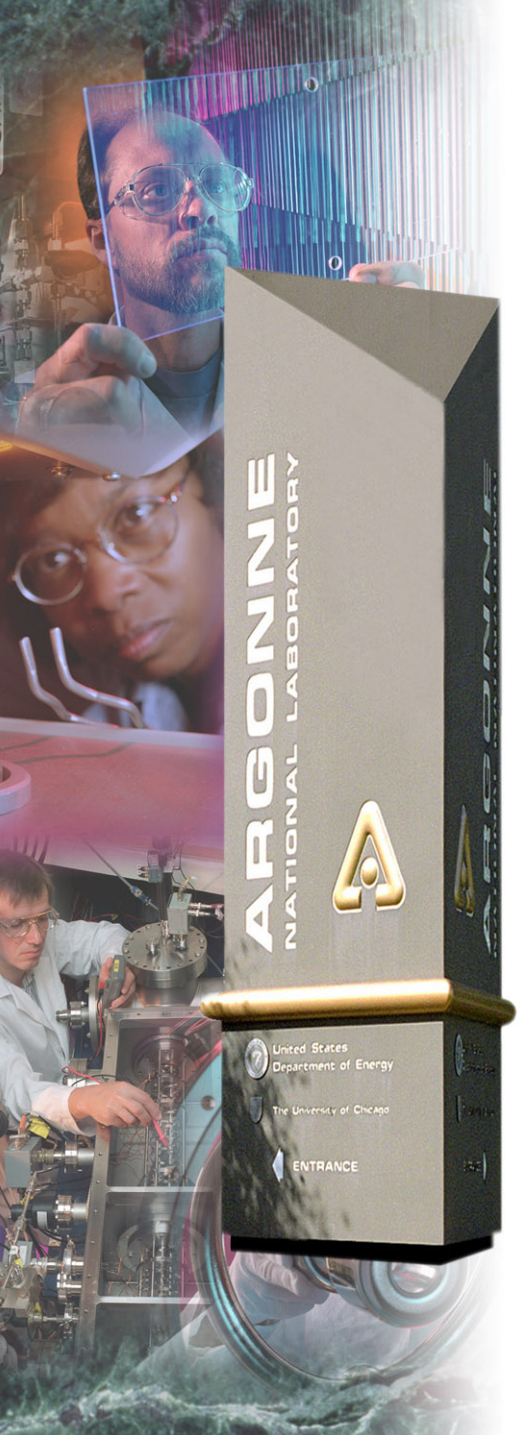

High Power ISOL Target Development – R&D Report

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(paper 3.2.1)

Physics Division and †Energy Technology Division
Argonne National Laboratory

Office of Science
U.S. Department of Energy

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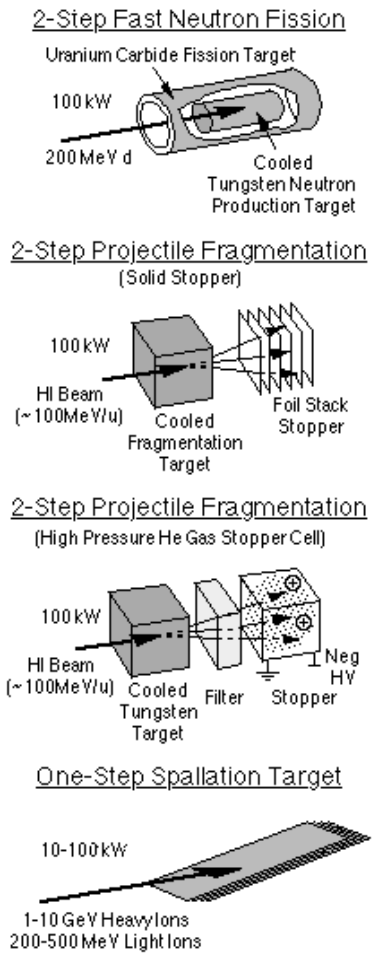
Office of Science Laboratory
Operated by The University of Chicago

Introduction

- **Introduction / Current Target Design**
  - two-step target / uranium carbide secondary target
  - UC_2 characterization / thermal simulations
- **Thermal Conductivity Measurements**
  - sample preparation / UC_2 production
  - experimental method / recent results
  - thermal evaluation of the method
- **Additional Thermal Analysis - Sintering**
  - independent analysis - ORNL
- **Release Studies - UNISOR**
- **Alternate Approaches**
  - spray coatings
  - Atomic Layer Deposition
- **What Still Needs to be Done**

Introduction

- One of the proposed production schemes for RIA will be a two-step ISOL target employing the compounds of uranium to produce ion species far from stability [1].
- An alternate design involves a tilted foil spallation target composed of uranium or a refractory metal. A substitute design would be a uranium compound coating on a metal or carbon substrate.



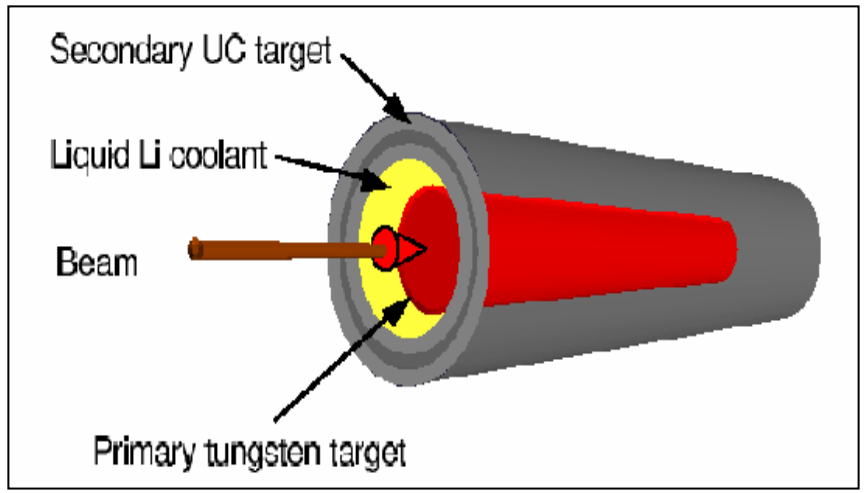
[1]Report to ATLAS Users Facility, ANL-ATLAS-99-1, March (1999)

Current Target Design – UC_2

- Why UC_2 ?
- Refractory
- Fissionable
- Favorable Density

- **Energy deposition and calculated fission rates determined that a density of 3.3 g/cm^2 or greater would reduce the volume of the secondary target (UC_2) while keeping the fission rate constant.**

- **Fission rates calculated for the secondary UC_2 target, using the prototype target design give fission rates for UC_2 material of 0.0329 fissions/proton for 2.5 g/cm^3 density, 0.0698 fissions/p for 5.0 g/cm^3 density, and 0.1077 fissions/p for 7.5 g/cm^3 density.**

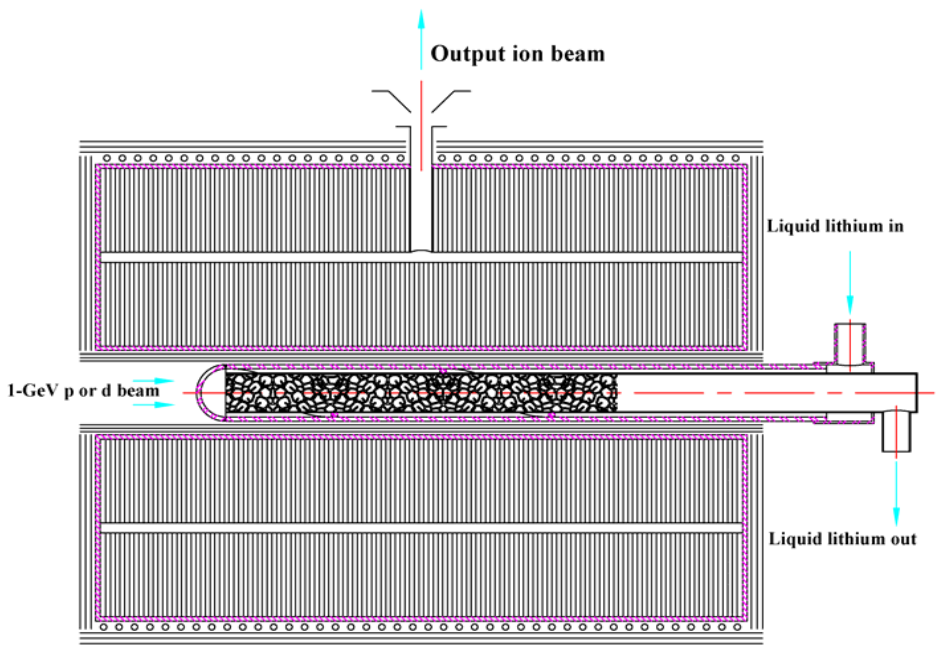


Current Target Design – UC_2

- **In our two-step target design, neutrons are first generated in a cooled, refractory, primary target which then induce fission in a surrounding assembly of uranium carbide.**

PROTOTYPE TARGET
UC_2 pressed annular disks
6-cm OD 3-cm hole
1-1.5 mm thick
100 disks

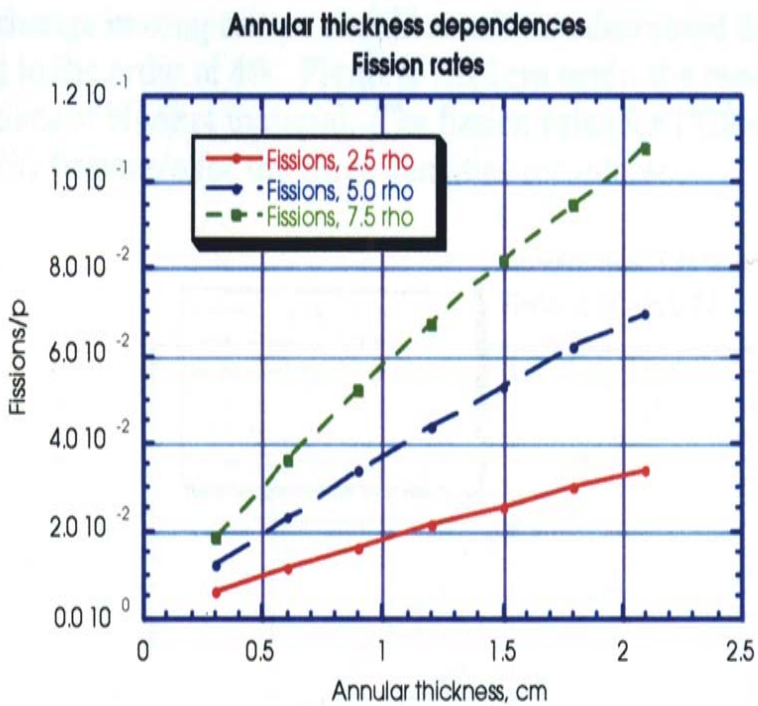
- Why Pressed Powder?
- Porosity
- Fine Grain Size
- Manufactured into Shape



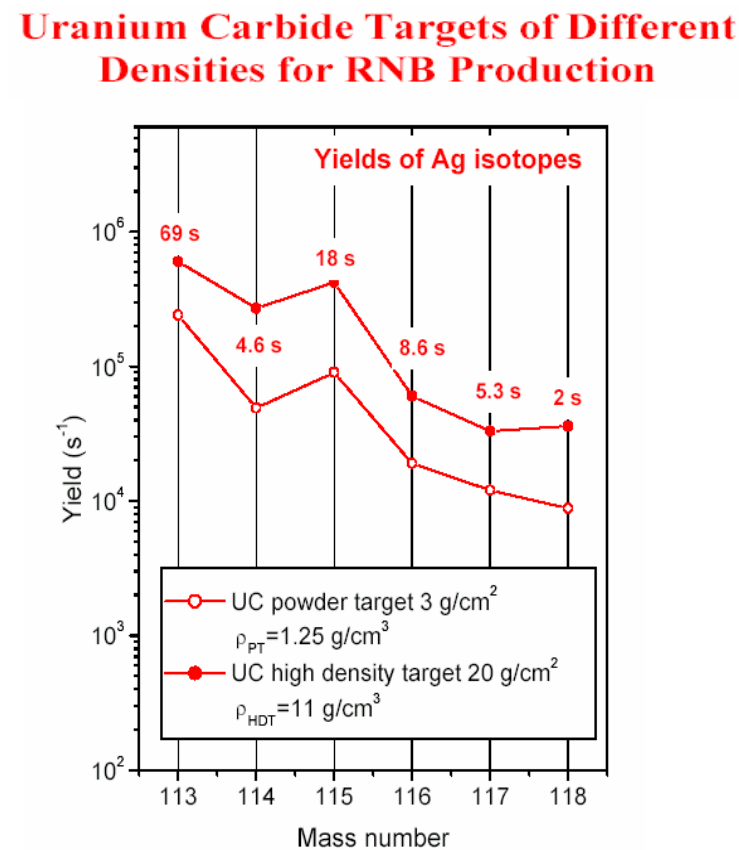
Prototype being developed by W. Talbert, et al., TechSource, Inc. (SBIR Grant).
Fine-grained, high thermal conductivity UC_2 being developed at ANL.

Investigations of Uranium Carbide Properties

- High Density → Increased Fission Rates → Better Release Properties?



Ref. W. L. Talbert and H.-H. Hsu, "Effects of composition and density of U/C material on fission rates in two-step targets"



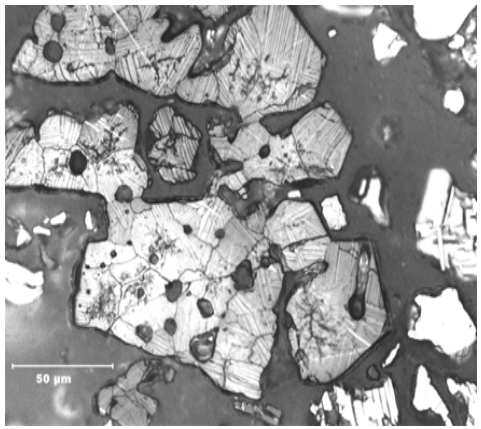
V.N. Panteleev, et. al. – PNPI, priv. comm.

Investigations of Uranium Carbide Properties

- Smaller Grain Size → Higher Density → Better Thermal Conductivity?

- **Comparison of UC_2 Powder Microstructures**

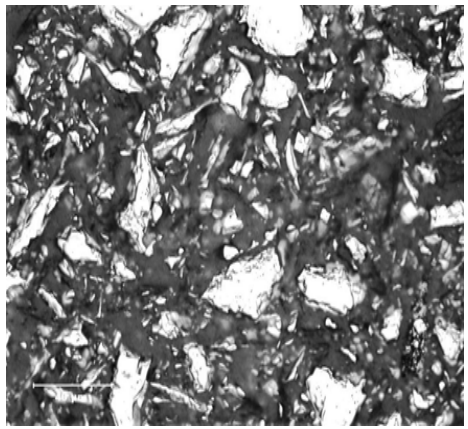
CERAC, Inc. material



Uranium Carbide powder (-60 mesh), 500x

Particle Size = 250 µm

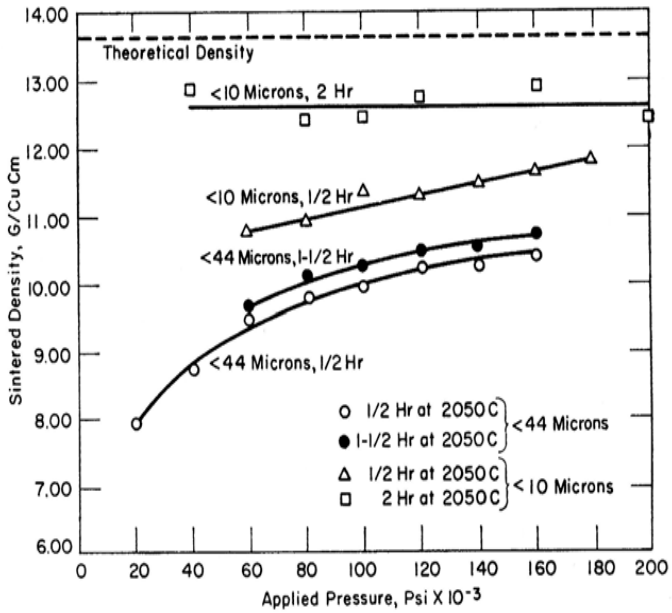
ANL material



Uranium Carbide powder (-325 mesh), x500

Particle Size = 44 µm

Density of uranium carbide as a function of grain size and compacting pressure.

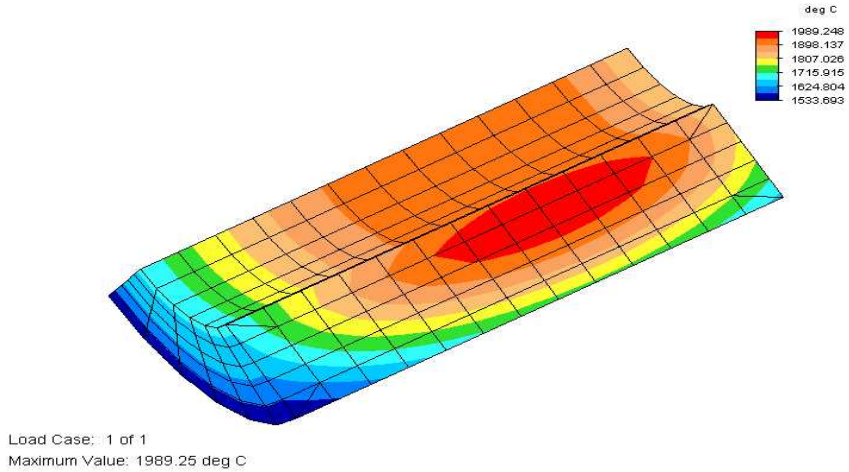


Ref. Ceramic Fuel Elements, Robert B. Holden, Gordon & Breach, New York 1966

Thermal Conductivity – Target Simulations

- Higher Thermal Conductivity → Uniform Heating → Better Fission Release?

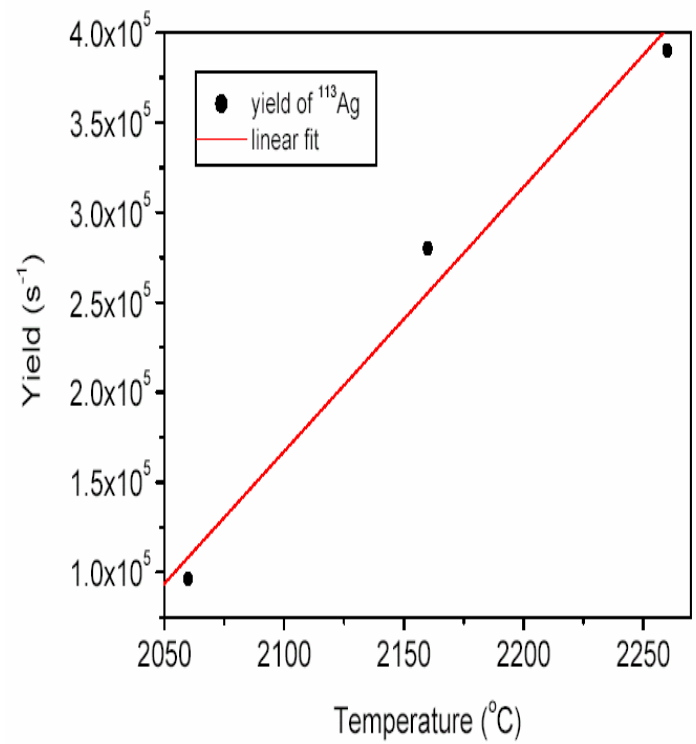
Temperature profiles for a target including heat shields



SBIR Memo #35 D. Drake & W. Talbert, 12 Feb. 2003

- **The small energy deposition rate in the surrounding UC_2 secondary target material is not sufficient to heat the target to the desired operating conditions necessary to promote effective release of fission products and therefore prompted the introduction of heat shielding into the secondary target design.**

Temperature Dependence of the Yield of ^{113}Ag



V.N. Panteleev, et. al. – PNPI, priv. comm.

Thermal Conductivity Measurements

- **For a thermally isolated disk with no internal heat sources, the steady-state rate of heat flow (q) into one face of the disk equals that out of the other face:**

$q/A = -k\,\Delta T/\Delta x$ (Eq. 1)

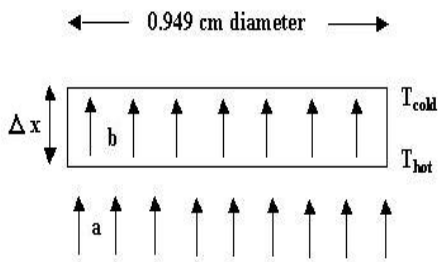
- **where**

A = area of the disk,

ΔT = temperature difference between the faces,

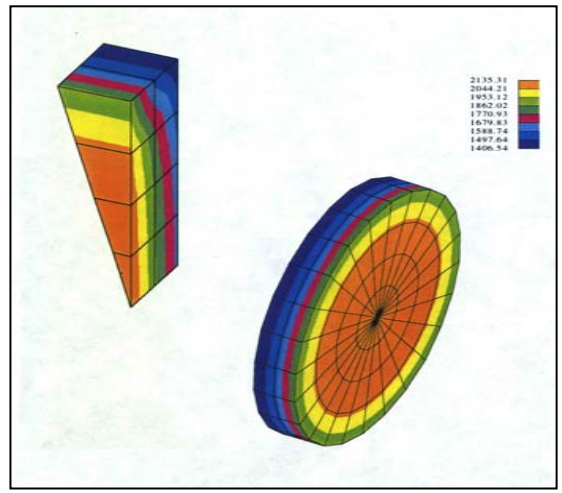
Δx = thickness of the sample,

k = thermal conductivity.



a) Electron beam heats lower side of the sample
b) Heat flux through the sample = heat radiated from upper side

Simulation of the temperature contour in the UC_x sample pellet using TAS-TRASYS.



Ref. M. Petra –ANL, priv. comm.

- **For a viable secondary target design containing internal heat shields surrounding the primary target as well as external shielding, thermal conductivities on the order of 2 W/m-K (or greater) over the temperature range 1600 to 2100 C are required.**

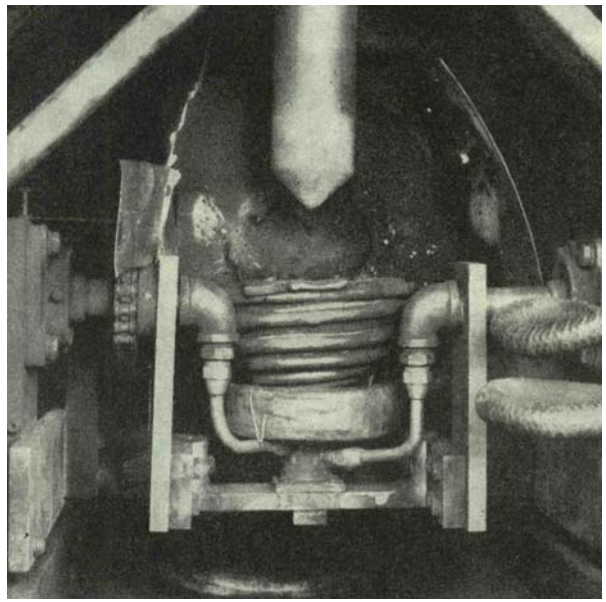
We need thin, pressed UC_2 pellets!

Uranium Carbide Manufacturing

- We have begun in-house preparation (ANL-E & ANL-W) of this material by the method of arc melting uranium metal combined with an excess of solid carbon [2]. Sample preparation using various carbon and/or graphite starting materials is being investigated.

- As grain size will prove to play an important role in ultimate densities and thermal conductivities achieved, this prepared material can be further characterized using sieves and new pellets pressed with enhanced properties. The desired release of the fission products produced under sample irradiation also needs to be explored as a function of density/grain size. Research into this aspect of our UC_2 samples as a step toward a RIA target is just now beginning.

Close-up view of arc melting furnace interior showing electrode tip, water-cooled crucible, and deflection coil at base of crucible.



Ref. Ceramic Fuel Elements, Robert B. Holden, Gordon & Breach, New York 1966

- An initial run at ANL-E produced a 25 g ingot which was crushed under a nitrogen atmosphere using a tool steel mortar & pestle to yield 20 g of powder (-325 mesh), grain size 44 μm. Investigations will begin employing samples prepared with this material.

[2] J. Crane, F.B. Litton and H.S. Kalish, *ASM Trans. Quarterly* 56 (1963) 176.

Sample Preparation Procedure

- The thermal conductivity measurement sample disks are prepared in a laboratory hood by first weighing out the UC_2 powder (-60 mesh) together with carbon (ratio of 8:1) in the form of high-purity synthetic graphite powder (-200 mesh).
- A small amount of albumin is added as a binder to the weighed material.
- The mixture is poured into a 10 mm compaction die and pressed to 5 tons (10,000 psi) using a laboratory press.

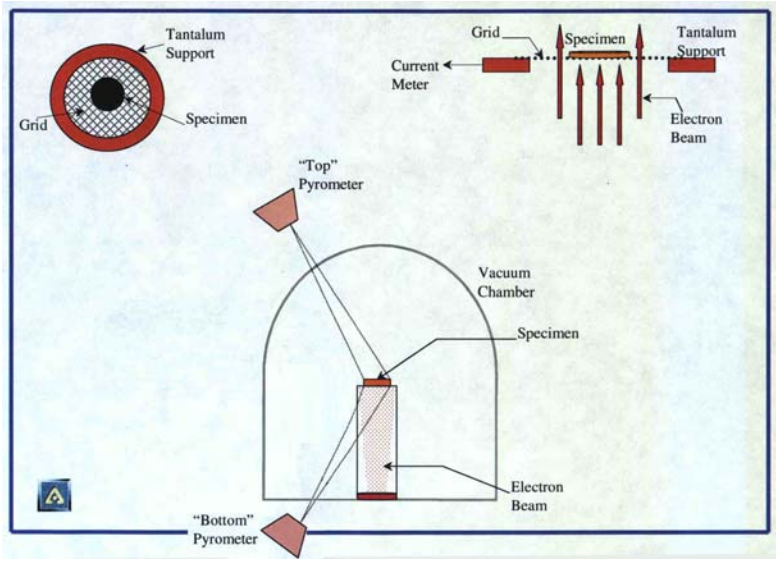
Radioactive materials hood for sample preparation.

10 mm sample die.

Carver, Inc., Model B Laboratory Press

Experimental Method

- The sample was heated on the bottom face by a vertical electron beam source installed within a vacuum evaporator. The sample was supported on a 95% transmission molybdenum mesh that was positioned central to the beam axis using a tantalum metal ring on top of a 6 in. glass tube where the beam spot size is approximately the size of the sample pellet.



Vacuum evaporator setup used for the measurements.

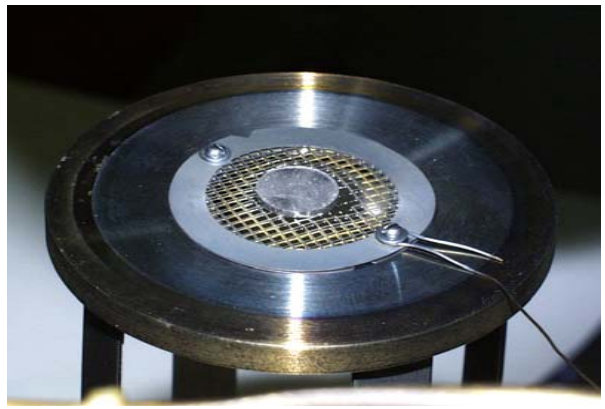
- After achieving thermal equilibrium, the temperature of both faces of the sample were measured with the aid of a two-color pyrometer first set up to measure the bottom face of the sample then re-positioned above to view the top face. For a complete measurement, the experiment consisted of the sample being irradiated twice.

Experimental Method

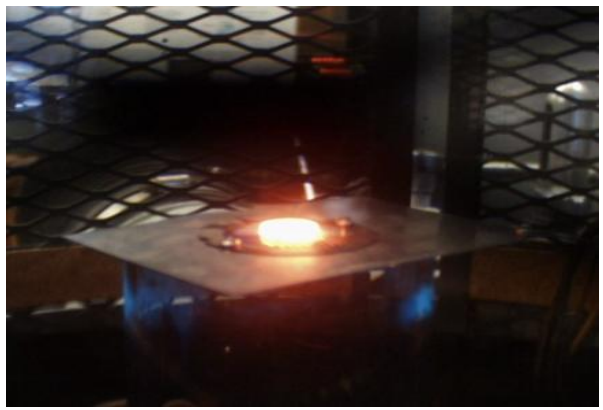
- **Sample Pellets of Uranium Carbide (10 mm ∅)**

Sample	Density (g/cc)
1	5.47
2	5.29
3	5.21
UC2 + C (8:1)	4.92
I (broken)	
II	4.97
III	4.78
IV	5.06
UC0130A	5.82
UC0130B	4.91
UC0130C	5.70
UC/C0130A	5.31
UC/C0130B	5.29
UC/C0130C	4.49
UC/0128A	5.21
UC/0128B	4.81
UC/C/ALB02/21 A	5.28
B	4.95
C	5.25
UC/C/ALB05/07 A	5.39

*Samples sent to ORNL for release studies at UNISOR.



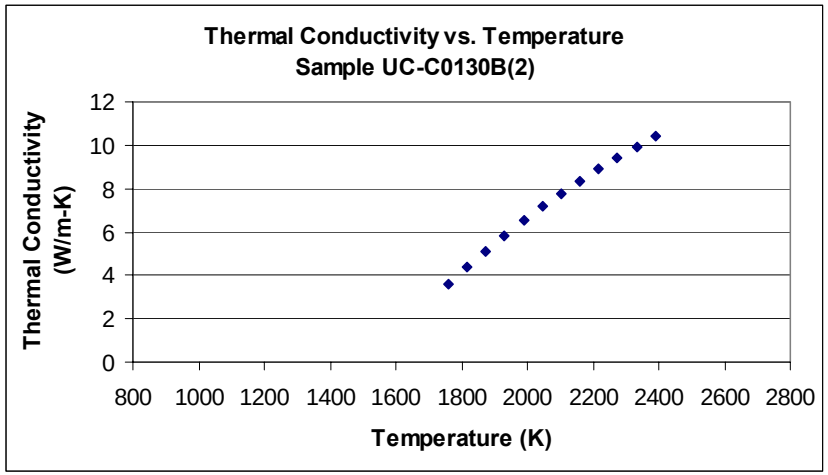
Photograph showing the uranium carbide disk above the electron beam source. Note the wire pick-up used to measure the electron beam current.

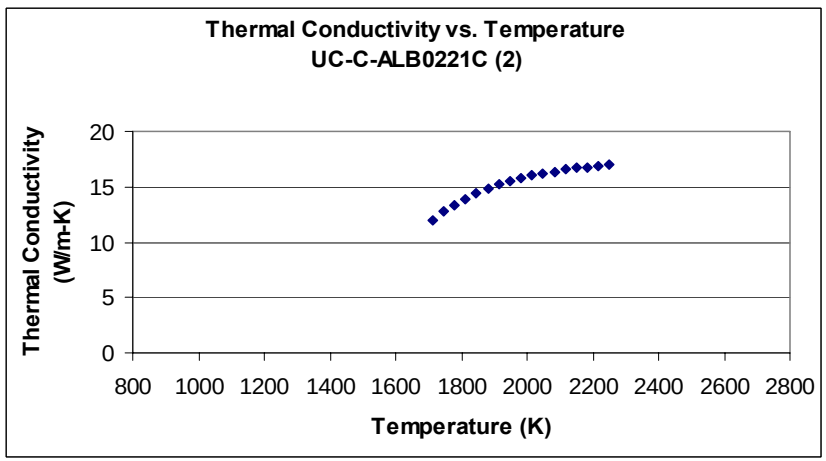


Photograph of the setup used to measure the thermal conductivity of UC_2. Shown is a 3/8" diameter UC_2 disk at approximately 1900°C supported on a Mo grid and being heated from below by the electron beam.

Initial Results and Comparison to Previous Work

Thermal conductivity of two recent UC_2 sample pellets



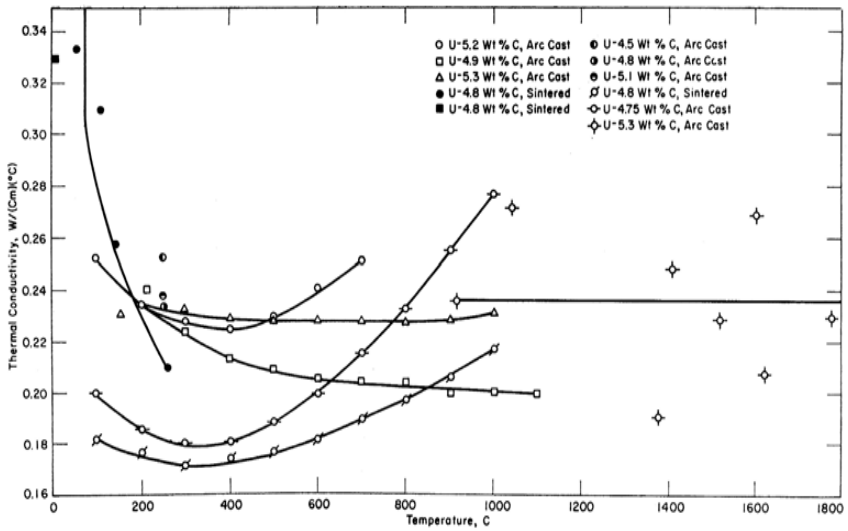


Previous work (ANL) on reduced UC_x.

Thermal Conductivity (k) Calculations	
Temperature (C)	Thermal Conductivity (W/cm K)
23	0.00414
100	0.00396
200	0.00355
300	0.00316

J. Greene, *et. al.*, INTDS Newsletter June (1997)

Thermal conductivity of uranium carbide reactor fuel pellets.

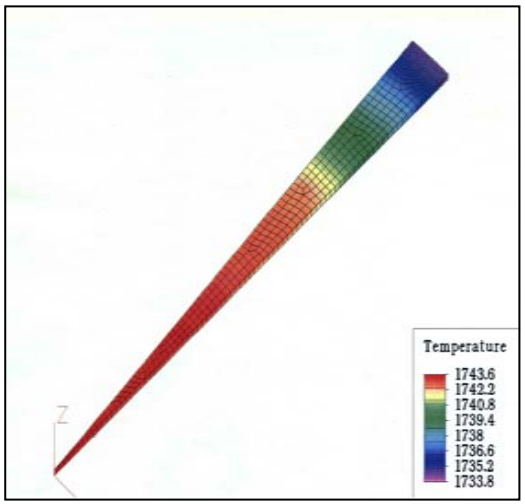


Ref. Ceramic Fuel Elements, Robert B. Holden, Gordon & Breach, New York 1966

Thermal Simulations –Evaluation of the Method

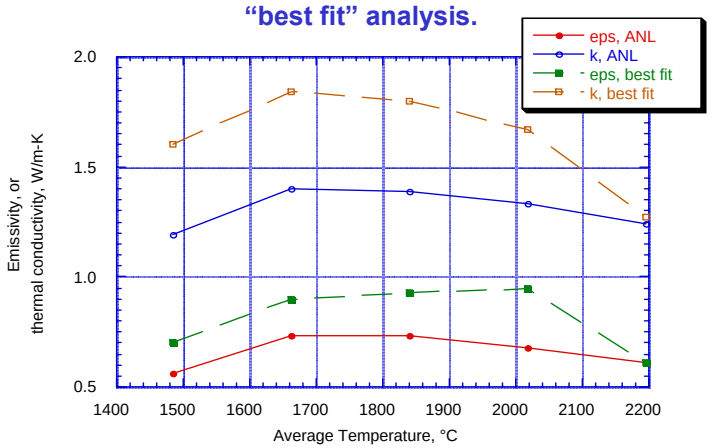
- **The method is still being refined, checked for accuracy and overall degree of reproducibility. The results for reduced UC_x samples prepared by the ISOLDE prescription give densities on the order of 3 g/cc with thermal conductivities of 1-2 W/m-K and emissivities of 0.7-0.8 in agreement with calculated thermal properties. Densities and thermal conductivities for the more recent UC_2 samples exhibit improved properties (k = 4-16 W/m-K), (ε = 0.4-0.5). This work is still in progress.**

Temperature distribution through a slice of the pellet.

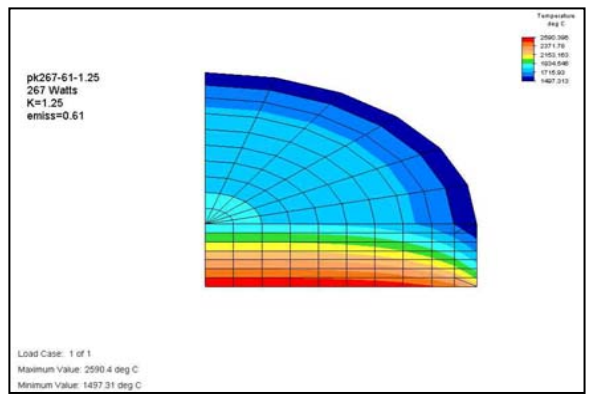


Ref. A. Levand – ANL, priv. comm.

Comparison of the thermal parameters of ANL results and "best fit" analysis.



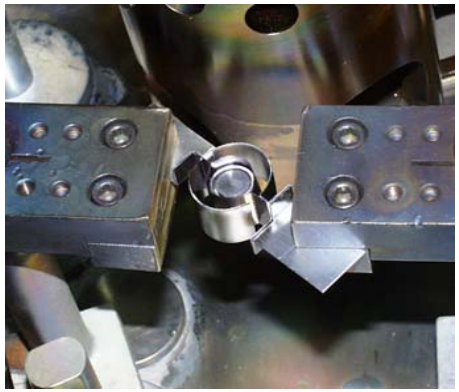
Evaluation of Argonne thermal conductivity measurement approach. Isothermal distribution for uniform beam profile.



Ref. Darrell Drake and Willard Talbert, SBIR Memo #47,

Thermal Analysis - Sintering

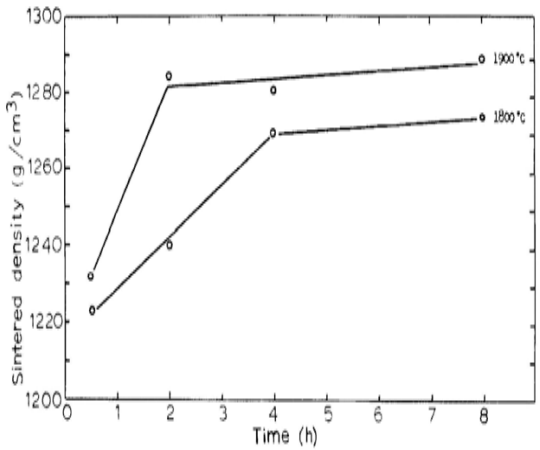
What happens at extended high temperatures?



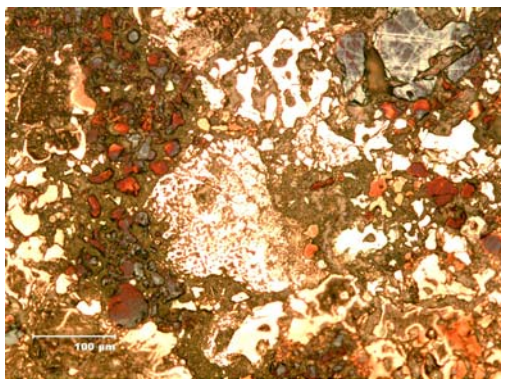
Sample for sintering within vacuum evaporator.

Sample being sintered in Ta crucible under vacuum.

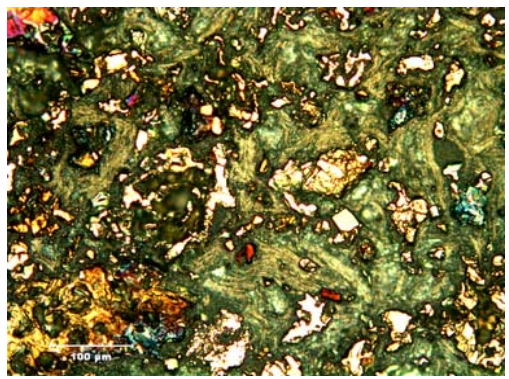
Density vs. Sintering Time



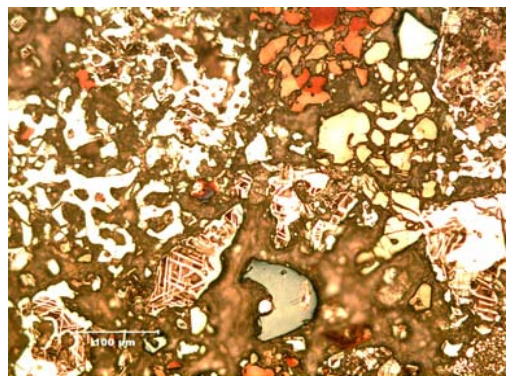
Ref. M.H. Rand and O. Kubaschewski, (Harwell) Report AERE-R 3487



Before Annealing



Annealing at 1100^{0}C



Annealing at 2000^{0}C

- **Microstructure of UC_2 samples prepared from powder (-60 mesh), graphite powder and albumin binder. (200x)**

Additional Thermal Analysis

- **Sample Pellets of Uranium Carbide (13 mm ∅)**

Sample	Density (g/cc)
UC/C/ACB05/07 A	5.12
B	5.14
UC/C/ACB01/28	4.96
A	4.88
B	5.08
UC/C/ACB03/05	5.69
A	4.53
B	5.53
UC/C/ACB03/05	4.98
A	5.31
B	5.05

Synthetic Graphite Binder

Sample	Density (g/cc)
UC/CA/ALB05.29 A	2.85
B	2.85
C	3.14
UC/CA/ALB05.29 A	3.14
B	2.88
C	2.37
UC/CA/ALB05.29 A	3.26
B	2.91
C	3.06
UC/CA/ALB05.29 A	2.67
B	3.16
C	3.37

Activated Carbon Binder

- We need access to:
- Vacuum Furnace
- Larger Press

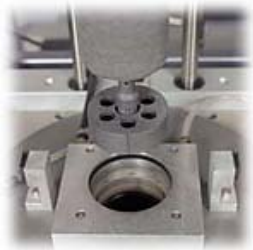
*Samples sent to ORNL for an independent thermal analysis.

Additional Thermal Analysis - ORNL

Thermophysical Properties User Center

Laser Flash Thermal Diffusivity System

- Duplicate samples have been forwarded to the Thermophysical Properties Users Center at Oak Ridge National Laboratory (ORNL) for independent thermal conductivity determinations. Two sets of six sample pellets are being measured using their Laser Flash Thermal Diffusivity System. In this method, a short pulse of heat is applied to the front face of the pellet using a laser, with the temperature change of the rear face measured with an infrared detector to determine the thermal diffusivity.



6 Sample Carousel

- To determine the specific heat, a Differential Scanning Calorimeter is used to measure the thermal response of the UC_2 pellet as compared to a standard while heating uniformly at a constant rate. These measurements taken together are then used in the determination of the thermal conductivity.

Differential Scanning Calorimeter

$$k = \alpha C_p \rho \qquad \text{(Eq. 2)}$$

- where

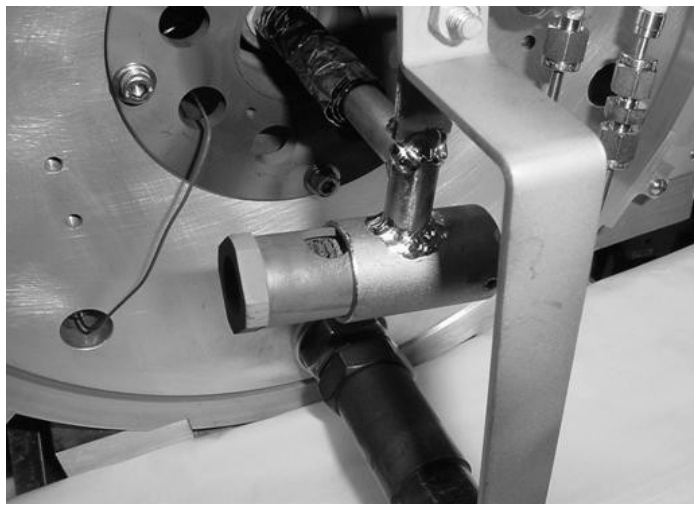
k = thermal conductivity
α = thermal diffusivity
C_p = specific heat
ρ = bulk density

Ref. G. Alton & R. Dinwiddie – ORNL, priv. comm.

Release Studies at UNISOR

- The previously shipped samples have been baked out for several hours at 1950 C to remove contaminants.
- Measurements of the release of radioactive ions from these pellets will be performed in mid to late September after completion of the modifications to the UNISOR beam line.

Photo of the UC targets in graphite holder



D. Stracener – ORNL, priv. comm.

- Sample Pellets Reduced From the Oxide

Sample	Density (g/cc)
1	
2	
3	2.26
4	2.85
5	2.72
6	2.84
7	-
8	2.72
9	2.76
10	2.93
11	3.04
12	2.99
A	3.01
B	3.14
C	2.99
D	2.94
E	3.19
F	2.95
G	-
H	3.15
I	3.3
J	3.3

*Samples sent to ORNL for a release studies at UNISOR.

Multi-Foil Target Approach – Spray Painting

- **In addition to using the properties of metal foils for ISOL targets, coatings with enhanced properties may be applied to the foils to improve production of rare ion species using various techniques.**

TRIUMF

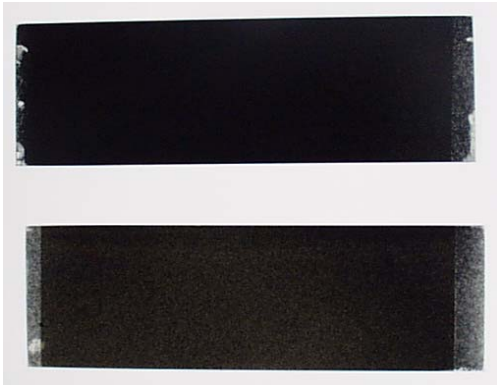
- **Collaborating with Alton at ORNL with UC_2 painting technique**

- **SiC #4 20.4 g SiC/cm^2:**

In place from November 5, 2002 – Dec 20, 2002
425 composite foils of SiC deposited on exfoliated graphite sheet.
Graphite thickness: 0.13 mm; presintered SiC thickness: 0.23 mm.
Length of foils stack: 18.6 cm.
Received 19397 uA-hrs of proton beam = 4.4 x 10^{20} protons.
45 uA maximum beam current.
On January 16, 2003 a ^{22}Na beam from the target was used to
produce a 4 x 10^4 Bq implanted source.

- **ZrC #1 37.6 g ZrC/cm^2:**

In place from March 12, 2003 – April 27, 2003
435 composite foils of ZrC deposited on exfoliated graphite sheet.
Graphite thickness: 0.13 mm; presintered ZrC thickness: 0.24 mm.
Length of foils stack: 18.6 cm.
Received 17556 uA-hrs of proton beam = 4.0 x 10^{20} protons.
50 uA maximum beam current.

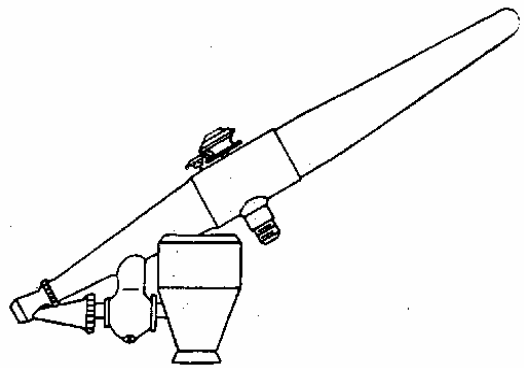


MoS_2 coatings on carbon-coated microscope slides applied using the spray-coating technique

MoS_2 GAMMASPHERE Target Wheel

paasche

H# and HS# Airbrushes

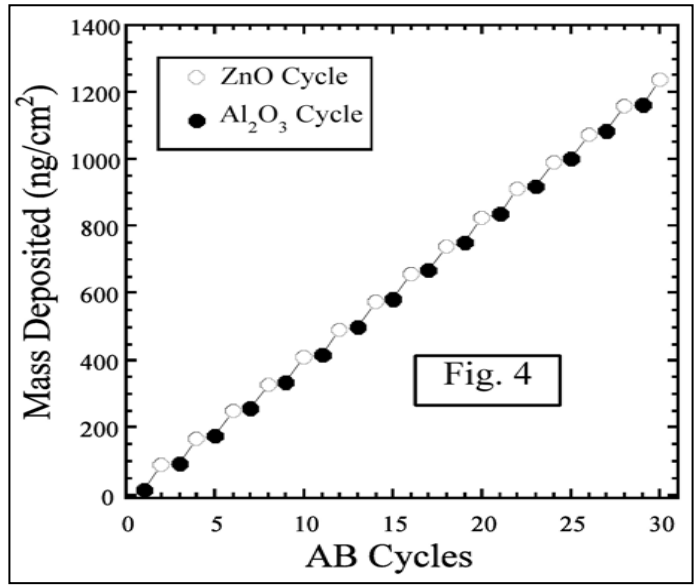


http://www.triumf.ca/people/marik/Targets.html

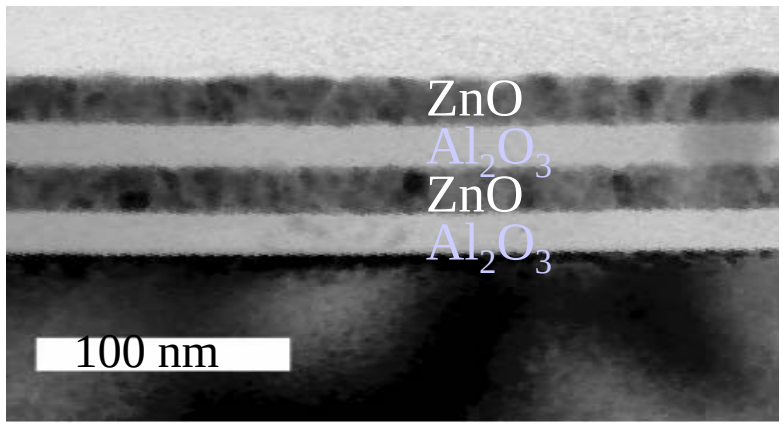
J. Greene & C. Lister, NIM A480 (2002) 79-83

Atomic Layer Deposition (ALD)

- **Working with the Material Science Division on a new deposition technique employing Chemical Vapor Deposition (CVD) onto metal (or foam) substrates using Atomic Layer Deposition – ALD.**
- **In this method monolayer films are grown using alternating reactive gas phases in a small furnace under computer control.**
- **In theory the process can be applied to uranium compounds – UO_2 but more promising with UN.**



M.Pellin –ANL, priv. comm.



ALTERNATE TARGET APPROACH
12 micron Ta foil substrates
12 micron UC coating (each side)
12 micron spacing
2000 layers

What Still Needs to Be Done

- **Uranium Carbide Pellets**
  - new powder characterization
  - more pellets, additional binders (access to larger press!)
  - scale up to secondary target annular disks (need dies)
- **Thermal Conductivity Measurements**
  - continue and refine the measurements
  - new values → thermal simulations → improve design
- **Sintering** (access to vacuum furnace!)
- **Thermal Analysis** (waiting to hear from ORNL?)
- **Release Studies** (soon!)
- **Spray Coatings** (many ideas to try)
- **Atomic Layer Deposition?**
  - chemical analogue to uranium
  - build a dedicated reactor system ($100k)