Nuclear Fuel Activities

Current Research Activities

Spark Plasma Sintering of Oxide, Nitrides and Refractory Metal Fuels-LDRD
W. Windes, D. Burkes, S. Howe, M. Frary, D. Butt, B. Pesic, T. Hartman…

Synthesis of Actinide Nitrides-DOE NERI
D. Butt, B. Jaques

Modeling of Vaporization of Carbide Fuels in Hydrogen
D. Butt

Developing Research Activities

Advanced Radioactive Materials Processing-NSF I/UCRC
D. Butt, B. Stucker, M. Frary, R. Grosshans…

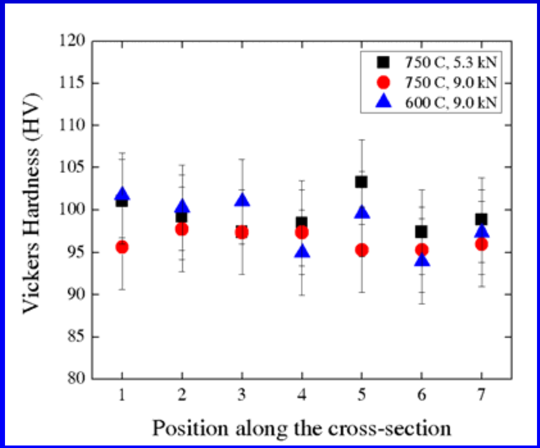
Direct Oxidation of Electrorefined Uranium
D. Butt, M. Frary, M. Simpson, …

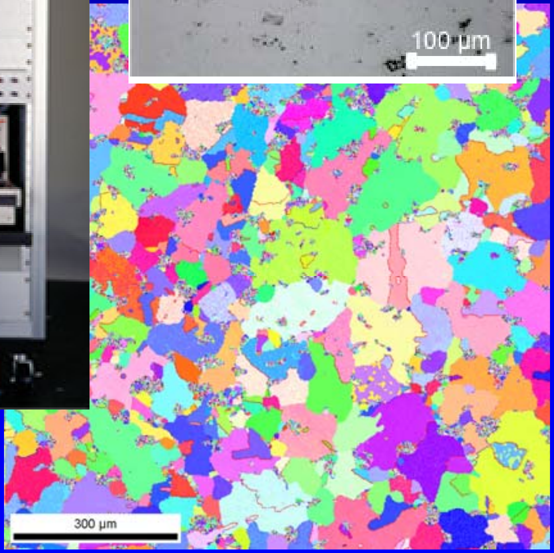
Thermodynamic Assessment of Actinide-Lanthanide-Rare Earth Metal Fuels
D. Butt, T. Hartman, R. Kennedy…

Spark Plasma Sintering

W. Windes, D. Burkes, S. Howe, M. Frary, D. Butt, B. Pesic, T. Hartman…

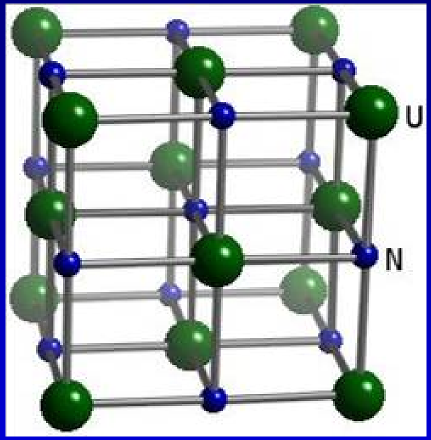
Recent Efforts:
Oxides
Nitrides
Refractory Metal Fuels
Nickel and Model Alloys





Mononitride Fuels

- Pros:
  - Candidates for 3 of 6 Gen. IV reactors
  - High actinide density
  - Lower enrichment
  - Higher thermal conductivity
    When compared to oxide fuels (X 5-10)
    - Lower “burning” temperatures
    - Decreased cladding compatibility concerns



www.webelements.com

- Cons:
  - Difficult to handle (Susceptible to oxidize)
  - Difficult to synthesize
  - Potentially large process footpring
  - Transmutation of ^{14}N to ^{14}C
    (a long-lived radiotoxic isotope)

http://www.gnep.energy.gov/

BOISE STATE UNIVERSITY

[1] S. T. Voit, *et al*. LANL, 2006. [2] S. P. Willson, *et al*. Trans. Amer. Nucl. Soc. 2004. **91** [3] R. Margevicius, *et al*. LANL, 2001.

Mononitride Fuels

Candidate non-fertile and low-fertile fuels considered for the AFCI

1 - **Non-fertile In[illegible]**
($Pu_{0.50}$, Am

2 - **Low-Fer[illegible]**
($U_{0.50}$, $Pu_{0.25}$

Non-Fertile Fuels [1-3]
$Pu_{0.48}$, $Am_{0.12}$, $Zr_{0.40}$
$(Pu_{0.50}, Am_{0.50})N$ + 36-wt% ZrN*
$(Pu_{0.20}, Am_{0.80})O_2$ + 65[illegible]% MgO
$(Pu_{0.50}, Am_{0.50})O_2$ + 70-vol% MgO
$(Pu_{0.23}, Am_{0.25}, Zr_{0.52})O_2$ + 60-vol% Mo^{92}
$(Pu_{0.50}, Am_{0.50})O_2$ + 60-vol% Mo^{92}
Low-Fertile Fuels [1-3]
$U_{0.35}$, $Pu_{0.29}$, $Am_{0.04}$, [illegible]$_{0.02}$, $Zr_{0.30}$
$(U_{0.50}, Pu_{0.25}, Am_{0.15}, Np_{0.10})N$*

Example Systems

Actual Fuel = Surrogate Fuel

Inert Matrix Fuel (IMF)

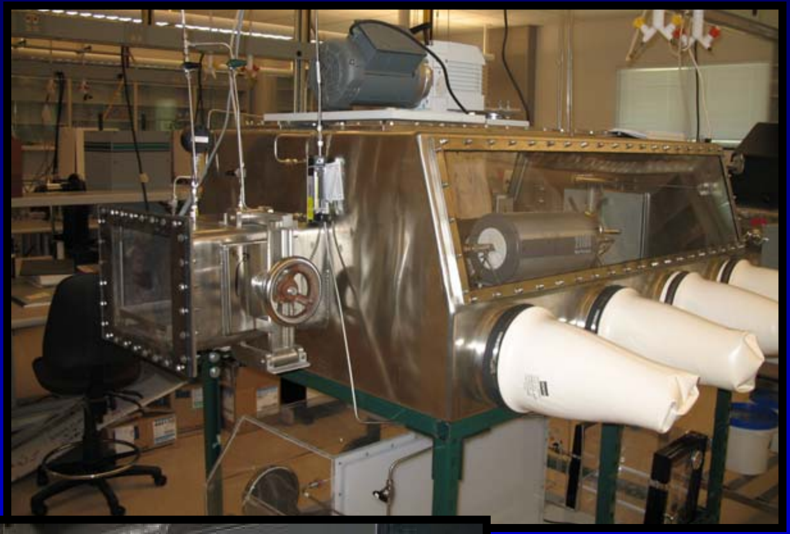
$(Pu_{0.50}, Am_{0.50})N$ + 36wt%-ZrN = $(Ce_{0.50}, Dy_{0.50})N$ + 36wt%-ZrN

Nitride Fuel

$(U_{0.50}, Pu_{0.25}, Am_{0.15}, Np_{0.10})N$ = $(U_{0.50}, Ce_{0.35}, Dy_{0.15})N$

*Based on the comparisons of;
- Melting Point,
- Thermal Conductivity
- Thermal Expansion Coefficient
- Theoretical Density
- Crystal Structure

Experimental: Powder Handling



Inert atmosphere glove-boxes for material handling and processing

Experimental: Controlled Atmosphere, Vacuum Tube Furnace

Hood for ammonia or hydrogen processing gases

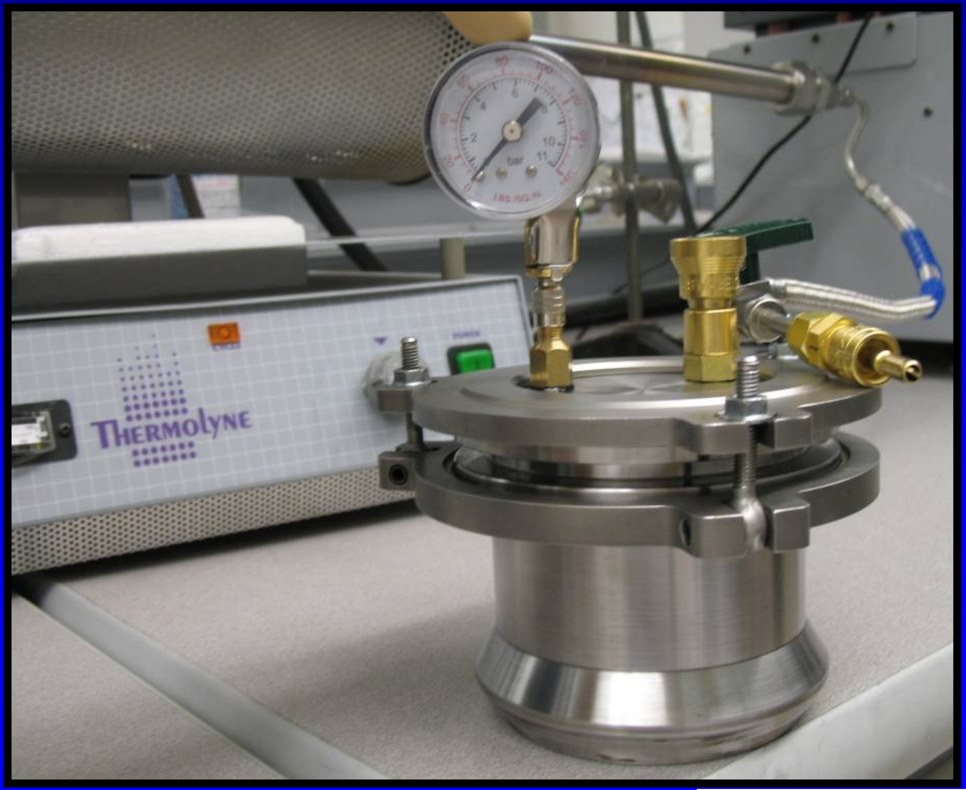
Turbo molecular pump for low pressure apps. ($5*10^{-6}$ Torr)

High temperature (1750°C) tube furnace for synthesis and sintering nitride materials

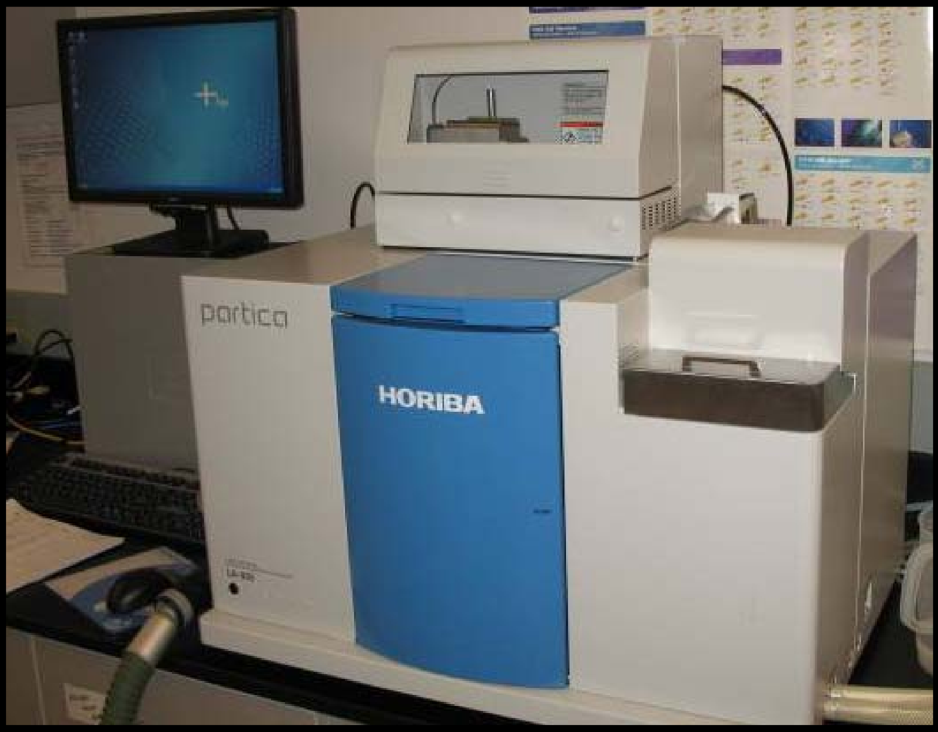
Experimental: Atmosphere Controlled Planatary Ball Mill

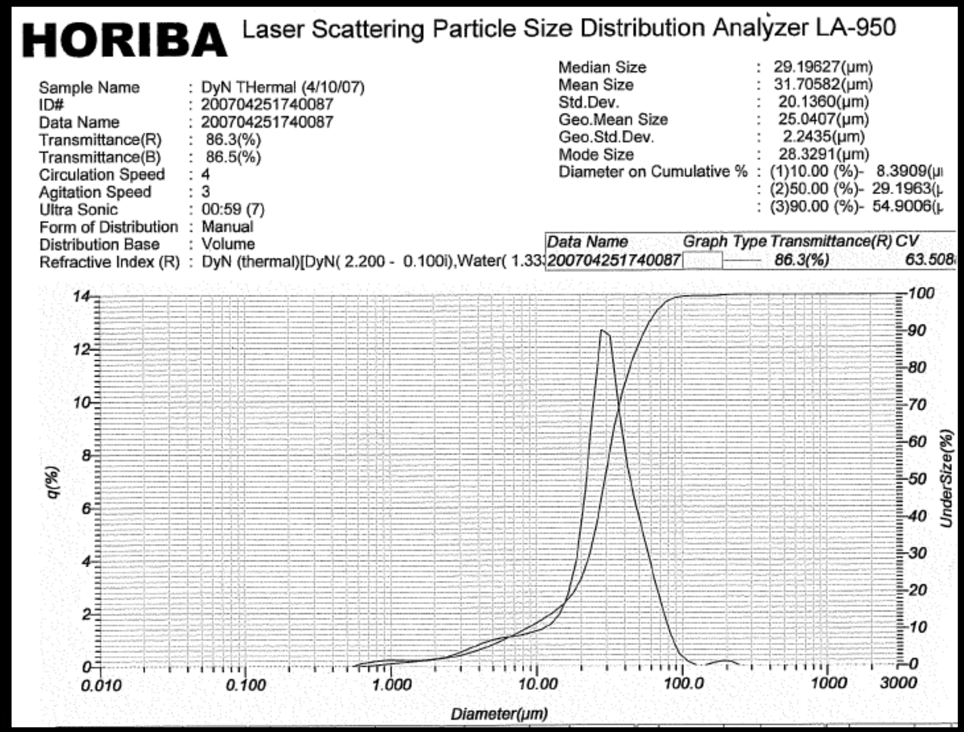
Planetary ball mill

Controlled atmosphere vessels (allows for <0.01 ppb O_2)



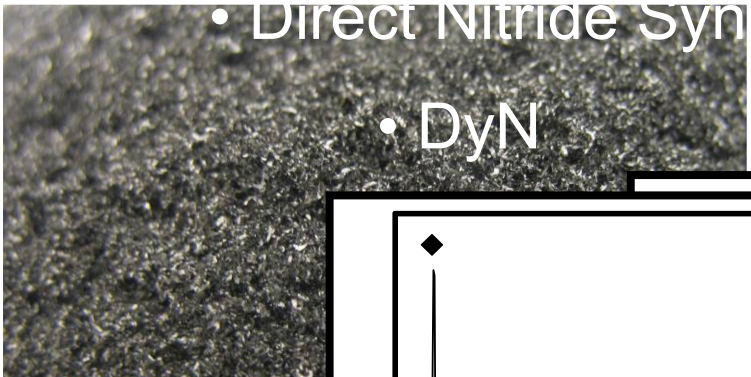
Experimental: Laser Scattering Particle Size Analysis



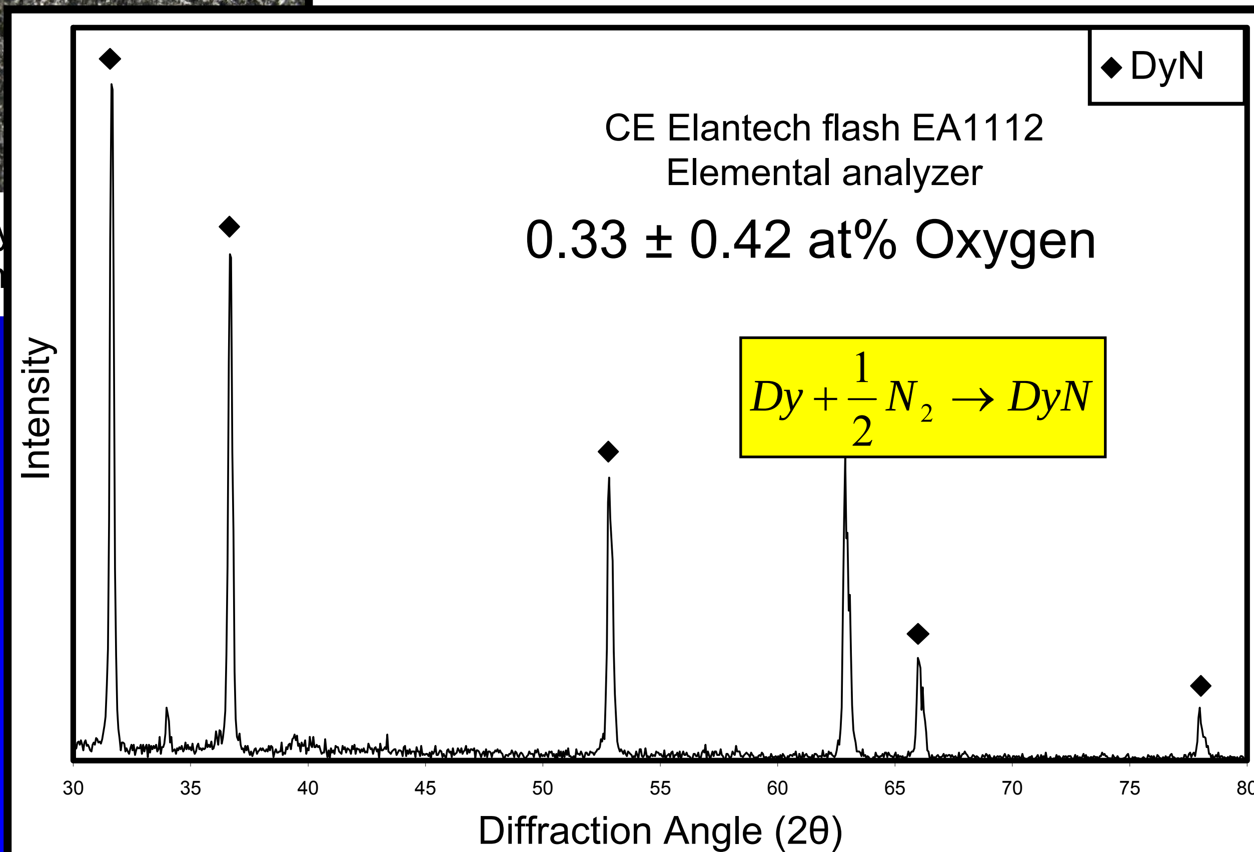


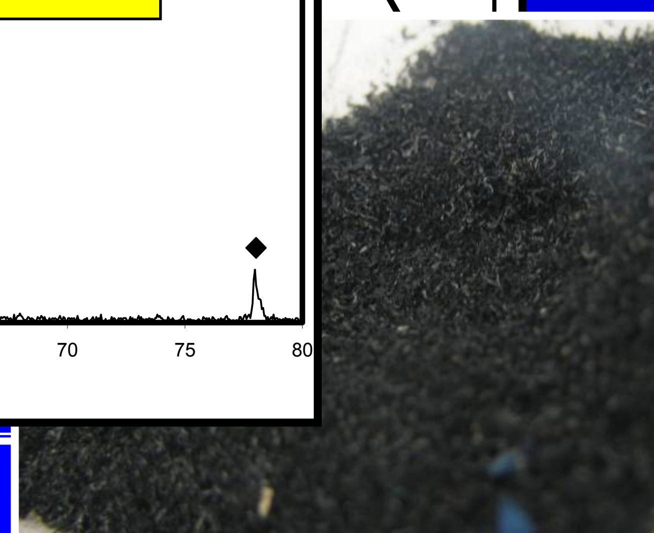
Nitride Synthesis Methods

- Direct Nitride Synthesis Route
  - DyN



10 g Dy
(ESPI, -40 n

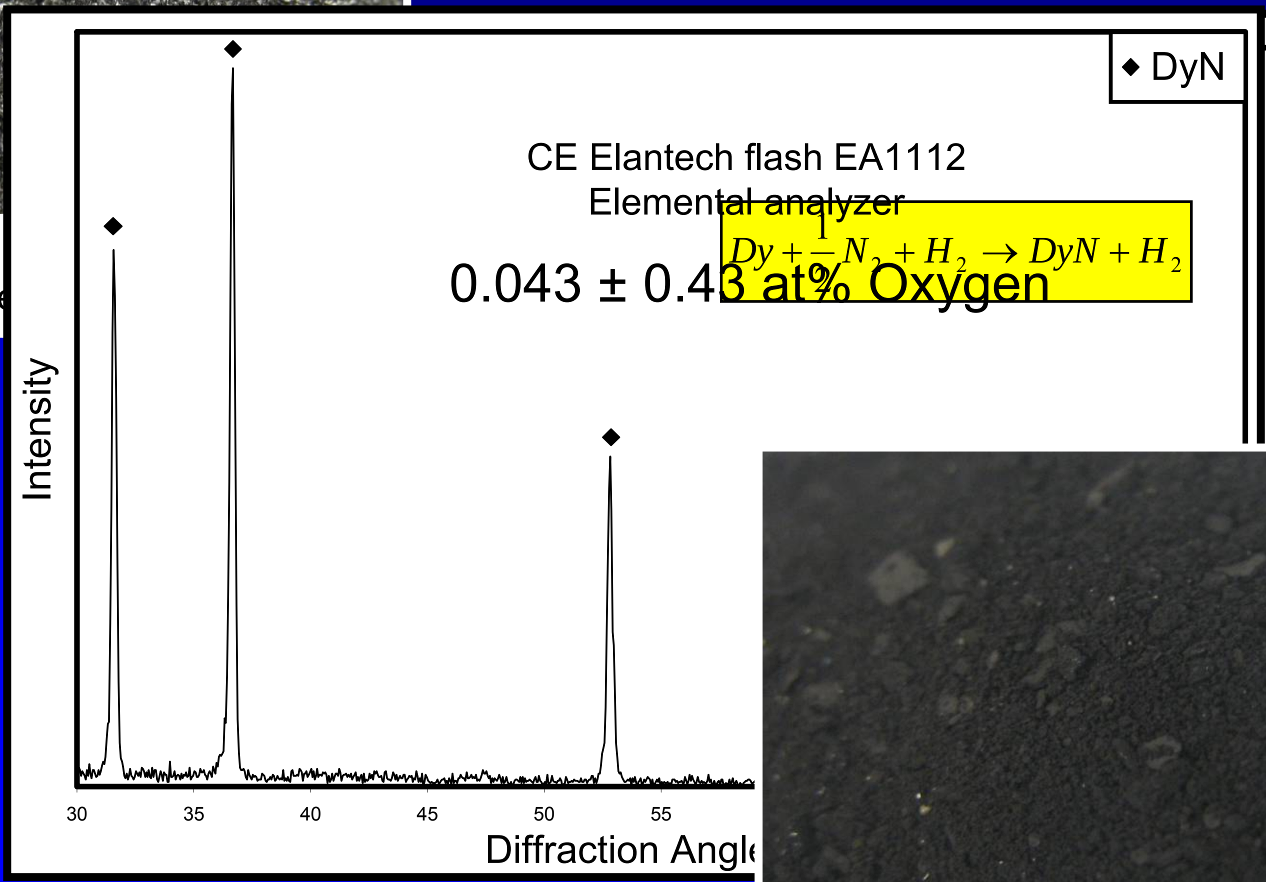




Nitride Synthesis Methods

- Hydride-Nitride Synthesis Route
  - DyN

10 g Dy
(ESPI, -40 me



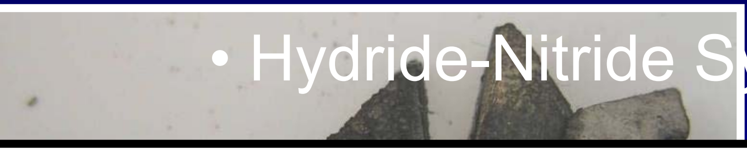
CE Elantech flash EA1112
Elemental analyzer

0.043 ± 0.43 at% Oxygen

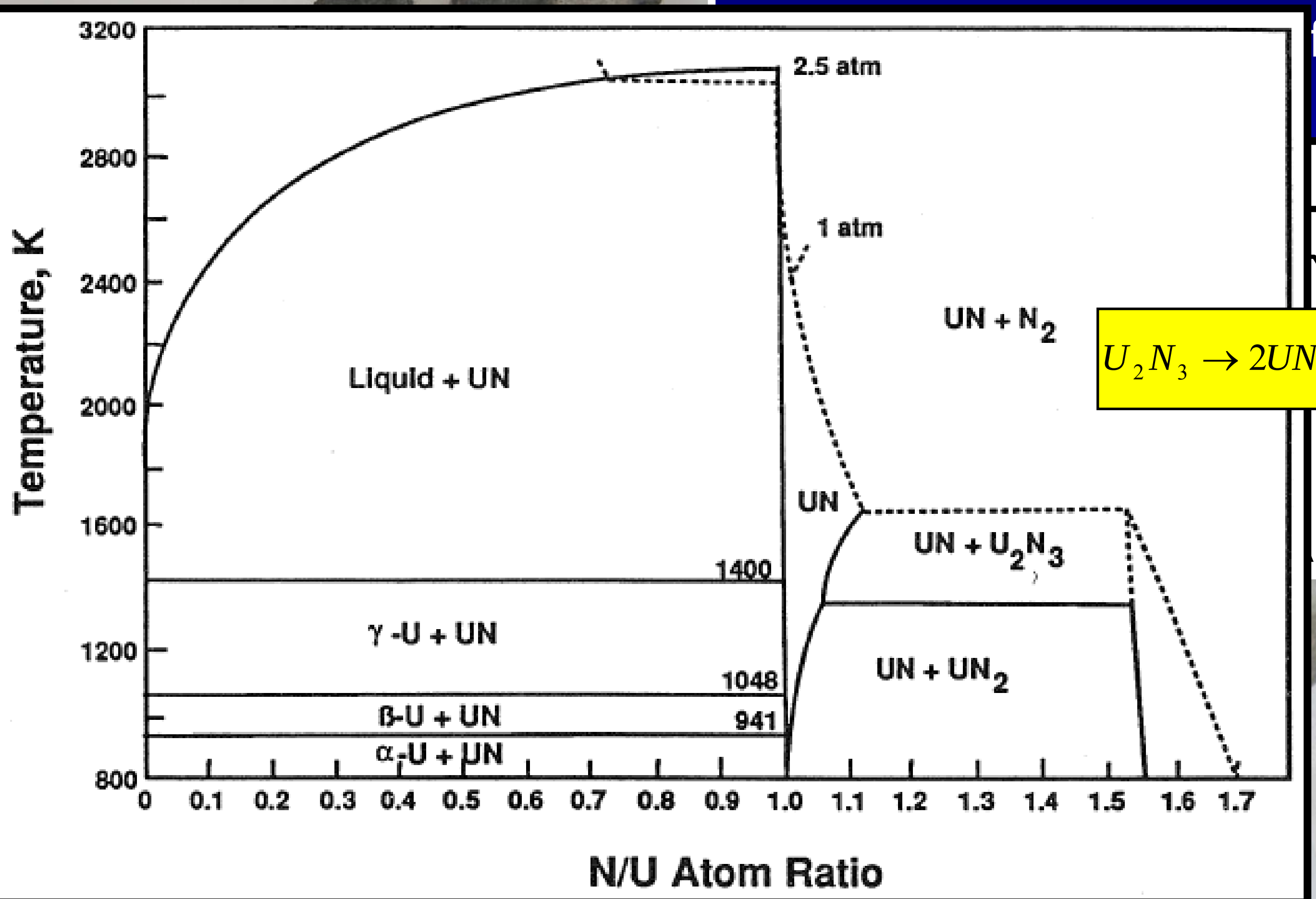
$$Dy + \frac{1}{2}N_2 + H_2 \rightarrow DyN + H_2$$

Nitride Synthesis Methods

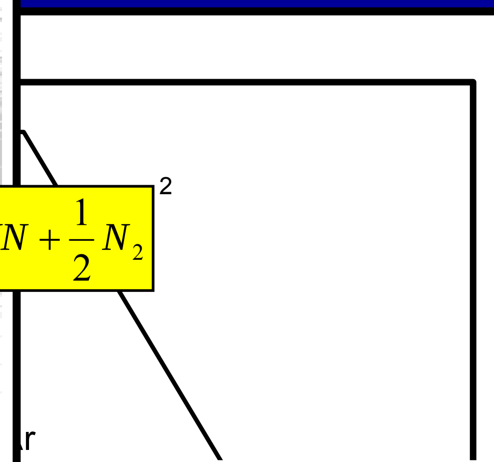
- Hydride-Nitride Synthesis Route



(from XRD)



Matthews, R.B., et al., J. Nucl. Mater, 1988. **151**(3)



$$U_2N_3 \rightarrow 2UN + \frac{1}{2}N_2$$

2

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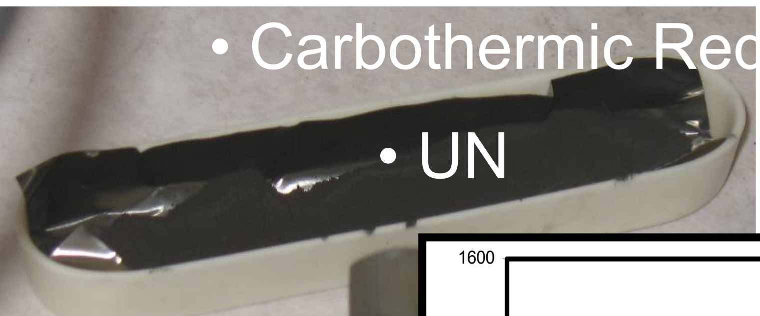
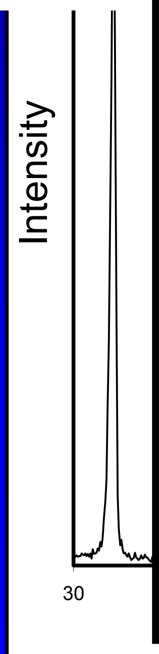
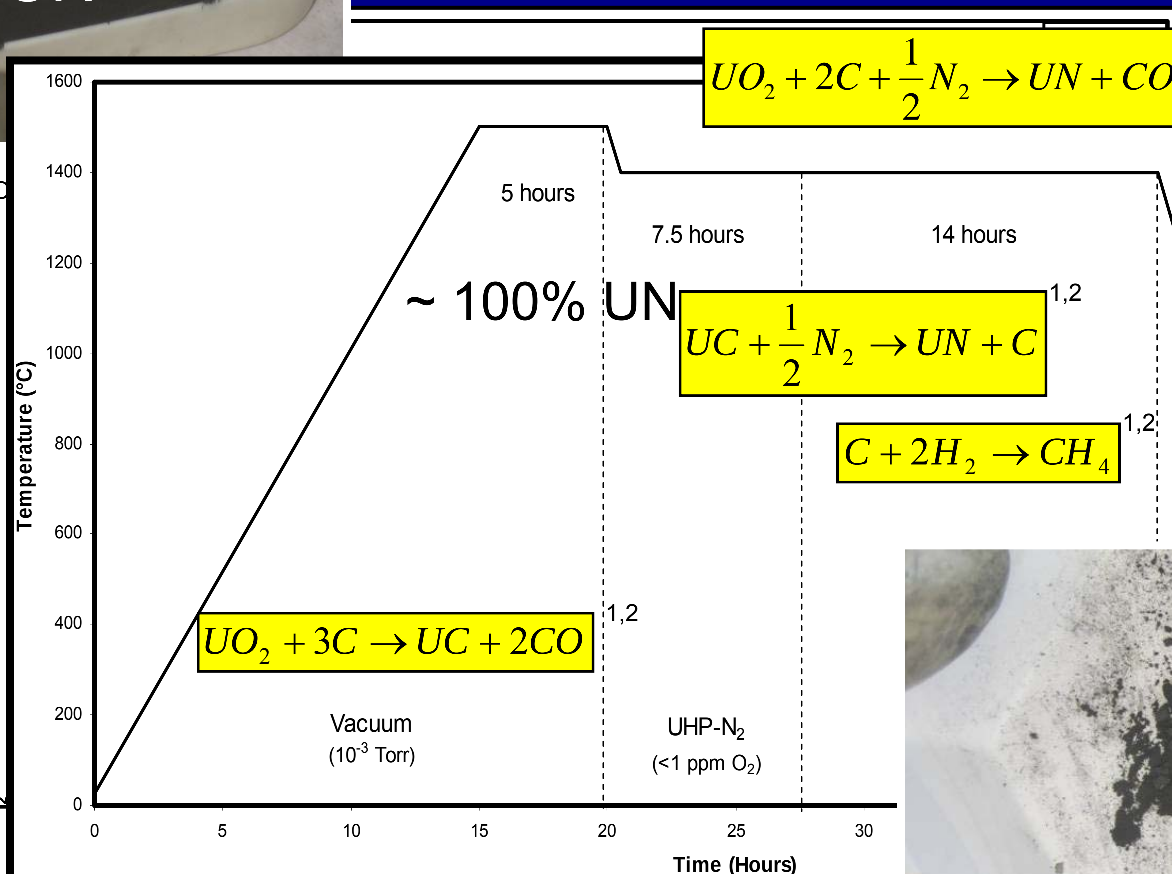
Nitride Synthesis Methods

- Carbothermic Reduction – N_2 Route
  - UN



Thoroughly mixed C/D-UO... with ball...





[1]Matthews, R.B., et al., J. Nucl. Mater, 1988. **151**(3)
[2]Arai, Y. *Proceedings of the workshop on manufacturing technology and process for reactor fuels*. 1995. Tokai, Japan.

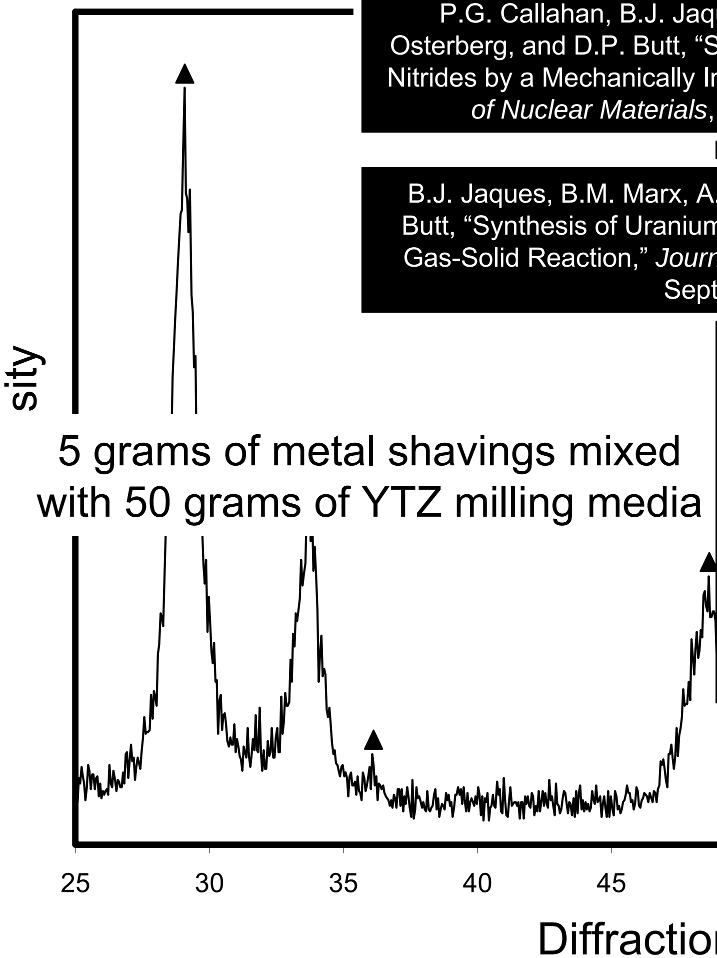
P.G. Callahan, *et al*. J. Nucl. Mat'ls, Submitted, September 2007. B.J. Jaques, *et al*. J. Nucl. Mat'ls, Submitted, September 2007.

Nitride Synthesis Methods

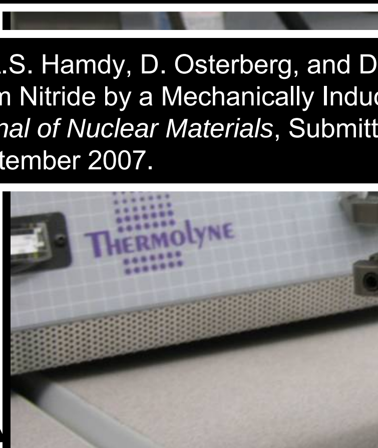
- Reactive Milling Route
  - U_2N_3 and DyN

P.G. Callahan, B.J. Jaques, B.M. Marx, A.S. Hamdy, D. Osterberg, and D.P. Butt, "Synthesis of Dysprosium and Cerium Nitrides by a Mechanically Induced Gas-Solid Reaction," *Journal of Nuclear Materials*, Submitted, September 2007.

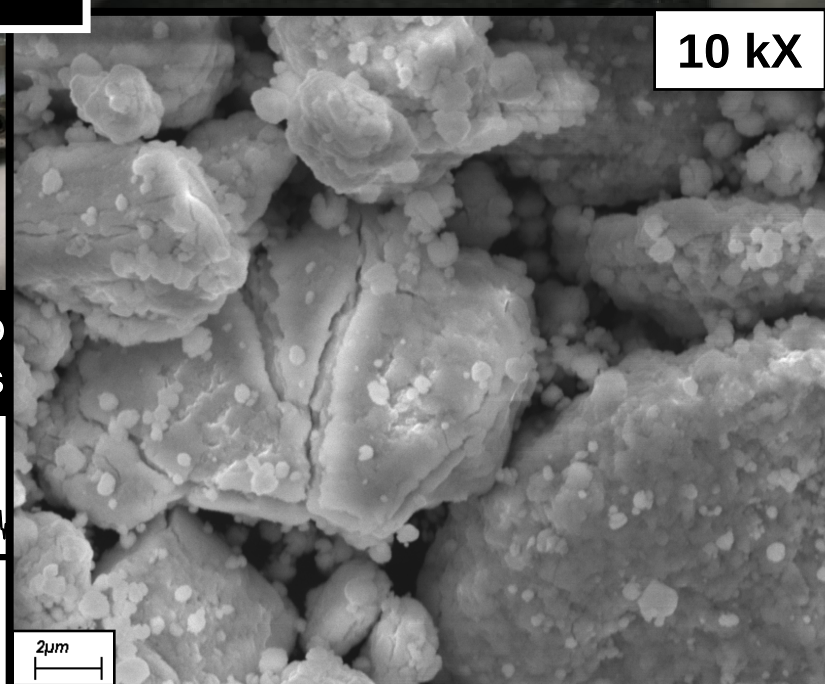
B.J. Jaques, B.M. Marx, A.S. Hamdy, D. Osterberg, and D.P. Butt, "Synthesis of Uranium Nitride by a Mechanically Induced Gas-Solid Reaction," *Journal of Nuclear Materials*, Submitted, September 2007.



5 grams of metal shavings mixed with 50 grams of YTZ milling media



N_2 with < 0.1 ppb
420 kPa (60 ps



Synthesis Summary

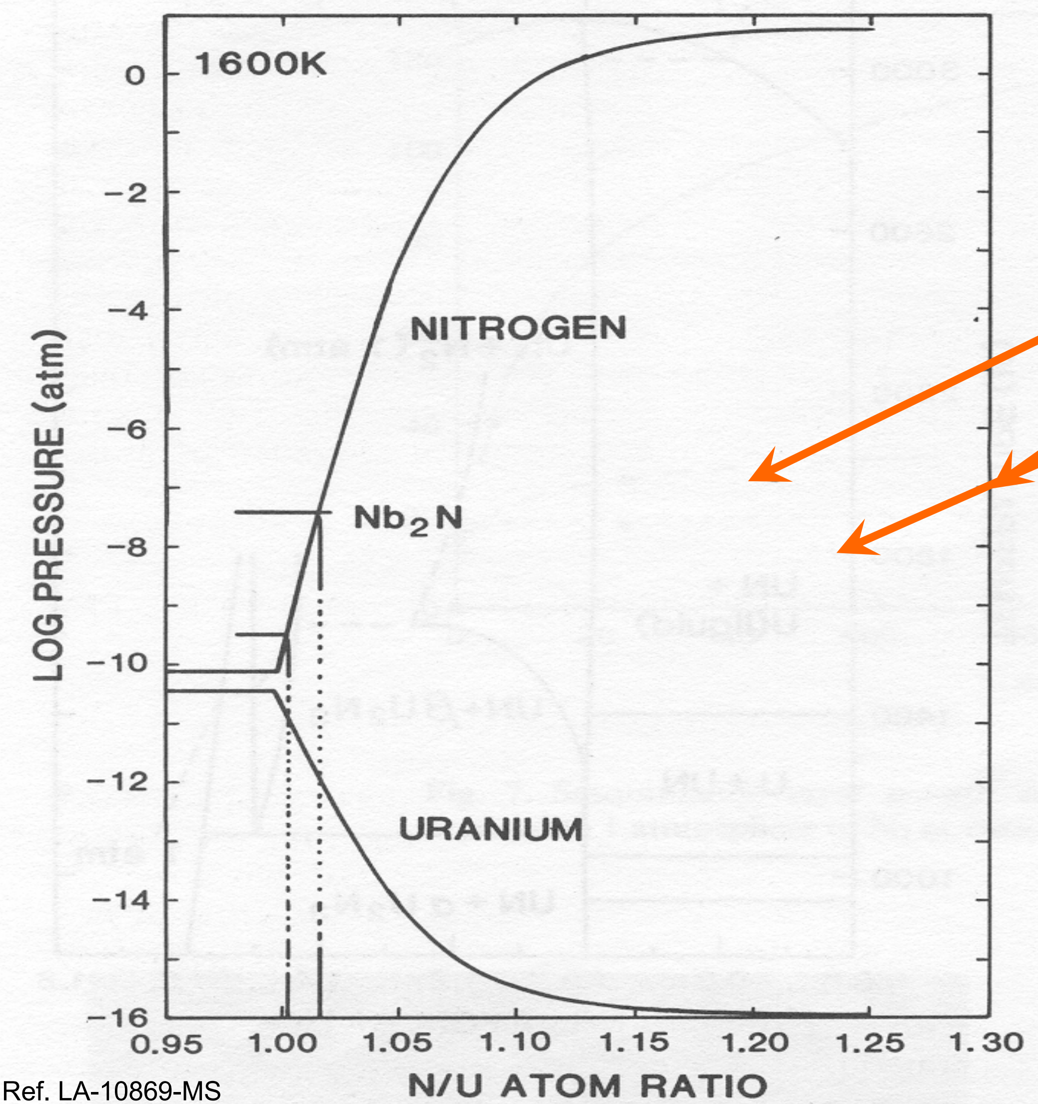
- Ball Milling Synthesis Route
  - U_2N_3 ~ 100%
  - DyN ~ 100%
- Carbothermic Reduction – N_2 Route
  - UN $UO_2 + 2C + \frac{1}{2}N_2 \rightarrow UN + CO$ ~ 100%
- Hydride-Nitride Synthesis Route
  - UN $U + \frac{1}{2}N_2 \rightarrow UN$ ~ 92% UN, 8% UO_2
  - DyN $Dy + \frac{1}{2}N_2 + H_2 \rightarrow DyN + H_2$ 0.043 ± 0.43 at% Oxygen
- Direct Nitride Synthesis Route
  - DyN $Dy + \frac{1}{2}N_2 \rightarrow DyN$ 0.33 ± 0.42 at% Oxygen

Initial Sintering Parameters

- Target green density is ~ 60% T.D.
- Target sintered density is ~ 85% T.D.
- $1.0 \leq {}^{Length}/_{Diameter} \leq 1.2$
- Compositional range (0 – 0.3 mol% DyN in UN matrix)
- Compaction pressure (420 MPa or 60 ksi) * On ¼" pellet
- Sintering temperatures (1450 - 1650°C, 50°C)
- Sinter times at temperature (2, 12, 24 hours)
- Oxygen-gettered argon atmosphere
- $250\ {}^{°C}/_{hour}$ ($\sim 4\ {}^{°C}/_{min}$) ramp rate

Continued . . .

The acceptable composition range for UN is ~ N/U = 1.002-1.015



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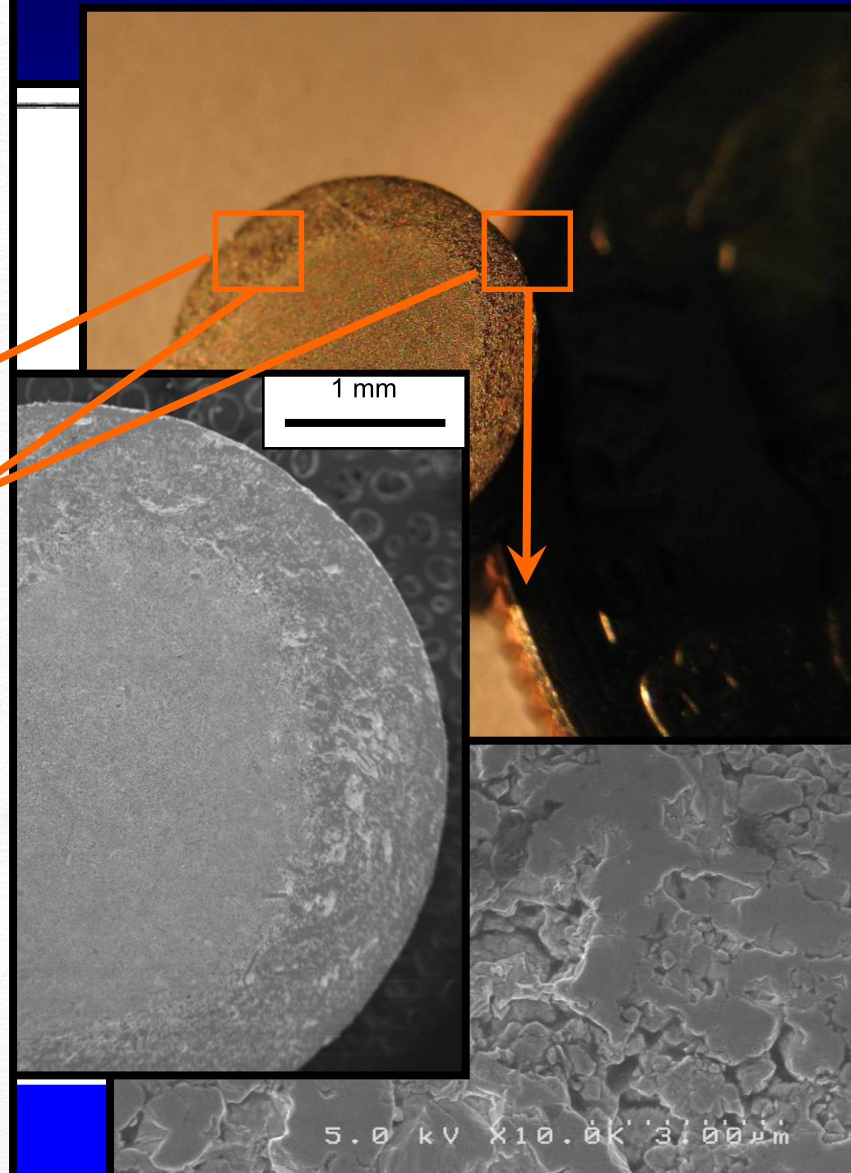
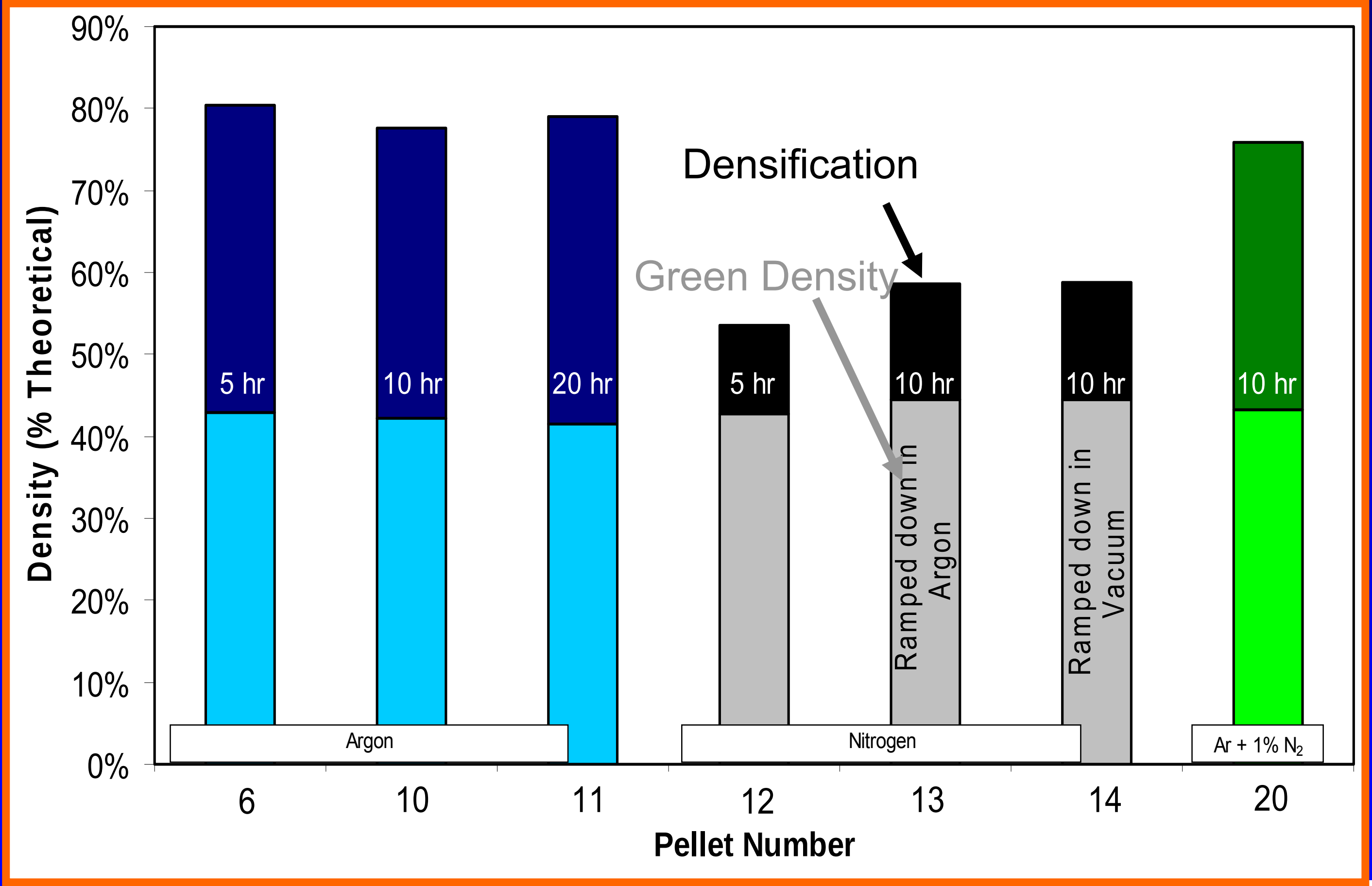


Atm. & Sinter Time vs. Density

1550°C, 24 hour dry ball mill



Vision:

Low-cost processing facility with a small footprint

Pyroprocessing

Metal

Direct Conversion to Nitride

Ball milling

Am retention

Low-Cost Forming

Cold-pressing

Low-Cost Sintering

Tube-furnace