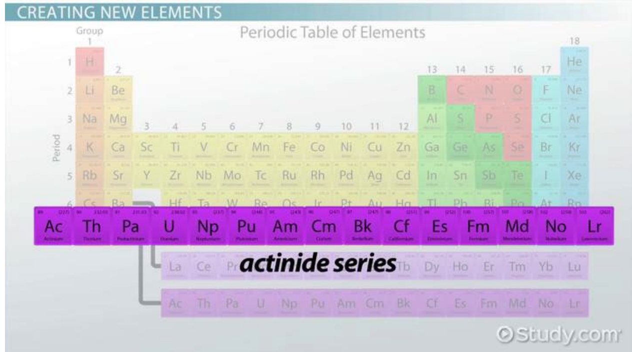
Novel Actinide Target Making Method: Spin Coating Assisted Combustion Synthesis

Ashabari Majumdar

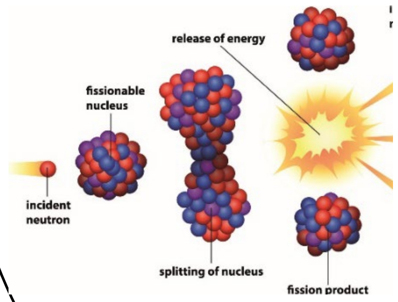
Funded by NNSA, Grant # DE-NA0003888 and DE-NA0004093)

Actinide Research

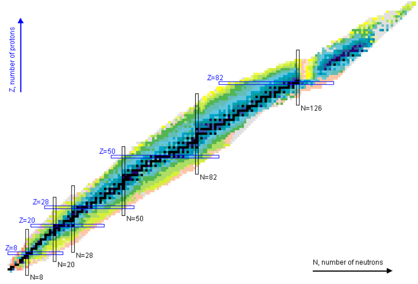


Fundamental Science

Elemental abundances



Fission and neutron capture



Nuclear structure and properties

Applications

Stockpile stewardship program

Nuclear medicine

Nuclear power plants

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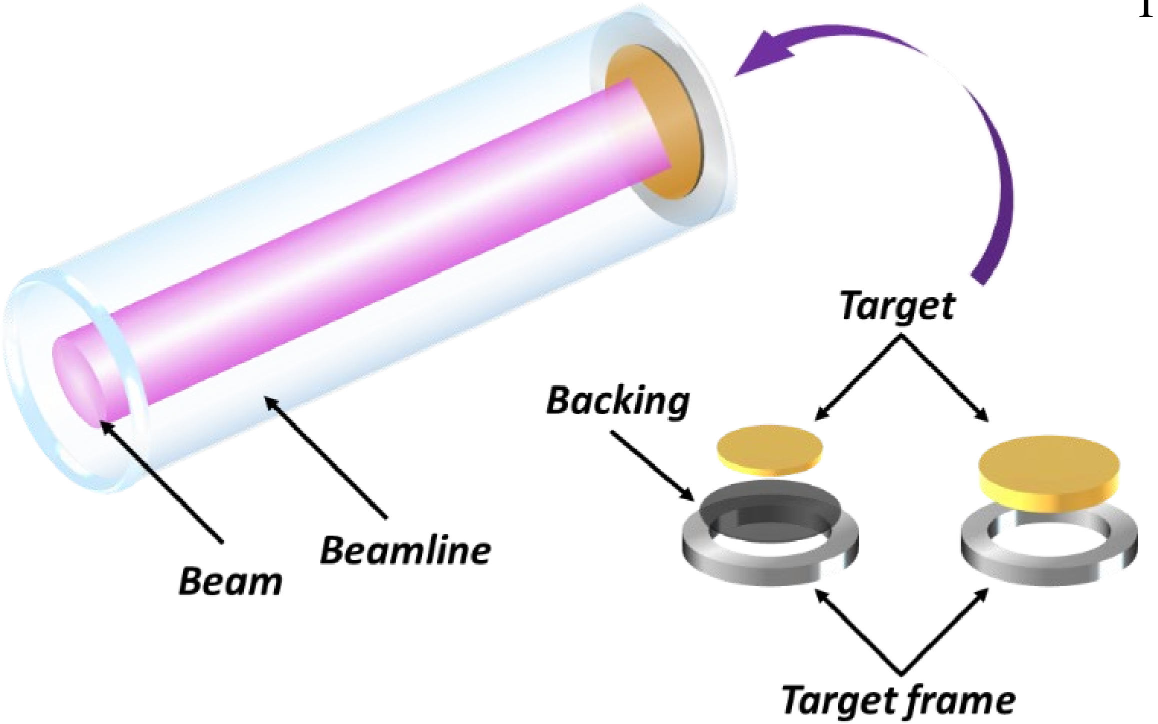
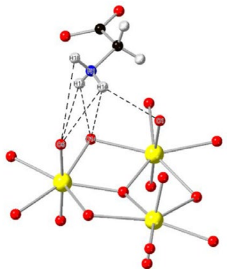
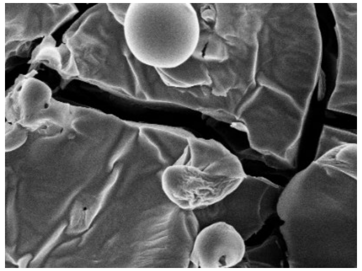
Importance of Targets



S. Dede et al., Actinide Target Materials for Nuclear Physics Measurements: A Review article (in preparation)

Ideal targets are

1. Highly uniform 2. Stable under irradiation 3. Cost efficient

Actinide Properties	Target Making Limitations
	
	
	

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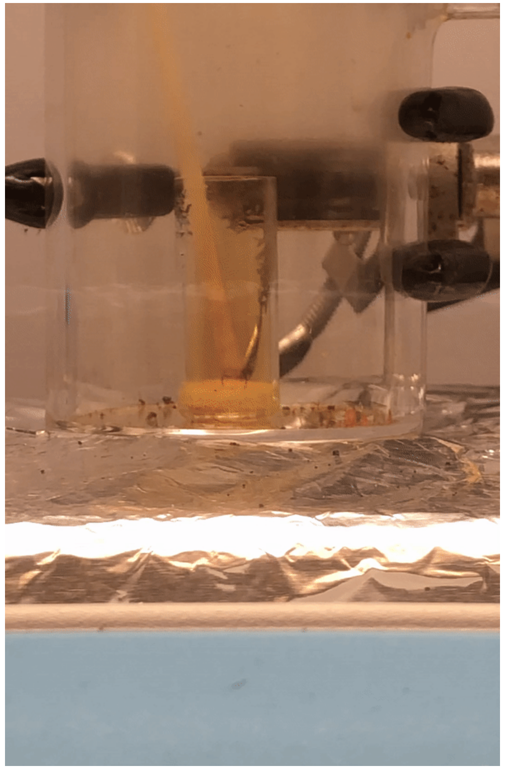
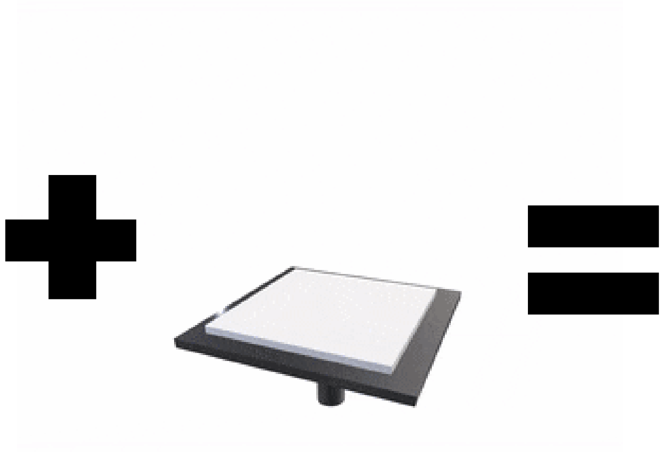
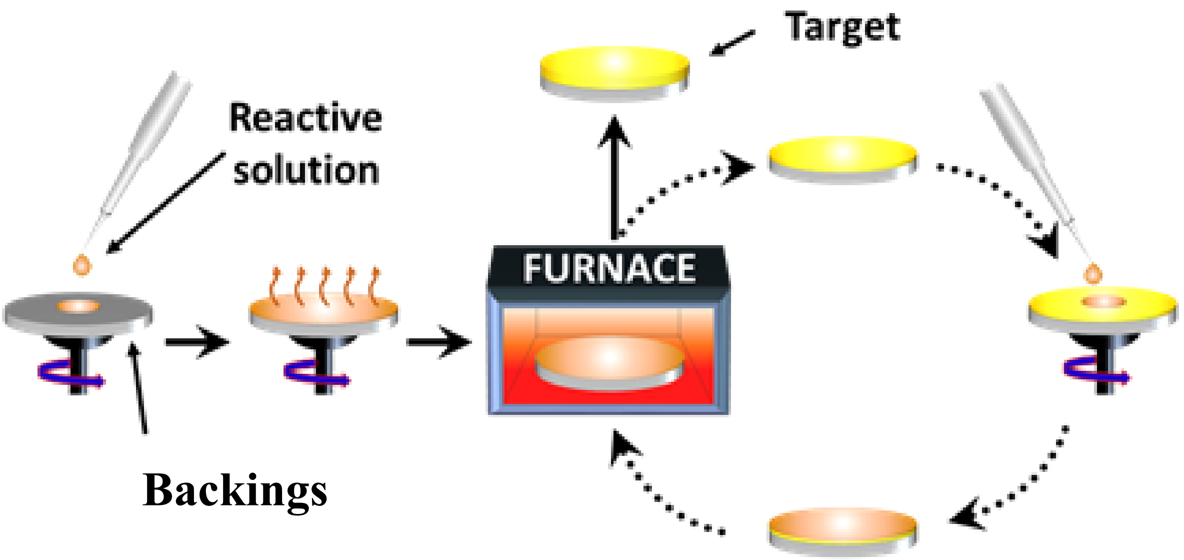
Novel Method for Target Making



Solution Combustion Synthesis



Spin Coating



Spin Coating Assisted Combustion

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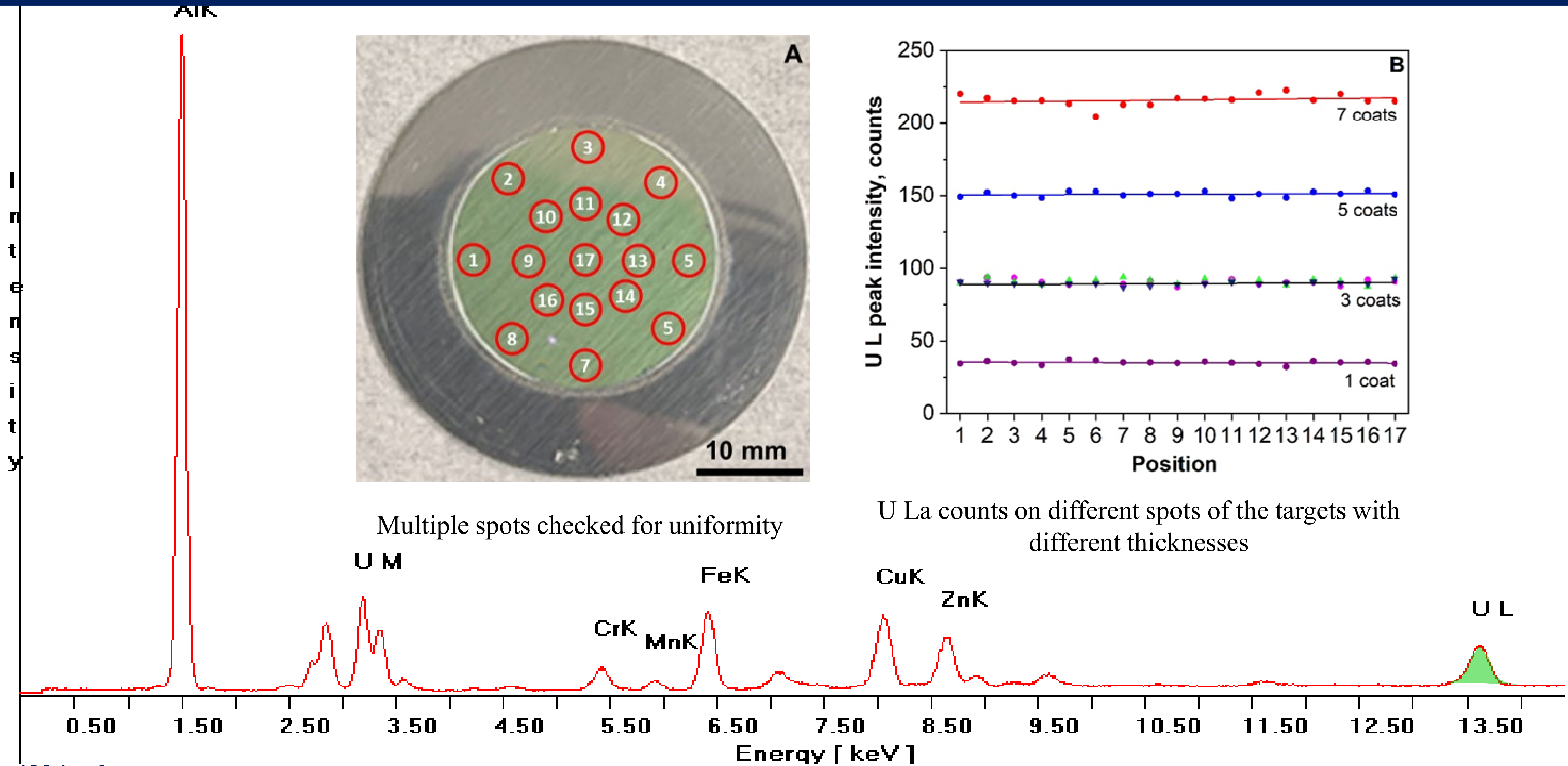
Uniform and Reproducible



Multiple spots checked for uniformity

U La counts on different spots of the targets with different thicknesses

X-ray fluorescence spectra showing presence of U and other elements in the Al alloy backing

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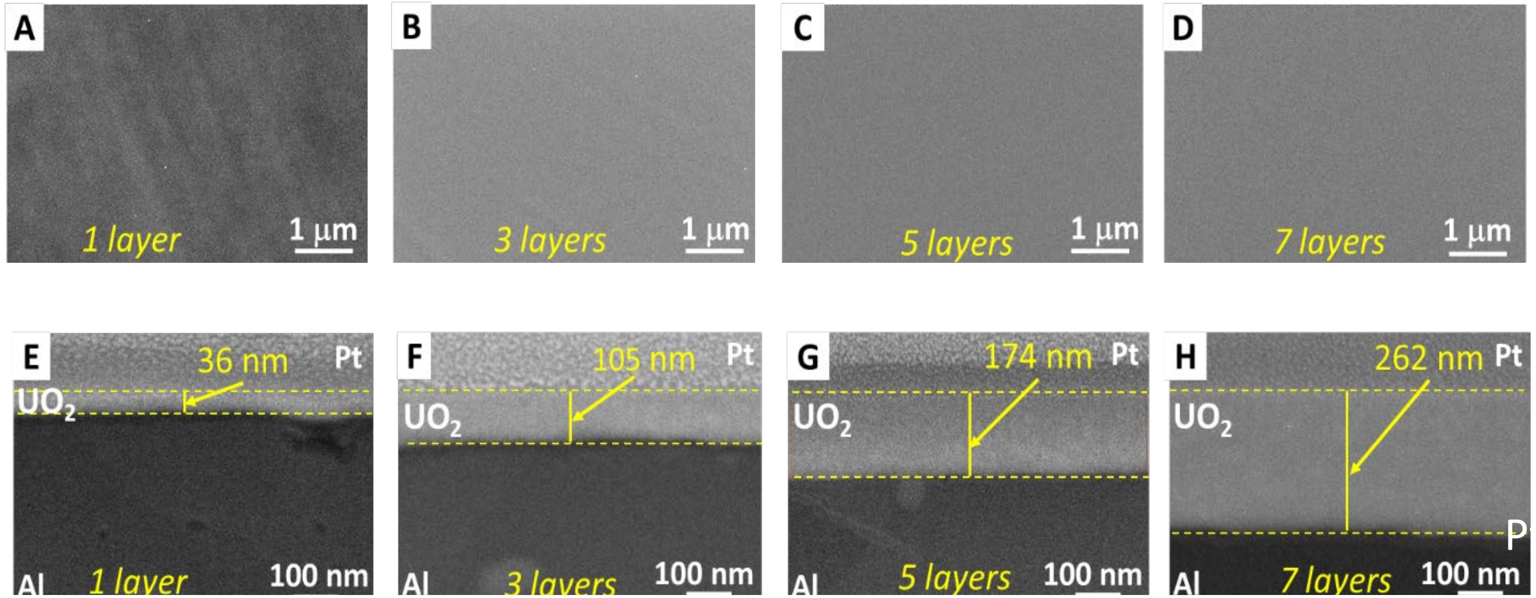
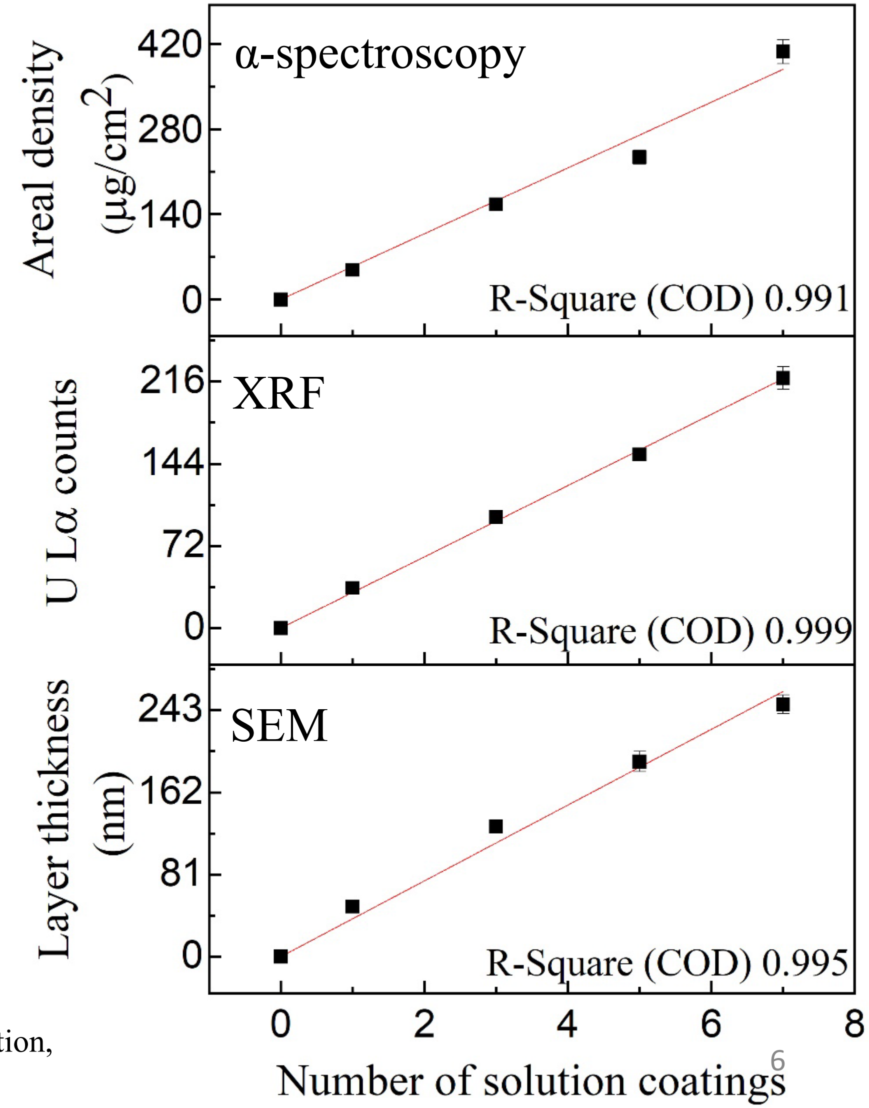
Controlled Thickness



Surface (A-D) and cross-sectional (E-H) SEM images of samples with increasing UO_2 layer thickness on Al substrate.



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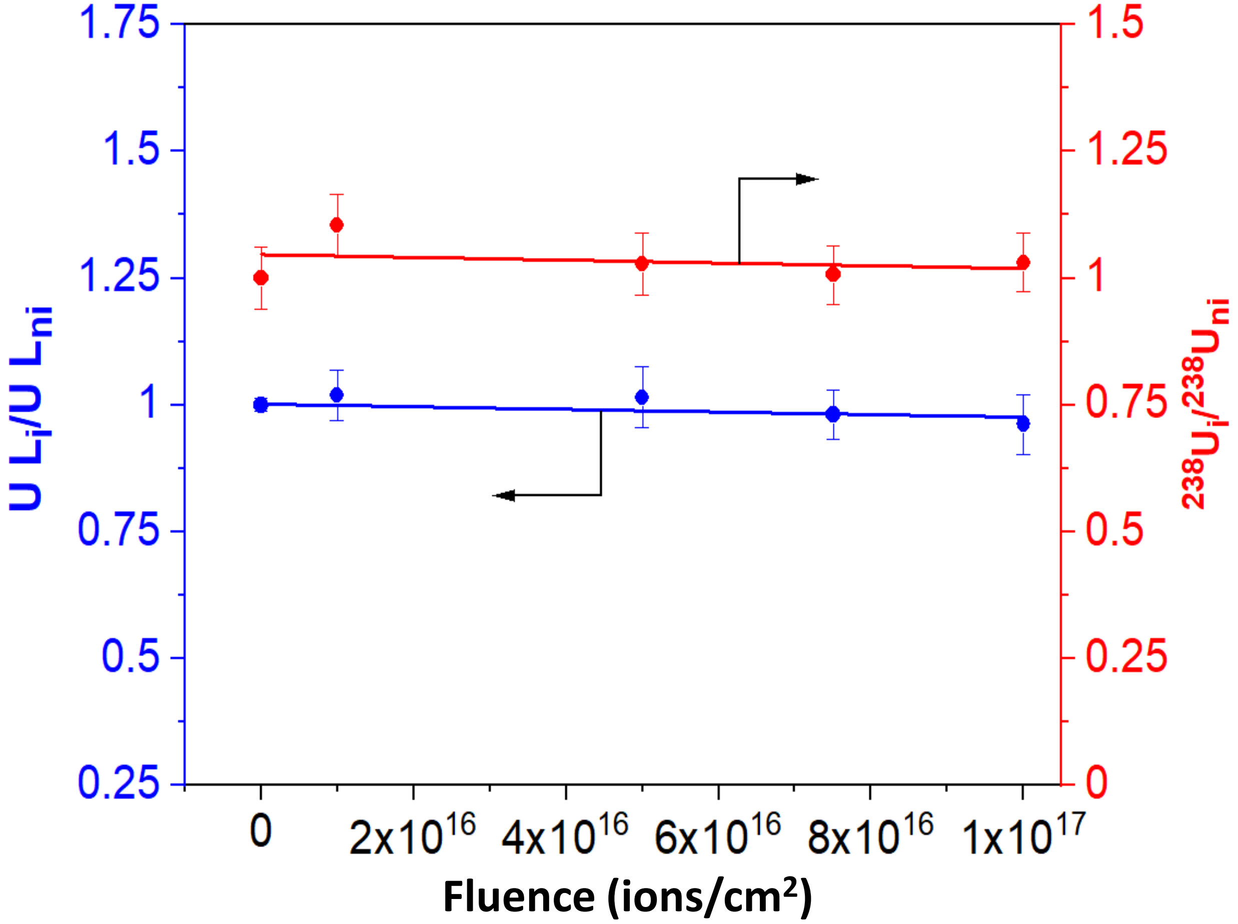
Majumdar et al., Irradiation Driven Restructuring of UO_2 thin films: Amorphization and Crystallization, ACS Appl. Mater. & Interfaces, (2021).

Robust Under Ion Irradiation

Ar^{2+} beam at energy 1.7 MeV
Fluence up to 10^{17} ions/cm^2

5U St. Ana accelerator
University of Notre Dame

$U_{irradiated}/U_{non\text{-}irradiated}$
X-Ray Fluorescence Spot scans (left Y axis)
α-spectroscopy (right Y-axis)



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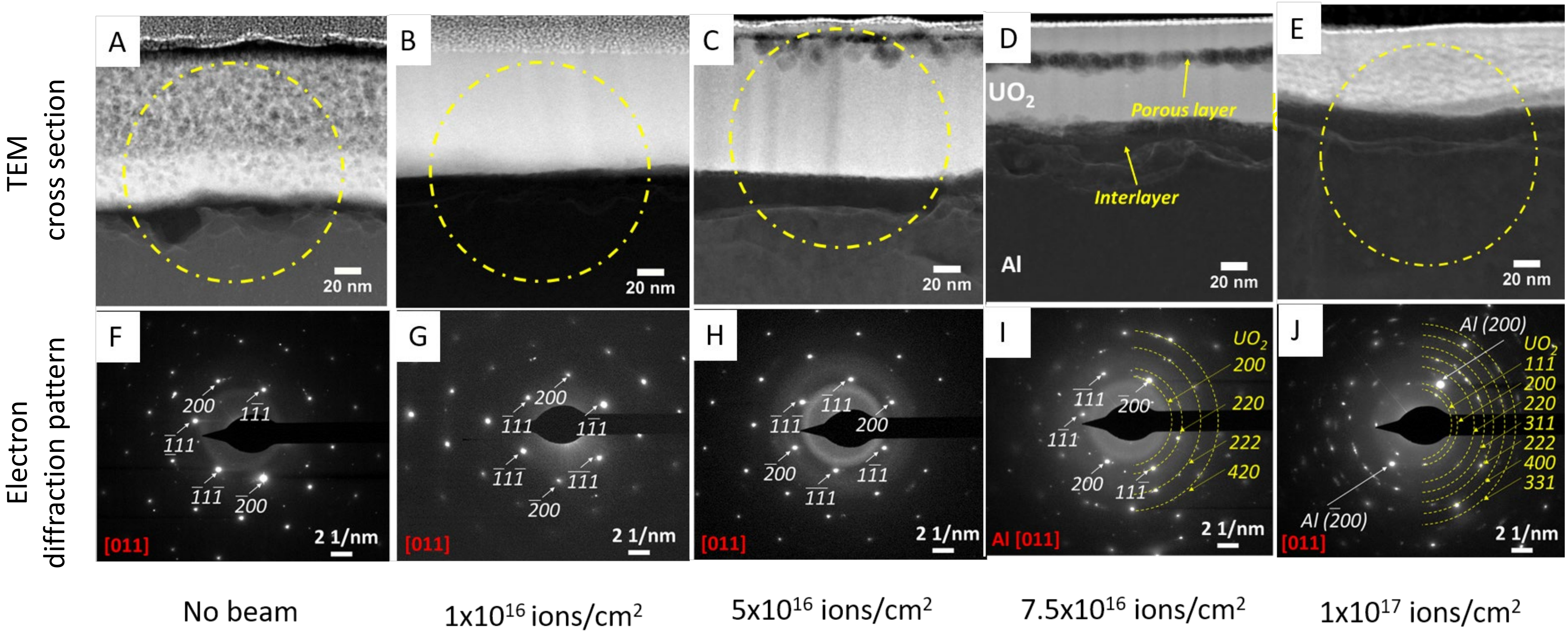
Chemical and Structural Changes



Irradiation-Driven Restructuring of UO_2 Thin Films: Amorphization and Crystallization

Ashabari Majumdar, Khachatur V. Manukyan*, Stefania Dede, Jordan M. Roach, Daniel Robertson, Peter C. Burns, and Ani Aprahamian

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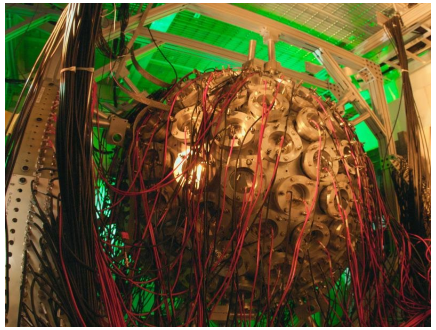
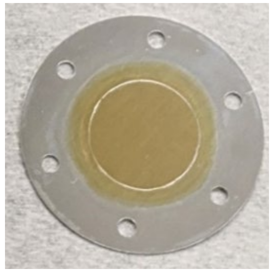
Publication Date: July 16, 2021
https://doi.org/10.1021/acsami.1c08682

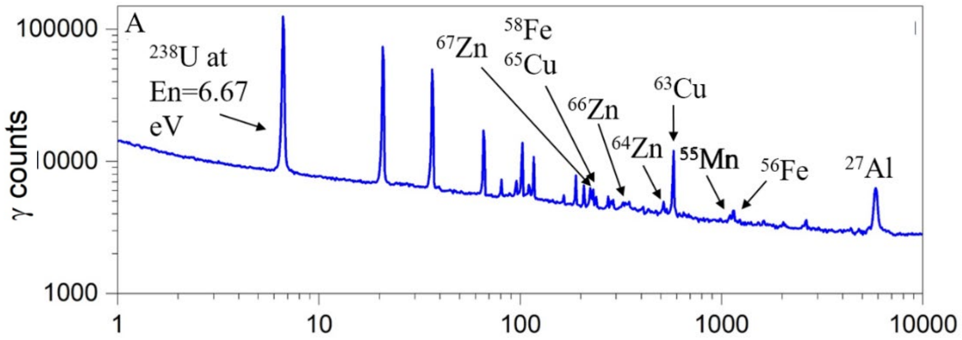
SUBJECTS: Deposition, Grain,

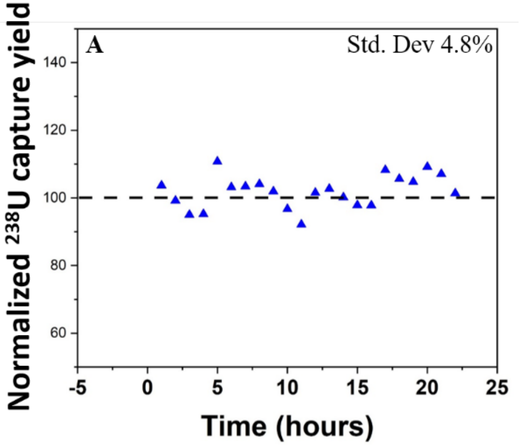
Target Purity and Consistency

Beam: neutrons
Detectors: DANCE
At Los Alamos National Laboratory US

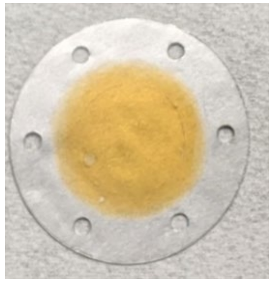


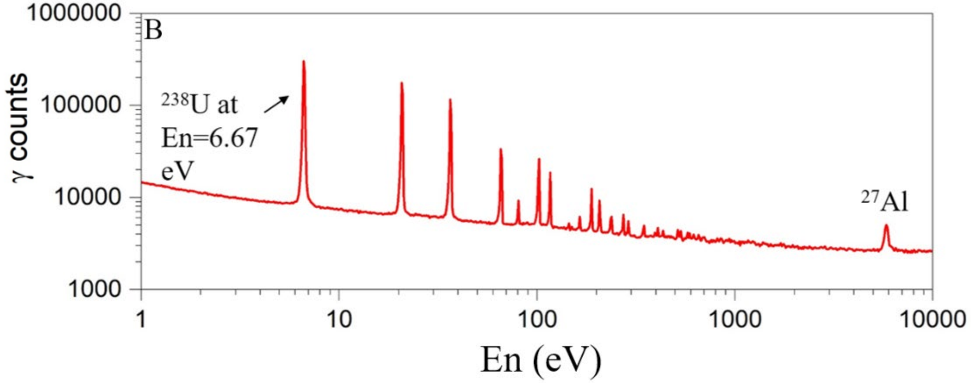


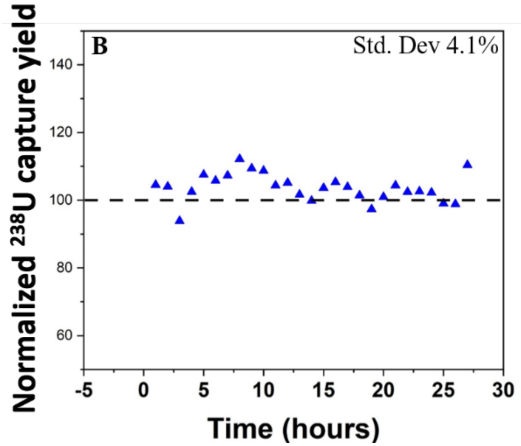




400 $\mu g/cm^2$ UO_2 on 96.75% pure Al alloy







1000 $\mu g/cm^2$ UO_2 on 99.997% pure Al

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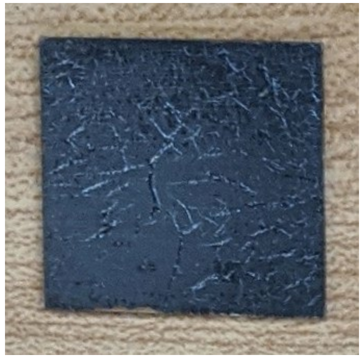
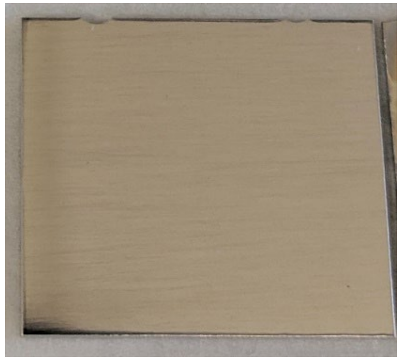
Homogeneous on Variety of Backings

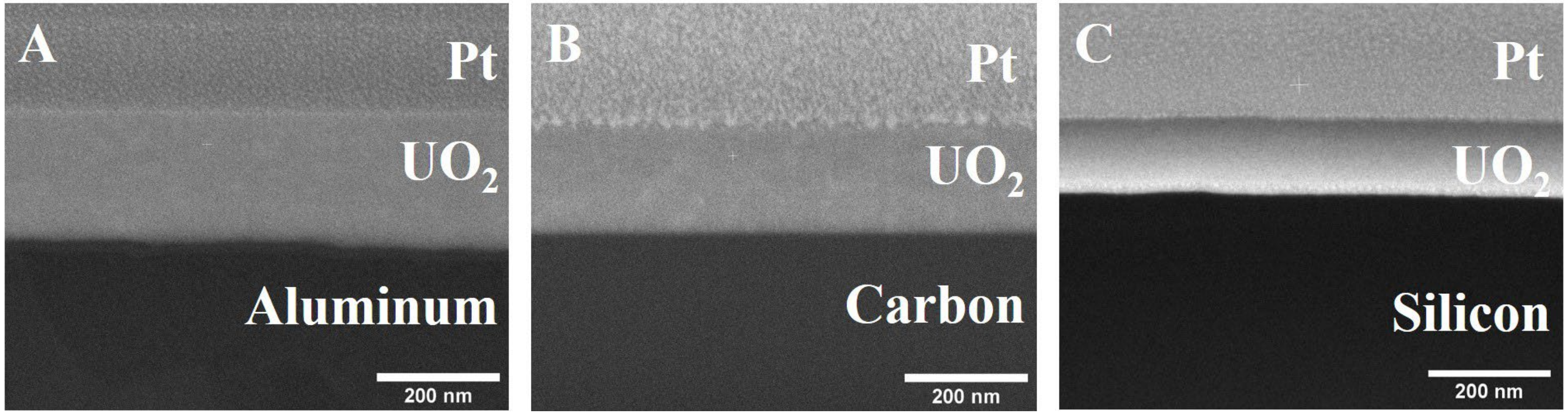
Aluminum



Carbon



Silicon



Cross-sectional SEM images of UO_2 thin films on different backings- aluminum (A), carbon (B) and silicon (C)

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Summary

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Novel actinide target making method:

Spin coating assisted solution combustion synthesis

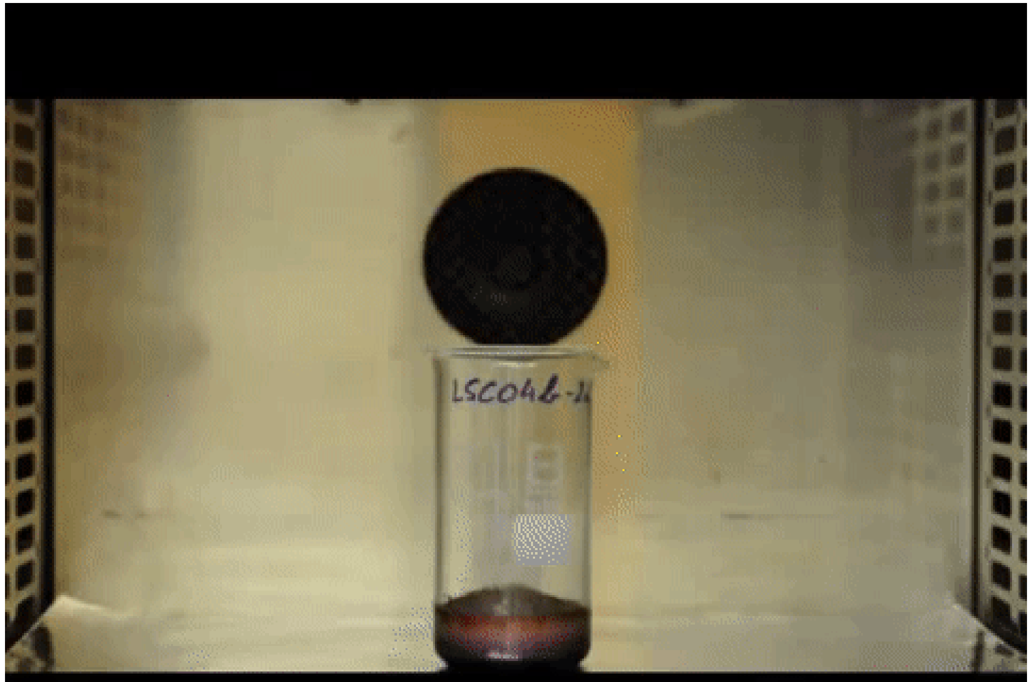
Characteristics of the method:

1. Simple set-up
2. Any solid backing can be used
3. Precise control over target thickness (30-1000 μg/cm^2)
4. Targets are- a. uniform (<5% thickness deviation) b. reproducible and c. does not degrade under ion and neutron irradiation
5. No contamination found

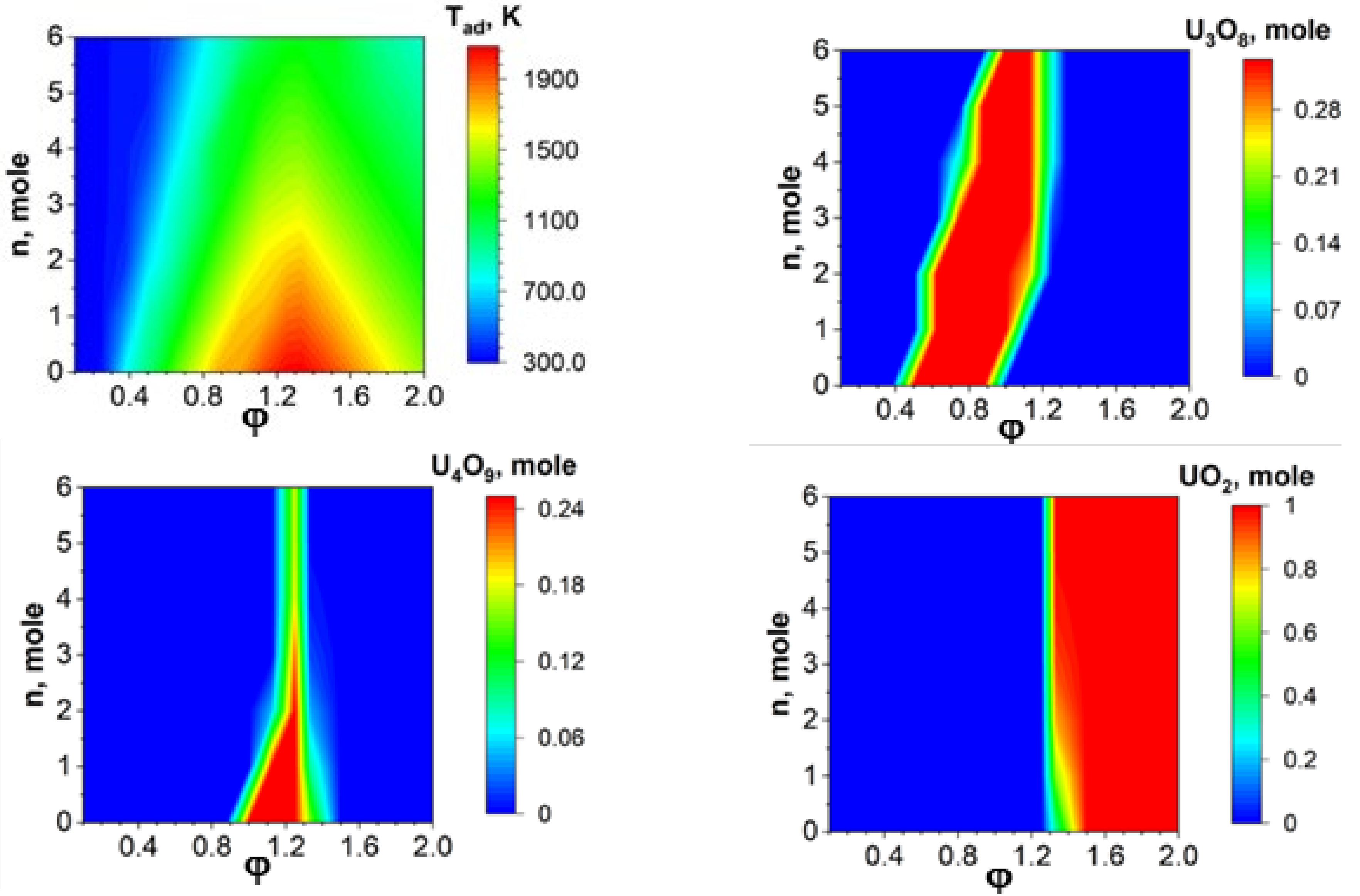
Backup Slides

Preparation method: Solution Combustion Synthesis

Oxidizers	Fuel	Solvents
Metal nitrate $Me^n(NO_3)_n$ where Me = Fe, Ni, Zn, Al, Cu, Sr, La, Bi, Zr, Li, Co, U, Ce, etc. n - metal valence	• Acetylacetone $C_5H_8O_2$ • Citric Acid, $C_6H_8O_7$ • Glycine, $C_2H_5NO_2$ • Glucose, $C_{12}H_{22}O_{12}$ • Hexamethylenetetramine, $C_6H_{12}N_4$	• Water • Ethanol • Methanol • 2-methoxy ethanol

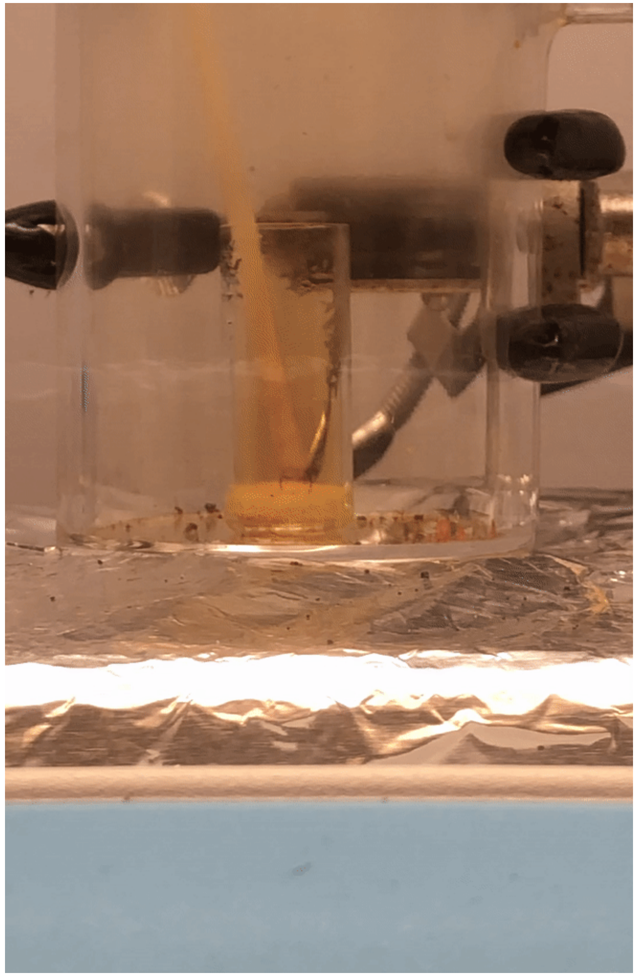


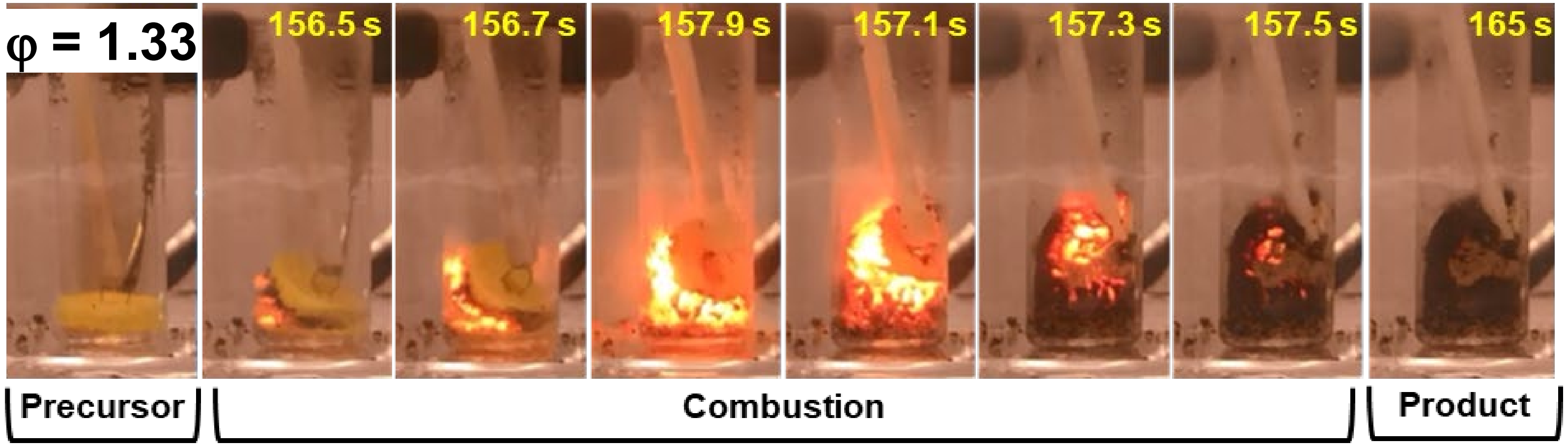
Thermodynamic modeling of SCS for uranium oxides

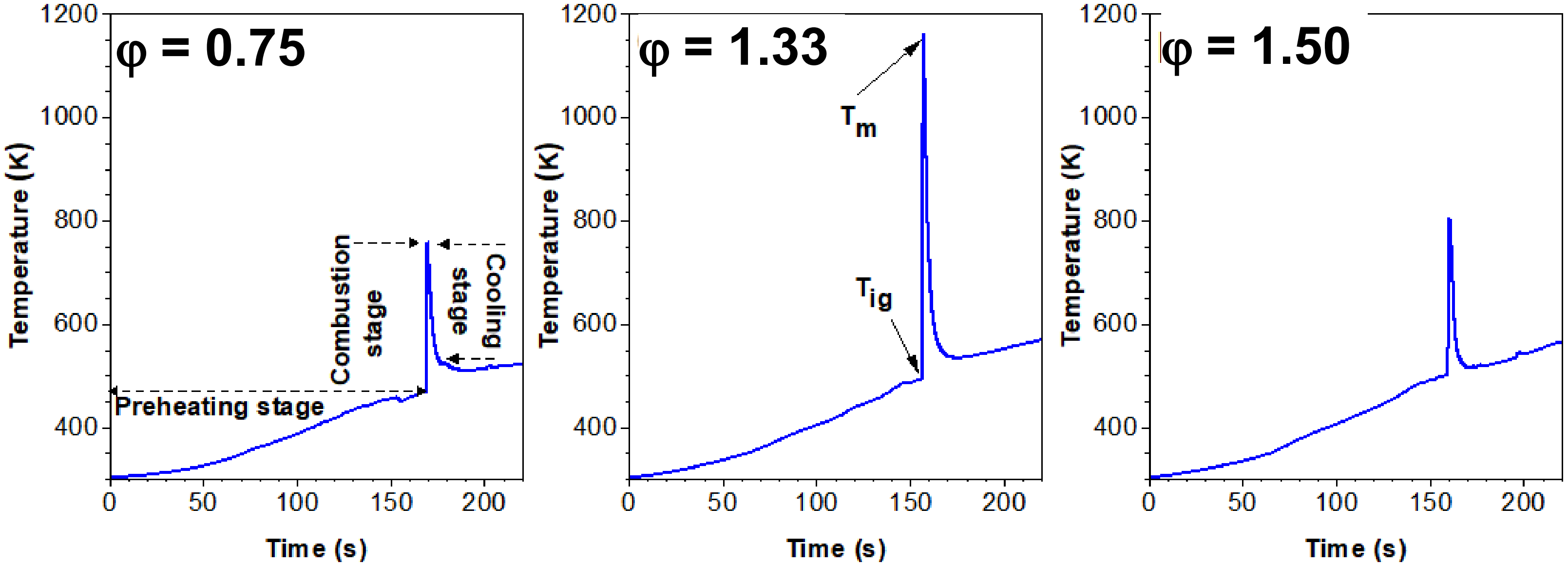


Calculated adiabatic temperature (T_{ad}) and composition of equilibrium solid products for the $UO_2(NO_3)_2$ + $\varphi C_2H_5NO_2$ + nH_2O system

SCS in the $UO_2(NO_3)_2$ + $\varphi C_2H_5NO_2$ bulk system



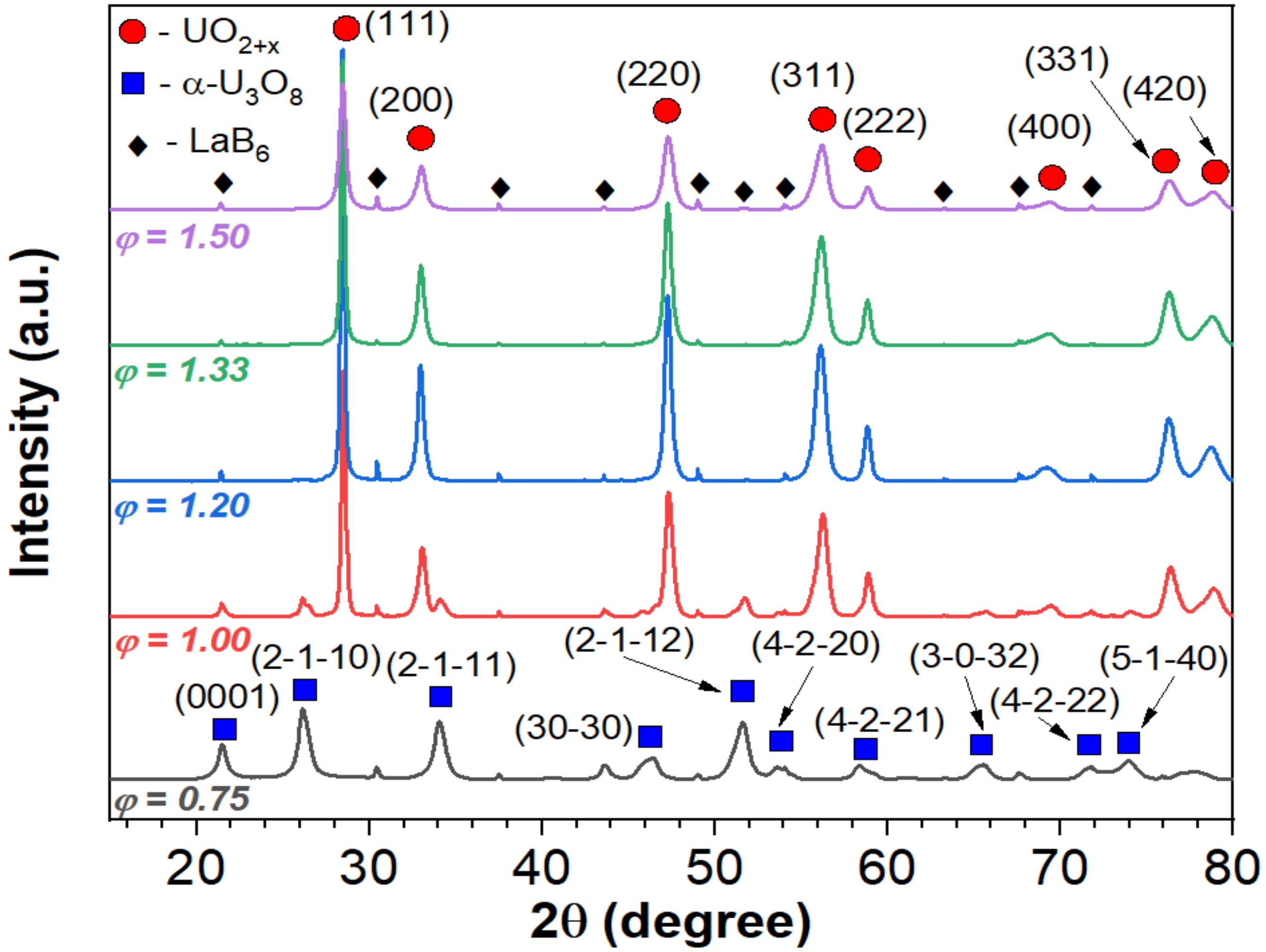




Time – temperature profiles for combustion for $UO_2(NO_3)_2$ + $\varphi C_2H_5NO_2$ system

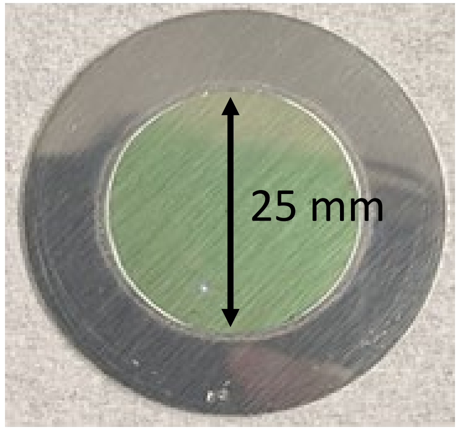
ACS Inorg. Chem. 60, 18938−18949 (2021)

The phase composition of bulk products

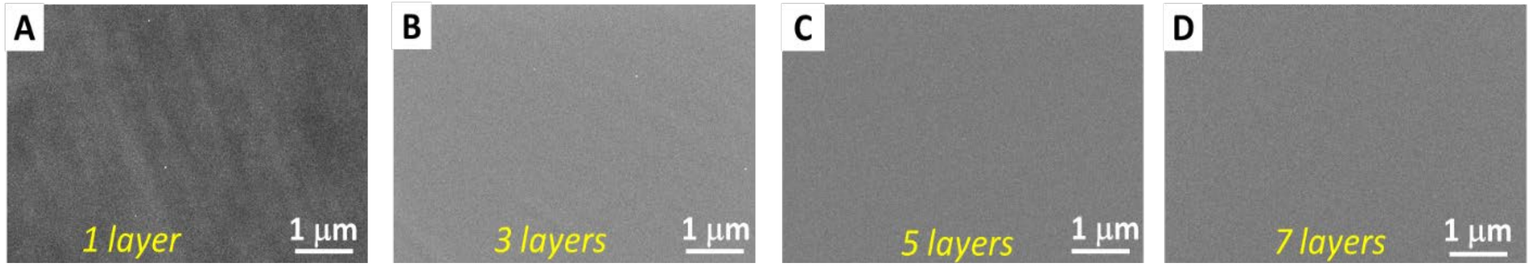


XRD patterns of uranium oxides prepared by combustion of $UO_2(NO_3)_2$ + $\varphi C_2H_5NO_2$ precursors with different φ.

Homogenous surface



A typical depleted UO_2 target on Al backing



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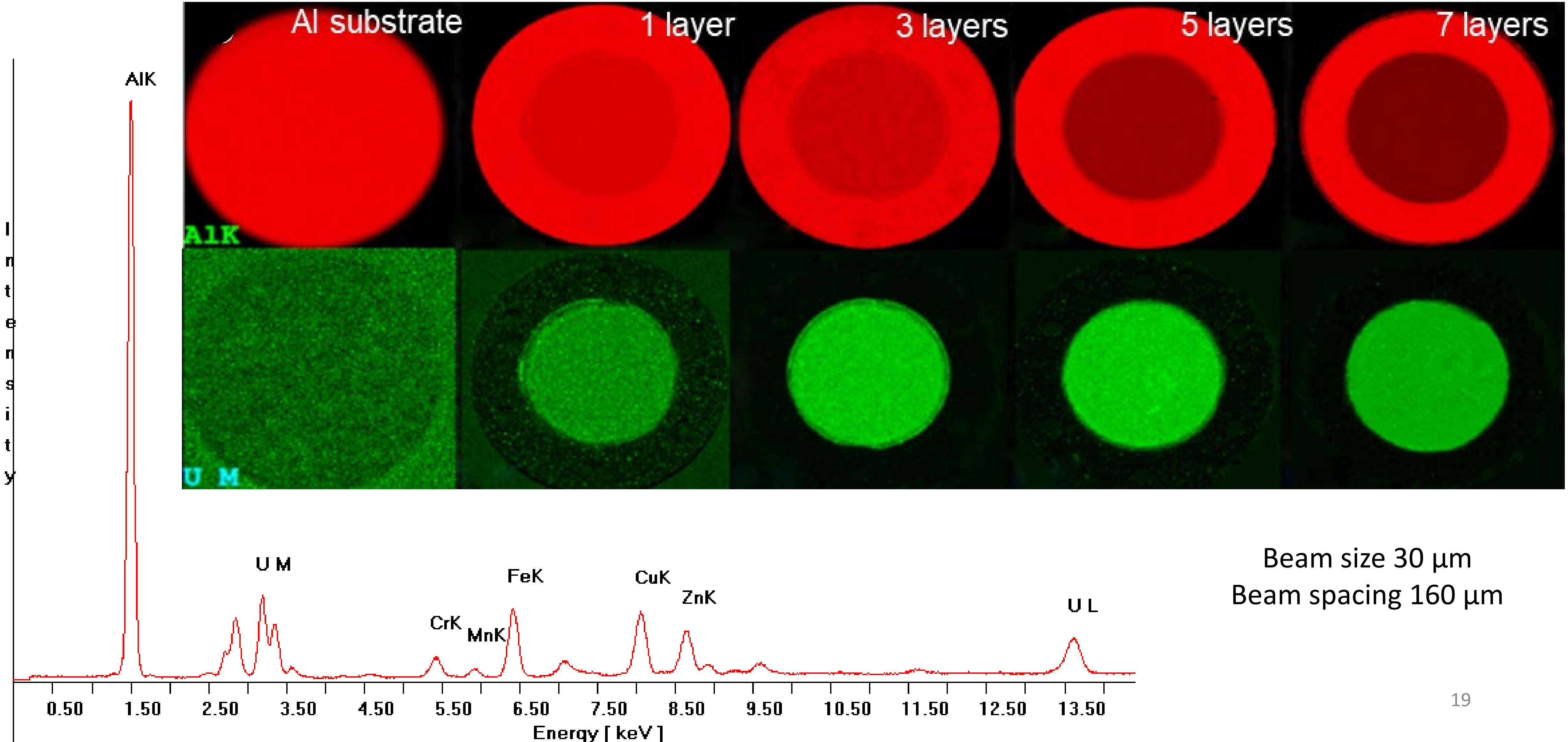
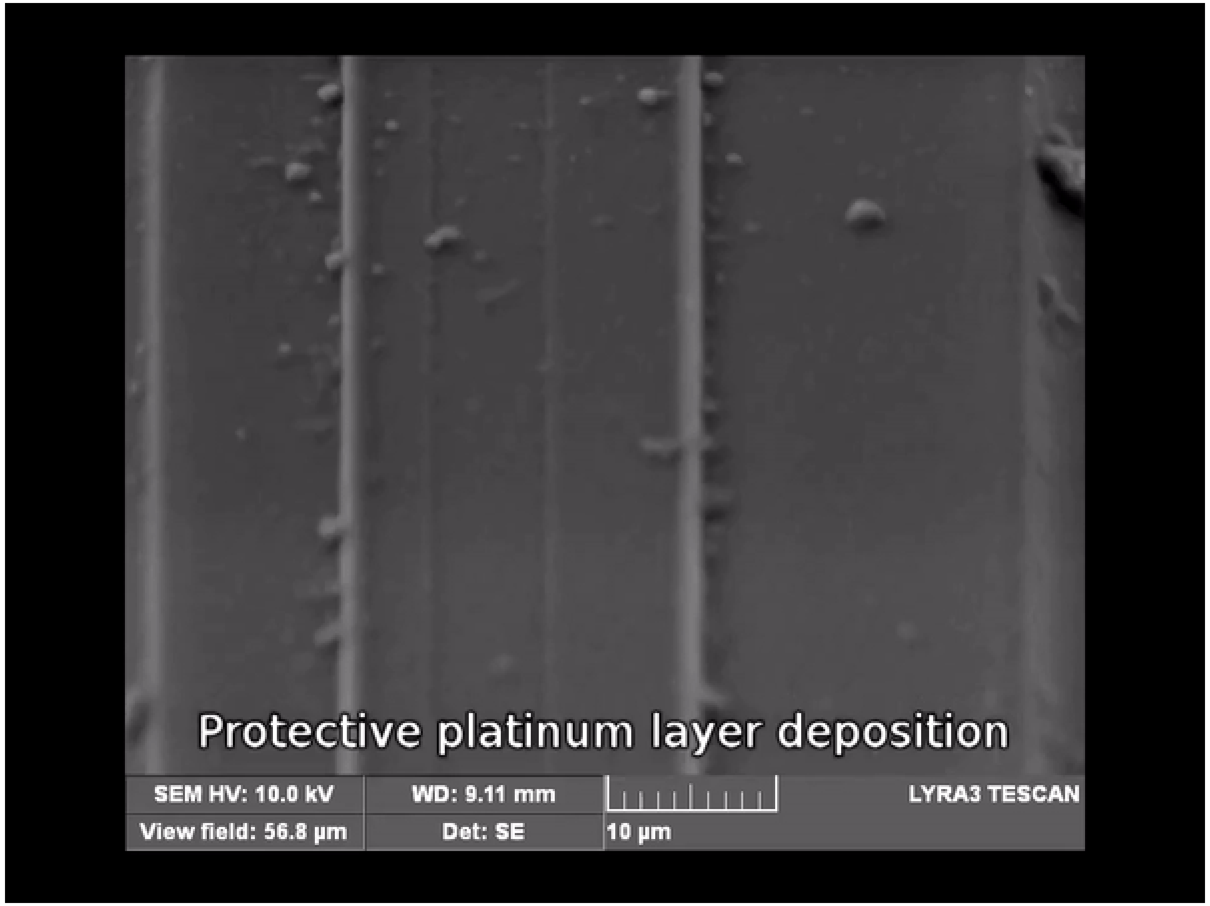
Macroscopic Uniformity



Beam size 30 µm
Beam spacing 160 µm

Thickness and Quality Investigation

Focused Ion Beam Milling Assisted Scanning Electron Microscopy



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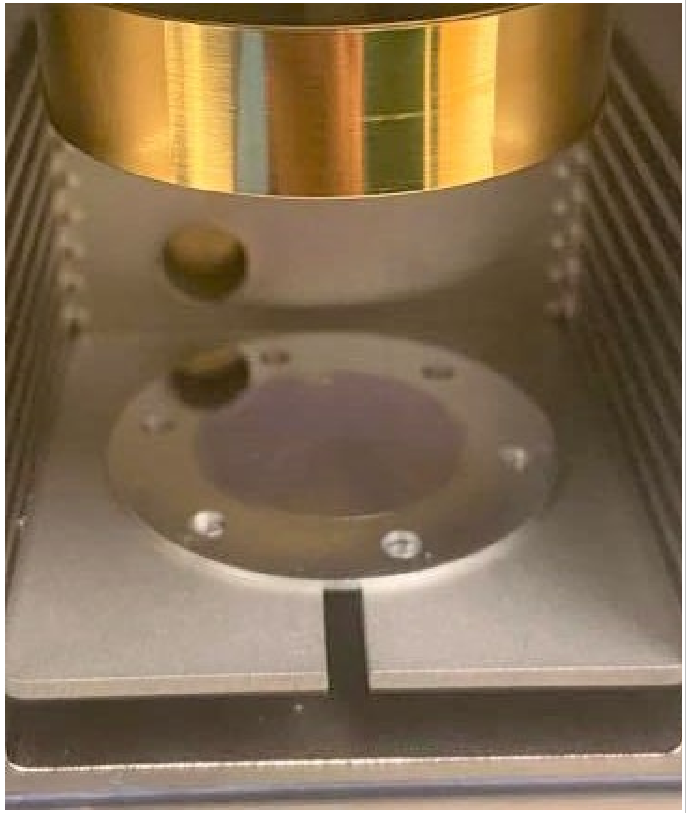
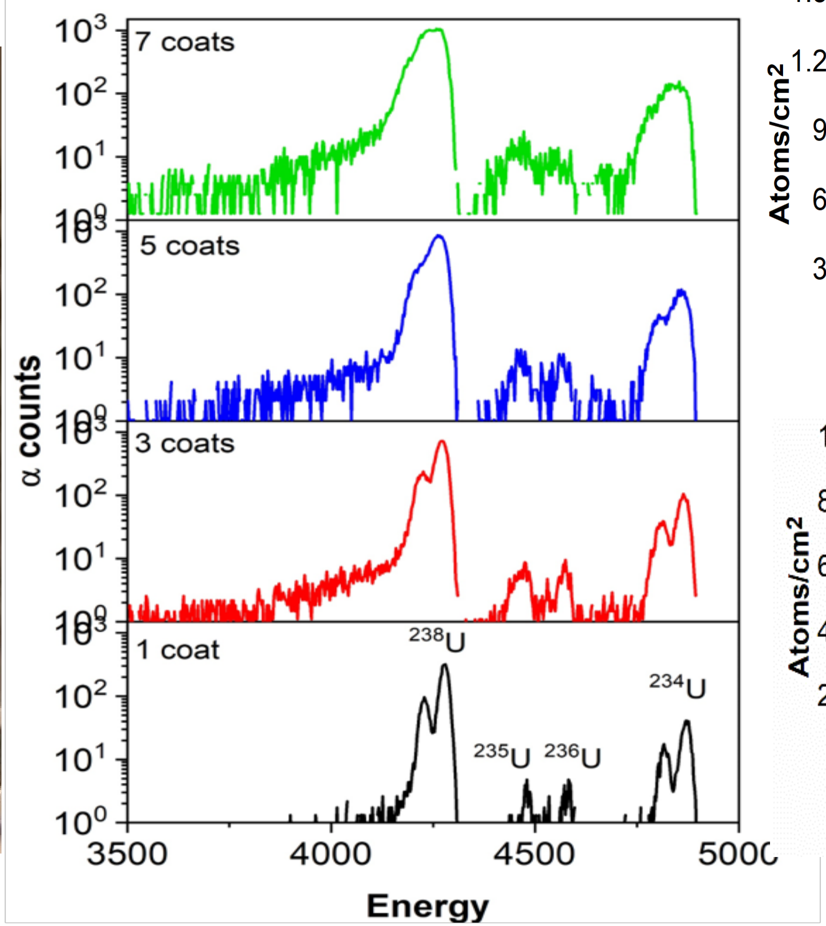
Controlled Uranium Quanity



ORTEC alpha spectrometer with Silicon detector



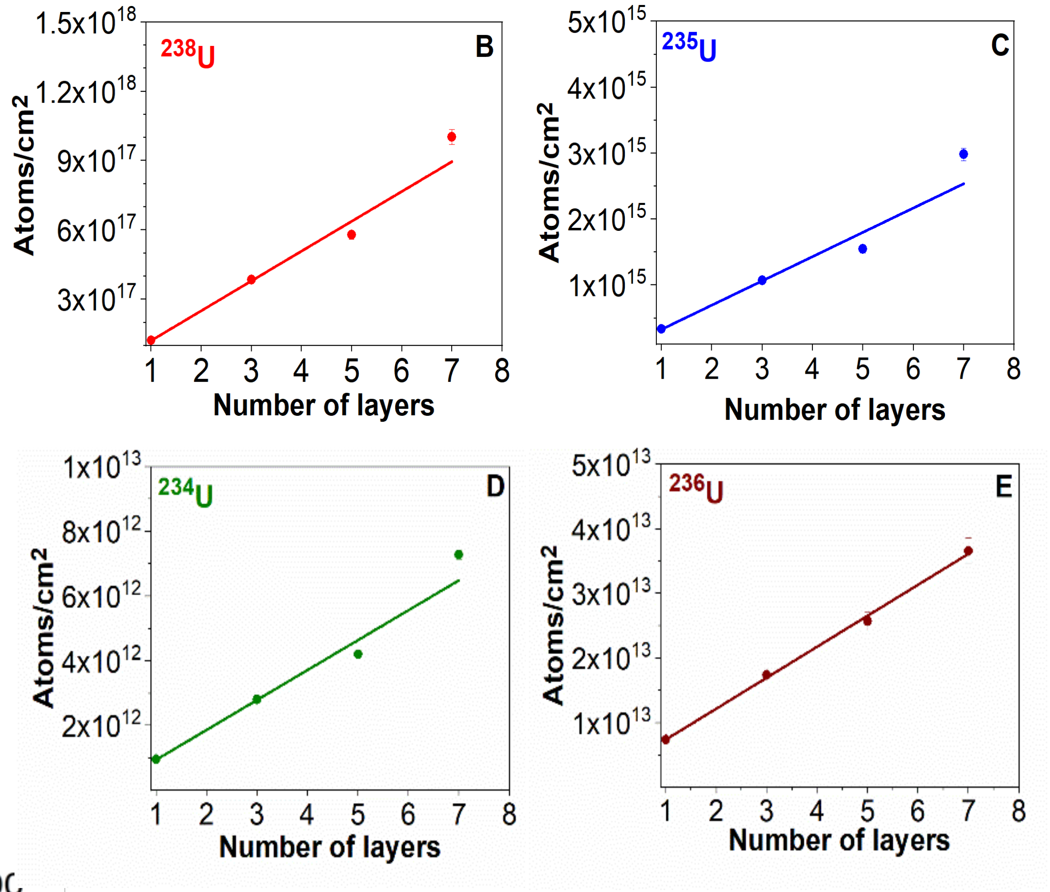
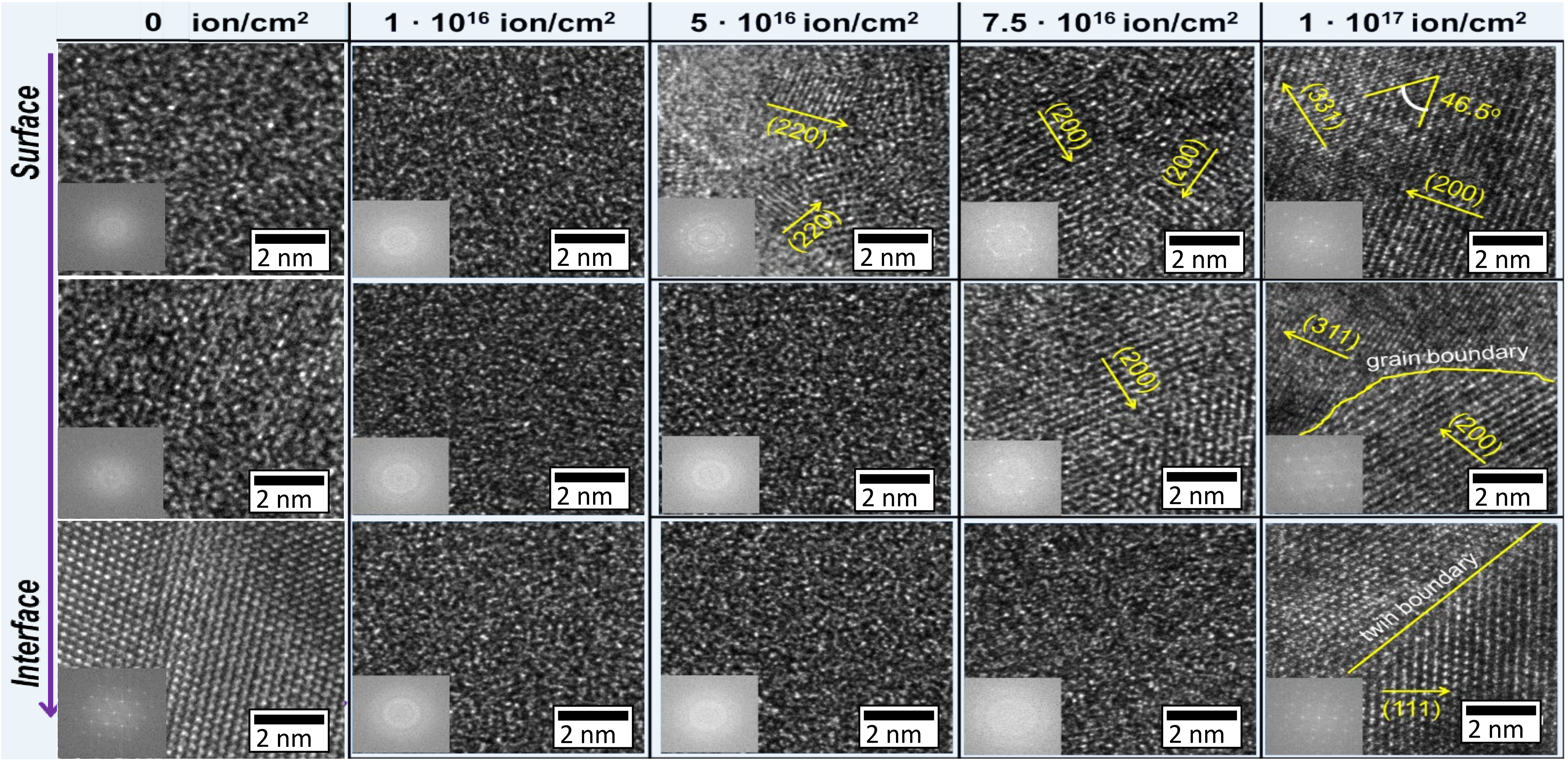


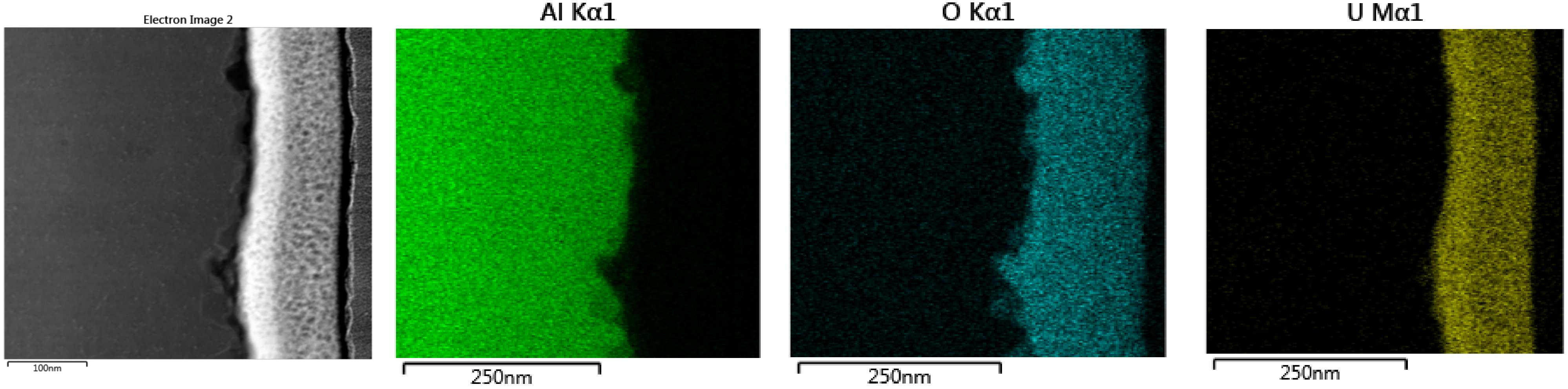
Photo Credits: Stefania Dede

High resolution TEM images

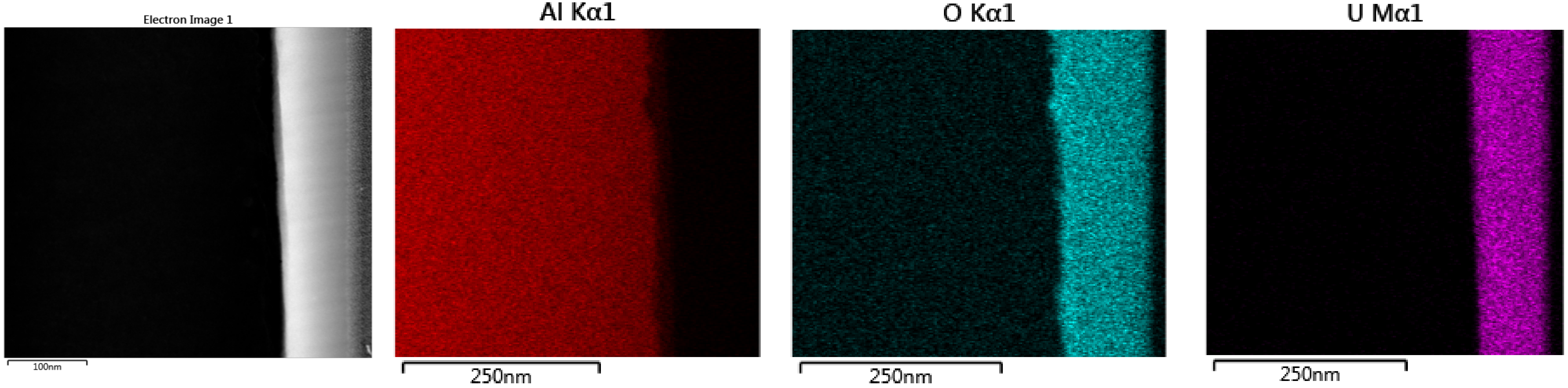


EDS elemental mapping

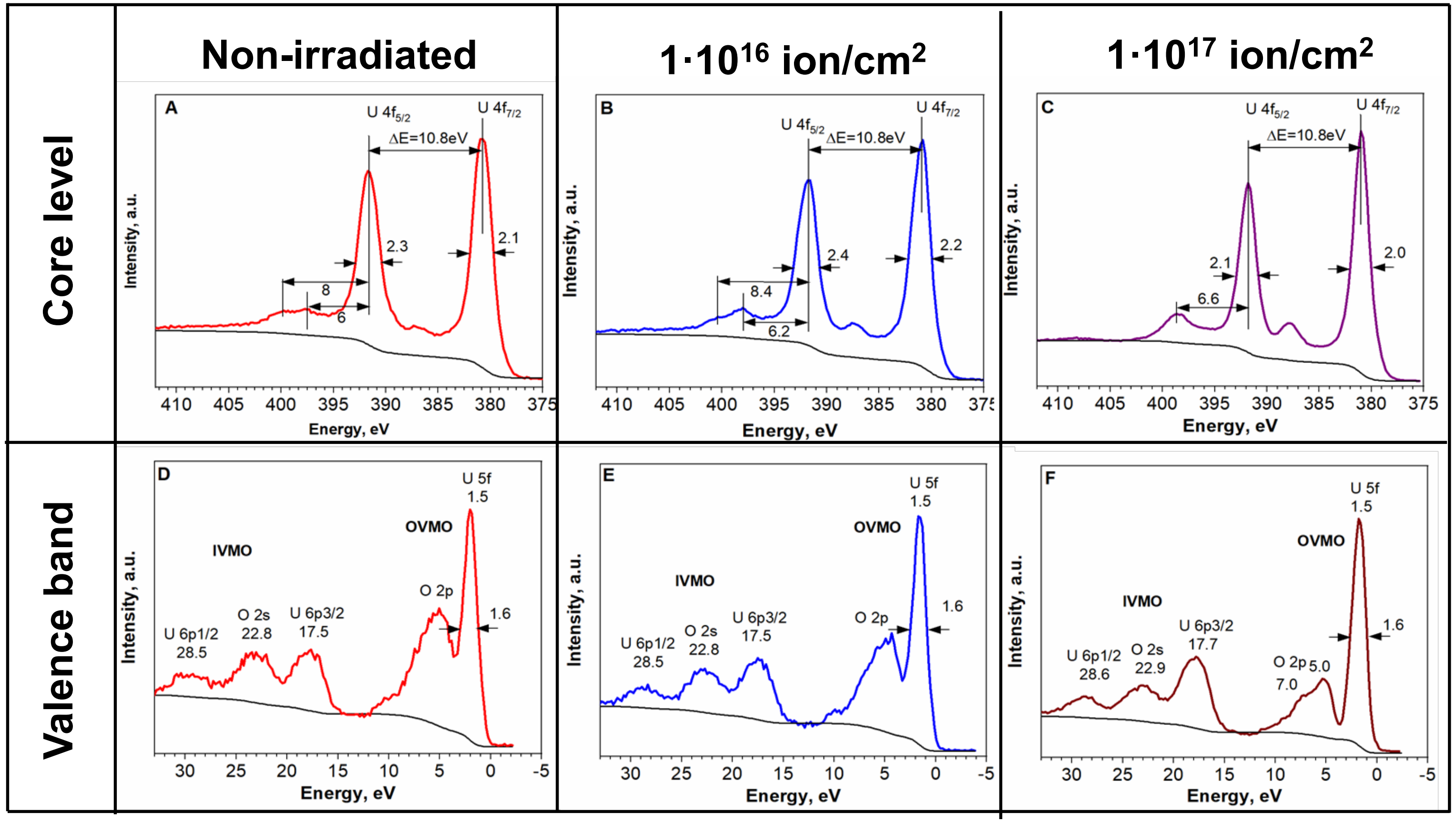
Before Irradiation



After Irradiation ($1x10^{16}$ ions/cm^2)



Stoichiometry of UO_{2+x} Thin Films: X-ray Photoelectron Spectroscopy



Oxygen coefficient (k_0)

K_0=2+x

$$\frac{U\,5f}{U\,4f_{7/2}} = I_1 = 5.366k_0^{-7.173}$$

$$I_2 = -0.0383k_0 + 0.1149$$

$$I_3 = 2I_1 - I_2$$

$$v_1(U^{4+}) = \frac{I_3}{0.0383}$$

$$v_2(U^{5+}) = \frac{2(I_2 - I_1)}{0.0383}$$

$$v_3(U^{6+}) = \frac{0.0383 - I_2}{0.0383}$$

Table 1 Oxygen coefficient (k_o) and surface ionic composition of UO_{2+x} films

Irradiation fluence, ion/cm²	I_1	k_0=2+x	I_2	I_3	ν, %		
					U^{4+}	U^{5+}	U^{6+}
0	0.0324	2.039	0.037	0.028	73	23	4
$1·10^{16}$	0.0317	2.045	0.037	0.027	70	25	5
$1·10^{17}$	0.0345	2.021	0.038	0.032	82	16	2

Inorg. Chem. **2016**, *55* (16), 8059–8070

Carbon targets after floating