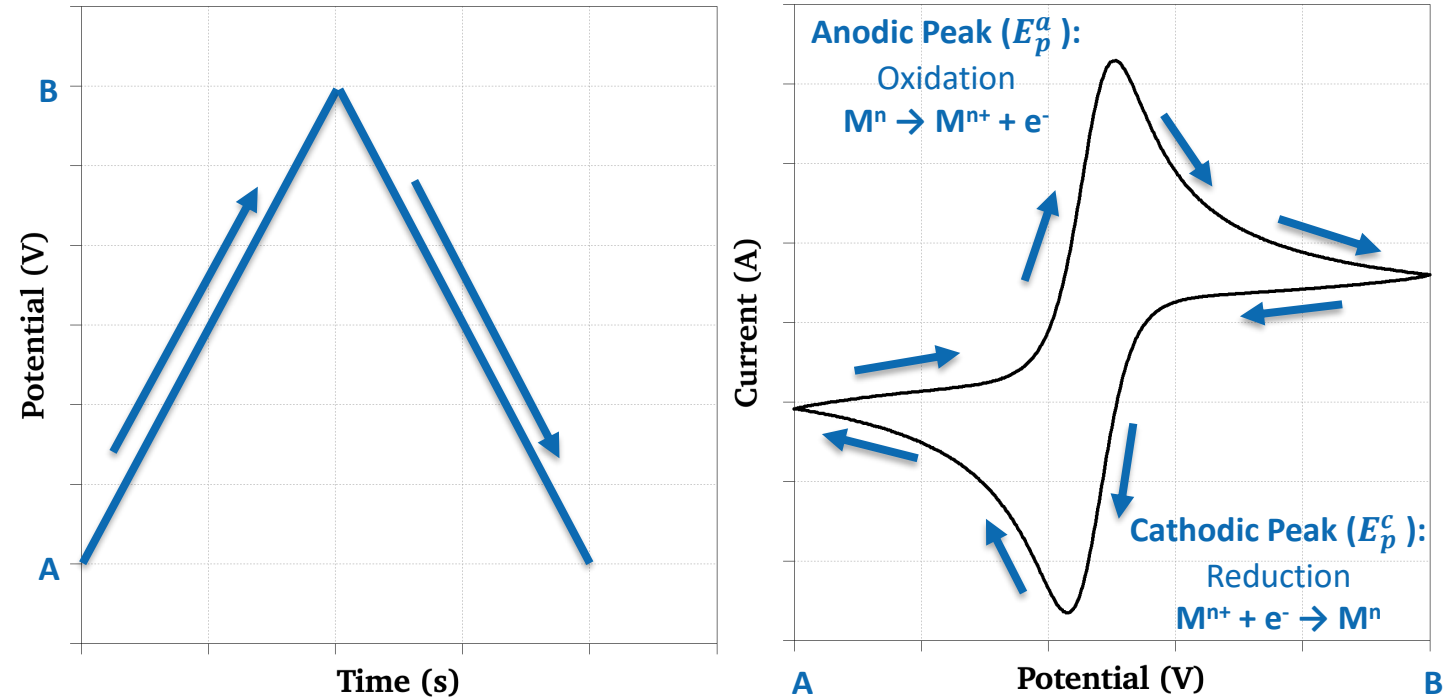
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Cyclic Voltammetry



Electron Transfer Kinetics:

$$k = 2.18 \sqrt{D_O(\alpha n_\alpha) \frac{vF}{RT}} \cdot e^{\left\{ \frac{\alpha^2 nF}{RT} (E_{p,c} - E_{p,a}) \right\}}$$

$$\psi = \frac{\sqrt{\frac{D_O}{D_R}}}{\sqrt{\frac{\pi D_O n F v}{RT}}} k$$

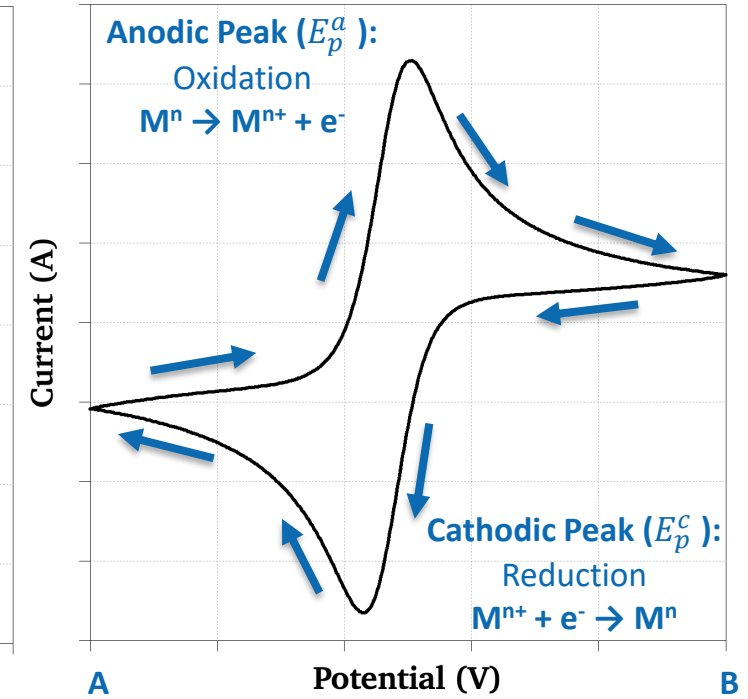
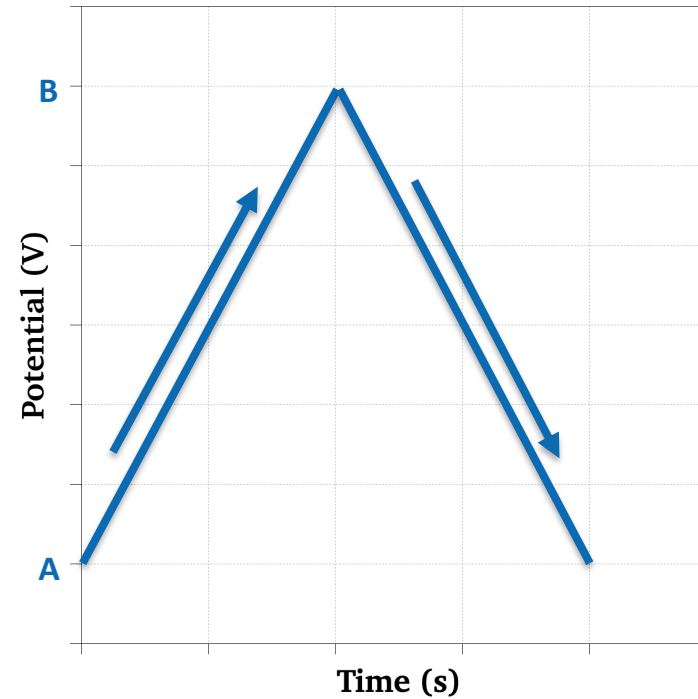
$$\alpha n_\alpha = \frac{1.857RT}{|E_p - E_{p/2}| F}$$

Electroanalytical Chemistry

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- Fundamental Characterizations
 - E^o , D , k , ΔG^o , ΔH^o , ΔS^o
- Elemental Analysis
- Corrosion Kinetics
- High Sensitivity

Cyclic Voltammetry



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- Working electrode *resilience*
- Reference electrode *stability*
- Matrix variations and *purification*

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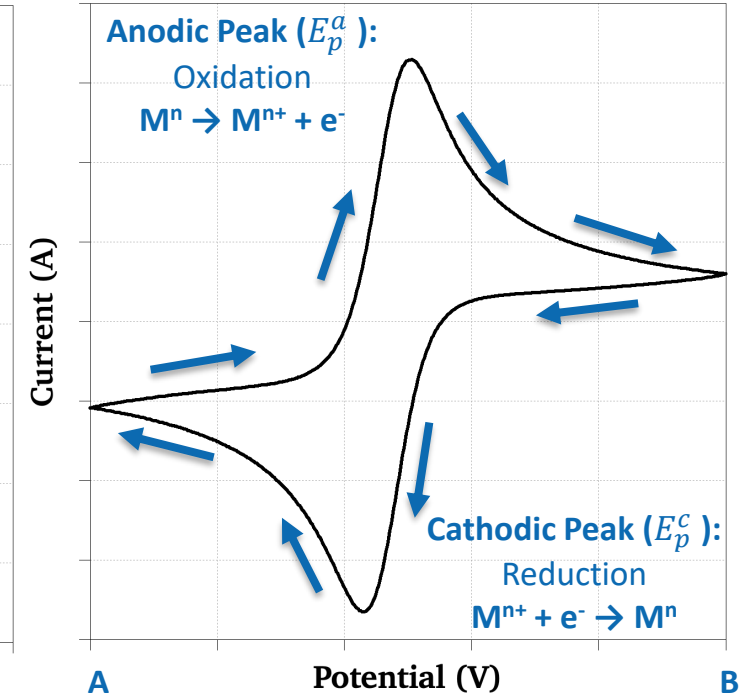
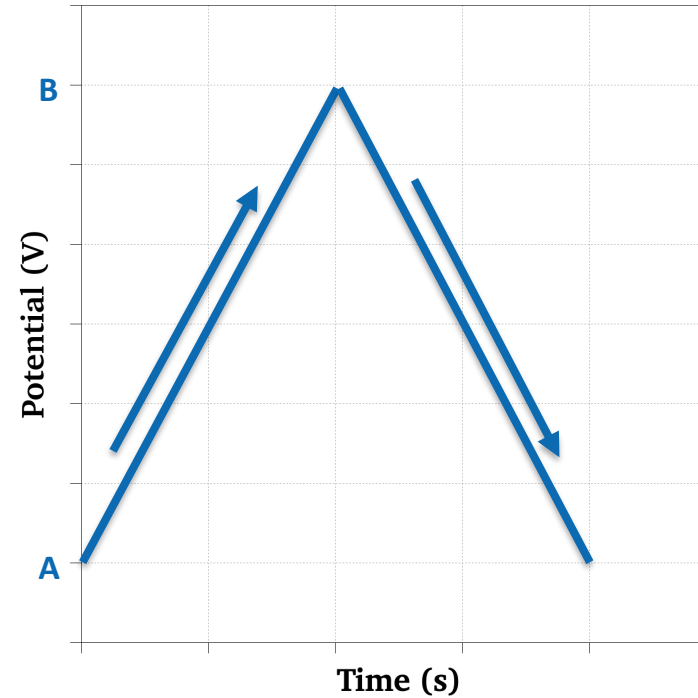
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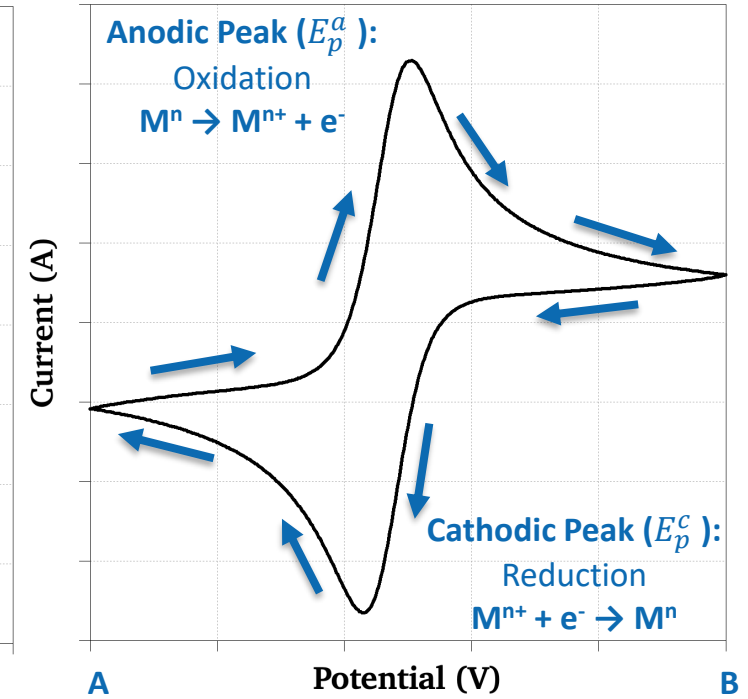
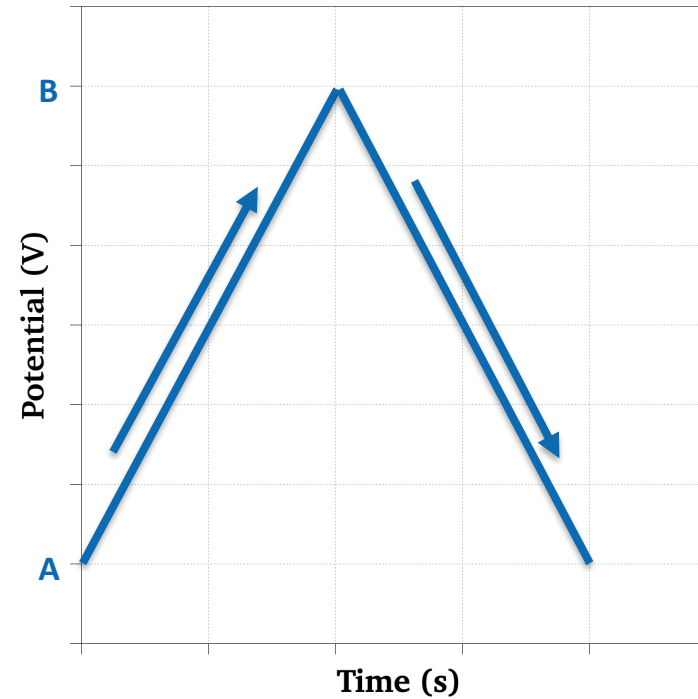
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The Mission:

Use *oxidation* to detect Pu in
MSR fuels with diamond

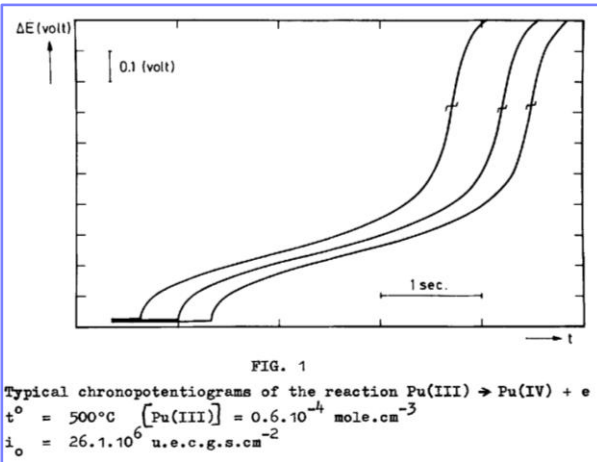
The White Whale: Pu^{4+/3+}

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Does it exist?

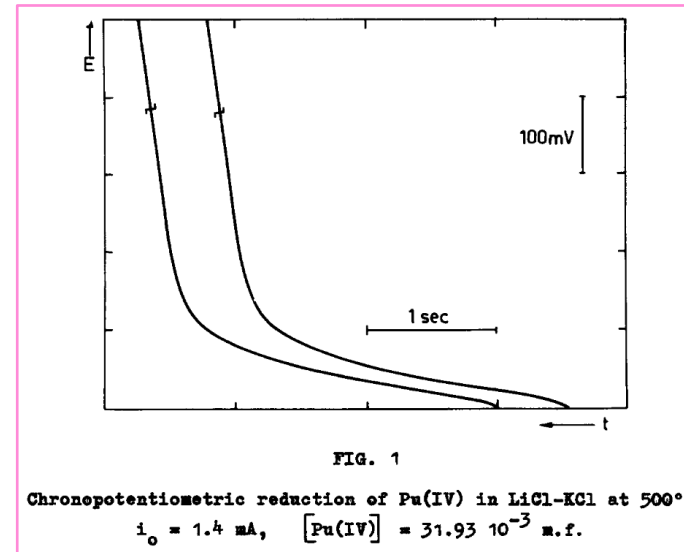
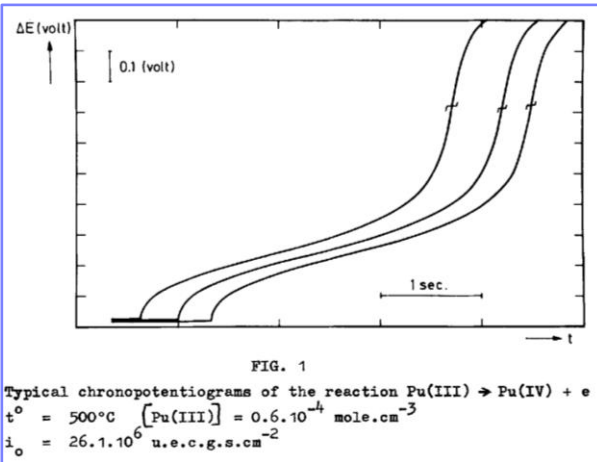
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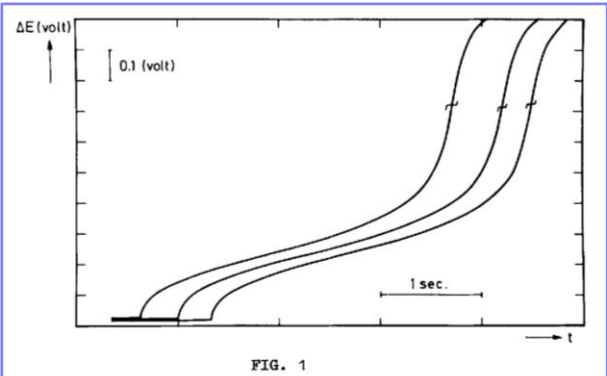


FIG. 1
Typical chronopotentiograms of the reaction $\text{Pu(III)} \rightarrow \text{Pu(IV)} + e$
 $t^{\circ} = 500^{\circ}\text{C}$ $[\text{Pu(III)}] = 0.6 \cdot 10^{-4} \text{ mole.cm}^{-3}$
 $i_o = 26.1 \cdot 10^6 \text{ u.e.c.g.s.cm}^{-2}$

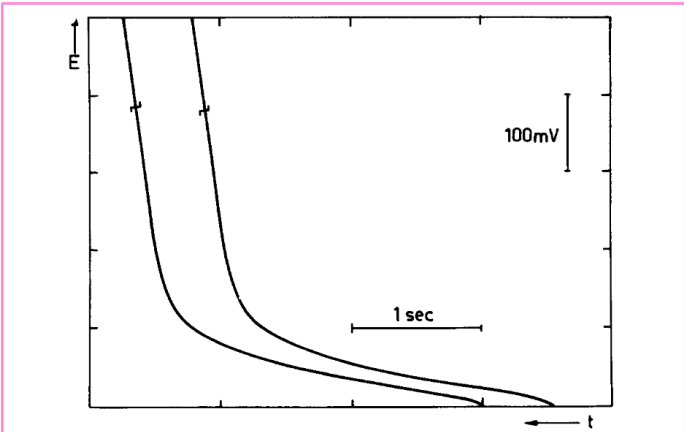


FIG. 1
Chronopotentiometric reduction of Pu(IV) in LiCl-KCl at 500°C
 $i_o = 1.4 \text{ mA}$, $[\text{Pu(IV)}] = 31.93 \cdot 10^{-3} \text{ m.f.}$

TABLE I
Diffusion coefficients $D \cdot 10^6 \text{ (cm}^2 \text{ sec}^{-1}\text{)}$ of tetravalent actinide ions in LiCl-KCl

$t(^{\circ}\text{C})$	U(IV)	Np(IV)	Pu(IV)
400	4.45 ± 0.2	4.65 ± 0.2	4.2 ± 0.2
450	8.0 ± 0.15	8.1 ± 0.1	7.2 ± 0.2
500	12.15 ± 0.2	11.7 ± 0.2	11.2 ± 0.25
550		15.75 ± 0.25	16.2 ± 0.25
600			22.4 ± 0.28
650			30.0 ± 0.5

TABLE II
Activation energy, $\Delta H \text{ (K.cal.mole}^{-1}\text{)}$, for the diffusion of tetravalent actinide ions in LiCl-KCl

$t(^{\circ}\text{C})$	U(IV)	Np(IV)	Pu(IV)
425	11.1	11.0	10.3
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575			9.2
625			9.3

The White Whale: Pu^{4+/3+}

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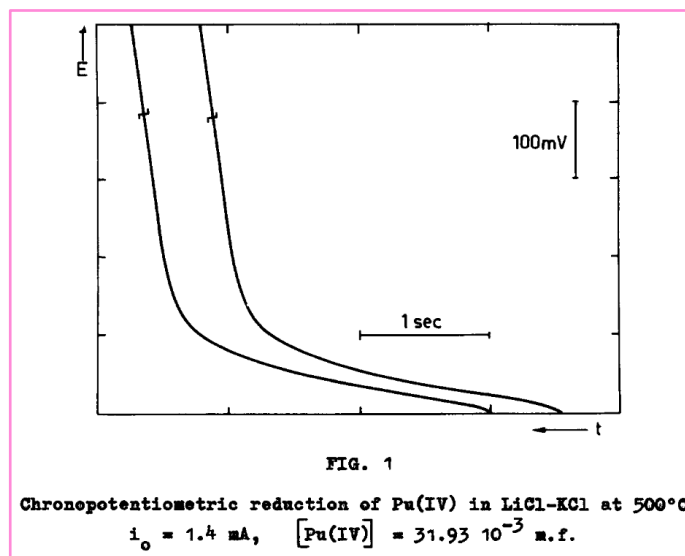
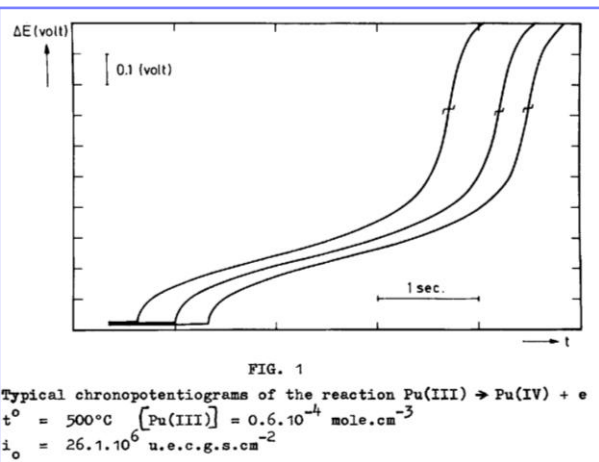


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TABLE XXII. STANDARD POTENTIAL FOR Pu(IV)|Pu(III) IN LiCl-KCl

t (°C)	$E^0_{\text{Pu(IV)} \text{Pu(III)}} - E^0_{\text{Cl}_2 2\text{Cl}^-}$ (V)
400	1.024
450	0.994
500	0.967

The standard potential for Pu(IV)|Pu(III) in LiCl - KCl has been recently measured by Martinot and Duyckaerts (Table XXII) and the thermodynamic quantities for the reaction $\text{AgCl} + \text{PuCl}_3 \rightarrow \text{Ag}^0 + \text{PuCl}_4$ were calculated:

	400°C	450°C	500°C
$-\Delta F^0$ (kcal/mole)	16.5	15.9	15.3
$-\Delta H^0$ (kcal/mole)	24.4	24.4	24.4
$-\Delta S^0$ (e.u.)	11.5	11.5	11.5

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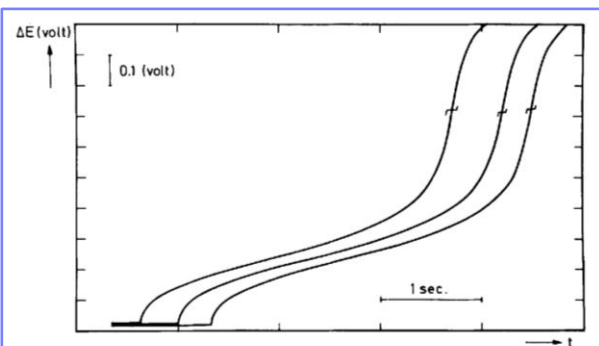


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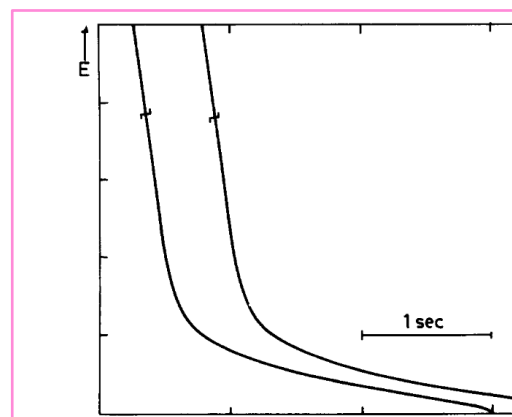


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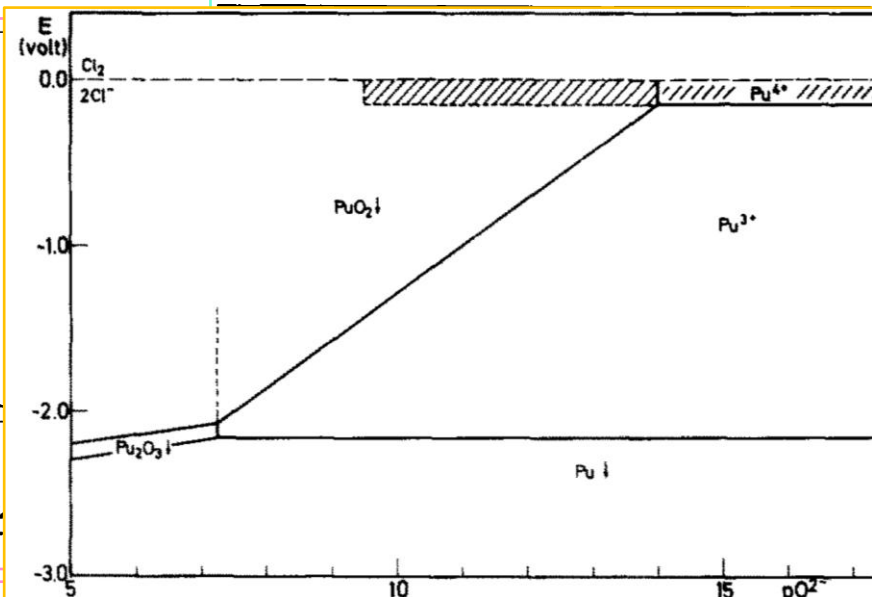


Fig. 4. Potential- $p\text{O}_2^-$ diagram of plutonium in (Li-K)Cl at 450 °C: $[\text{Pu}^{3+}] = [\text{Pu}^{4+}] = 10^{-2} \text{ mol l}^{-1}$.

This diagram must be considered only as tentative because a large error is involved in the value of $L(\text{PuO}_2)$.

our equilibrium diagram (Fig. 4). The standard potential of the couple $\text{Pu}^{3+}-\text{Pu}^0$ is equal to -2.86 V versus Cl_2-2Cl^- at 450 °C [20]. The only acceptable value for $E^0_{\text{Pu}^{4+}-\text{Pu}^{3+}}$ has been deduced from spectroscopic investigations [6] and found equal to -0.050 V versus Cl_2-2Cl^- at 400 °C; our determination by cyclic voltammetry gives $E^0_{\text{Pu}^{4+}-\text{Pu}^{3+}} = -0.15 \text{ V}$ at 450 °C. We use the latter value in Fig. 4.

The White Whale: Pu^{4+/3+}

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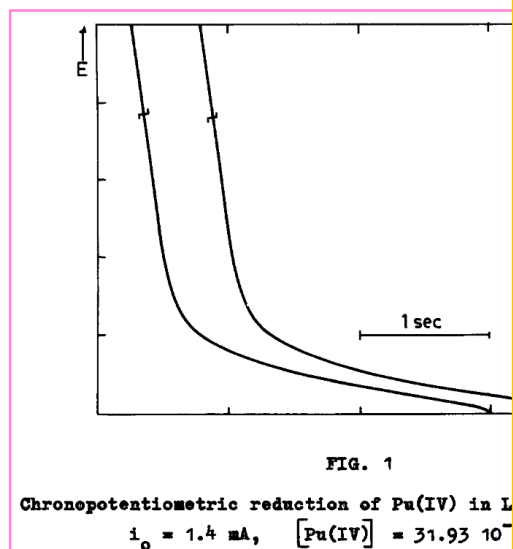
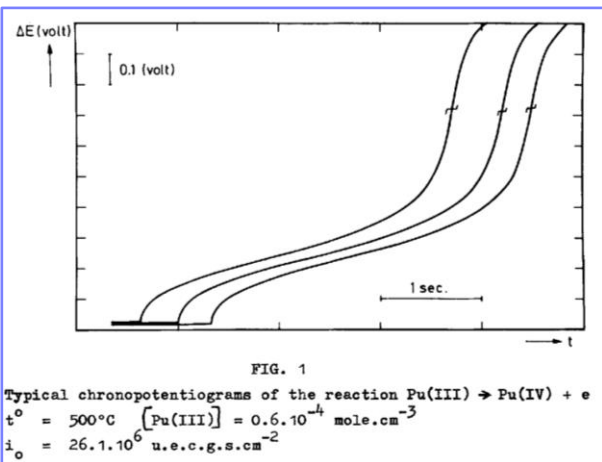


TABLE II
Activation energy, ΔH (K.cal.mole⁻¹), for the diffusion of tetravalent actinide ions in LiCl-KCl

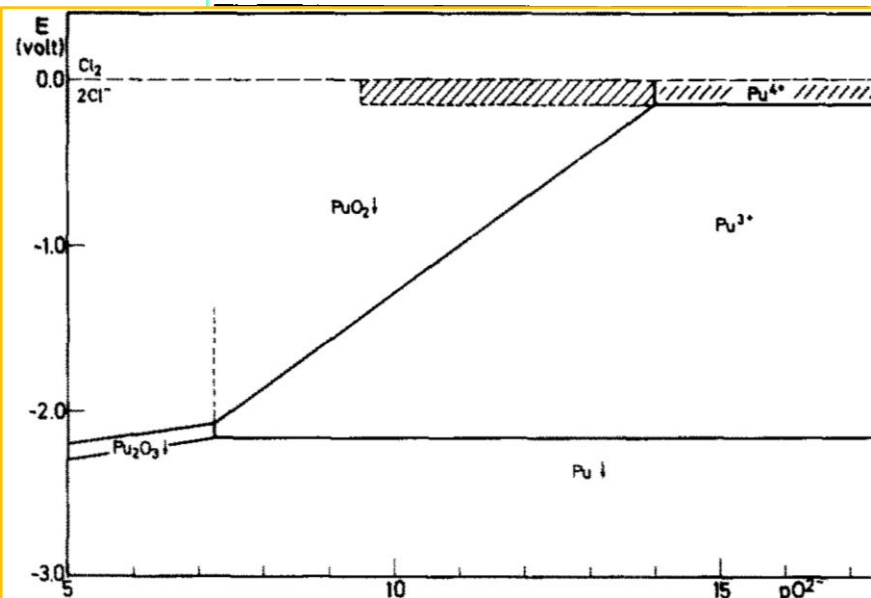
t(°C)	U(IV)	Np(IV)	Pu(IV)
425	11.1	11.0	10.3
475	9.4	9.5	9.7
525		7.5	9.5
575			9.2
625			9.3

TABLE I
Diffusion coefficients $D \cdot 10^6$ (cm² sec⁻¹) of tetravalent actinide ions in LiCl-KCl

t(°C)	U(IV)	Np(IV)	Pu(IV)
400	4.45 ± 0.2	4.65 ± 0.2	4.2 ± 0.2
450	8.0 ± 0.15	8.1 ± 0.1	7.2 ± 0.2
500	12.15 ± 0.2	11.7 ± 0.2	11.2 ± 0.25
550		15.75 ± 0.25	16.2 ± 0.25
600			22.4 ± 0.28
650			30.0 ± 0.5

TABLE XXII. STANDARD POTENTIAL FOR Pu(IV)|Pu(III) IN LiCl-KCl

t (°C)	$E^\circ_{\text{Pu(IV)} \text{Pu(III)}}$ (V)	$-E^\circ_{\text{Cl}_2 2\text{Cl}^-}$ (V)
-----------	---	--



This diagram must be considered only as tentative because a large error is involved in the value of $L(\text{PuO}_2)$.

our equilibrium diagram (Fig. 4). The standard potential of the couple $\text{Pu}^{3+}-\text{Pu}^0$ is equal to -2.86 V versus Cl_2-2Cl^- at 450 °C [20]. The only acceptable value for $E^\circ_{\text{Pu}^{4+}-\text{Pu}^{3+}}$ has been deduced from spectroscopic investigations [6] and found equal to -0.050 V versus Cl_2-2Cl^- at 400 °C; our determination by cyclic voltammetry gives $E^\circ_{\text{Pu}^{4+}-\text{Pu}^{3+}} = -0.15 \text{ V}$ at 450 °C. We use the latter value in Fig. 4.

The White Whale: Pu^{4+/3+}

Does it exist?

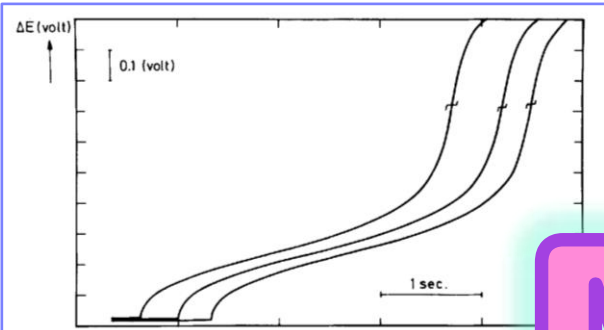


FIG. 1
Typical chronopotentiograms of the reaction Pu(III) → Pu(IV)
 $t^0 = 500^\circ\text{C}$ $[\text{Pu(III)}] = 0.6 \cdot 10^{-4} \text{ mole} \cdot \text{cm}^{-3}$
 $i_0 = 26 \cdot 10^{-6} \text{ u.e.c.g.s.cm}^{-2}$

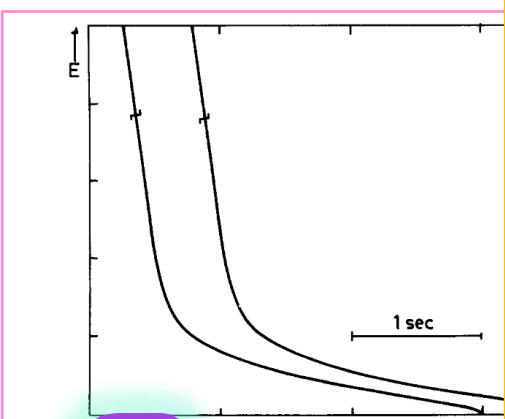


FIG. 1
Chronopotentiograms of the reaction Pu(III) → Pu(IV)
at $t^0 = 500^\circ\text{C}$ and $[\text{Pu(III)}] = 0.6 \cdot 10^{-4} \text{ mole} \cdot \text{cm}^{-3}$
for different current densities i_0 (u.e.c.g.s.cm⁻²):
 $i_0 = 26 \cdot 10^{-6}$ (—), $52 \cdot 10^{-6}$ (---), $78 \cdot 10^{-6}$ (· · · · ·)

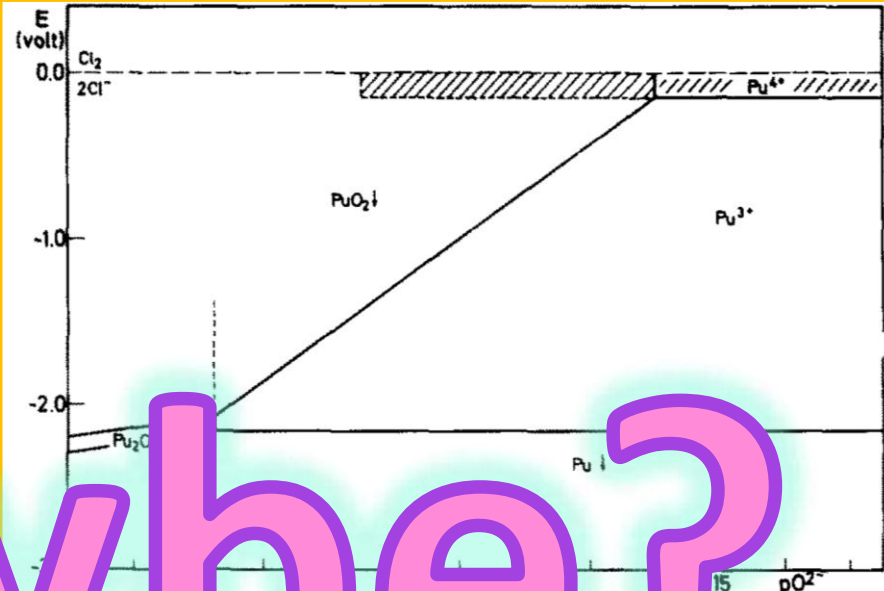
TABLE I
Diffusion coefficients $D \cdot 10^6 \text{ (cm}^2 \text{ sec}^{-1})$ of
tetravalent actinide ions in LiCl-KCl

$t(^{\circ}\text{C})$	U(IV)	Np(IV)	Pu(IV)
400	4.45 ± 0.2	4.65 ± 0.2	4.2 ± 0.2
450	8.0 ± 0.15	8.1 ± 0.1	7.2 ± 0.2
500	12.15 ± 0.2	11.7 ± 0.2	11.2 ± 0.25
550		15.75 ± 0.25	16.2 ± 0.25
600			22.4 ± 0.28
650			30.0 ± 0.5

Maybe?

TABLE XXII. STANDARD POTENTIAL FOR Pu(IV)|Pu(III) IN LiCl-KCl

t ($^{\circ}\text{C}$)	$E^0_{\text{Pu(IV)} \text{Pu(III)}} - E^0_{\text{Cl}_2 2\text{Cl}^-}$ (V)
-------------------------------	--



4. Potential-pO₂ diagram of plutonium in (Li-K)Cl at 450 °C: $[\text{Pu}^{3+}] = [\text{Pu}^{4+}] = 10^{-4} \text{ mole} \cdot \text{cm}^{-3}$

This diagram must be considered only as tentative because a large error is involved in the value of $L(\text{PuO}_2)$.

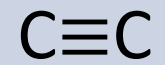
our equilibrium diagram (Fig. 4). The standard potential of the couple $\text{Pu}^{3+}-\text{Pu}^0$ is equal to -2.86 V versus Cl_2-2Cl^- at 450°C [20]. The only acceptable value for $E^0_{\text{Pu}^{4+}-\text{Pu}^{3+}}$ has been deduced from spectroscopic investigations [6] and found equal to -0.050 V versus Cl_2-2Cl^- at 400°C ; our determination by cyclic voltammetry gives $E^0_{\text{Pu}^{4+}-\text{Pu}^{3+}} = -0.15 \text{ V}$ at 450°C . We use the latter value in Fig. 4.



The Key: Boron Doped Diamond

Diamond & Carbon Hybridization

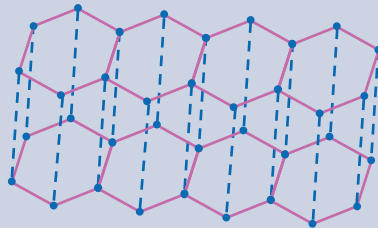
sp



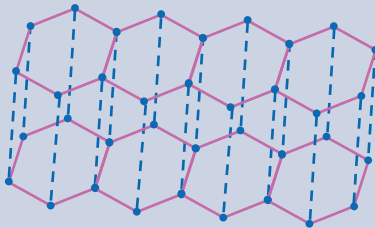
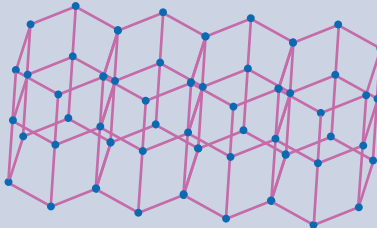
$1\ \sigma, 2\ \pi$



Diamond & Carbon Hybridization

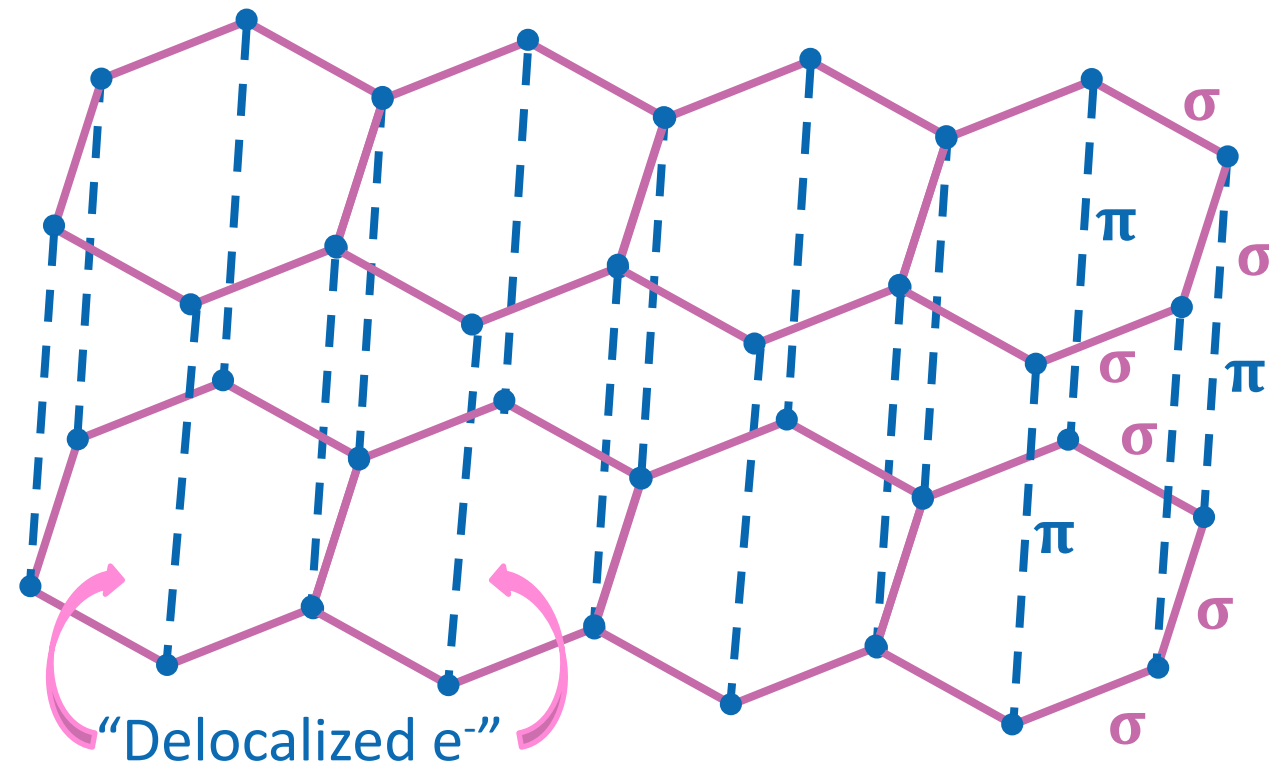
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$\text{C}\equiv\text{C}$	$\text{C}=\text{C}$	$\begin{array}{c} \text{C}-\text{C} \\ \quad \\ \text{C}-\text{C} \end{array}$
1 σ , 2 π	1 σ , 1 π	1 σ , π plane
$\text{H}-\text{C}\equiv\text{C}-\text{H}$		

Diamond & Carbon Hybridization

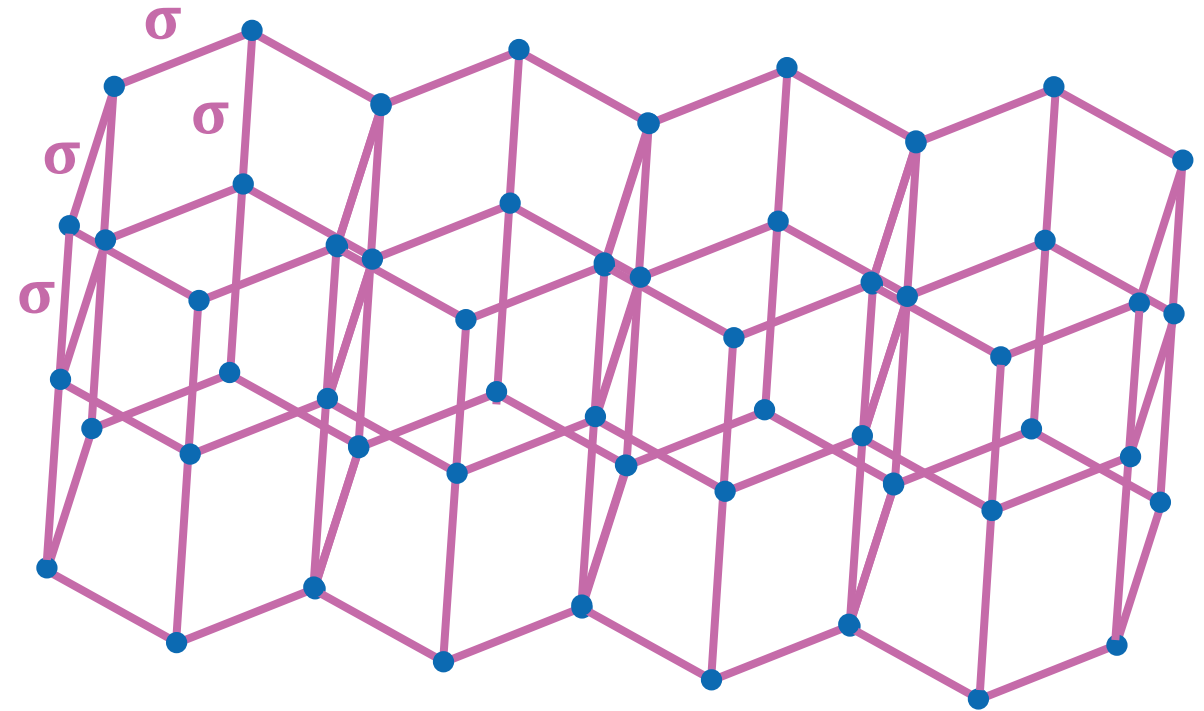
sp	sp ²		sp ³	
$\text{C}\equiv\text{C}$	$\text{C}=\text{C}$	$\begin{array}{c} \text{C}-\text{C} \\ \quad \\ \text{C}-\text{C} \end{array}$	$\text{C}-\text{C}$	
1 σ , 2 π	1 σ , 1 π	1 σ , π plane	1 σ	
$\text{H}-\text{C}\equiv\text{C}-\text{H}$			$\begin{array}{c} \text{H} \quad \text{H} \\ \quad \\ \text{H}-\text{C}-\text{C}-\text{H} \\ \quad \\ \text{H} \quad \text{H} \end{array}$	

Diamond & Carbon Hybridization

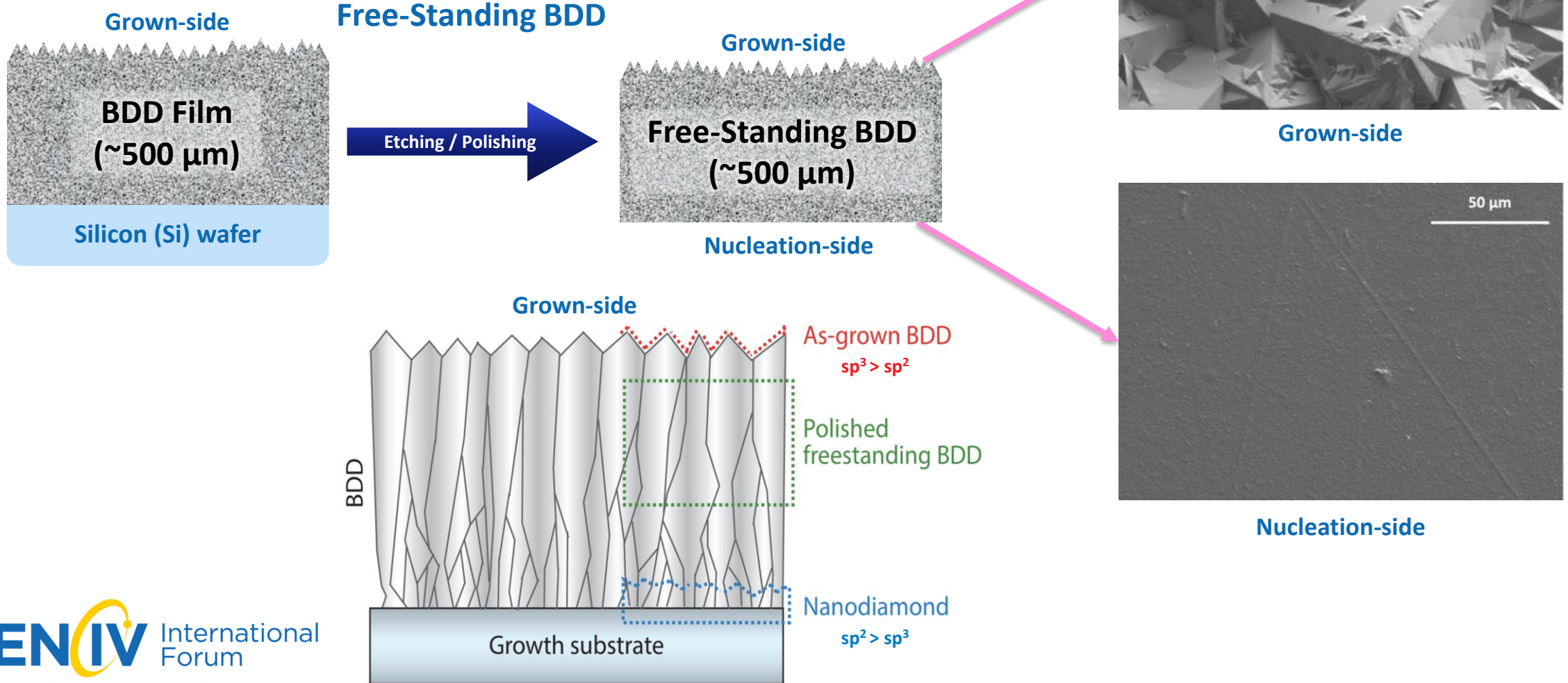
Graphite Sheets



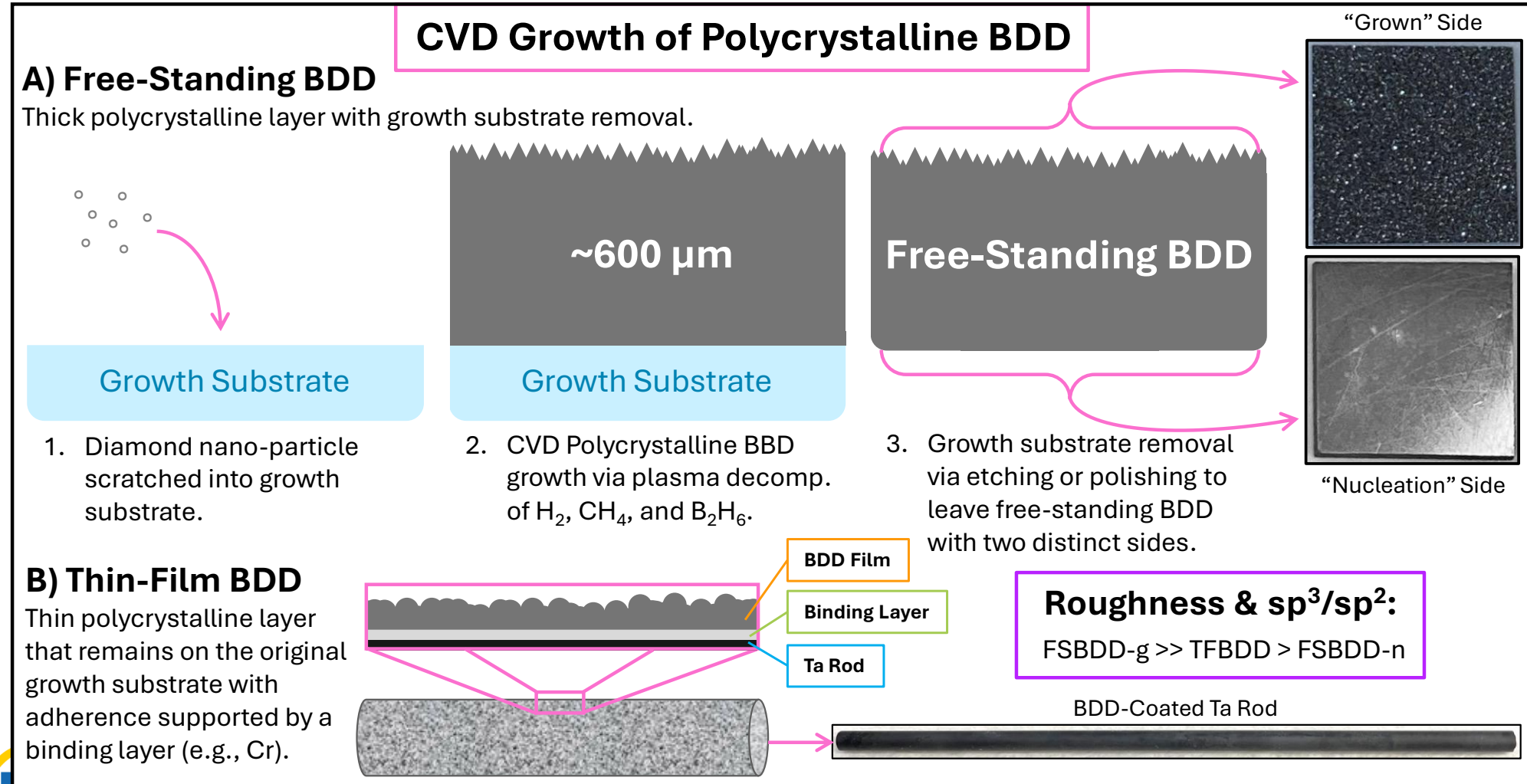
Diamond Crystal



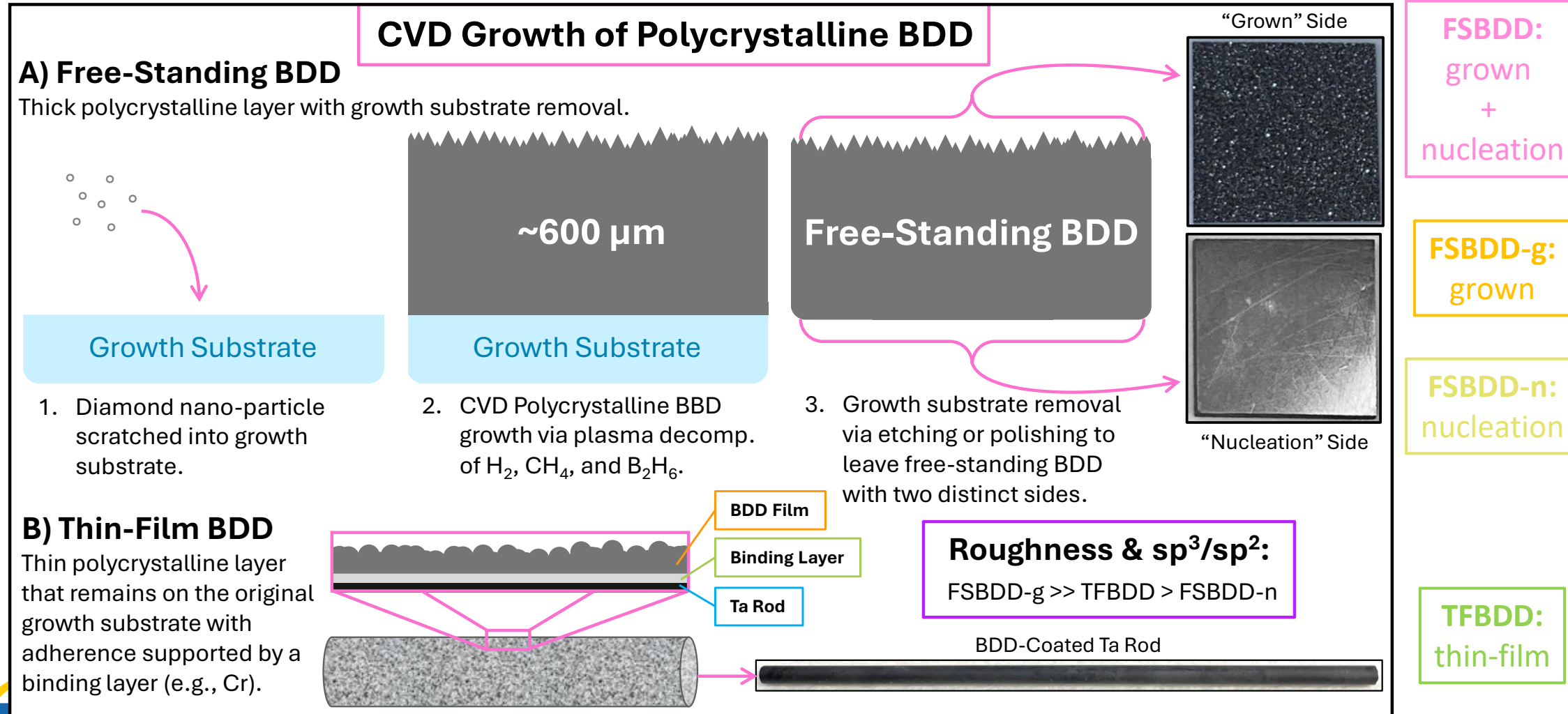
Boron Doped Diamond



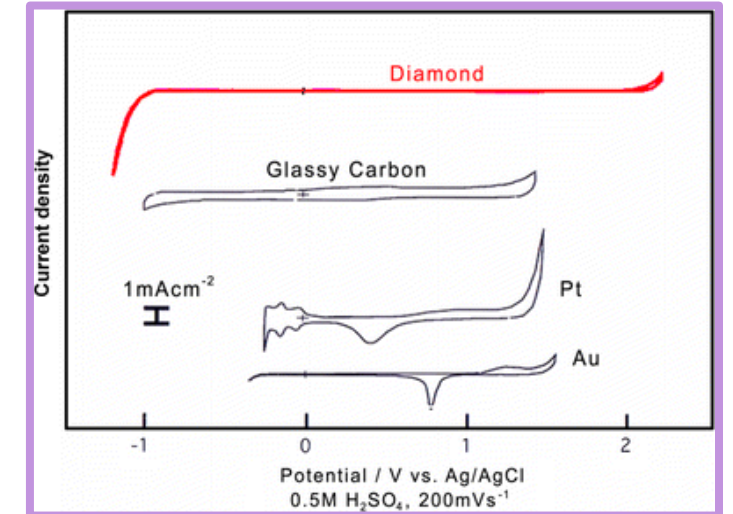
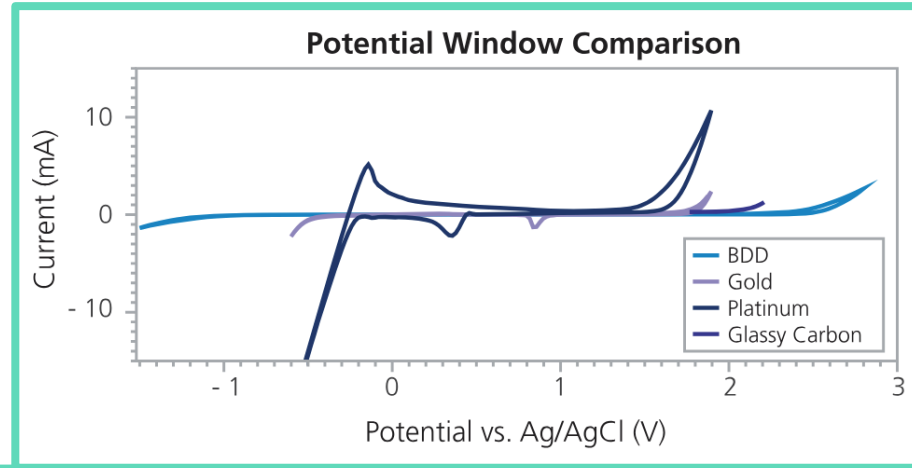
Boron Doped Diamond (BDD): Chemical Vapor Deposition



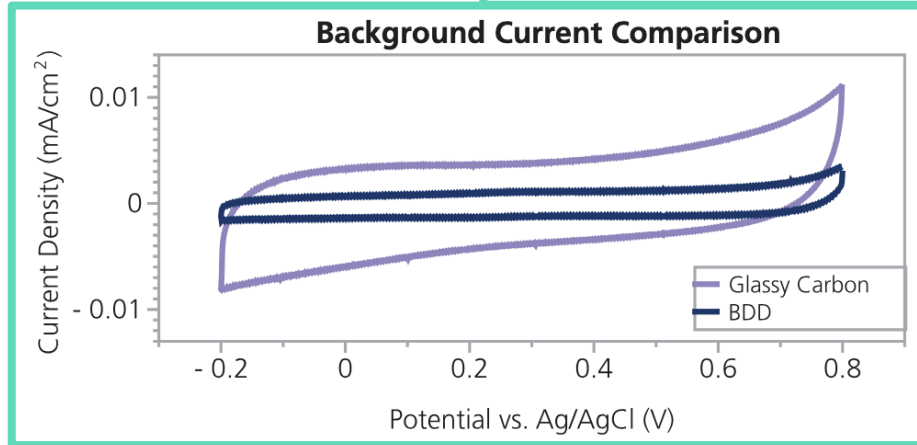
Boron Doped Diamond (BDD): Chemical Vapor Deposition



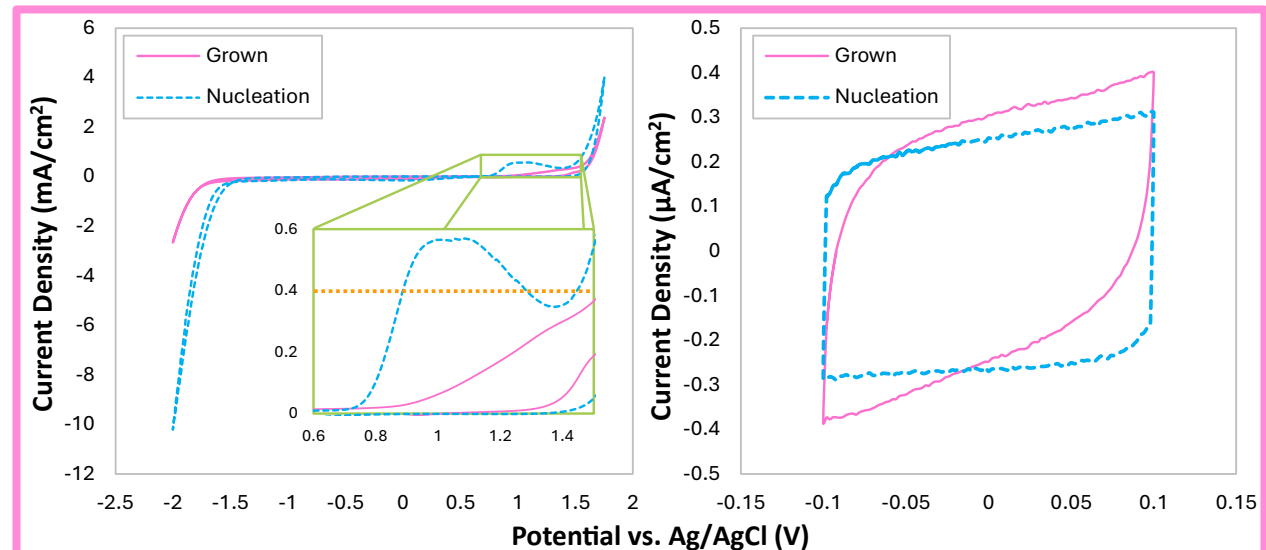
Boron Doped Diamond (BDD): Comparative Performance



Einaga, Y. J. *App. Electrochem.*, 40, 1807 (2010).



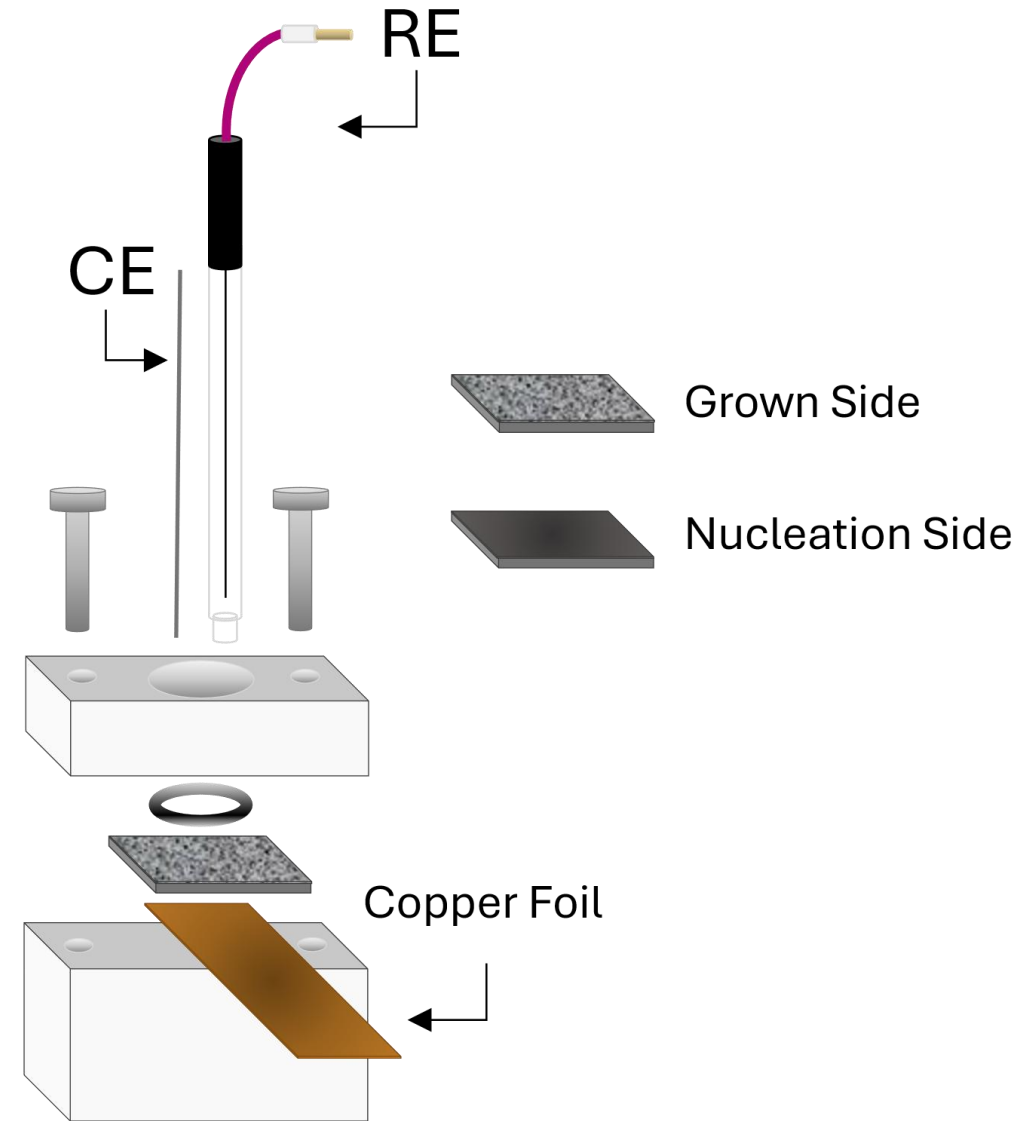
Fraunhofer USA



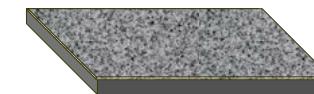
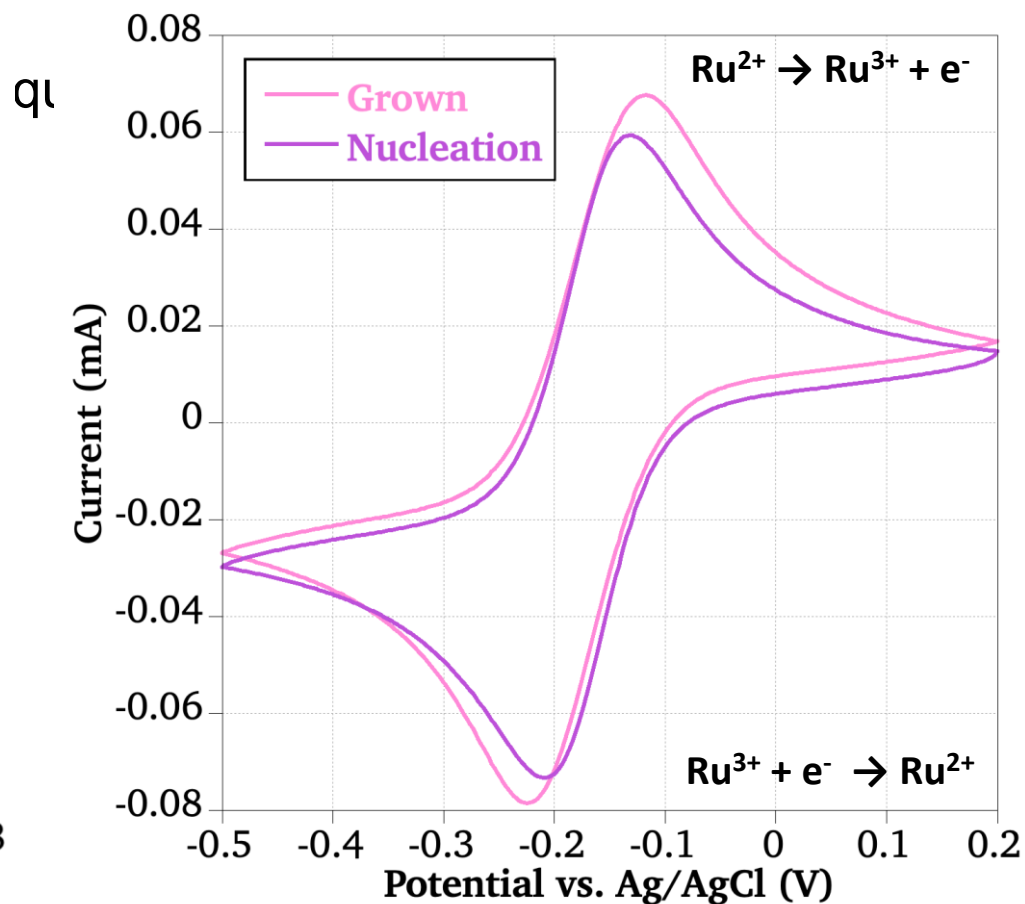
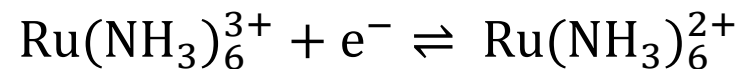
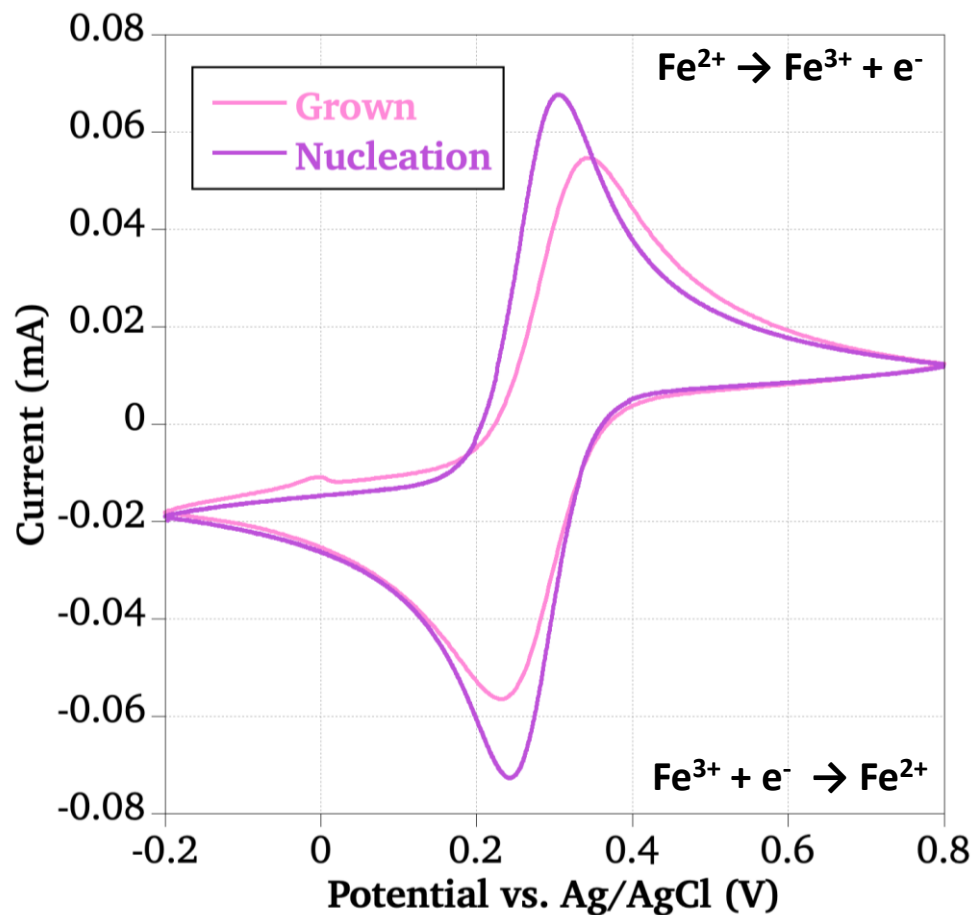
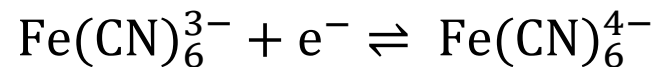
Patenaude et al., in preparation

Aqueous Cyclic Voltammetry

- **Side-Isolating Cell**
- **WE:** Grown or Nucleation Side, BDD
- **RE:** Ag/AgCl (3.5 M KCl)
- **CE:** Pt Wire
- **Sample Solutions**
 - 1 mM $\text{K}_3\text{Fe}(\text{CN})_6$ (1 M KCl)
 - 1 mM $\text{Cl}_3\text{Ru}(\text{NH}_3)_6$ (1 M KCl)
- **Scan Rate Studies**
 - 10, 25, 50, 75, 100 mV/s



Aqueous Cyclic Voltammetry



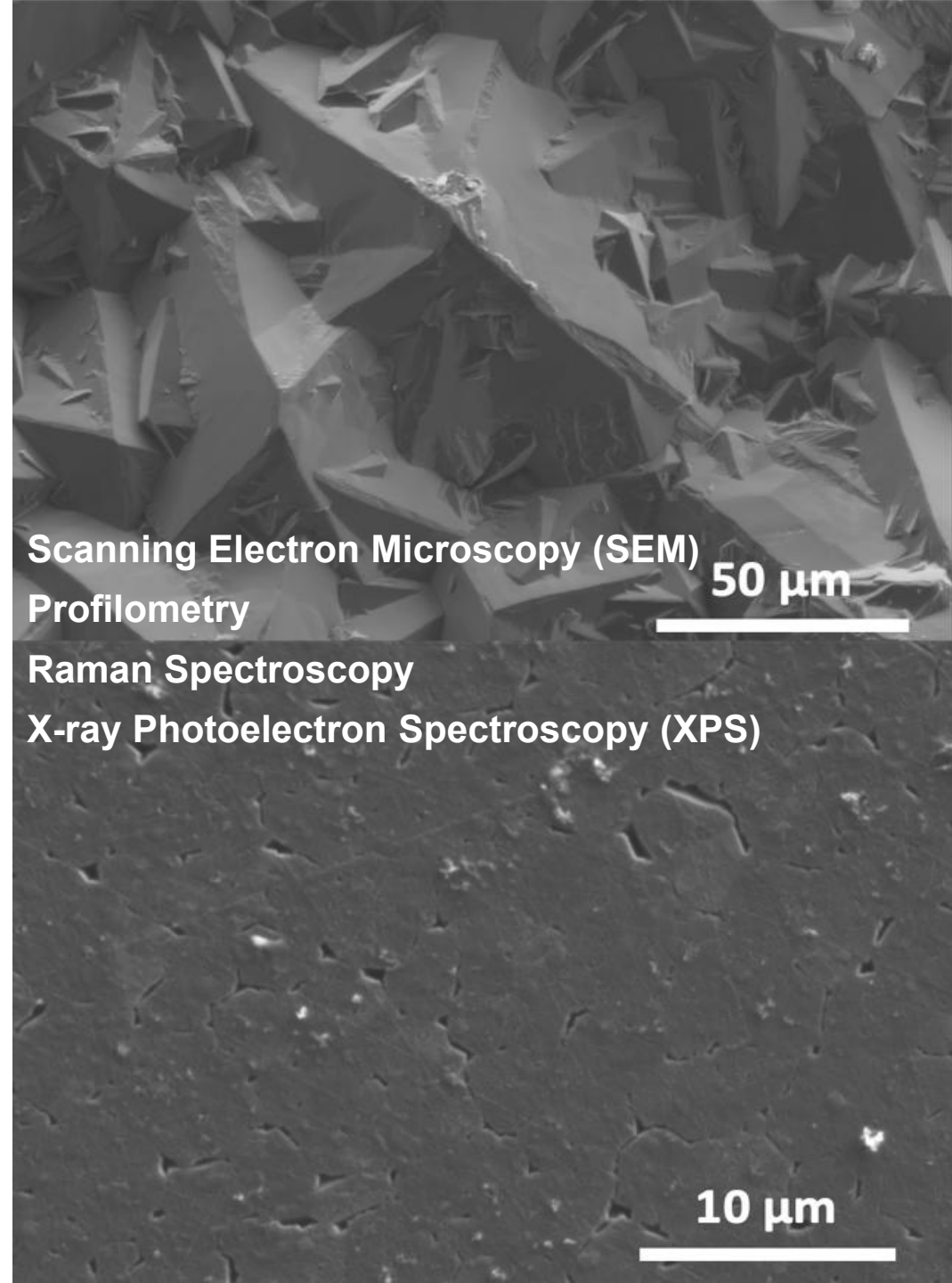


Exposure Studies: Does BDD Survive Molten Salts?

Salt Exposure

NaCl-KCl	FLiNaK
50-50 mol%	46.5-11.5-42 mol% LiF-NaF-KF
700 °C	500 °C

- 10 Day Hold
- 'As received' salt:
 - Furnace in fume hood
- Thermally dried salt
 - Vacuum oven antechamber
 - Furnace in glovebox
 - Ar(g), <0.5 ppm O₂, <0.5 ppm H₂O

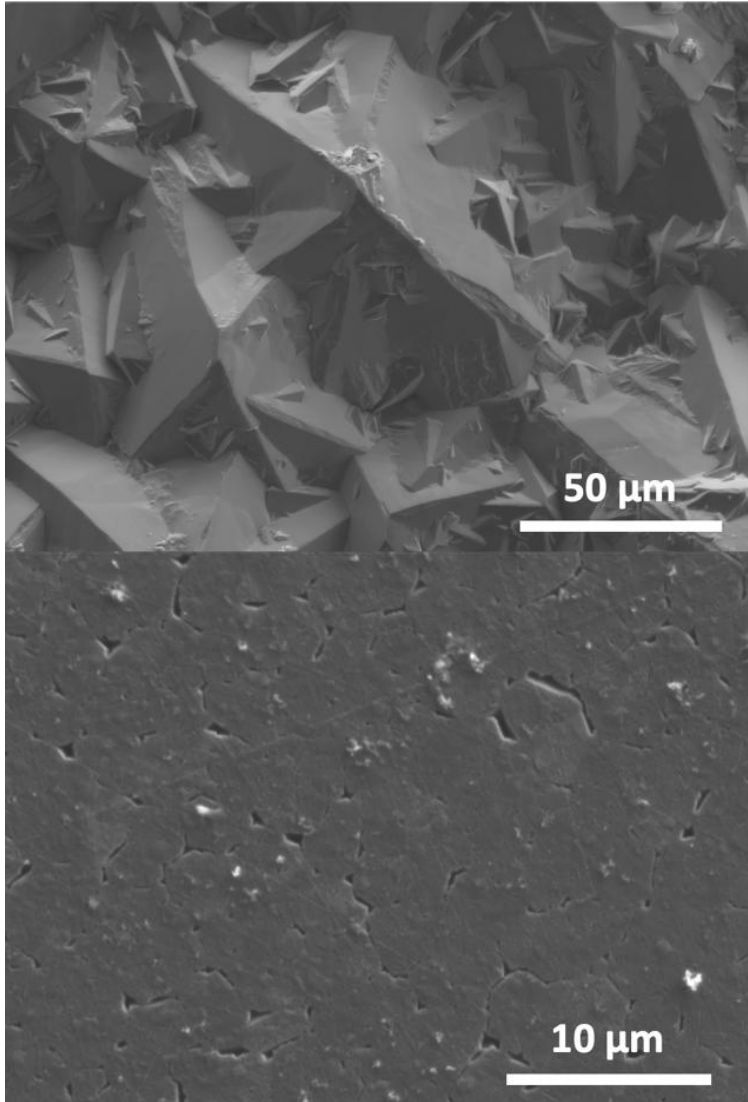


Scanning Electron Microscopy (SEM)
Profilometry

Raman Spectroscopy

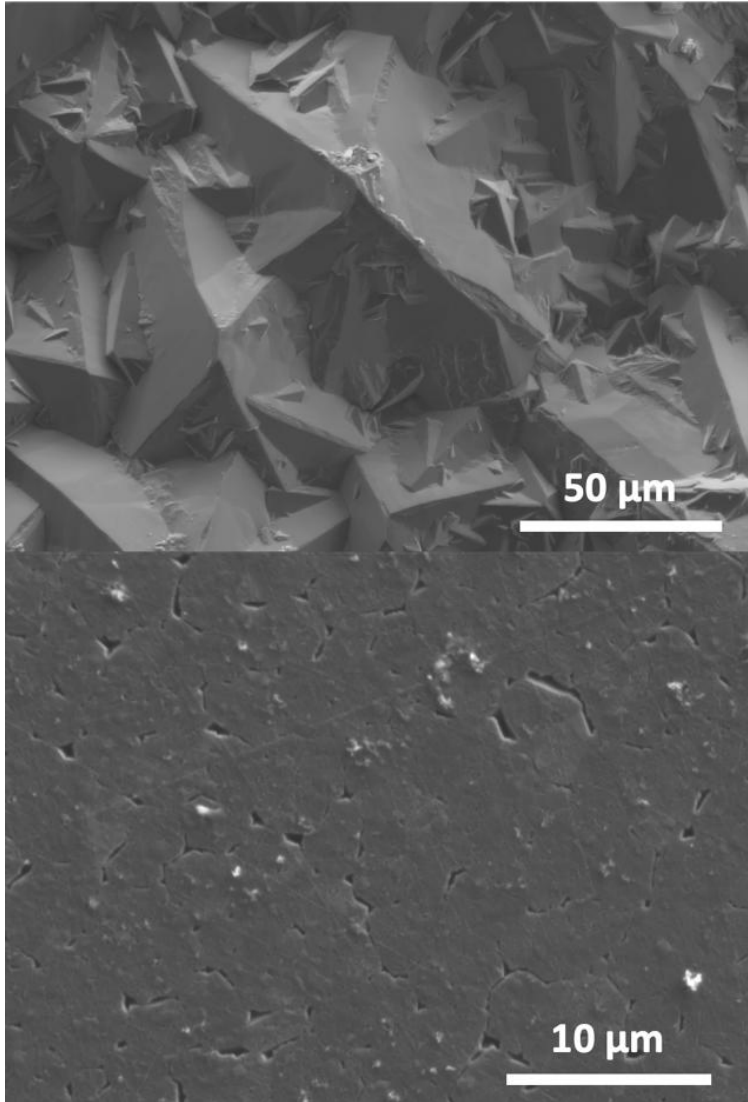
X-ray Photoelectron Spectroscopy (XPS)

Scanning Electron Microscopy (SEM): NaCl-KCl Exposure

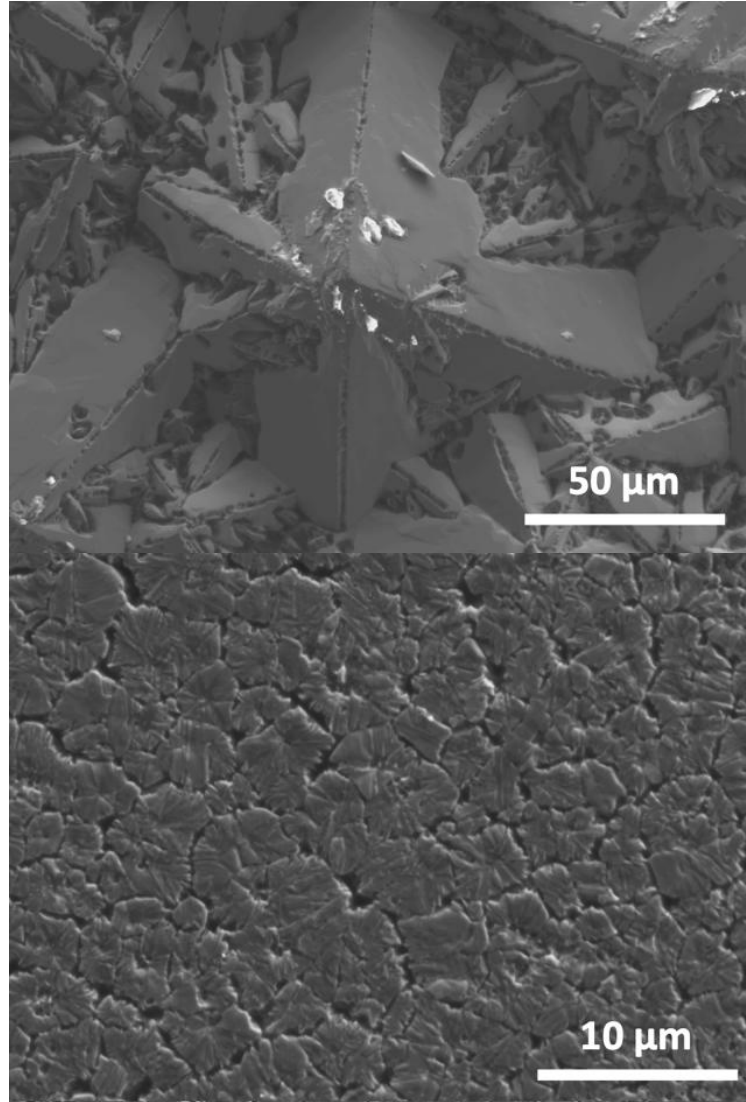


New

Scanning Electron Microscopy (SEM): NaCl-KCl Exposure

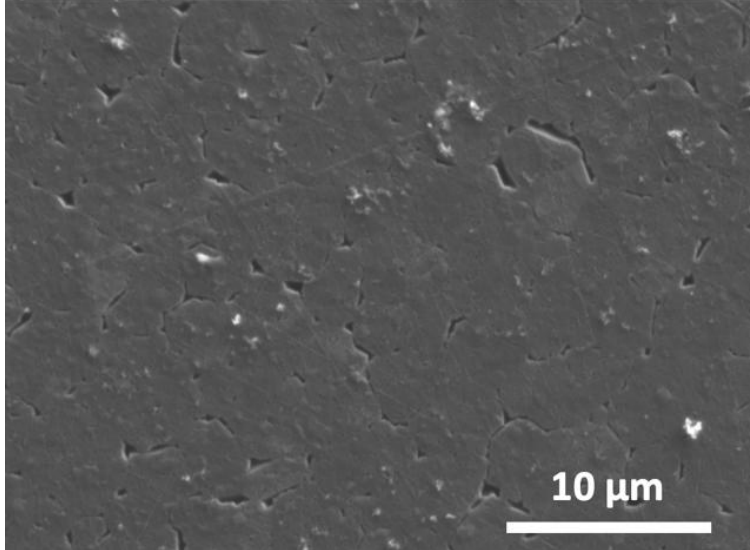
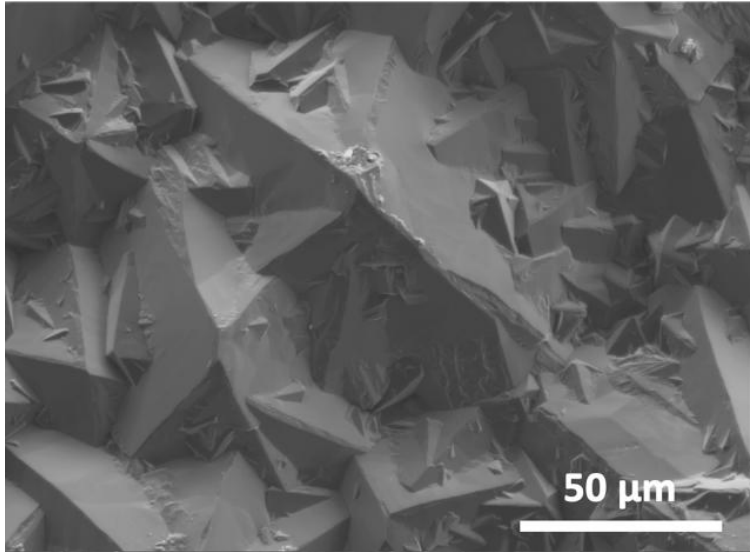


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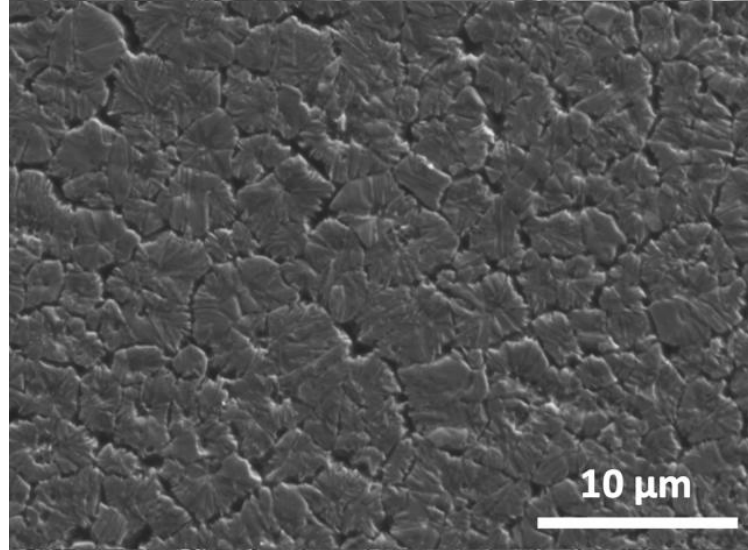
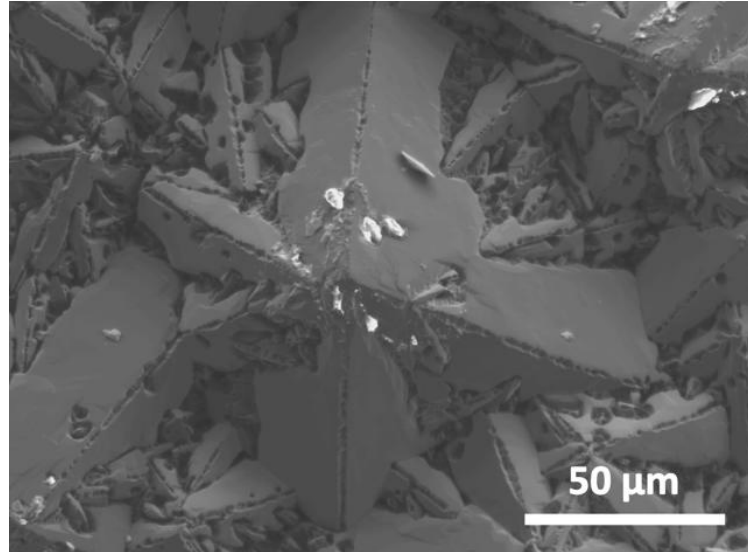


Not Dried | Fume Hood

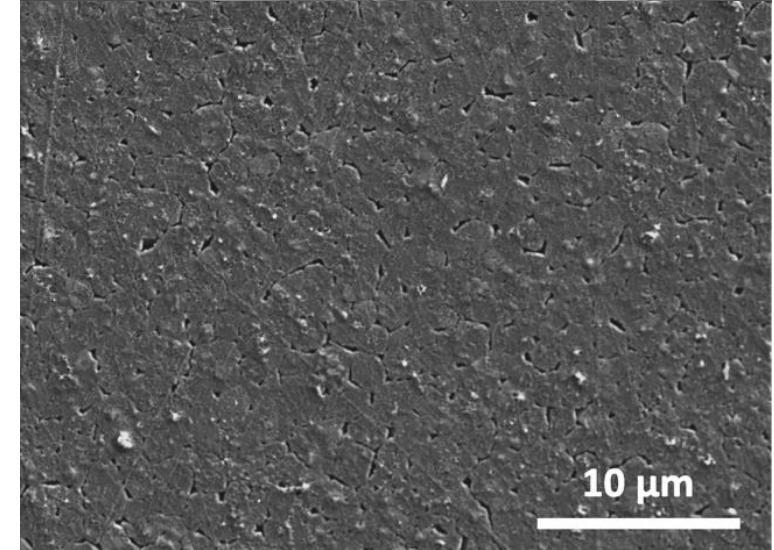
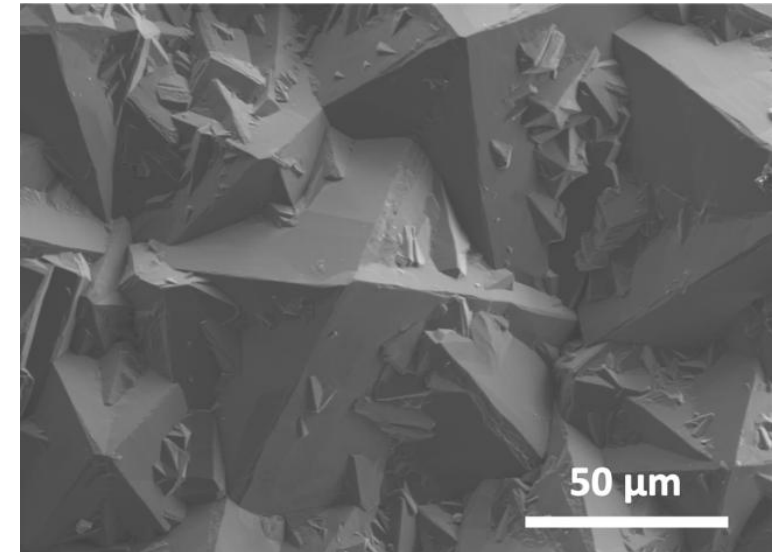
Scanning Electron Microscopy (SEM): NaCl-KCl Exposure



New

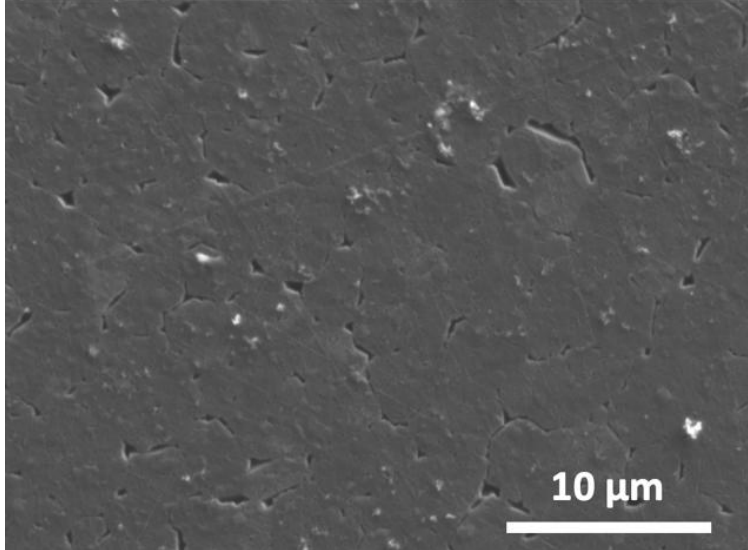
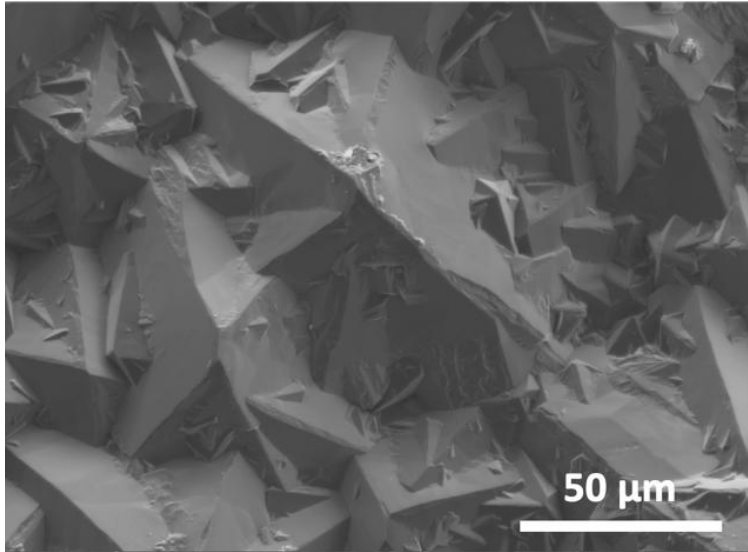


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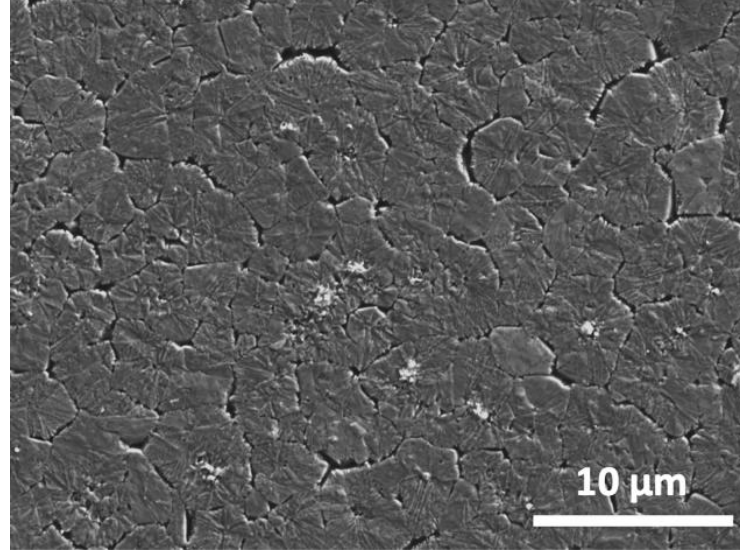
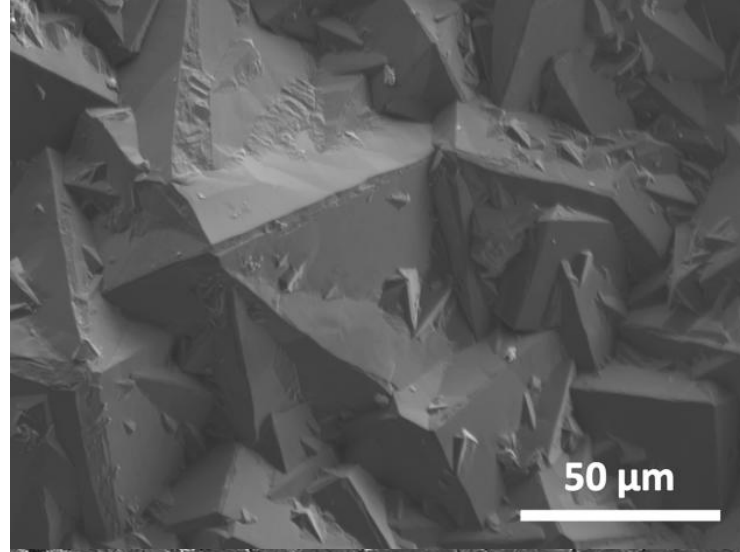


Dried | Glovebox

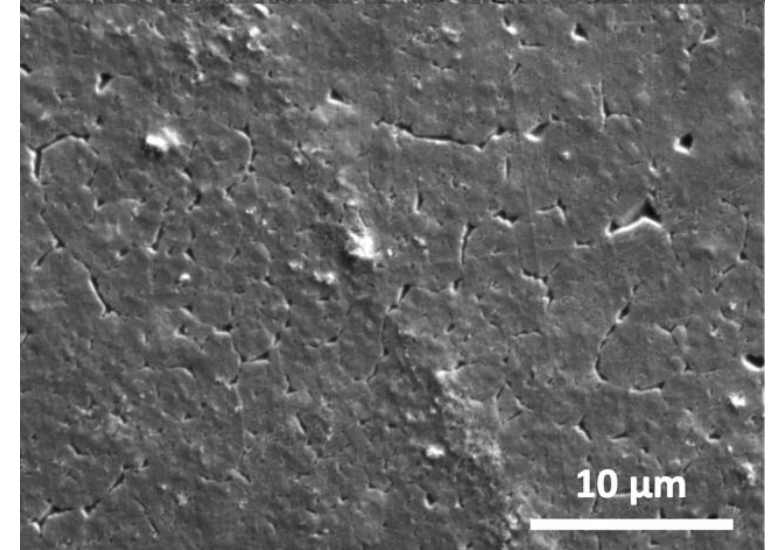
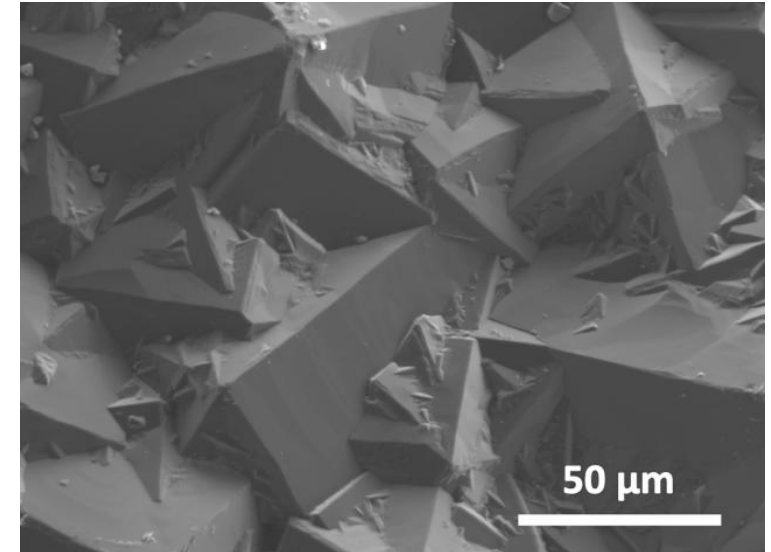
Scanning Electron Microscopy (SEM): FLiNaK Exposure



New



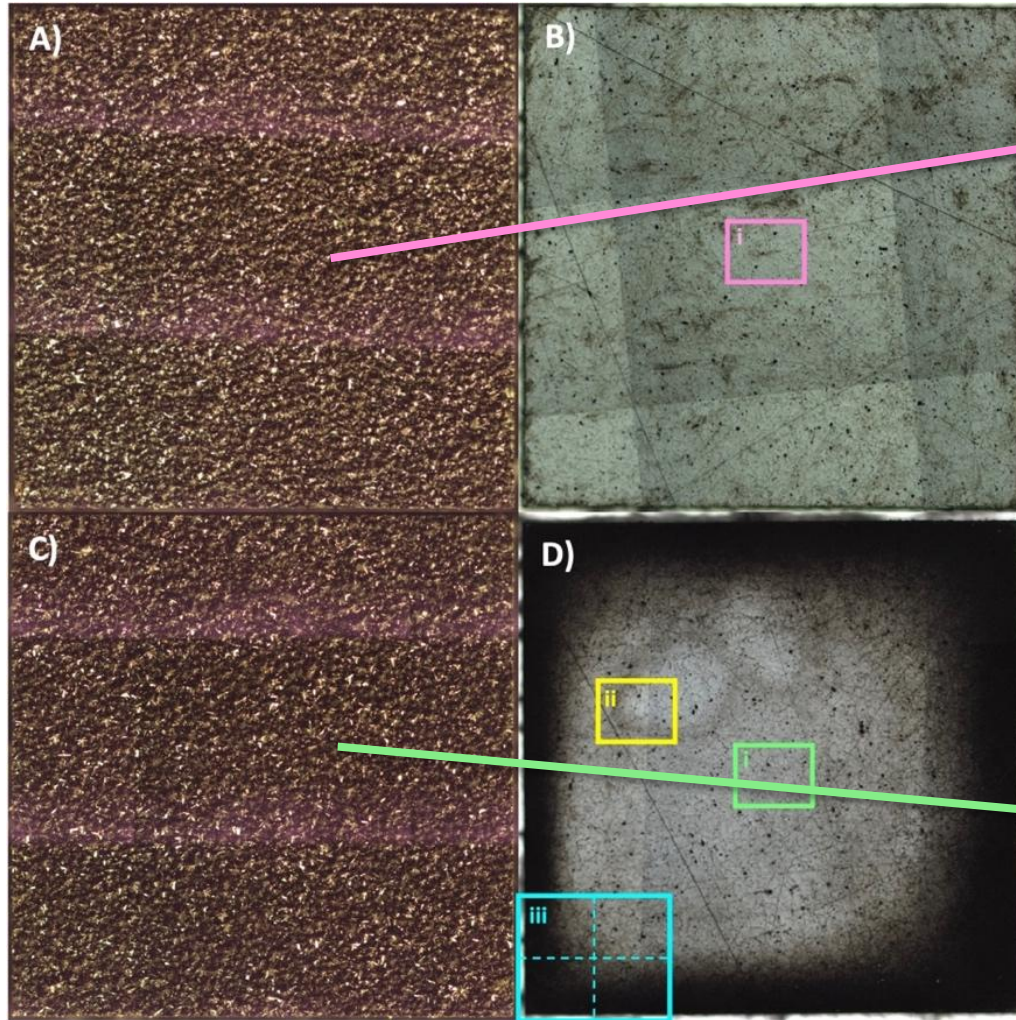
Not Dried | Fume Hood



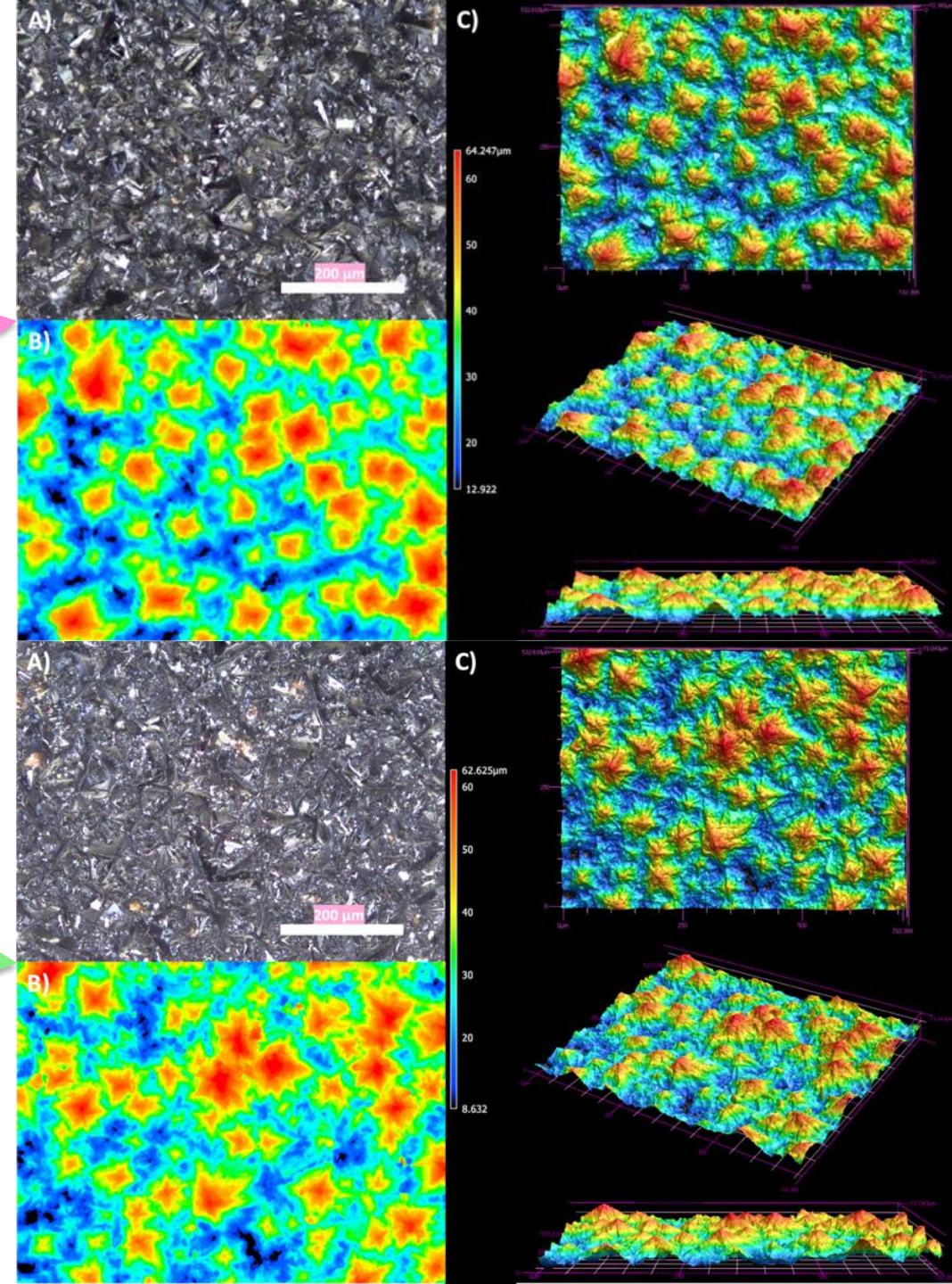
Dried | Glovebox

Profilometry

New BDD

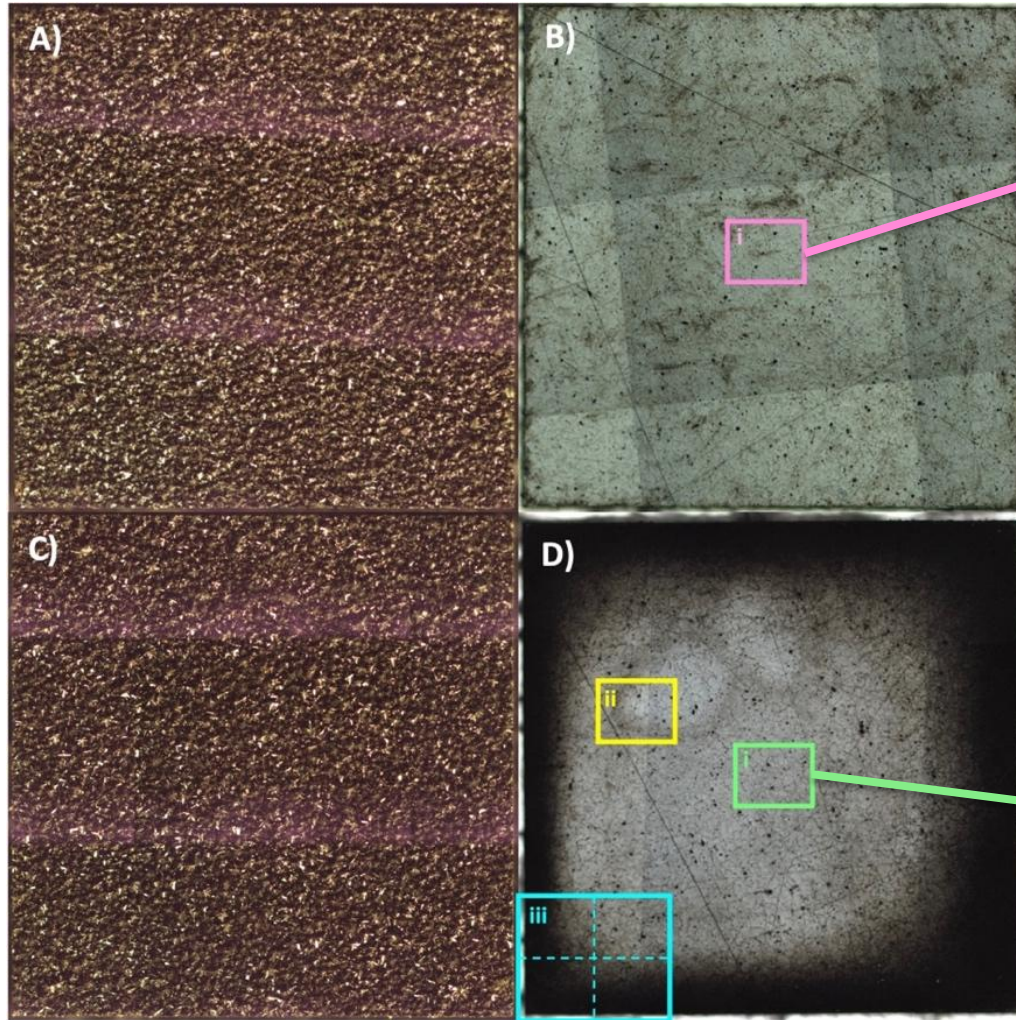


NaCl-KCl (Corroded) BDD

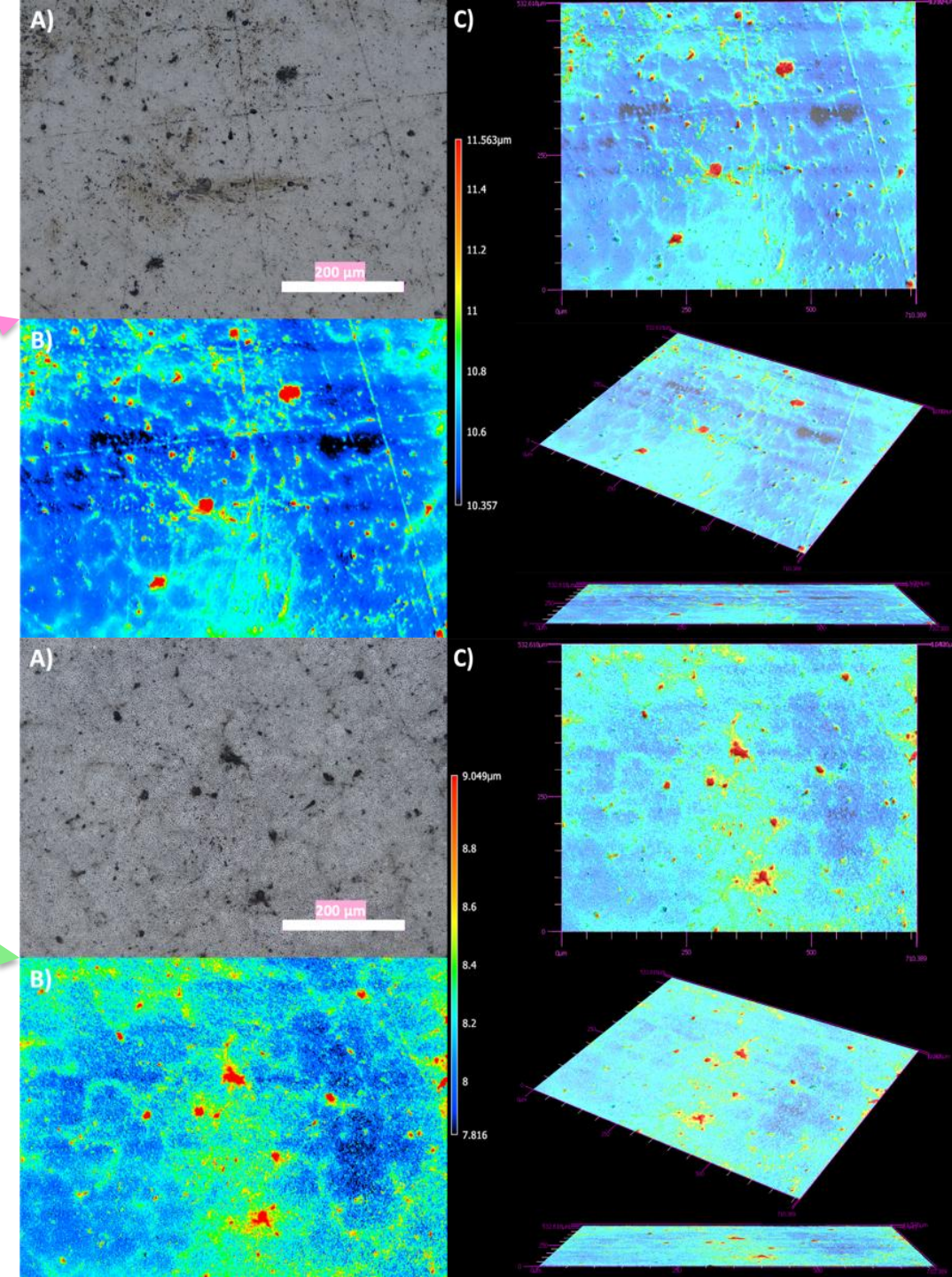


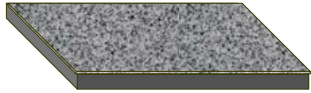
Profilometry

New BDD



NaCl-KCl (Corroded) BDD

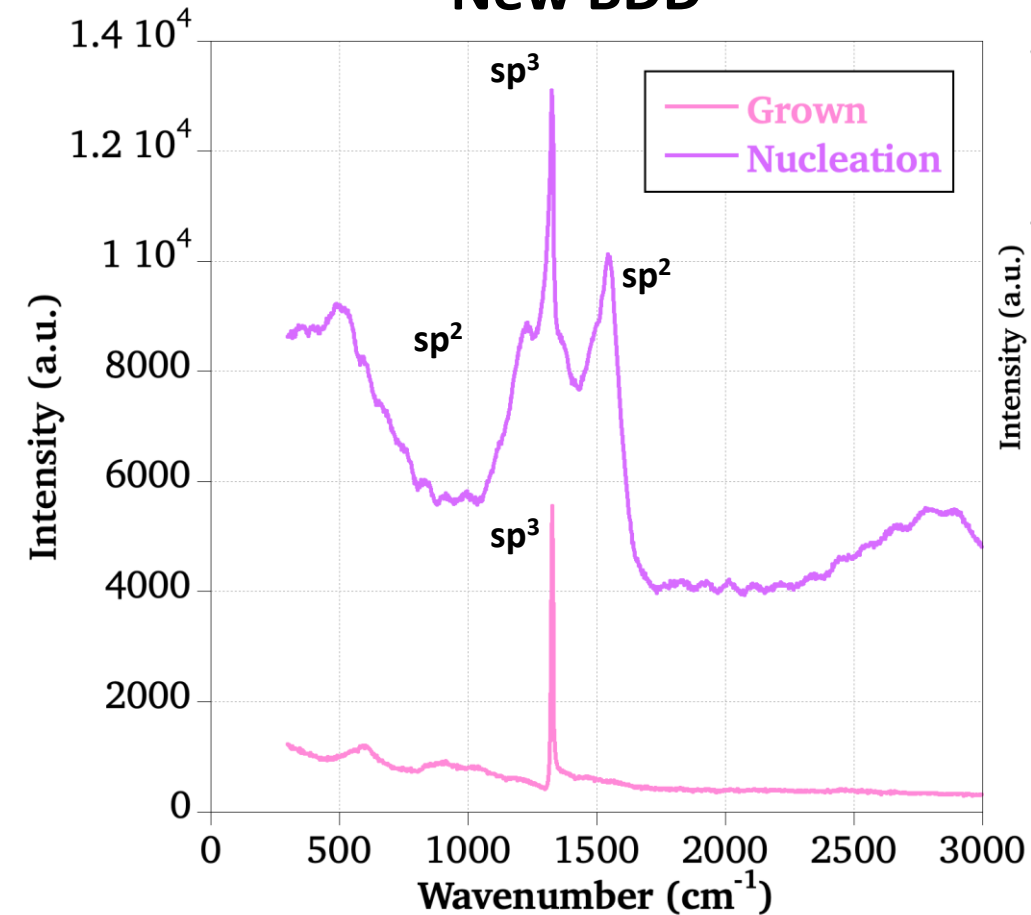




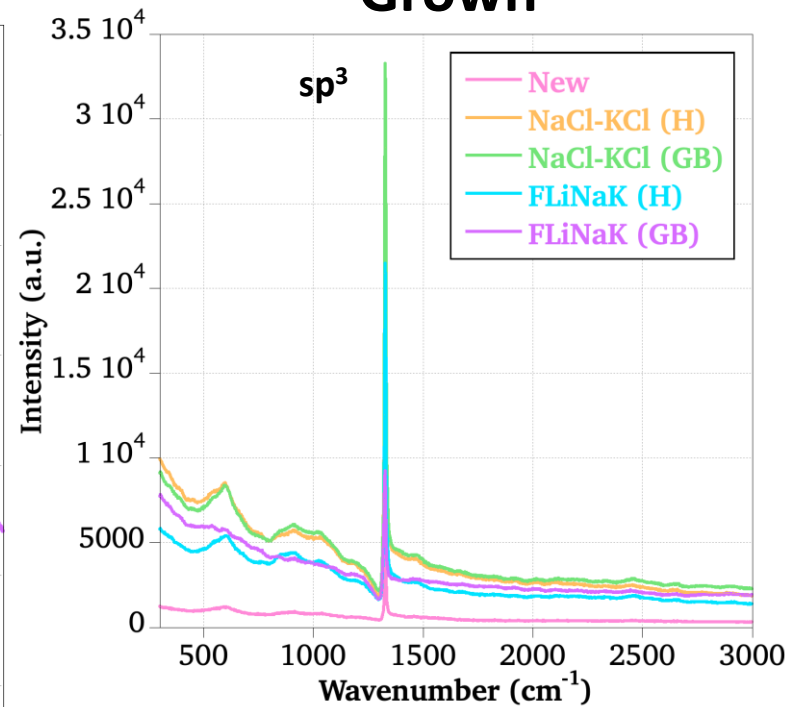
Raman Spectroscopy



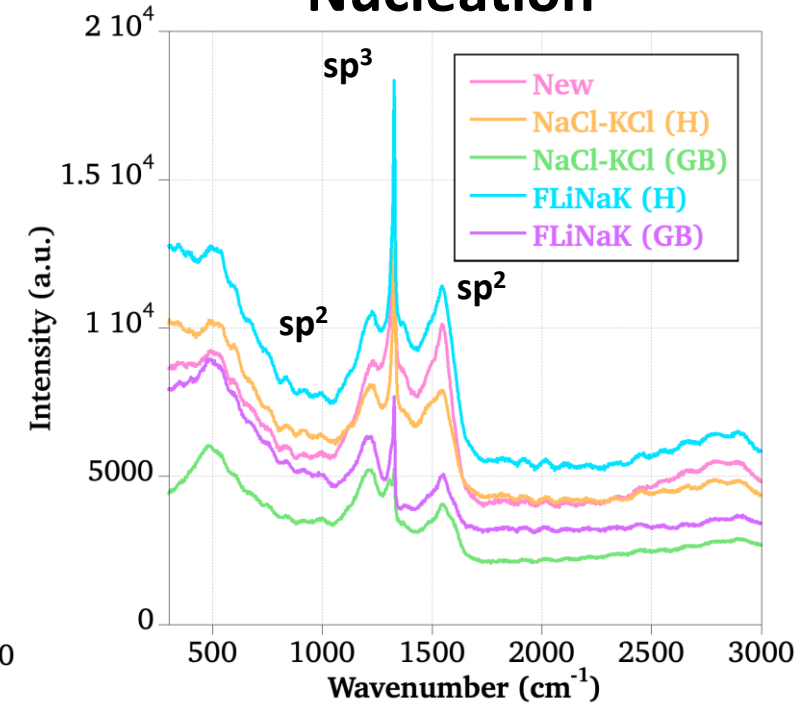
New BDD



Grown



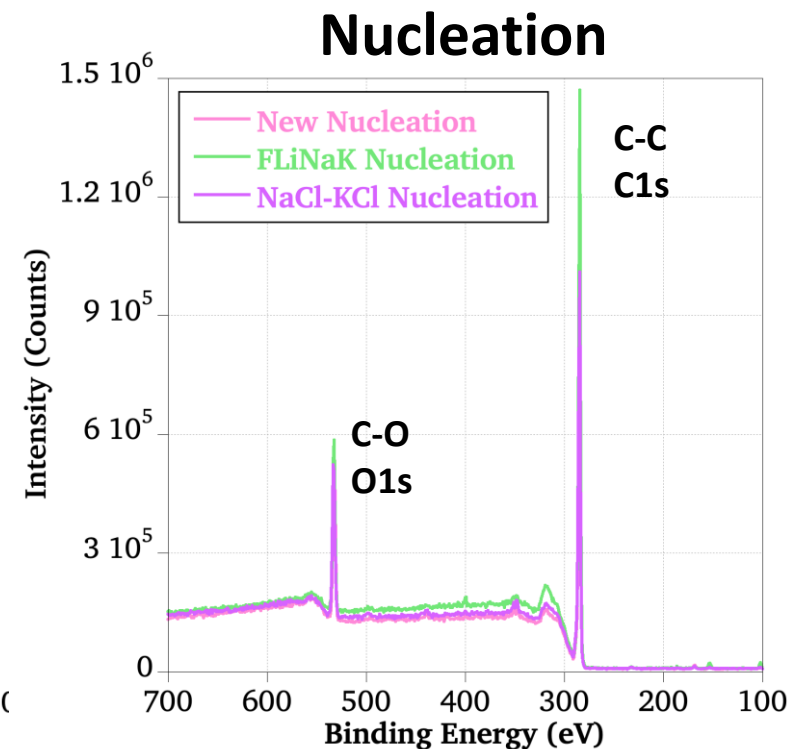
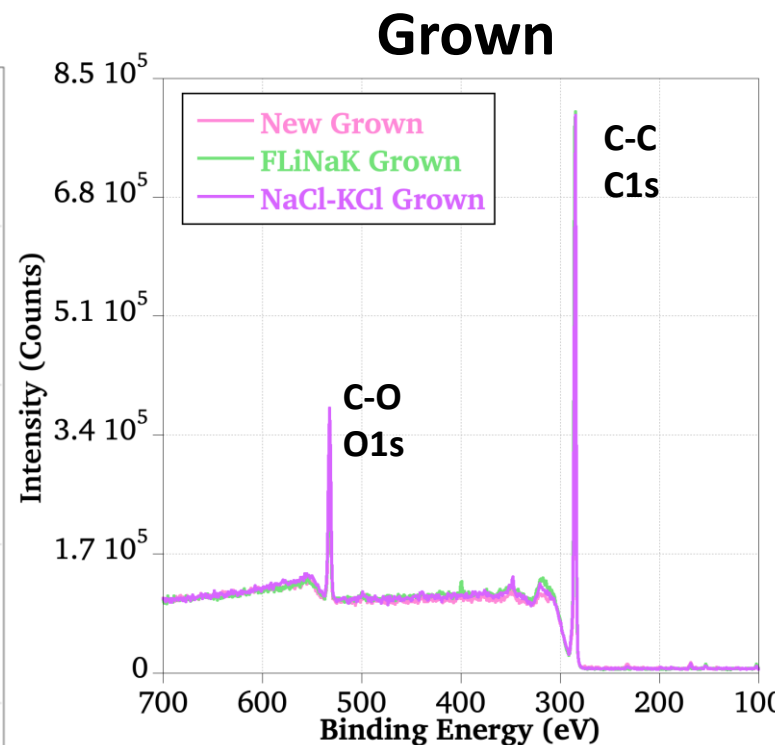
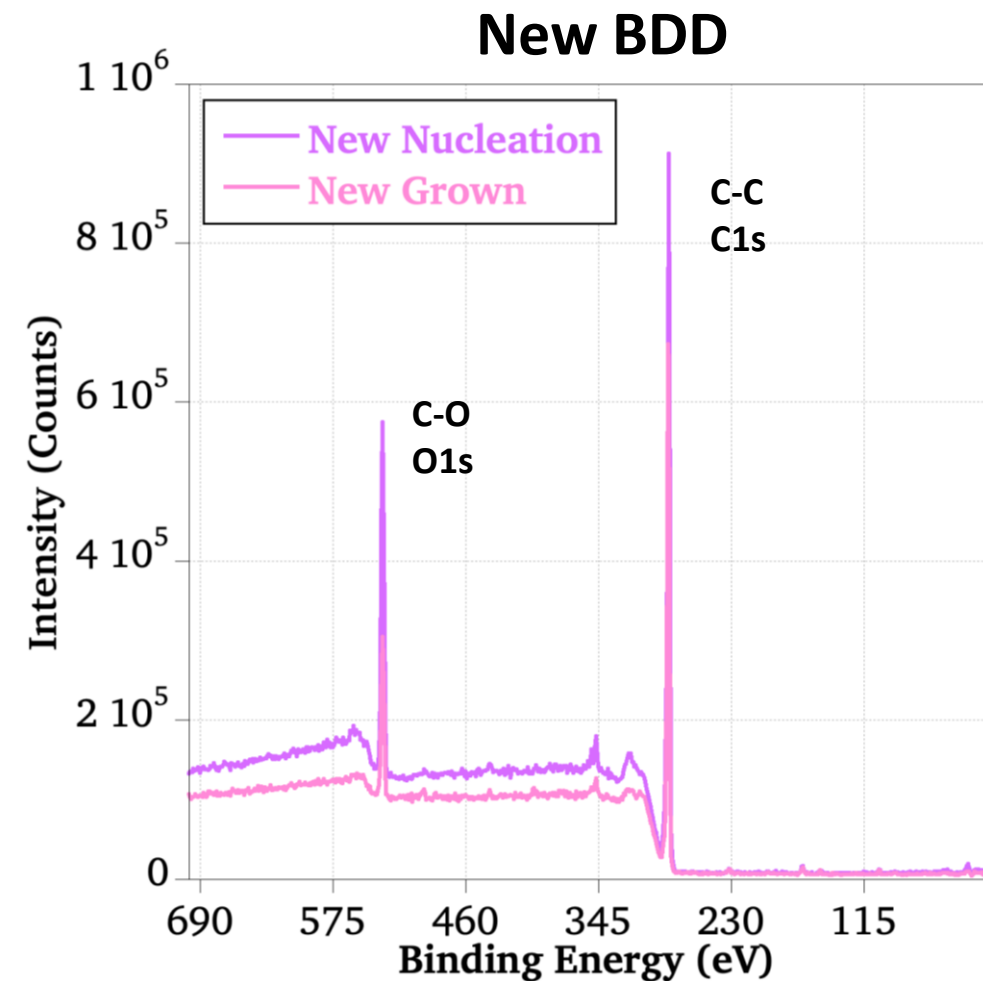
Nucleation



- ★ Only sp³ & sp² C
- ★ No evidence of F or Cl
- ★ No impact from inert vs. non-inert



X-ray Photoelectron Spectroscopy (XPS)



- ★ Only C1s and O1s
- ★ No evidence of F1s or Cl2p
- ★ Only measured inert exposure samples
- ★ No difference between grown and nucleation

Kuo et al., *ESL.*, 2(6), 1999.

Kondo et al., *Electrochim. Act.*, 52, 2007.

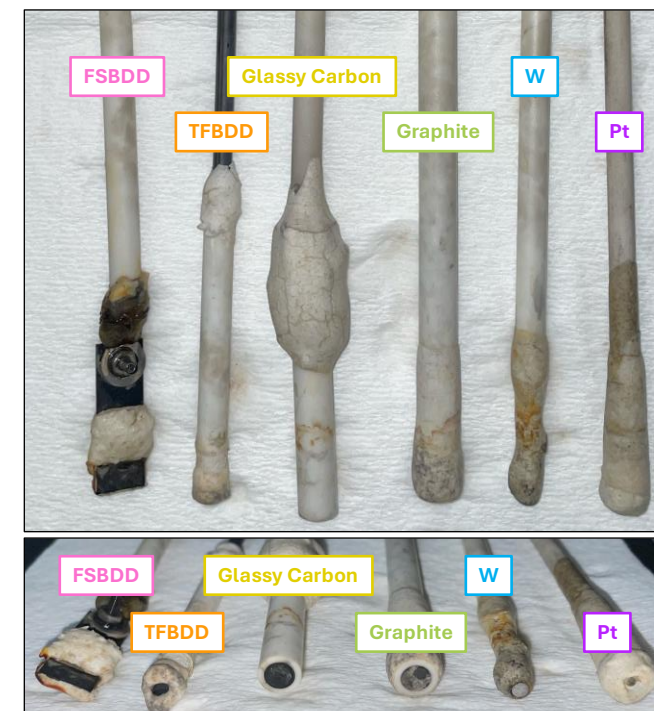
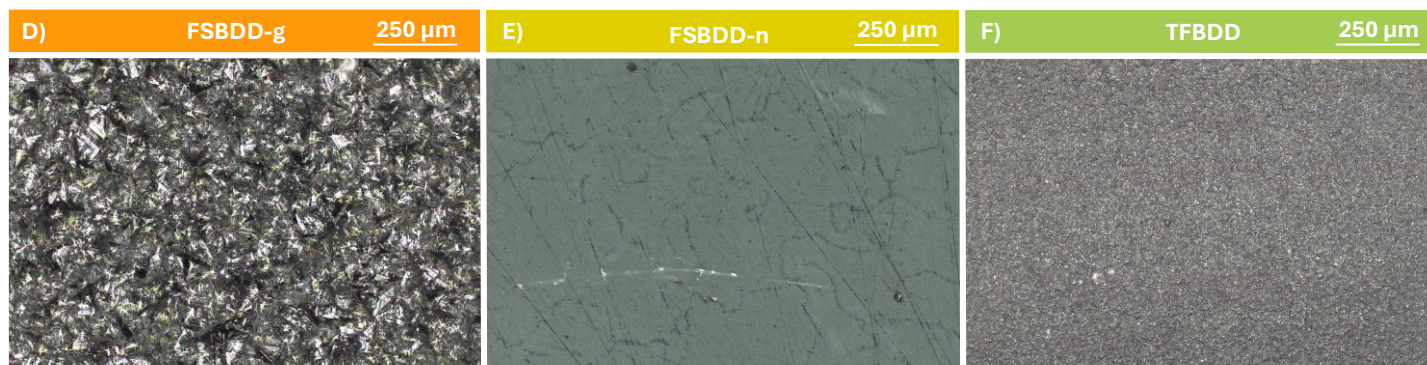
Goeting et al., *Diam. Relat. Mater.*, 9, 2000.



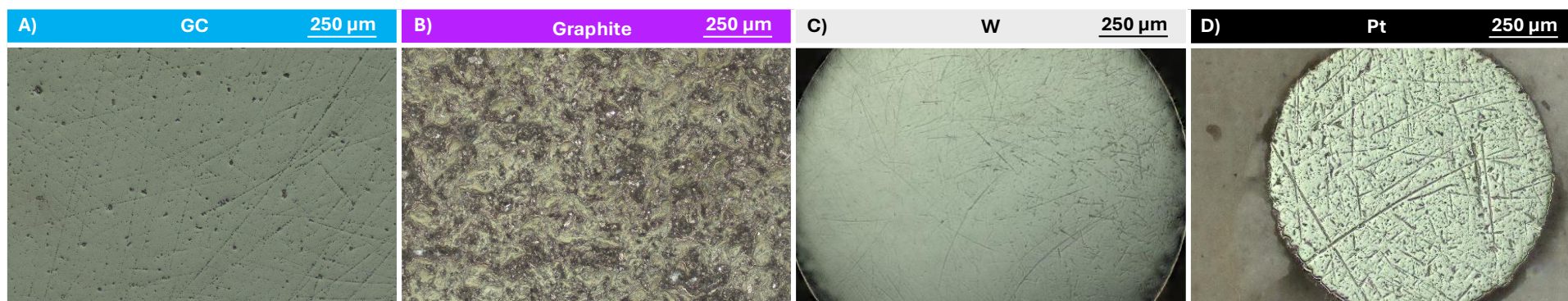
BDD Performance: Molten Salt Environments

Comparing a Series of Electrodes

Various BDD Morphologies

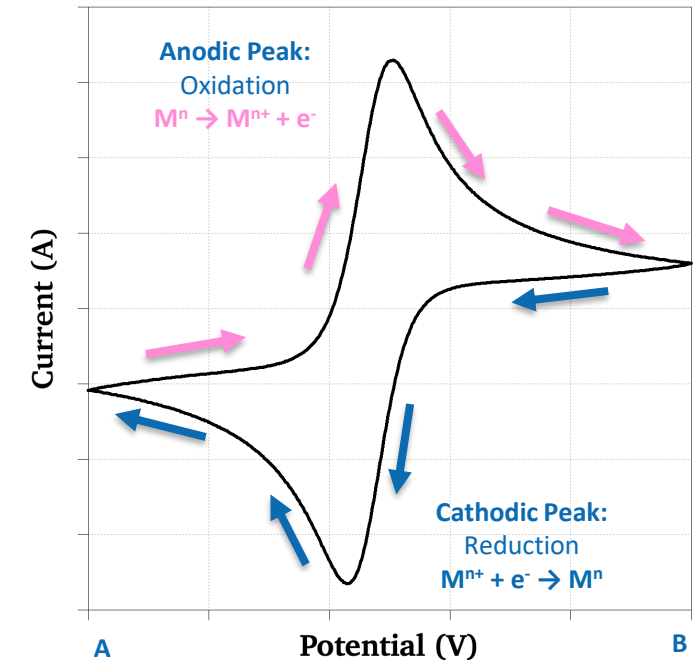
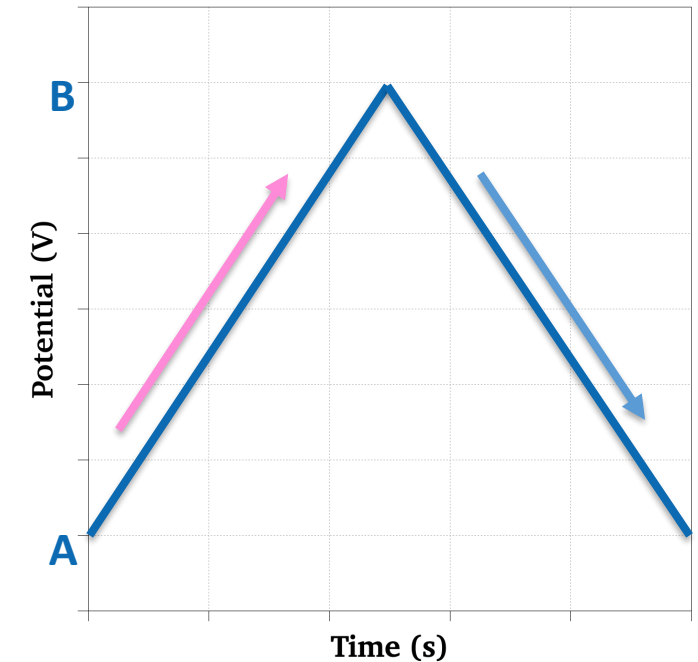


Common Molten Salt Electrodes



Experimental Design

- **Simple:**
 - Different Electrodes
 - ✓ Same Electrolyte
 - ✓ Same Analyte Concentration
 - ✓ Same Temperature
 - ✓ Same techniques
 - ✓ Same Scan Rates
 - ✓ All in One Run
- **Cyclic Voltammetry**
 - NOT ideal for reporting novel information about an analyte
 - Valuable for rapid, relative comparison within the same system



Electrode Comparisons

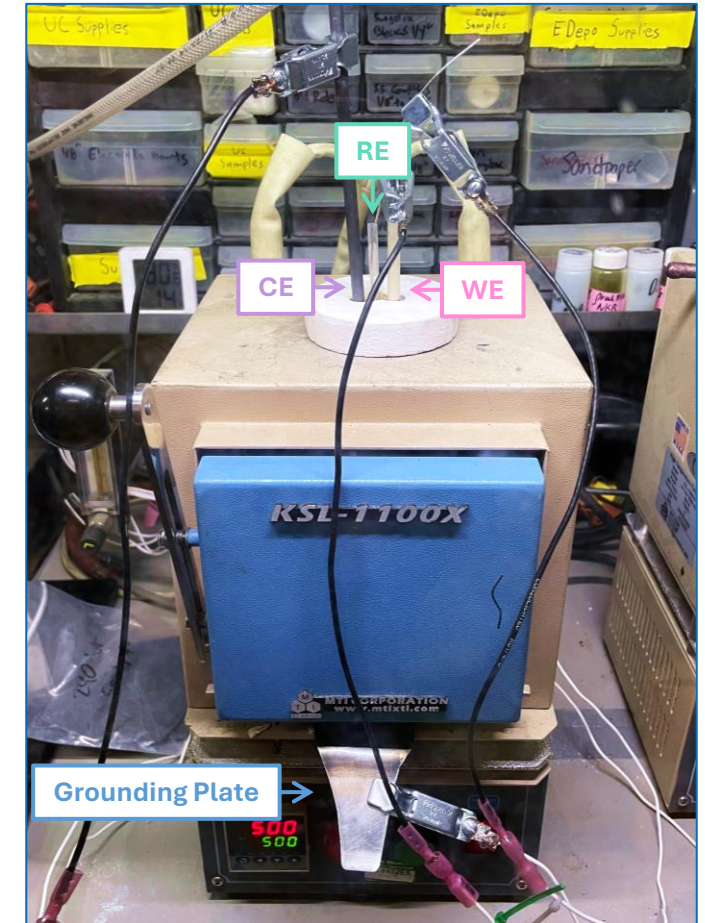
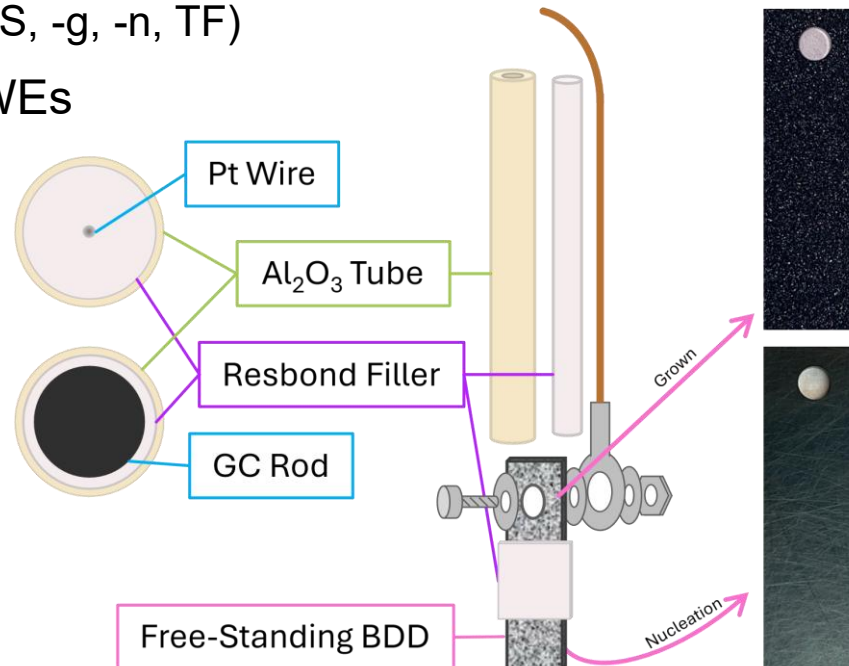
- **8 WE Materials:**

- Boron-Doped Diamond (BDD)
 - Four different morphologies (FS, -g, -n, TF)
- Four “common” molten salt WEs
 - Glassy Carbon (GC)
 - Graphite
 - Tungsten (W)
 - Platinum (Pt)

- **Surface Area**

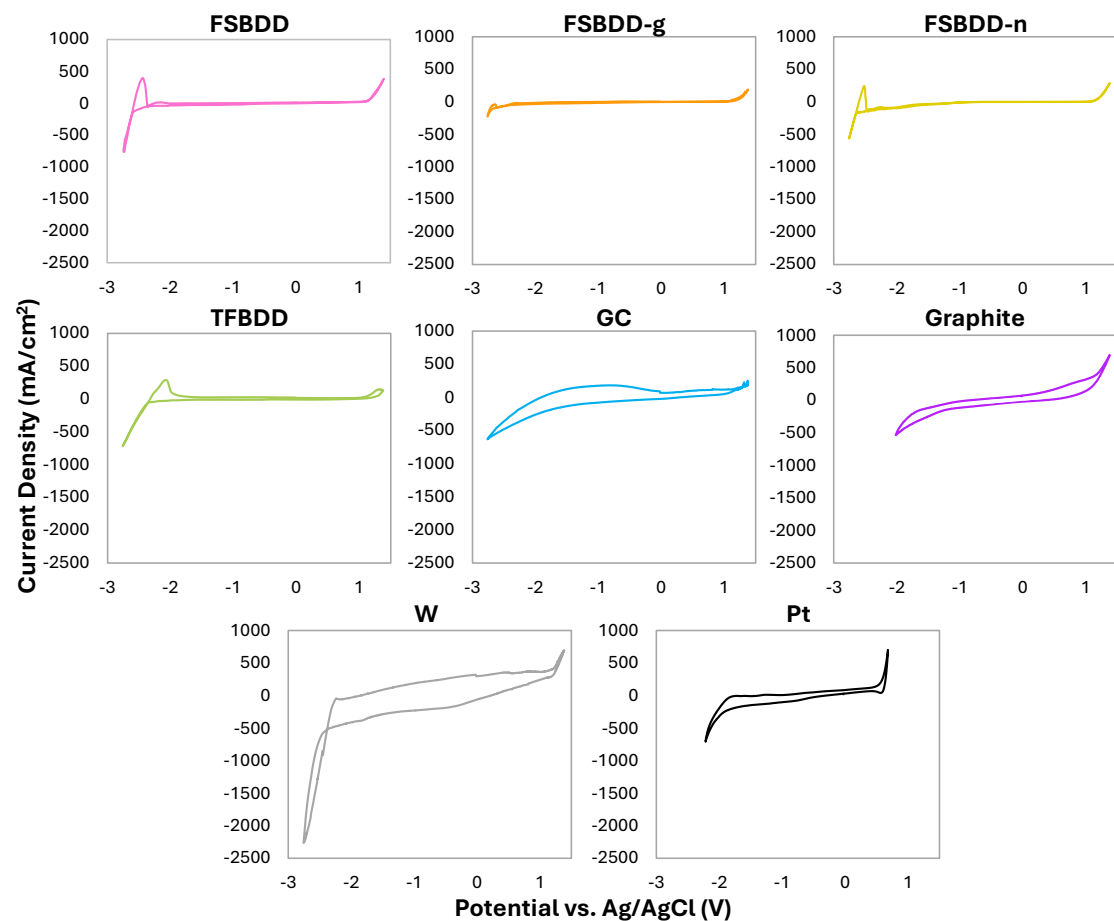
- Controlled
- Minimized
- Measured (via profilometry)

- **Ceramic Epoxy & Insulation**

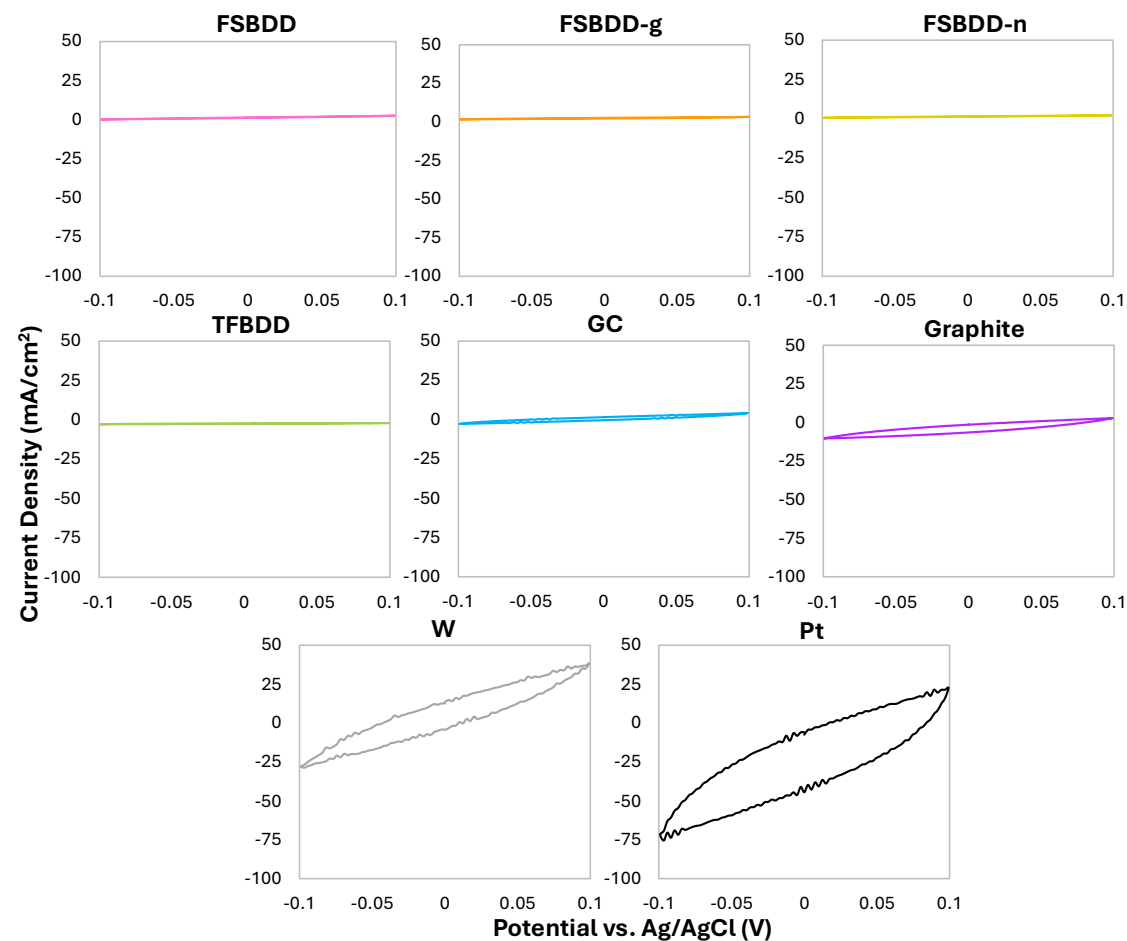


LiCl-KCl | 454 °C

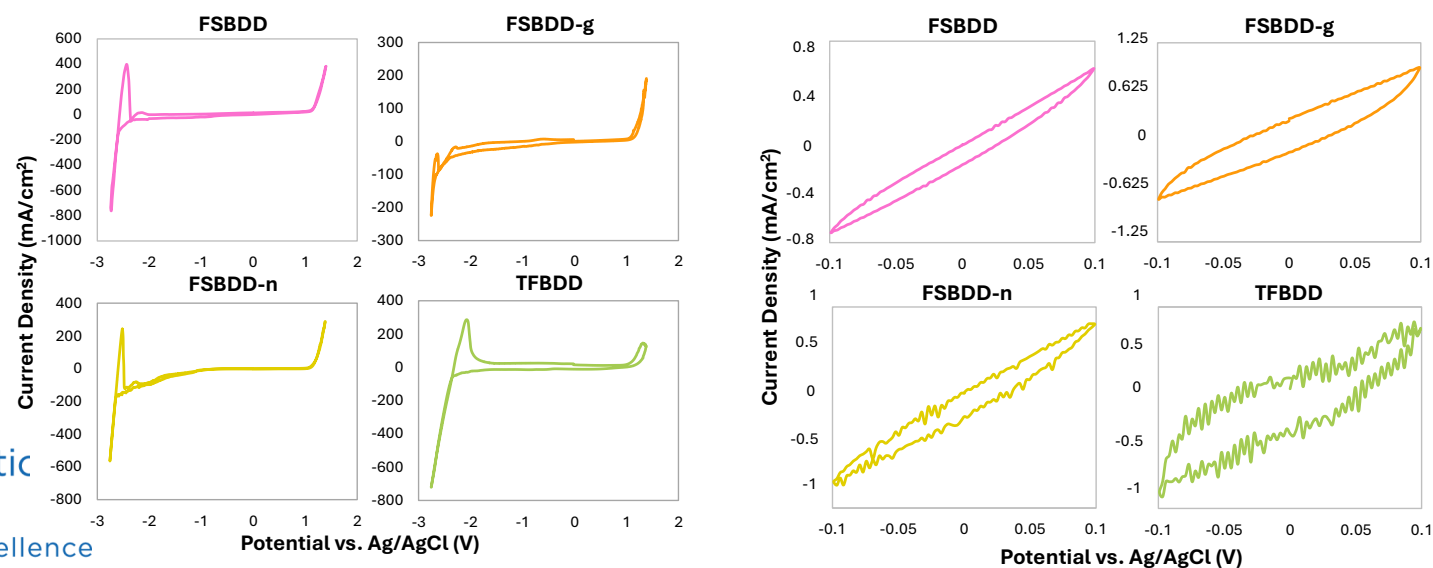
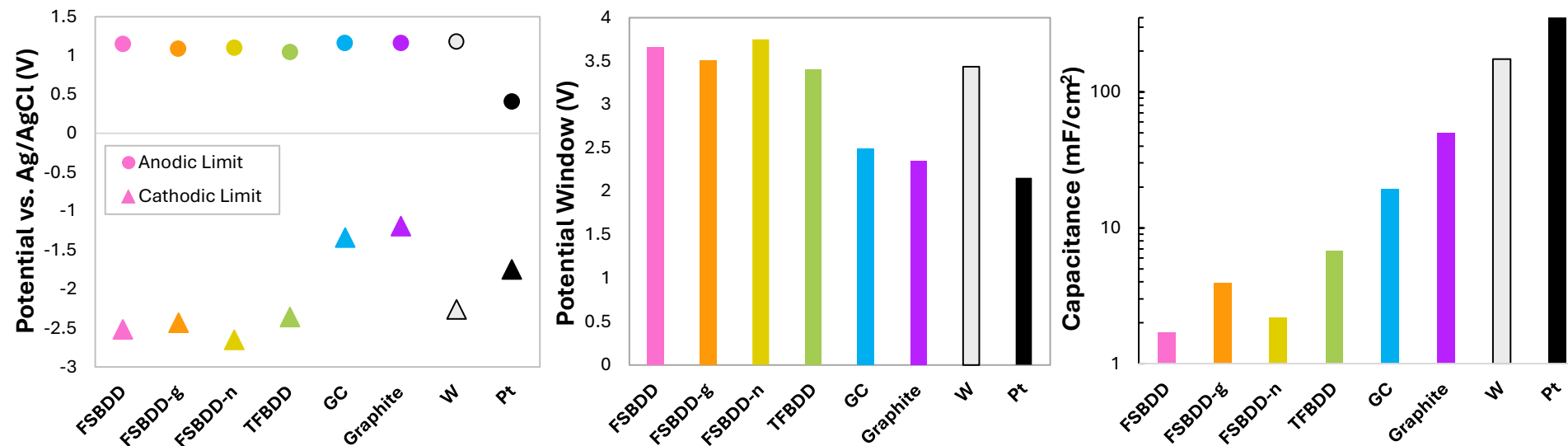
Potential (“Solvent”) Window:
Width of Usable Potential



Capacitance:
“Background” Current or Charge Stored at the Surface

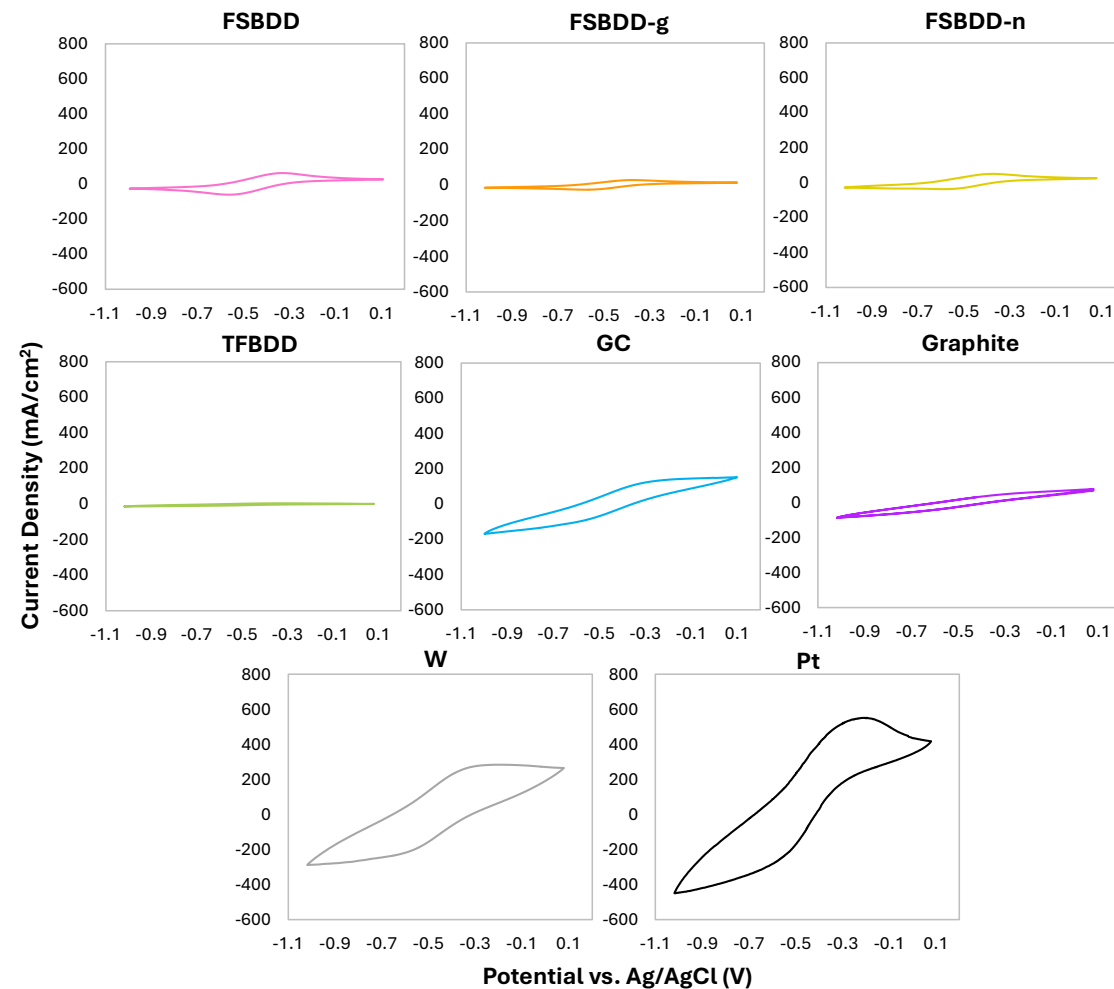
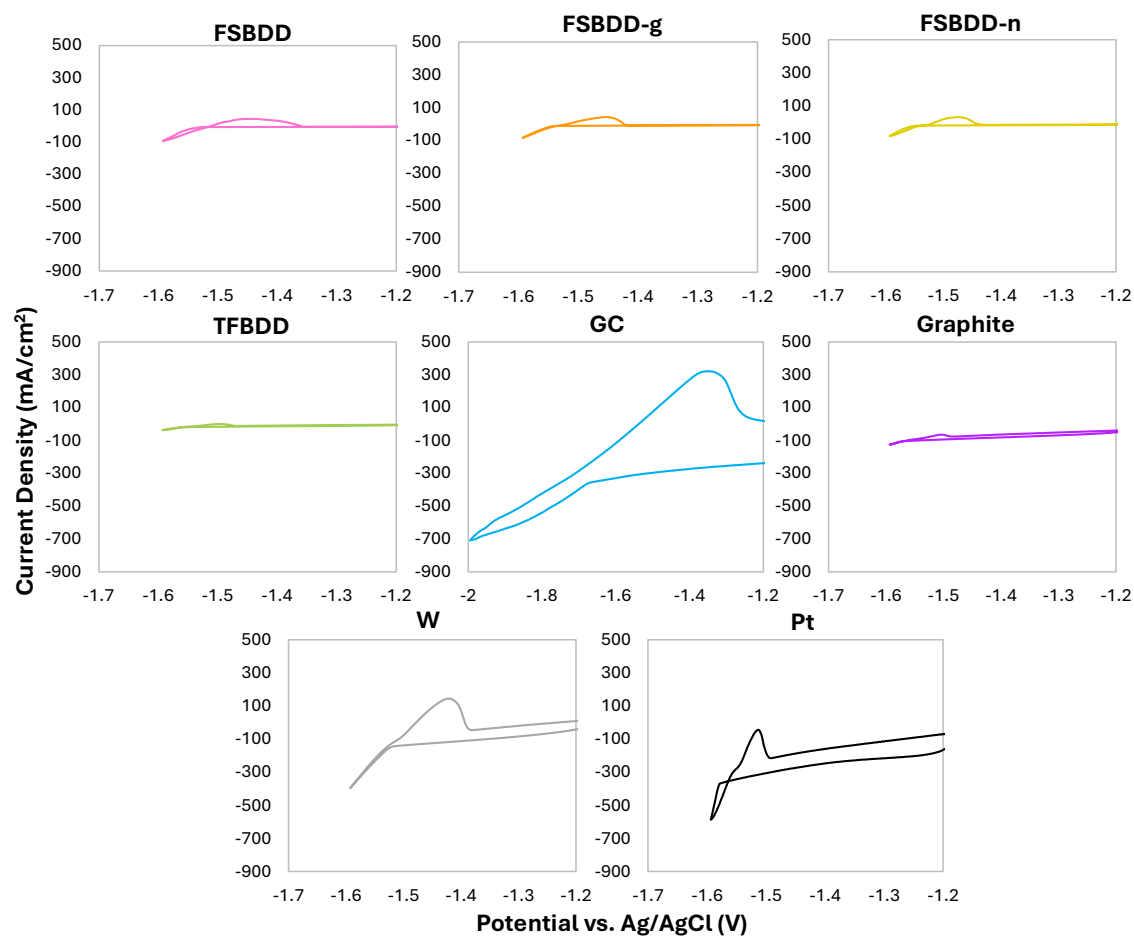


LiCl-KCl | 454 °C



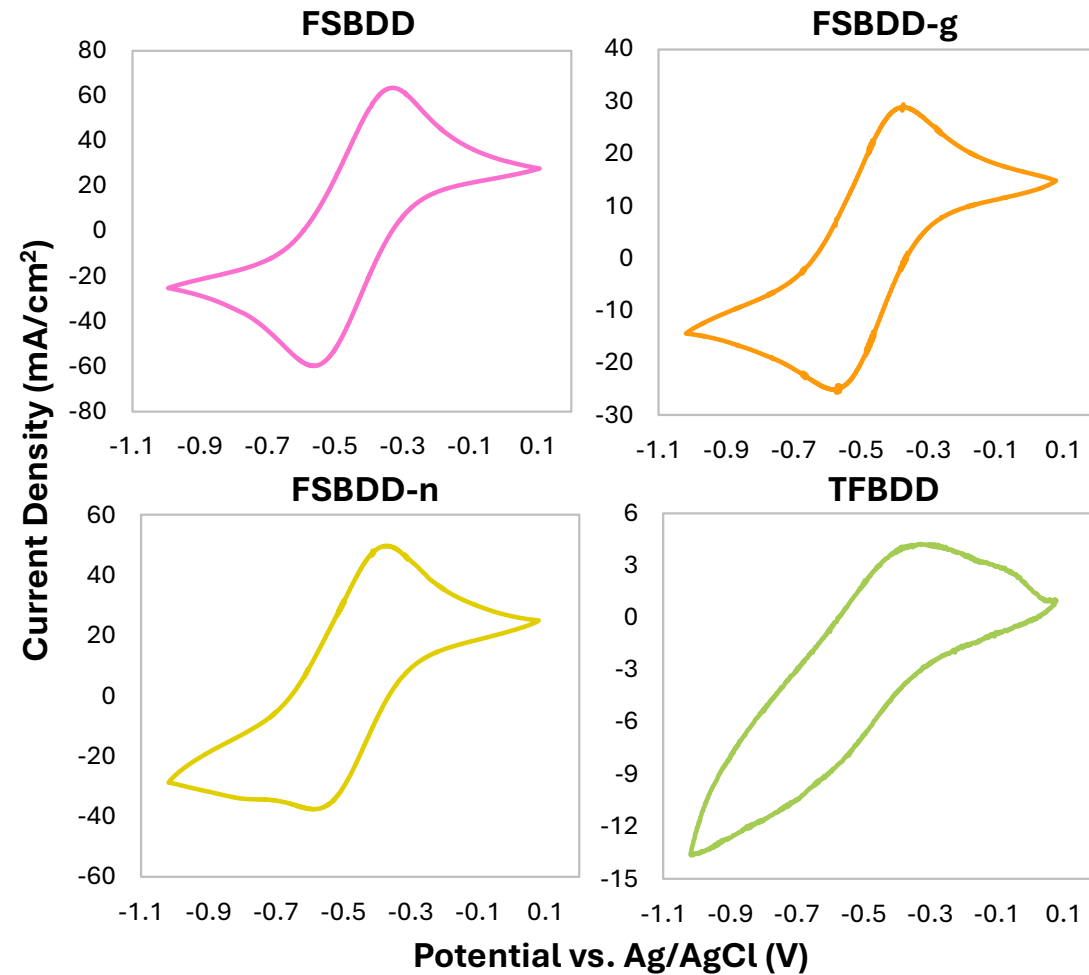
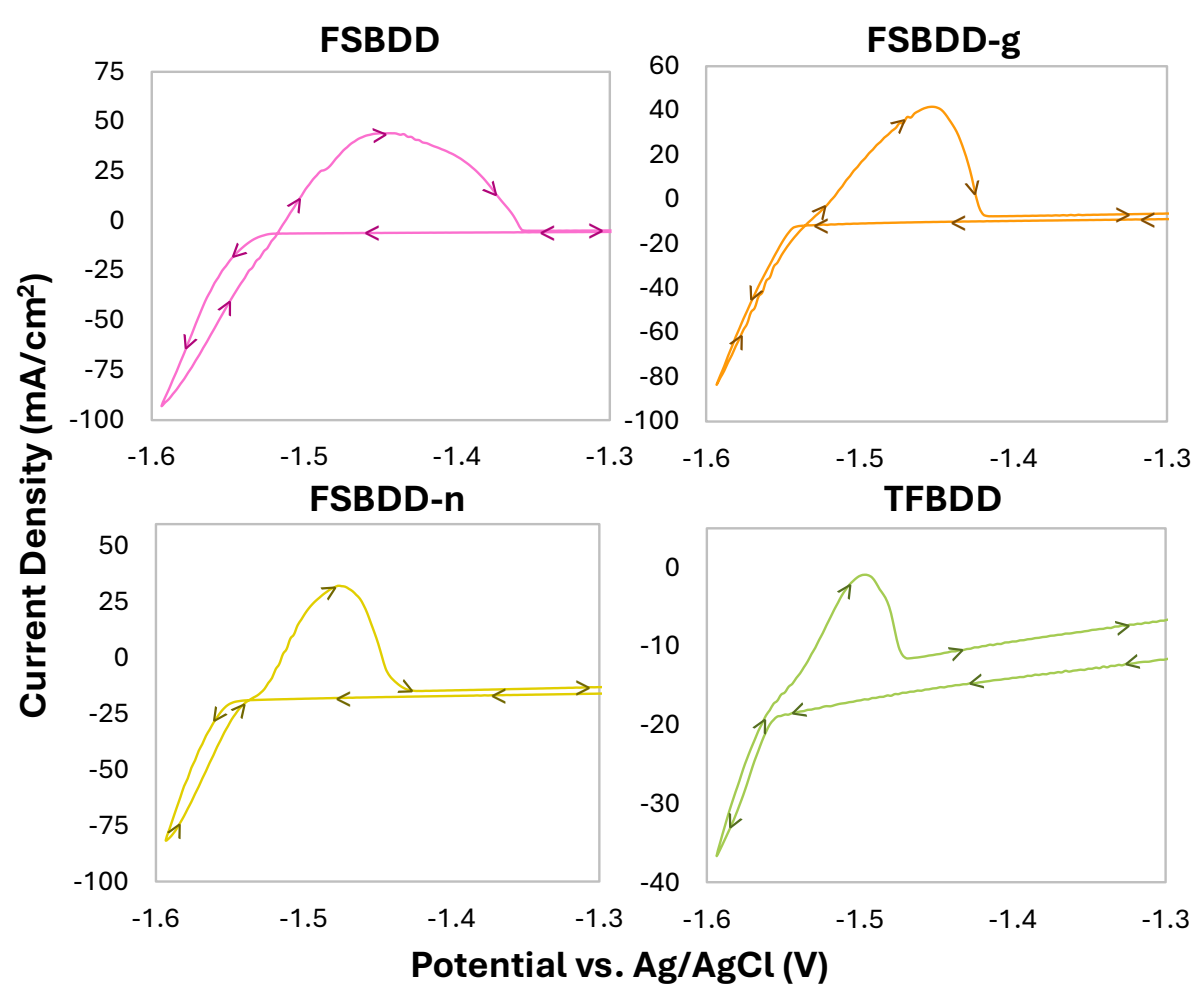
$U^{3+/0}$ & $U^{4+/3+}$:

1.47 mol% UCl_3 | $LiCl-KCl$ | 446 °C



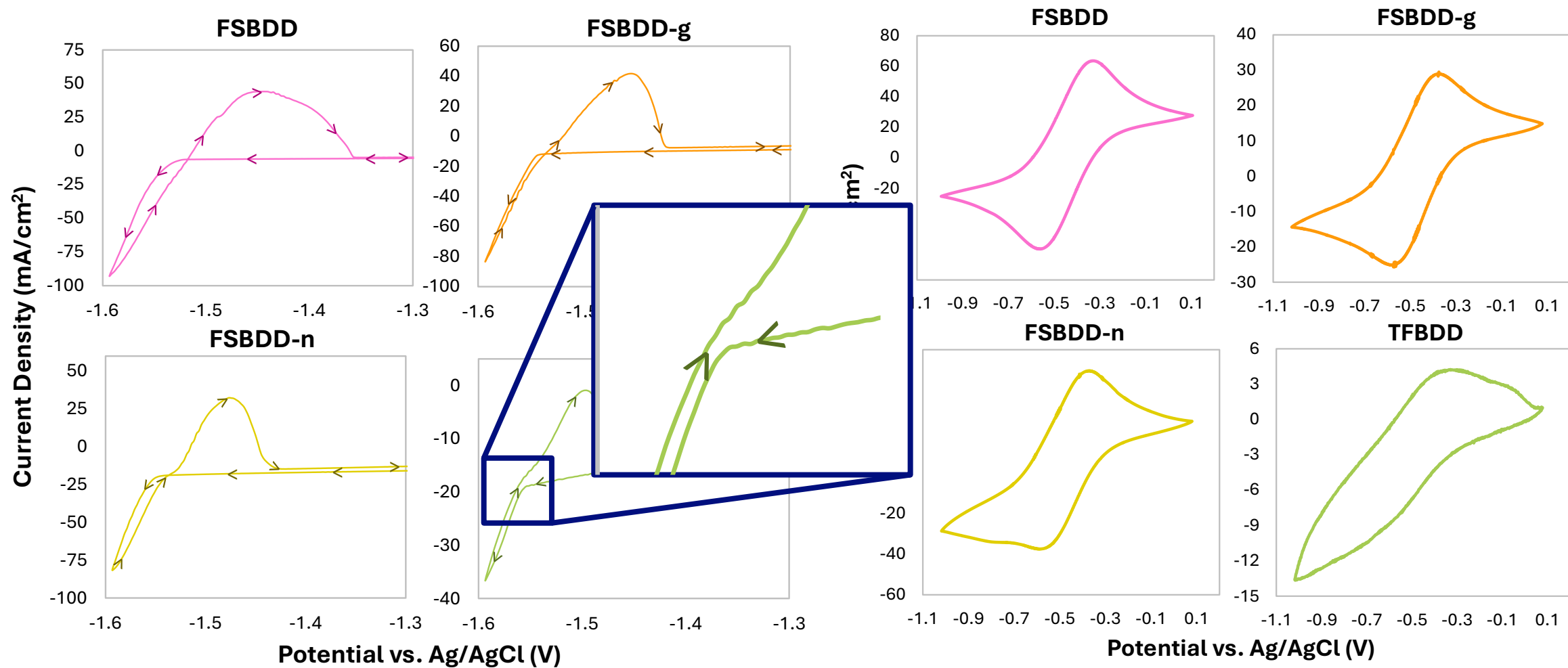
$U^{3+/0}$ & $U^{4+/3+}$:

$1.47 \text{ mol\% } UCl_3 \mid LiCl-KCl \mid 446 \text{ } ^\circ C$



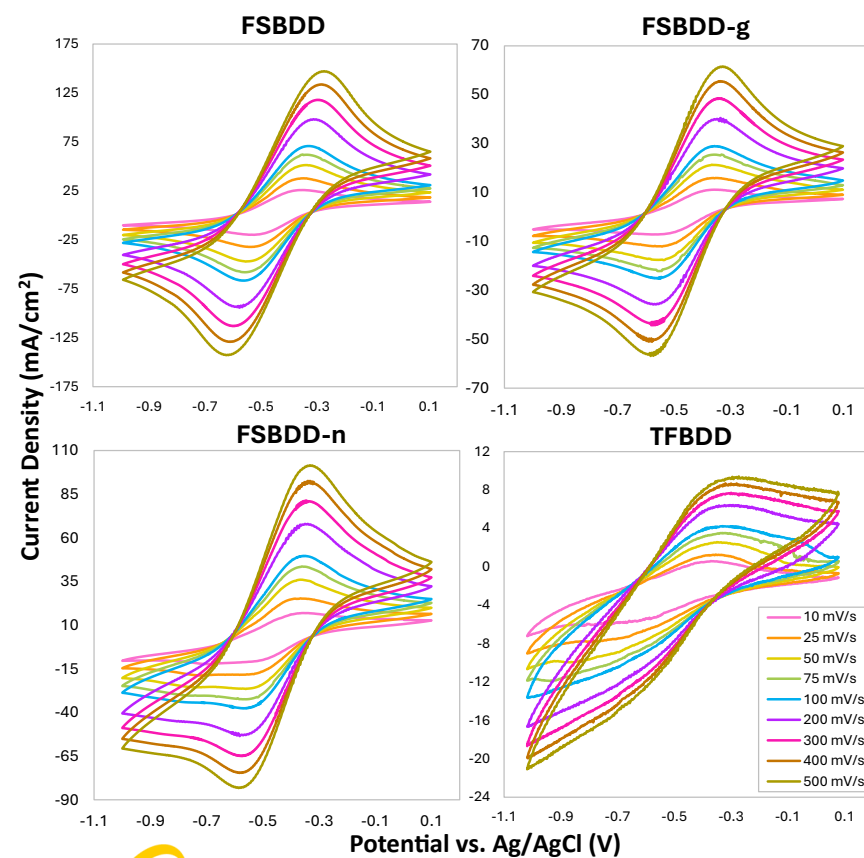
$U^{3+/0}$ & $U^{4+/3+}$:

$1.47 \text{ mol\% } UCl_3 \mid LiCl-KCl \mid 446 \text{ }^\circ\text{C}$



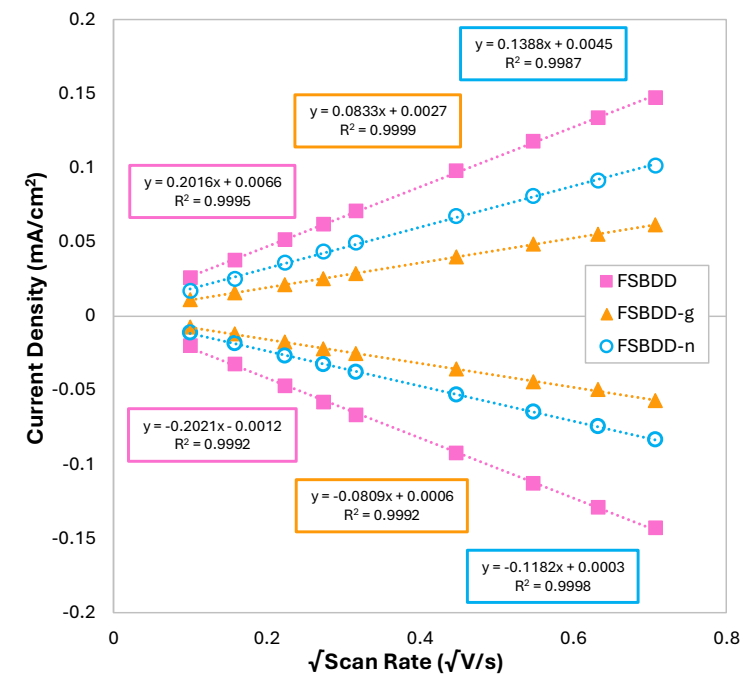
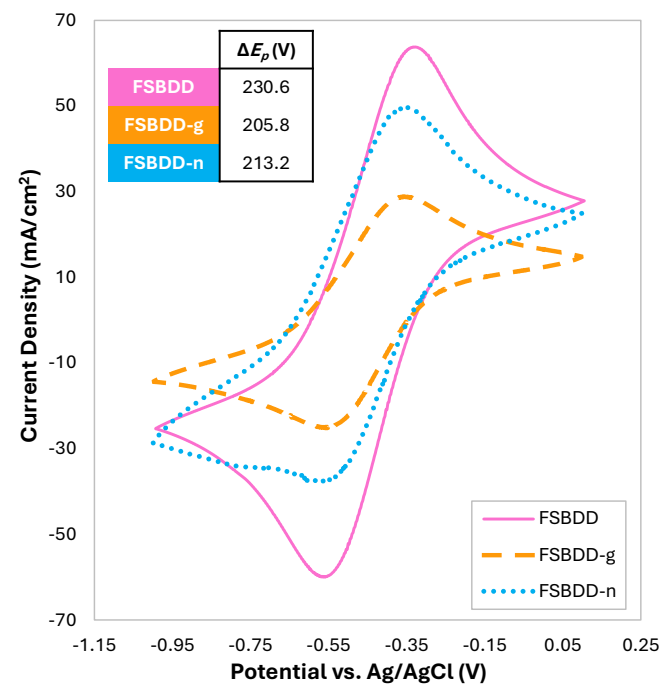
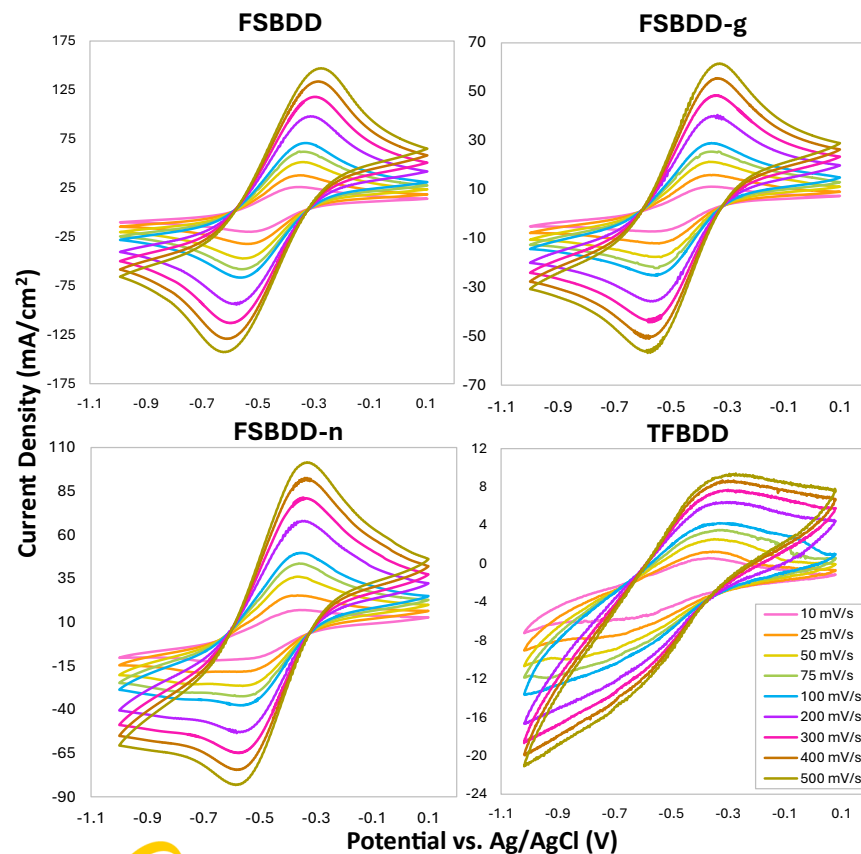
U^{3+/0} & U^{4+/3+}

1.47 mol% UCl₃ | LiCl-KCl | 446 °C



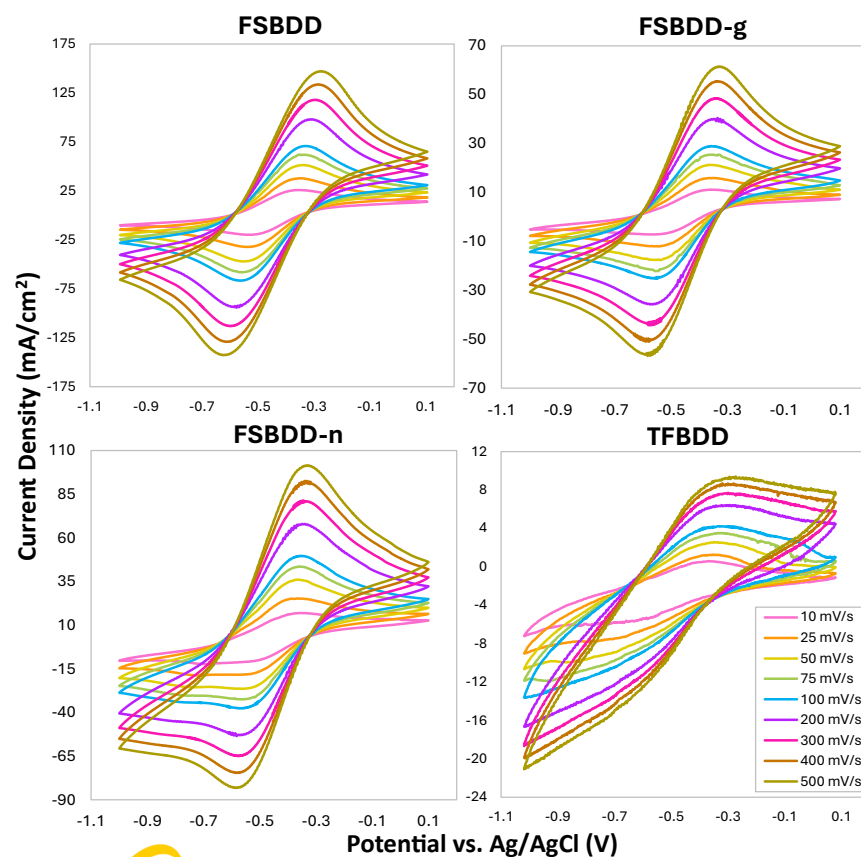
U^{3+/0} & U^{4+/3+}

1.47 mol% UCl₃ | LiCl-KCl | 446 °C

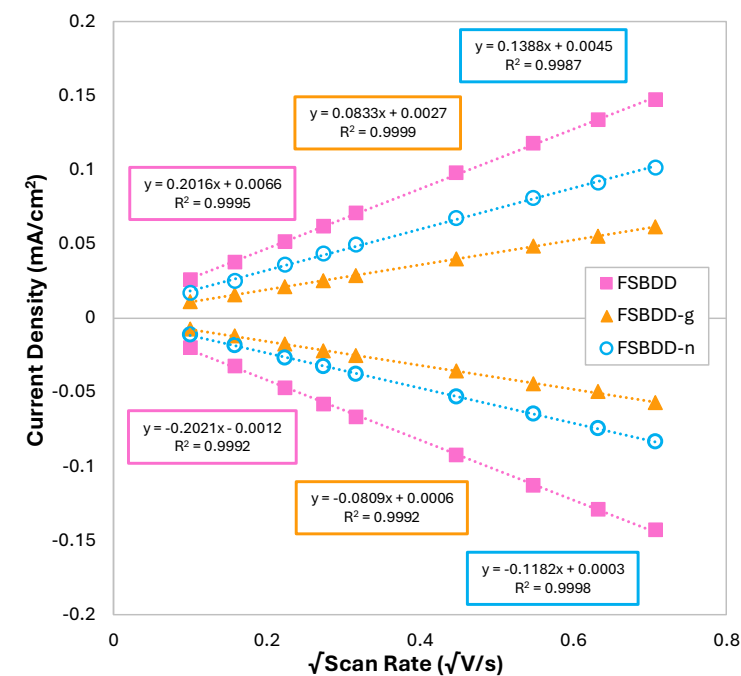
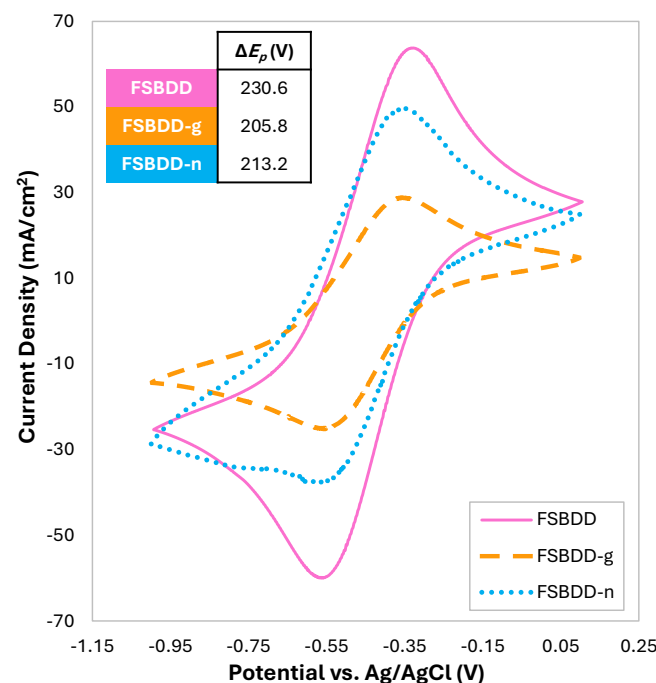


$U^{3+/0}$ & $U^{4+/3+}$

1.47 mol% UCl_3 | LiCl-KCl | 446 °C



- **Can't** solve for D due to high concentration
 - Among other assumptions...
- **Can** confirm semi-infinite linear diffusion
 - Linearity of regressions
- **Can** compare magnitude of current responses
 - Indication of WE-dependence on response
- **Can** describe “apparent” (relative) kinetics based on peak separations and peak shapes

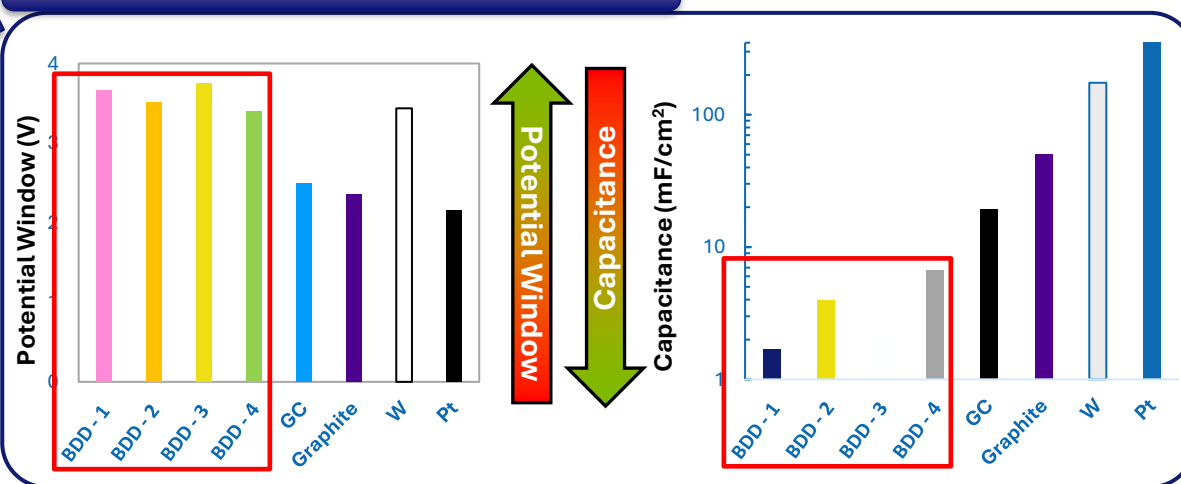


Summary of BDD Performance in Molten Salts

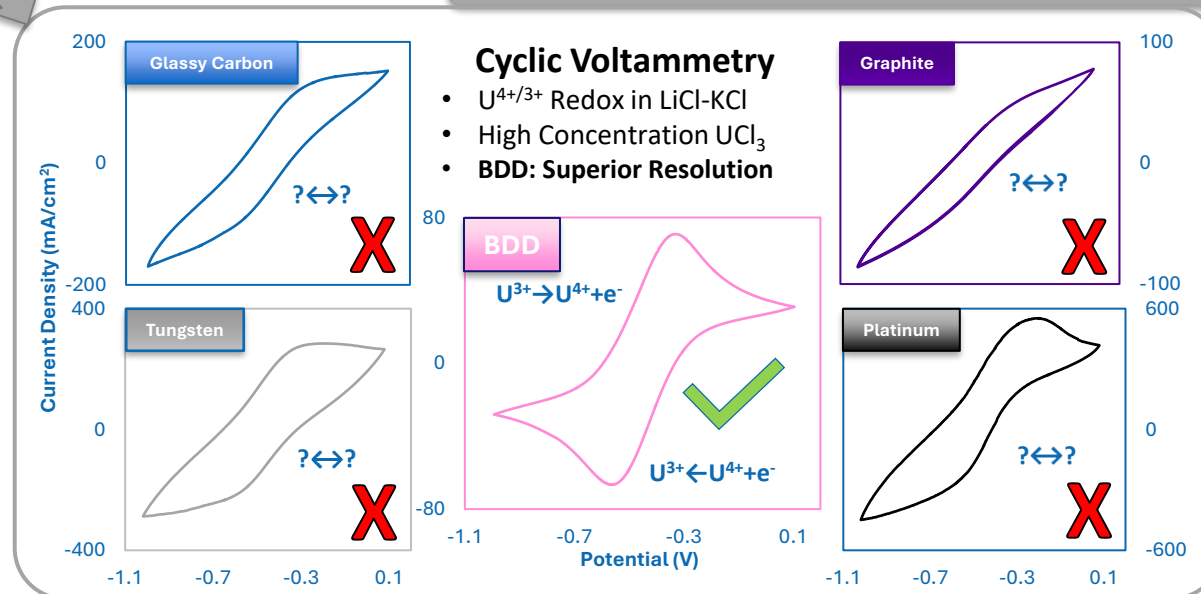
- ✓ Survived **exposure studies** in Cl and F molten salts
- ✓ Successful **redox measurements** in Cl and F molten salts
- ✓ **Out-performs** standard molten salt electrodes

- Wider working windows
- Lower background
- Enabled novel investigations of 'fuel-like' chloride salts

Inherent Improvements to Sensitivity



Exceeds Capabilities of Standard Materials



Molten Salt BDD Electrochemical Sensor Development Stage:

TRL 2 (fluorides) & 3 (chlorides)

Conclusions

- **WE selection** measurably affects electrochemical response
- **Novel materials** (e.g., BDD) may offer significant advantages
- **Electrode fabrication** deserves greater attention in molten salt electrochemistry

Conclusions

- **WE selection** measurably affects electrochemical response
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- **Electrode fabrication** deserves greater attention in molten salt electrochemistry

Future Work

- Expansion to measurements of **fluorides**
 - (e.g., FLiNaK, FLiBe)
- Exploration of **Pu redox chemistry**
 - New materials may unlock new capabilities
- **Application:** Online monitoring
 - MSR applications for electroanalysis
 - Novel sensor development

Acknowledgements



Franz Freibert
Mentor



Marisa Monreal
Mentor



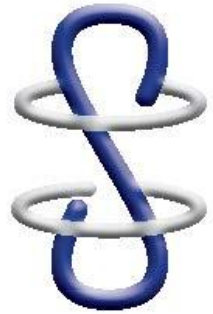
Scott Parker
Mentor



Jarom Chamberlain
Partner in ~~Crime~~ Molten Salt Chemistry



Clare Hatfield
Lab Queen & Electrochemistry Genius



THE GLENN T.
SEABORG
INSTITUTE



*Diamonds are a Girl's
Best Friend*

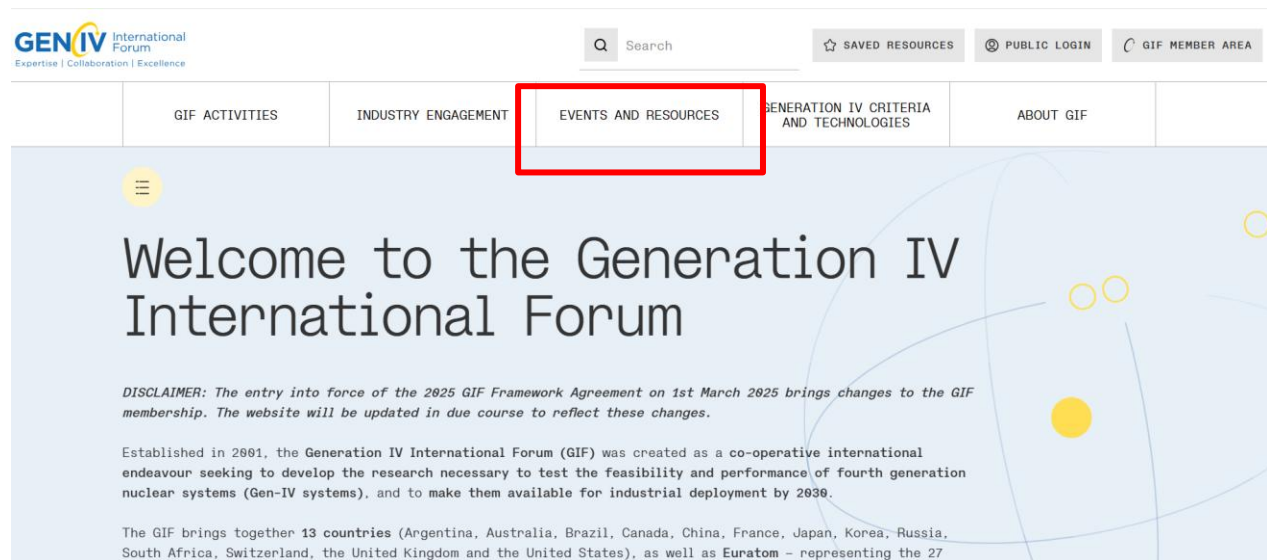
^
Reactor's



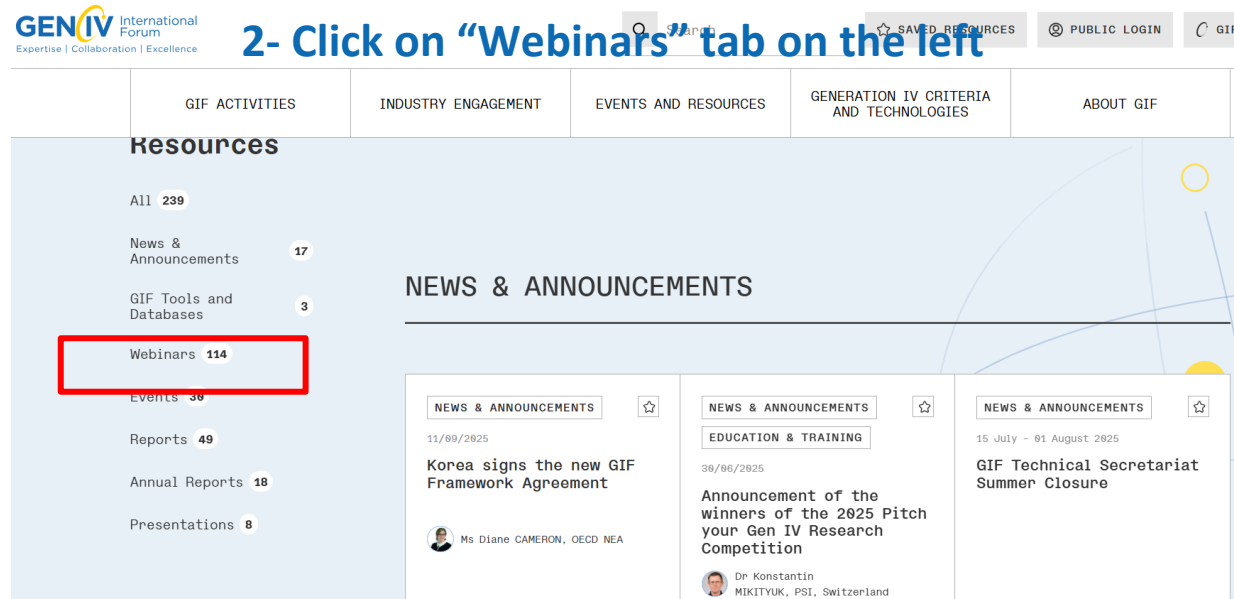
Upcoming Webinars

Date	Title	Presenter
10 September 2026	Thermal properties of molten salts: crucial data for the development of MSR	Prof. Juliano Schorne-Pinto, University of South Carolina, USA
15 October 2026	Temperature-based approach to estimate decay heat in sodium-cooled fast reactors and validate associated computational tools	Dr. Dorian Da Cunha, CEA, France
12 November 2026	MELCOR: The evolution of nuclear safety analysis technology to enable nuclear innovation	Dr. Matthew Christian and Dr. David Luxat, Sandia National Laboratories, USA

1- Click on “Events and Resources Tab



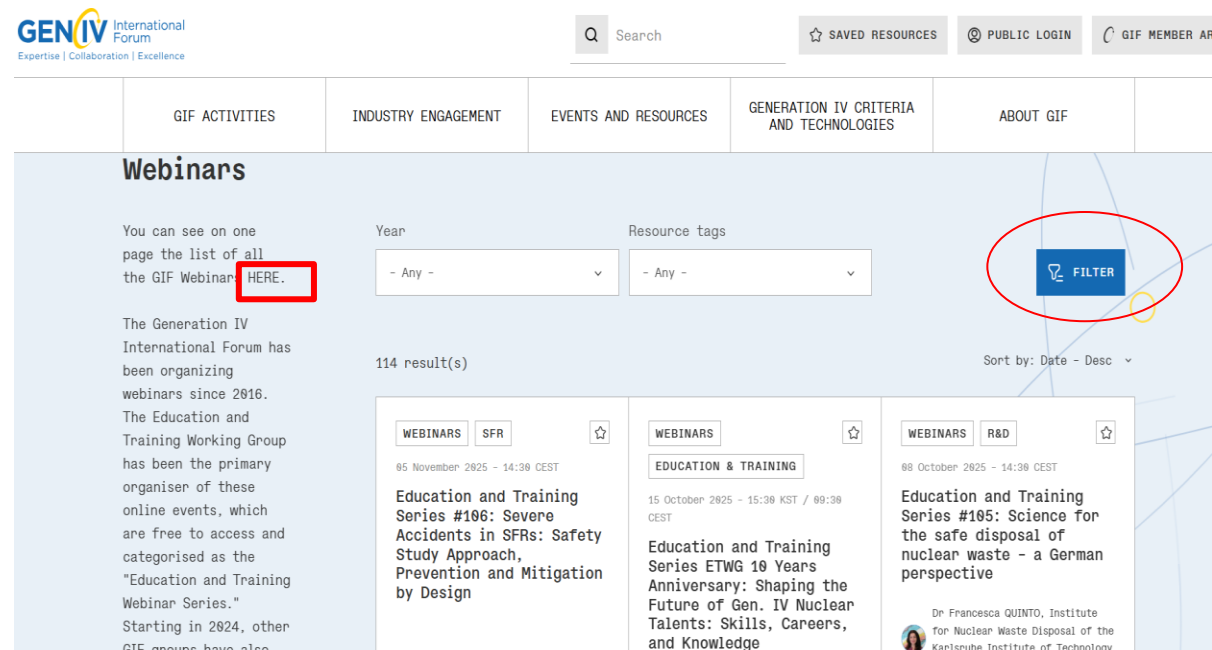
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