A Novel Technique for the Production of Robust Actinide Targets

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The **GOAL** of this project is to develop new approaches for the preparation of actinide targets that are isotopically pure, cost-efficient, reliable, robust, and highly uniform with controlled thicknesses.

The program relies on the use of rapid **solution combustion synthesis** (SCS) processes between actinide metal nitrates with different organic compounds for the preparation of thin films as targets for nuclear science measurements.

**OBJECTIVES:**

1. Investigate chemical reactions between oxidizers and organic compounds
2. Investigate thin films deposition
3. Investigate electro-spraying techniques with actinide-oxide clusters
4. Create actinide targets by electro-spraying
5. Characterization and testing of targets
6. Modernization of target preparations

**MILESTONES**

- Determine the dynamics and kinetics of SCS for actinides  
  September 30, 2019
- Apply the method to produce uranium (U) targets and study characteristics  
  September 30, 2020
- Extend the procedure to other actinides such as Pu and Am.  
  September 30, 2021
Team

Graduate Students

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Articles & Talks

Articles:
5. P. Sapkota, A. Aprahamian, K.Y. Chan, B. Frentz, K.T. Macon, S. Ptasinska, D. Robertson, K. Manukyan, Irradiation-induced reactions at the CeO₂/SiO₂/Si interface, Accepted to *Journal of Chemical Physics*

Presentations:
1. solution combustion synthesis; 2. Spin coating; 3. Glove boxes for Pu and Am; 4. Alpha spectrometer (6 simultaneous)
First step: SCS in surrogate systems

**Fe(NO_3)_3 + Hexamethylenetetramine (HMTA)**

- **HMTA/Fe(NO_3)_3 = 2**
  - $T_{\text{max}} = 1370^\circ C$

- **HMTA/Fe(NO_3)_3 = 3**
  - $T_{\text{max}} = 900^\circ C$

- **HMTA/Fe(NO_3)_3 = 5**
  - $T_{\text{max}} = 640^\circ C$

Calculated adiabatic temperature and distribution of equilibrium solid products

**Ni(NO_3)_2 + Glycine (C_2H_5NO_2)**

- **Temperature, K**
  - **HMTA**
  - **Glycine**

- **Fuel to oxidizer ratio**
  - (1) - Glycine, $\phi = 0.75$, $E = 107\pm13$ kJ/mol
  - (2) - Glycine, $\phi = 1.25$, $E = 54\pm8$ kJ/mol
  - (3) - HMTA, $\phi = 0.75$, $E = 140\pm18$ kJ/mol
  - (4) - HMTA, $\phi = 2.25$, $E = 110\pm25$ kJ/mol

- **Ln(νT)** vs. **Reciprocal temperature, K**
  - (1) - (2)

*Inorganic chemistry* 58 (9), 5583-5592 (2019)

*Article in preparation*
Second Step: SCS in $\text{UO}_2(\text{NO}_3)_2 \cdot n\text{H}_2\text{O} + \text{C}_2\text{H}_5\text{NO}_2$ system

**Calculated Combustion Temp**

**Calculated Product Composition**

**Ratio of glycine /Uranium Nitrite**

*Poster by Jordan Roach*

*Article in preparation*
Characteristics of uranium oxides

PXRD of Glycine:Uranyl Nitrate Combustion Products

20-50nm

SEM

20-50nm
Mechanism and Kinetics of UO$_2$(NO$_3$)$_2$·nH$_2$O + C$_2$H$_5$NO$_2$ (glycine) reaction

\[
\begin{align*}
\text{[\(UO_2\)\(_3\)(Gly)\(_2\)O\(_3\)(H_2O)\(_3\)NO}_3]
\end{align*}
\]

\[
P_2_1_2_1
\]

\[
a = 10.059(7) \, \text{Å} \\
b = 12.386(8) \, \text{Å} \\
c = 16.024(11) \, \text{Å} \\
\alpha = \beta = \gamma = 90^\circ
\]

\[
V = 1997(2) \, \text{Å}^3
\]

\[
Z = 4
\]

\[
R_1 = 4.56\%
\]

\[
wR_2 = 12.61\%
\]

Raman Spectra of Glycine-Uranil Nitrate Solutions with Increasing Molar Ratios

DSC Plots for Combustion Samples at Increasing Heating Rates

Peaks 1, 2, 3
Target preparation: Spin Coating of Reactive Solutions

1. Reactive solution
2. Reactive gel
3. Heat treatment
4. Actinide layer

Poster by Ashabari Majumdar
Target Characterization

XRF imaging

Cross-sectional SEM

RBS

α-particle spectroscopy

Preliminary
Target Characterization: TEM and EDS analysis

- Uranium oxide
- Aluminum

EDS analysis results:
- Al Kα1
- O Kα1
- U Lα1
Heating of Al sheets at 300°C before spinning

Heating time

30 min

60 min

90 min
Target preparation: Electrospraying of reactive solutions

- No surface treatment substrate
- Plasma and ozone treatment
- Heat and ozone treatment

Poster by Stefania Dede
Target preparation: Other Approaches

Dissolution – crystallization of uranium oxides.
Drop casting or spraying methods for thin target fabrication.

Synthesis of $\text{U}_3\text{O}_8$

Method 1:
Solution combustion of uranyl nitrate + glycine solutions

Method 2:
High-temperature calcination (HTC) of uranyl nitrate

Processing

- sonication of aqueous suspensions of $\text{U}_3\text{O}_8$ allowed to rest for 20 min (A and B).
- drop casting of suspended materials on Al (C)
- evaporation of solvent in air or humidity chamber (D)
Target Stability: Ion irradiation tests

- CeO$_2$/Si material
- Ar$^{2+}$ beam (0.8 - 1.6 MeV)
- 1·10$^{12}$ to 1·10$^{18}$ ion/cm$^2$
- Post-irradiation vacuum annealing at (25-500°C)
High-Resolution TEM Images & XPS spectra of Irradiated Targets

Non-irradiated CeO$_2$/Si (A), irradiated with $1\cdot10^{14}$ (B) and $1\cdot10^{16}$ ion/cm$^2$ (C) fluences, and annealed (D) after irradiation ($1\cdot10^{16}$ ion/cm$^2$)

De-convoluted XPS spectra for CeO$_2$ thin films deposited on Si: non-irradiated (a,e); irradiated with $1\cdot10^{14}$ ion/cm$^2$ (b,f) and $1\cdot10^{16}$ ion/cm$^2$ (c,g) fluences, as well as those samples annealed in vacuum at $500^\circ$C (d,h) after irradiation ($1\cdot10^{16}$ ion/cm$^2$).

Article is accepted (Journal of Chemical Physics)
Next steps

- Continue research in bulk combustion synthesis between oxidizers and organic compounds to tailor reactive solutions for target preparation.
- Continue studies of uranium oxide thin film deposition from reactive solutions on different backings using spin coating and electrospray methods.
- Investigate electro-spraying techniques with actinide-oxide clusters.
- Perform in-depth characterization of targets and test them after irradiation.
- Continue dissemination of results, facilitate the modernization of targetry, and target preparation.
• New instrumentation on combustion synthesis, spin coating, electrospray deposition methods has been developed.
• The students trained on a large variety of chemistry, materials science and nuclear physics techniques.
• The first milestone has been achieved. Essential information on the dynamics and kinetics of combustion synthesis reactions for depleted uranium oxides have been obtained.
• A large number of targets have been prepared and characterized by an array of spectroscopic, microscopic, and ion-beam analysis methods.