Feasibility study of the preparation of uranium layers on metallic backings

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Goedele Sibbens², André Moens², Roger Eykens²

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Outline

• Introduction
• My participation in the ERINDA project
• Experimental work
  – Natural uranium
  – Deposition on aluminium and stainless steel backings
  – Deposition from inorganic and organic media
  – Investigation of the deposited material
• Results
• Conclusion
Introduction

• The Target Preparation laboratory at IRMM produces and characterizes targets for measurements related to:
  • Basic research in nuclear physics and nuclear data standards
  • Nuclear data for radioactive waste management and safety of new reactor developments

• Target characteristics are an integral and important part of the measurements

→ continuation and improvement in target preparation and characterization require knowledge transfer and research!
My participation in the ERINDA project

• Visiting scientist at EC-JRC-IRMM
  – 8 weeks:
    • 2 weeks in March 2012
    • 6 weeks in November and December 2012

• Work programme
  – Preparation of actinide targets
    • Developing a procedure to prepare actinide layers up to mg/cm²
    • Investigation on the influence of the roughness of the backing on the quality of the deposited actinide layer
    • Feasibility on the preparation of actinide layers on thin polyimide foils
    • Joint paper
Experimental work

• Preparation of targets:
  – Uranium with natural isotopic composition

• Backings material:
  – aluminum: Ø = 25 mm, d = 0.4 mm
  – stainless steel: Ø = 25 mm, d = 0.4 mm

• Electrolyte solutions:
  – Inorganic: 5.7 % \((\text{NH}_4)_2\text{C}_2\text{O}_4\) in 0.3M HCl
  – Organic: 2-propanol, 2-butanol, DMF
Experimental work

- Determining the amount of the deposited uranium on the backings
  - Gamma-ray spectrometry: U-235 at 187.5 keV
  - Low-geometry alpha-particle counting

- Measuring the homogeneity of the deposited layer
  - Autoradiography

- Testing the deposited material on the backings
  - Energy dispersive X-ray fluorescence spectrometry
Experimental work

• JSI electrodeposition cell
  – 10 mL of electrolyte
  – spirally wounded Pt anode
  – distance: 8 – 10 mm
Experimental work

- IRMM molecular plating cell
  - 10 mL of electrolyte
  - rectangular rotating Pt anode
Uranium material

• IRMM

ISOTOPIC COMPOSITION
LOT 61B UO2 Powder

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Atom %</th>
<th>Mass %</th>
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<tbody>
<tr>
<td>$^{234}\text{U}$</td>
<td>0.00506</td>
<td>0.00498</td>
</tr>
<tr>
<td>$^{235}\text{U}$</td>
<td>0.72243</td>
<td>0.71337</td>
</tr>
<tr>
<td>$^{236}\text{U}$</td>
<td>&lt;0.0002</td>
<td>&lt;0.0002</td>
</tr>
<tr>
<td>$^{238}\text{U}$</td>
<td>99.2725</td>
<td>99.2817</td>
</tr>
<tr>
<td>Total</td>
<td>100</td>
<td>100</td>
</tr>
</tbody>
</table>

c = 386.1 mgU/mL

• JSI

– Ammonium diuranate (ADU)
$(\text{NH}_4)_2\text{U}_2\text{O}_7$

c = 82.3 mgU/mL
Electrodeposition

• Procedure 1:
  – 5.7% (NH₄)₂C₂O₄ in 0.3M HCl: 10 mL
  – Uranium solution: depends on required thickness
  – Time of electrodeposition: 2h
  – Current: 0.6 A
  – 1 min before stop add 1 mL of NH₄OH
  – Wash the electrodeposition cell:
    • 10 mL of distilled water which contains 5 drops of NH₄OH
    • 10 mL of ethanol which contains 5 drops of NH₄OH
  – Dry the disc with electroplated material
Molecular plating

• Procedure 2:

– 2-propanol, 2-butanol: 10 mL
– Uranium solution: depends on required thickness
– Time of molecular plating: 2h
– Voltage: 300 V
– Current: 1 – 3 mA/cm²
– Empty the molecular plating cell
– Dry the disc with deposited material
– Heat the disc with deposit at 100°C
## Results

An overview of prepared samples at IRMM and JSI

<table>
<thead>
<tr>
<th>Backing</th>
<th>No of prepared samples</th>
<th>Electrolyte</th>
<th>Areal density of deposited U nat ($\mu g/cm^2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aluminium</td>
<td>5</td>
<td>Oxalate</td>
<td>20 – 650</td>
</tr>
<tr>
<td>Stainless steel</td>
<td>25</td>
<td>Oxalate</td>
<td>50 – 1600</td>
</tr>
<tr>
<td>Aluminium</td>
<td>7</td>
<td>Iso-propanol</td>
<td>50 – 650</td>
</tr>
<tr>
<td>Aluminium</td>
<td>2</td>
<td>Iso-butanol</td>
<td>150, 500</td>
</tr>
<tr>
<td>Aluminium</td>
<td>1</td>
<td>Dimethyl formaldehyde</td>
<td>~ 500</td>
</tr>
<tr>
<td>Stainless steel</td>
<td>2</td>
<td>Iso-propanol</td>
<td>~ 1000</td>
</tr>
</tbody>
</table>

**Source area: 3.5 cm²**
Results

- Backing: Al
- Electrolyte: 5.7 % \((\text{NH}_4)_2\text{C}_2\text{O}_4\) in 0.3M HCl

blank ~ 400 µgU/cm² ~ 600 µgU/cm²
Results

- Backing: Al
- Electrolyte: Iso-propanol

blank ~ 150 µgU/cm²

~ 500 µgU/cm²
Results

- Backing: Stainless steel
- Electrolyte: 5.7 % \((\text{NH}_4)_2\text{C}_2\text{O}_4\) in 0.3M HCl

\(~ 150 \ \mu\text{gU/cm}^2~\)  \(~ 300 \ \mu\text{gU/cm}^2~\)
• Backing: Stainless steel
• Electrolyte: 5.7% (NH₄)₂C₂O₄ in 0.3M HCl

~ 1000 µgU/cm²

~ 1600 µgU/cm²
Results

- Backing: Stainless steel
- Electrolyte: Iso-propanol

~ 1000 µgU/cm² ~ 1000 µgU/cm²
U-235

http://www.nucleide.org/DD

6.2 Gamma Emissions

<table>
<thead>
<tr>
<th>Energy</th>
<th>Photons per 100 disint.</th>
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</thead>
<tbody>
<tr>
<td>31.60(5)</td>
<td>0.017 (6)</td>
</tr>
<tr>
<td>41.4(3)</td>
<td>0.020 (11)</td>
</tr>
<tr>
<td>42.01(6)</td>
<td>0.056 (9)</td>
</tr>
<tr>
<td>51.21(4)</td>
<td>0.034 (7)</td>
</tr>
<tr>
<td>54.1(1)</td>
<td>0.000115</td>
</tr>
<tr>
<td>54.2(5)</td>
<td>0.0285</td>
</tr>
<tr>
<td>64.45(2)</td>
<td>0.018</td>
</tr>
<tr>
<td>72.7(2)</td>
<td>0.116</td>
</tr>
<tr>
<td>74.9(3)</td>
<td>0.051 (6)</td>
</tr>
<tr>
<td>96.09(2)</td>
<td>0.091 (11)</td>
</tr>
<tr>
<td>97(4)</td>
<td>0.016 (4)</td>
</tr>
<tr>
<td>109.1(7)</td>
<td>1.66 (13)</td>
</tr>
<tr>
<td>115.45(5)</td>
<td>0.03 (1)</td>
</tr>
<tr>
<td>129.35(5)</td>
<td>0.026</td>
</tr>
<tr>
<td>136.55(5)</td>
<td>0.012</td>
</tr>
<tr>
<td>140.76(2)</td>
<td>0.20 (1)</td>
</tr>
<tr>
<td>142.40(5)</td>
<td>0.0051</td>
</tr>
<tr>
<td>143.76(3)</td>
<td>10.94 (6)</td>
</tr>
<tr>
<td>150.936(15)</td>
<td>0.09 (3)</td>
</tr>
<tr>
<td>163.356(3)</td>
<td>5.08 (3)</td>
</tr>
<tr>
<td>173(1)</td>
<td>0.006 (5)</td>
</tr>
<tr>
<td>182.62(5)</td>
<td>0.39 (5)</td>
</tr>
<tr>
<td>185.720(4)</td>
<td>57.0 (3)</td>
</tr>
<tr>
<td>194.94(6)</td>
<td>0.63 (1)</td>
</tr>
</tbody>
</table>
Results

Gamma-ray measurement

~ 1000 µgU/cm²
Results

- low-solid angle alpha-particle counting with LG3 spectrometer

\[
\begin{align*}
\text{counts} & \quad \text{channel} \\
0 & \quad 0 \\
50 & \quad 500 \\
100 & \quad 1000 \\
150 & \quad 1500 \\
200 & \quad 2000 \\
250 & \quad 2500 \\
300 & \quad 3000 \\
350 & \quad 3500
\end{align*}
\]

\sim 600 \text{ µgU/cm}^2
• low-solid angle alpha-particle counting with LG3 spectrometer

~ 1000 µgU/cm²
### Results

- **low-solid angle alpha-particle counting with LG3 spectrometer**

<table>
<thead>
<tr>
<th>TP no</th>
<th>Geom. unc 1/Ω</th>
<th>Integral counts ch500-2500</th>
<th>Time s</th>
<th>Backgr. cps</th>
<th>cps unc</th>
<th>activity Bq unc Bq</th>
<th>mass U µg unc µg</th>
<th>areal density µg/cm²</th>
</tr>
</thead>
<tbody>
<tr>
<td>SS 3</td>
<td>71.77 1%</td>
<td>56727</td>
<td>238000</td>
<td>0.0015</td>
<td>0.24 0.6%</td>
<td>17.0 1.2%</td>
<td>696.7 1.2%</td>
<td>201</td>
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<tr>
<td>SS 4</td>
<td>71.77 1%</td>
<td>11588</td>
<td>60000</td>
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<td>0.19 1.2%</td>
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<td>563.7 1.6%</td>
<td>163</td>
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<tr>
<td>SS 5</td>
<td>71.77 1%</td>
<td>23844</td>
<td>75000</td>
<td>0.0015</td>
<td>0.32 0.8%</td>
<td>22.7 1.3%</td>
<td>930.7 1.3%</td>
<td>269</td>
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<tr>
<td>SS 6</td>
<td>71.77 1%</td>
<td>33759</td>
<td>73708</td>
<td>0.0015</td>
<td>0.46 0.7%</td>
<td>32.8 1.2%</td>
<td>1342.8 1.2%</td>
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<tr>
<td>SS 7</td>
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<td>198257</td>
<td>300000</td>
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<td>1939.4 1.1%</td>
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<td>SS 8</td>
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<td>64039</td>
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<td>0.85 0.5%</td>
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<td>2507.1 1.1%</td>
<td>724</td>
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<td>SS 9</td>
<td>71.77 1%</td>
<td>89764</td>
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<td>0.0015</td>
<td>1.20 0.4%</td>
<td>85.8 1.1%</td>
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<tr>
<td>SS 10</td>
<td>71.77 1%</td>
<td>84114</td>
<td>75000</td>
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<td>1.12 0.4%</td>
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<td>951</td>
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<tr>
<td>SS 13</td>
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<td>261274</td>
<td>225000</td>
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<td>1.16 0.2%</td>
<td>83.2 1.0%</td>
<td>3411.2 1.0%</td>
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<tr>
<td>SS 14</td>
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<td>11397</td>
<td>150000</td>
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<td>0.07 1.6%</td>
<td>5.3 1.9%</td>
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<td>Al 16 IP</td>
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<td>14948</td>
<td>75000</td>
<td>0.0015</td>
<td>0.20 1.1%</td>
<td>14.2 1.5%</td>
<td>581.8 1.5%</td>
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</tr>
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<td>Al 20 IP</td>
<td>71.77 1%</td>
<td>47554</td>
<td>65715</td>
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<td>0.72 0.5%</td>
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<td>Al 21 IP</td>
<td>71.77 1%</td>
<td>42522</td>
<td>75000</td>
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<td>0.57 0.6%</td>
<td>40.6 1.2%</td>
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<tr>
<td>Al 22 IB</td>
<td>71.77 1%</td>
<td>34372</td>
<td>75000</td>
<td>0.0015</td>
<td>0.46 0.7%</td>
<td>32.8 1.2%</td>
<td>1343.6 1.2%</td>
<td>388</td>
</tr>
<tr>
<td>SS 24 Ox</td>
<td>71.77 1%</td>
<td>6903</td>
<td>75000</td>
<td>0.0015</td>
<td>0.09 1.8%</td>
<td>6.5 2.0%</td>
<td>266.3 2.0%</td>
<td>77</td>
</tr>
</tbody>
</table>
Results

• Autoradiography

Oxalate  SS

Iso-propanol  Al
Result

• Autoradiography: SS Oxalate

~ 600 µgU/cm²
Result

- Autoradiography: Al Iso-propanol

\[ \sim 600 \, \mu gU/cm^2 \]
Results

• X-ray fluorescence spectrum of U electroplated on stainless steel disc excited by Cd-109 excitation source

EDXRF analysis:

530 μgU/cm²

thickness: 0.28 μm

~ 600 μgU/cm²
Conclusion

- Stainless steel and aluminium backings were used for preparation of thick uranium layers
- Different electrolytes were tested
- The layers of uranium were characterized by
  - Gamma-ray spectrometry
  - Alpha-particle counting
  - Autoradiography
  - Energy dispersive X-ray spectrometry
- Up to 1 mg U/cm² could be deposited with sufficient homogeneity
Thank you