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Title: Radioisotopic Thermoelectric Generator (RTG) Surveillance

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Radioisotopic Thermoelectric Generator (RTG) Surveillance

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September 2016

Los Alamos

Core Surveillance is comprehensive preventative maintenance for nuclear weapons

- **All components of nuclear weapons are examined annually**
- **Design Agency issues clear directives**
- **Tests are prescribed and well-established**

They're well-kept, but will they run?

(and where will you get leaded gas?)

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SBSS: Science-Based Stockpile Stewardship

- Assessment & certification without nuclear testing
- Sustainment of weapons-in-being (at the next plateau / near-term)
- Responsiveness / adaptation to new requirements – could be weapons security, more margin, or a military effect not already stockpiled.

(Both the 1993 and 2001 Nuclear Posture Reviews required that the Complex be capable of new design "if required.")

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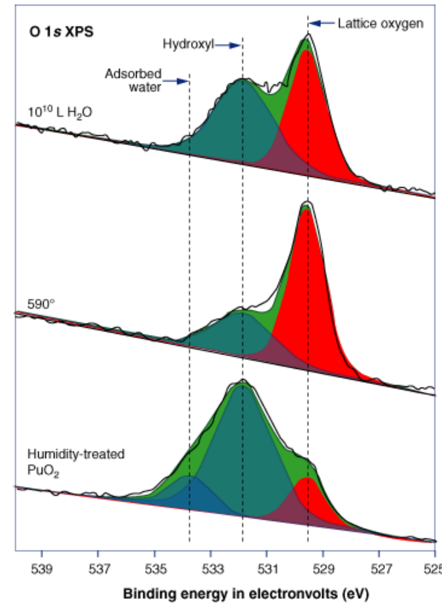
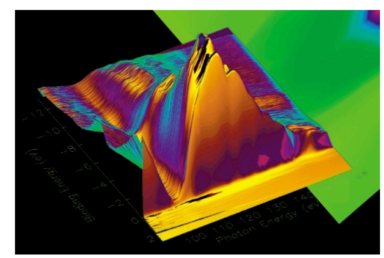
Enhanced Surveillance examines materials in depth

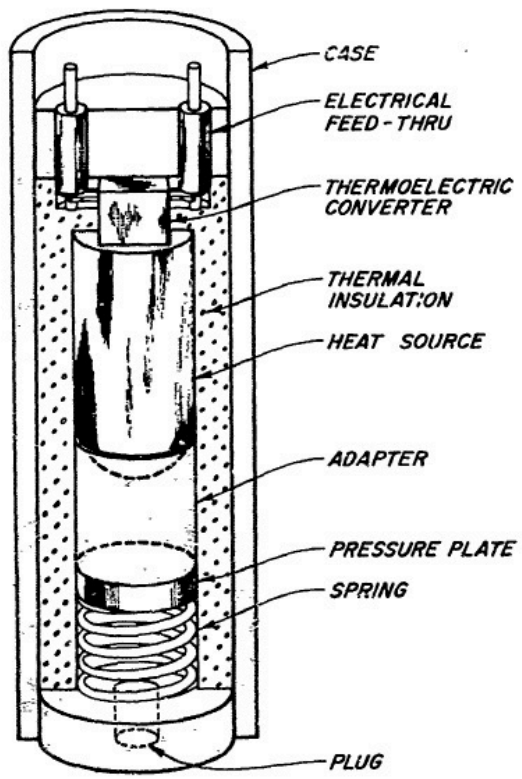
- **Identify and characterize aging signatures**
  - Stockpile aging (CS) & artificial aging studies
  - Provide science-based understanding (mechanisms)
- **Develop and implement age-aware predictive models**
  - Long-term materials behavior
  - Material lifetime estimates
- **Improve future stockpile through LEP support**
  - Replacement & reuse materials
- **Resolve unanticipated results**
  - Establish cause and effect



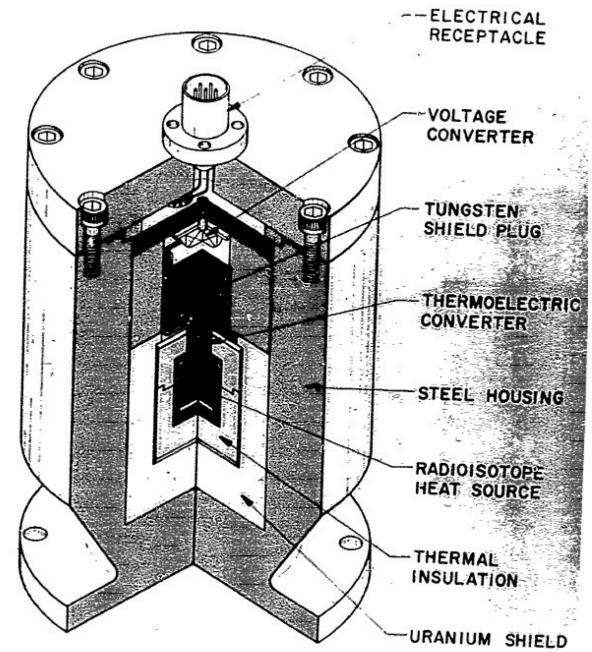
- **Determine and communicate engineering and physics impacts**
  - Delivery and Performance
  - Lifetime estimation
- **Produce innovative diagnostics**
  - New tools for core and enhanced surveillance programs
- **Maintain (and exercise) nuclear weapon subject matter expertise**



RTG The Radioisotope Thermoelectric Generator is a self-contained, long-lived battery



Navy superbattery, ca. 1978



Sentinel 25
$SrTiO_3$

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RTG The Radioisotope Thermoelectric Generator is a self-contained, long-lived battery

Medtronic heart pacemaker 1973-1985

As of 2011, these units were still in use

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A range of isotopes serve to power RTGs

Balance power and lifetime, power and shielding

^{238}Pu	α	0.54 W/gram	87.7 years
^{90}Sr	β (γ)	0.46 W/gram	28.8 years
^{210}Po	α	140 W/gram	138 days (0.5 g to 500 C)
^{241}Am	α (γ)	0.13 W/gram	432 years

also ^{244}Cm, ^{147}Pm, ^{137}Cs, ^{144}Ce, ^{106}Ru, ^{60}Co, ^{242}Cm, ^{171}Tm, ^{235}U

Sakhalin Light

Aneva Light, ^{90}Sr

Beta-M battery

http://www.michaeljohngrist.com/2011/08/abandoned-lighthouses-rubjerg-knude/

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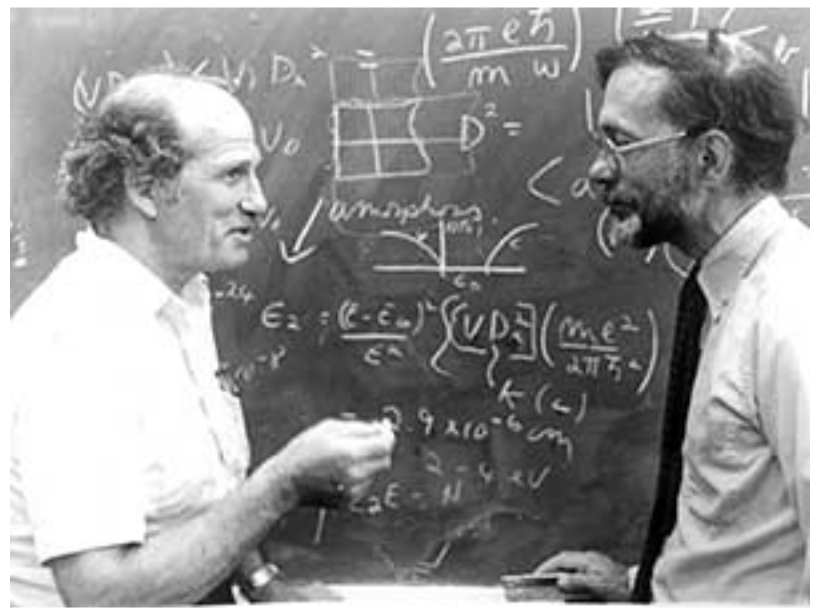
Silicon-Germanium Thermopile Converts Heat to Electricity

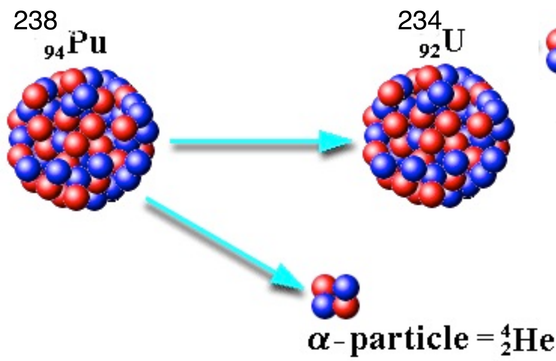
- **April 3 1965**
- **RCA Laboratory New Jersey**
- **Stable to 1000 C**
- **High electric conductivity**
- **Low thermal conductivity**
- **measurement was difficult**
- **Co-located n-type and p-type semiconductors (60% Si)**



Ben Abeles and George Cody

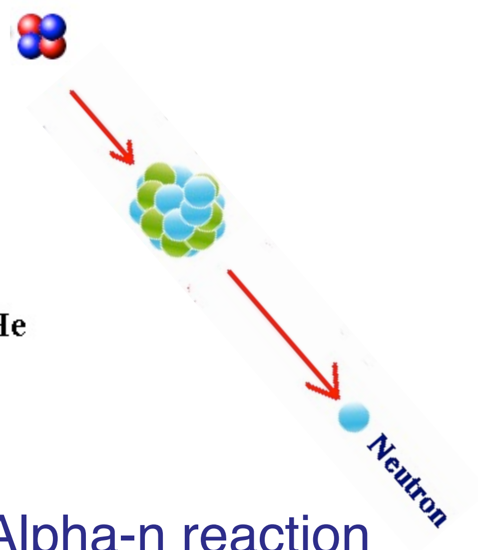
Other designs for heat sources have been proposed

"subcritical multiplicator" or "Advanced Subcritical Assistance" RTG

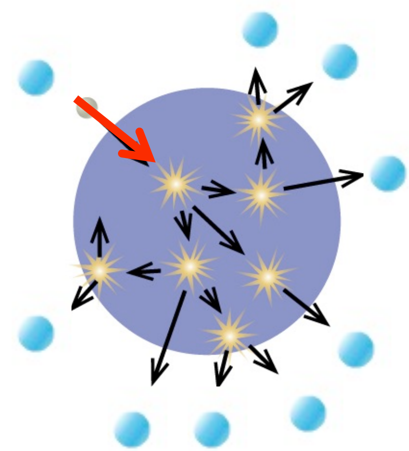


^{238}Pu produces alpha particle

Eneergy about 6MeV



Alpha-n reaction produces neutrons



Neutrons promote subcritical reaction fission energy 200 MeV

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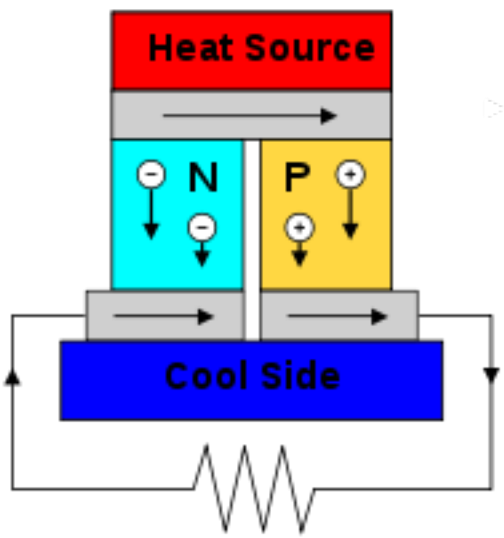
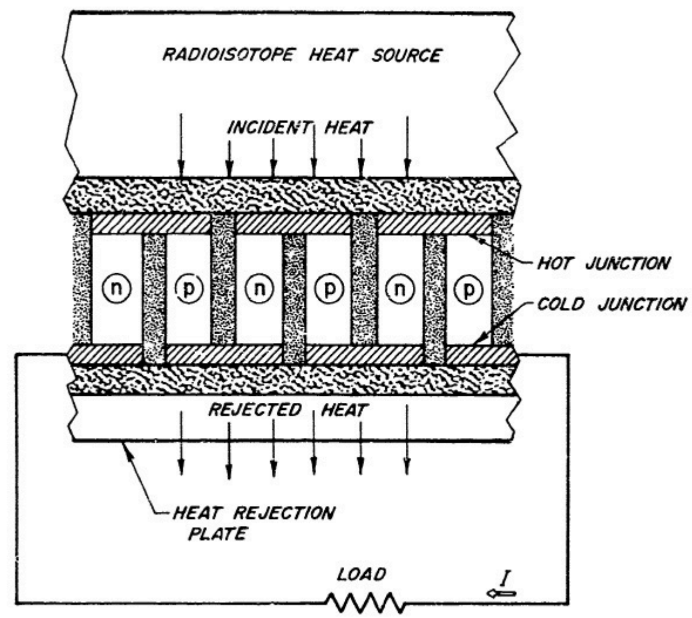
The Seebeck effect generates the electrical current

Electron mobility increases with increasing temperature

Voltage differential can be converted to current at a junction





Semiconductors can be coupled to maximize thermoelectric current

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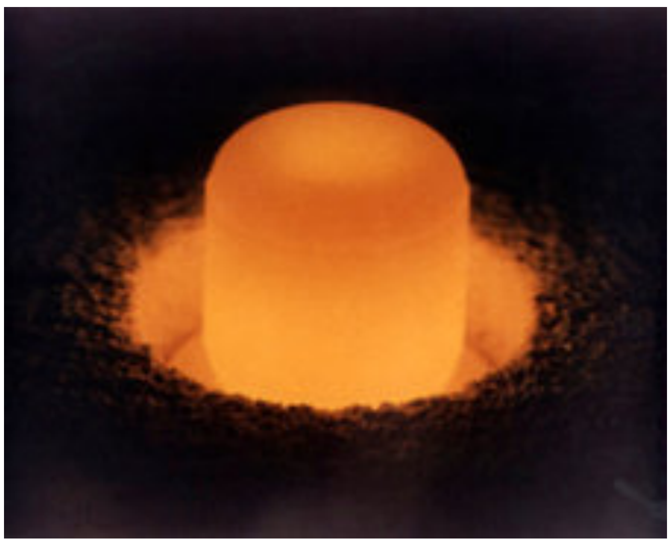
^{238}Pu Oxide gives off heat

^{238}Pu oxide pellet



87.7 year half life

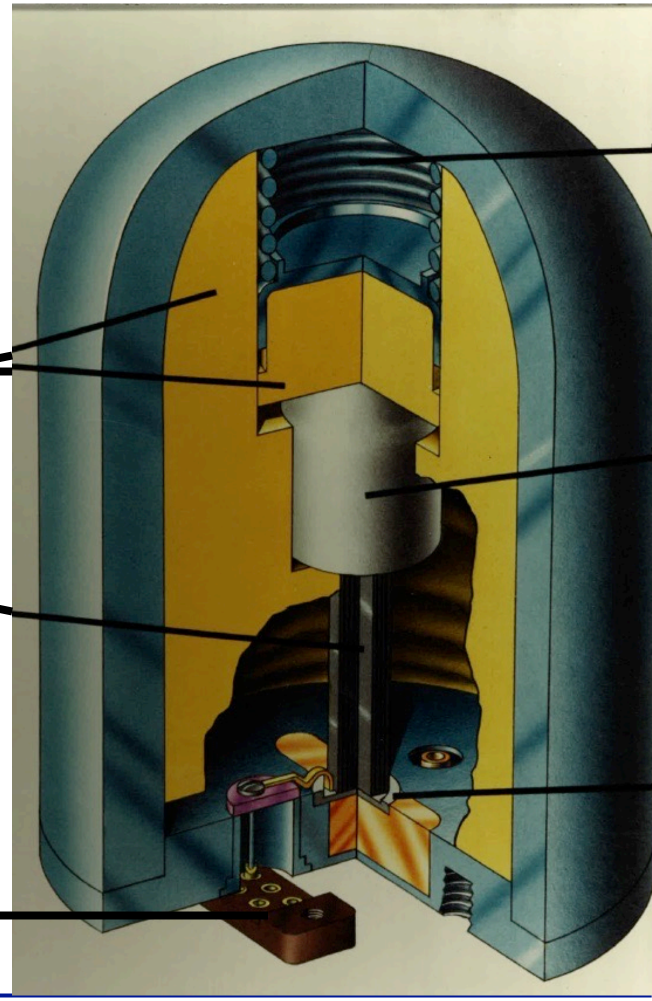
Alpha decay

0.4 Watts/gram output

Converted to electricity
using a thermopile
with 3% to 7% efficiency

Our RTGs are very robust

Text



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Several Required Tests are Performed

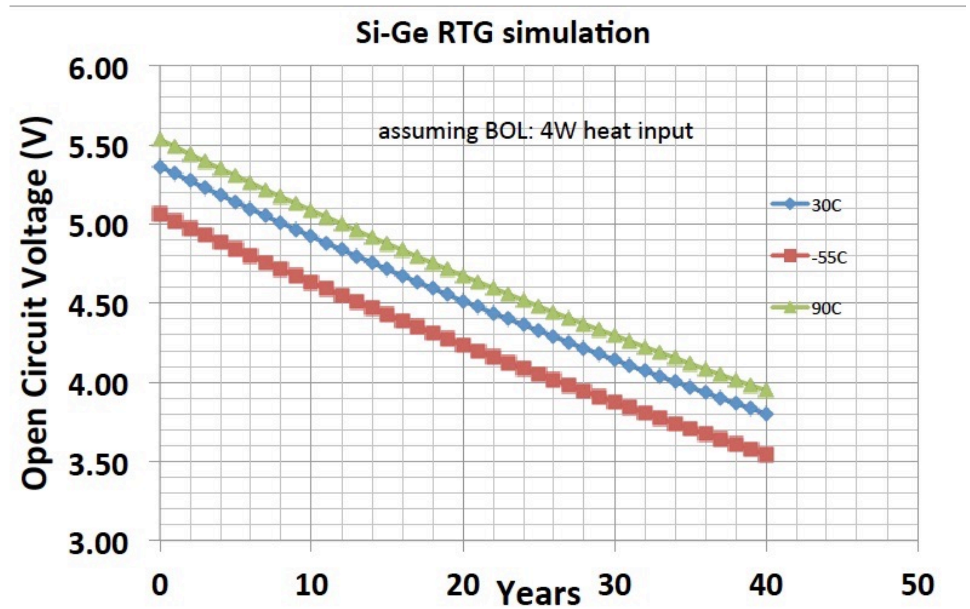
- **Electrical tests**
- **Disassembly "D-test" (destructive testing)**

- **Recovery of all parts**
- **Heat source tests**
- **Gas pressure, helium generated by decay of ^{238}Pu**
- **Weld integrity**

Fuel is recovered, sieved, and re-used

Four electrical tests are performed

- AC Impedance
- Open-circuit voltage
- Loaded Voltage
- Baseplate isolation resistance



Test at ambient (25° C) and at upper and lower temperature limits

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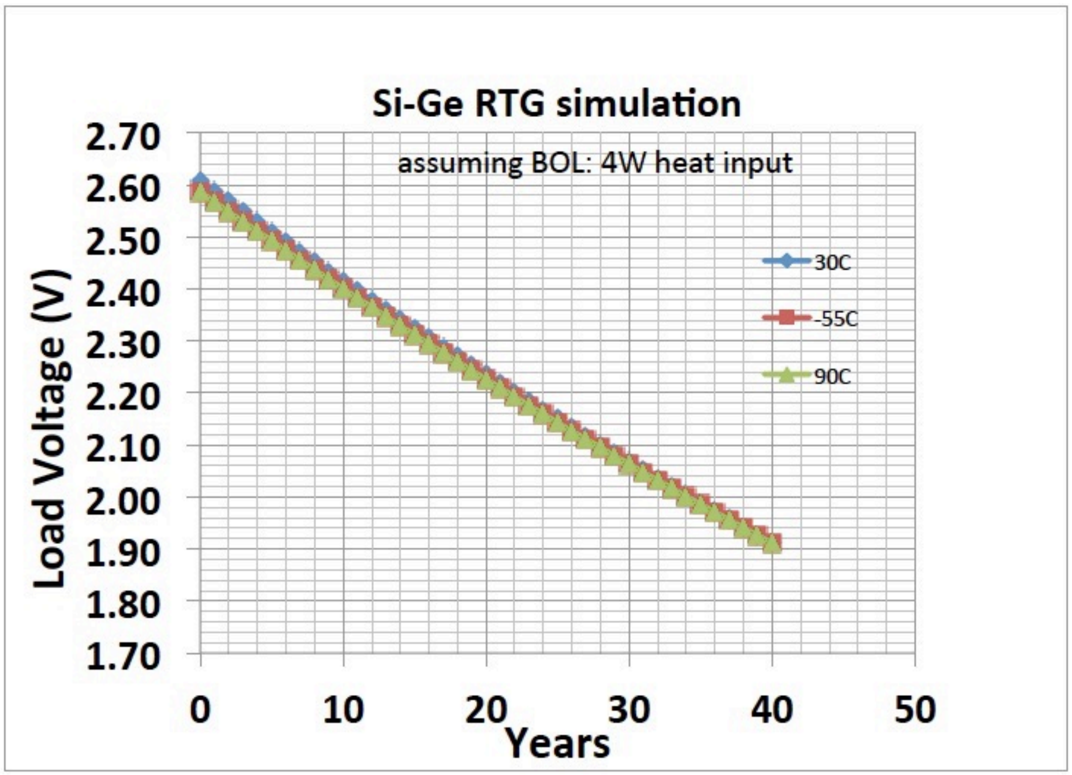
Results of electrical testing reflect radioactive decay

Voltage output decreases with time

$$V = V_0\, e^{-(t/\tau)}$$



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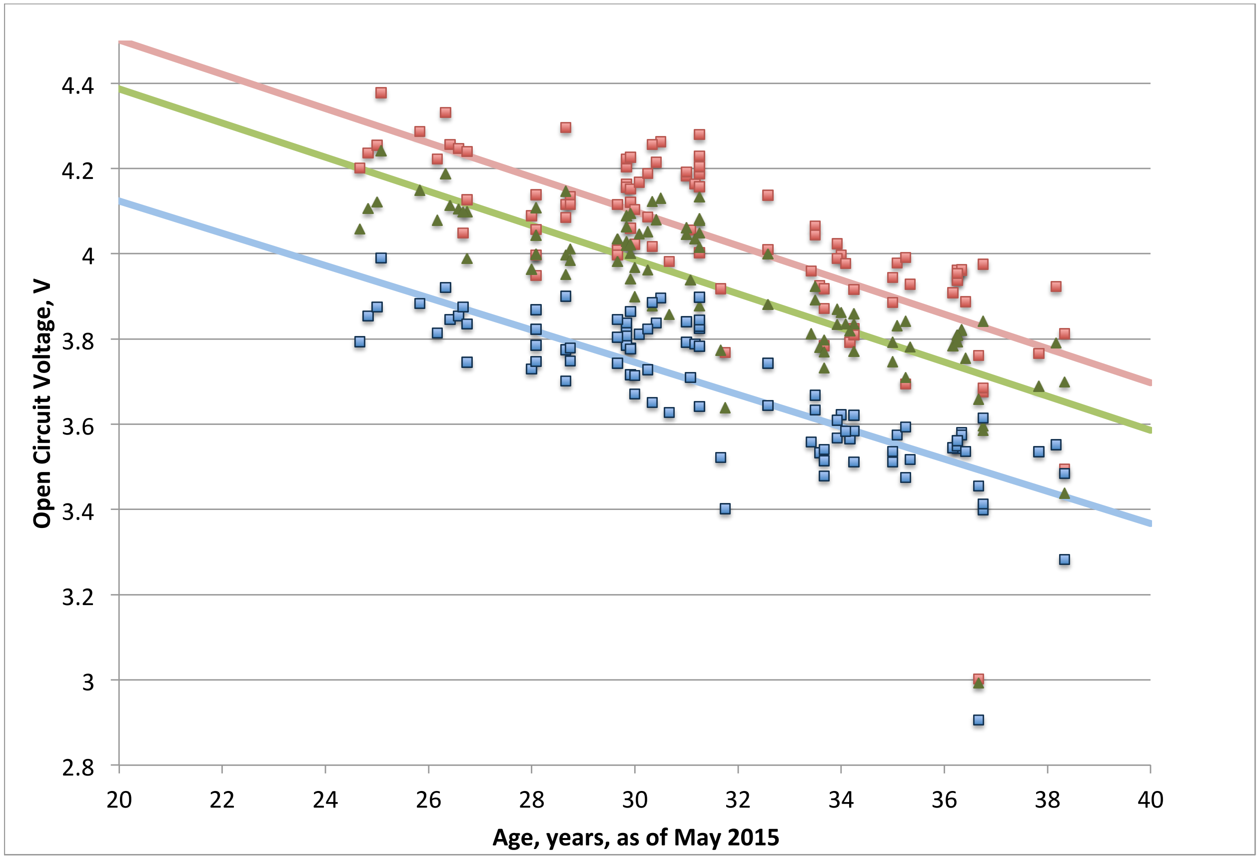
Measured voltage varies with quantity of fuel



Response of the thermoelectric varies with temperature

Measured data exhibits scatter

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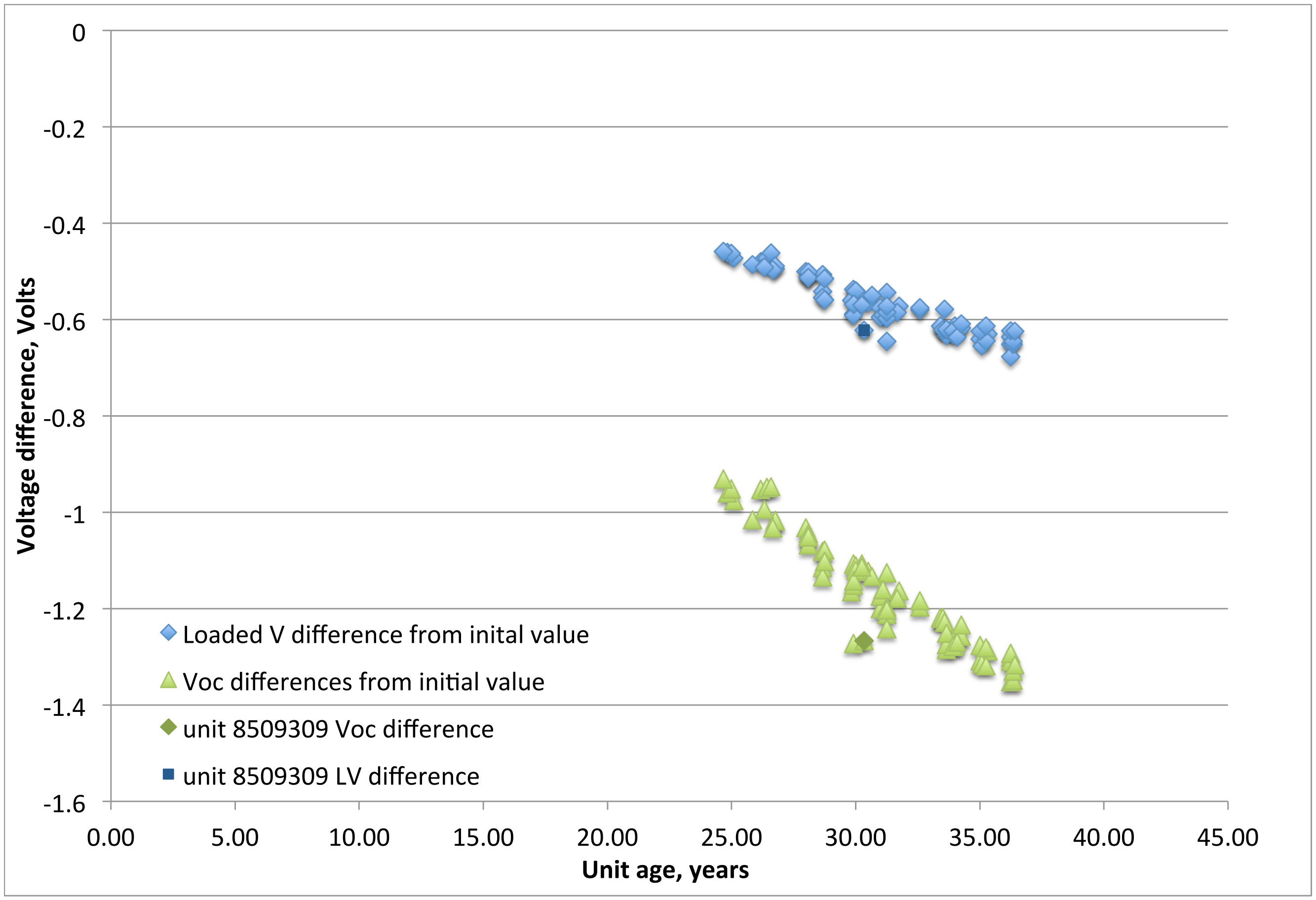
Subtraction of initial voltages reduces scatter in data

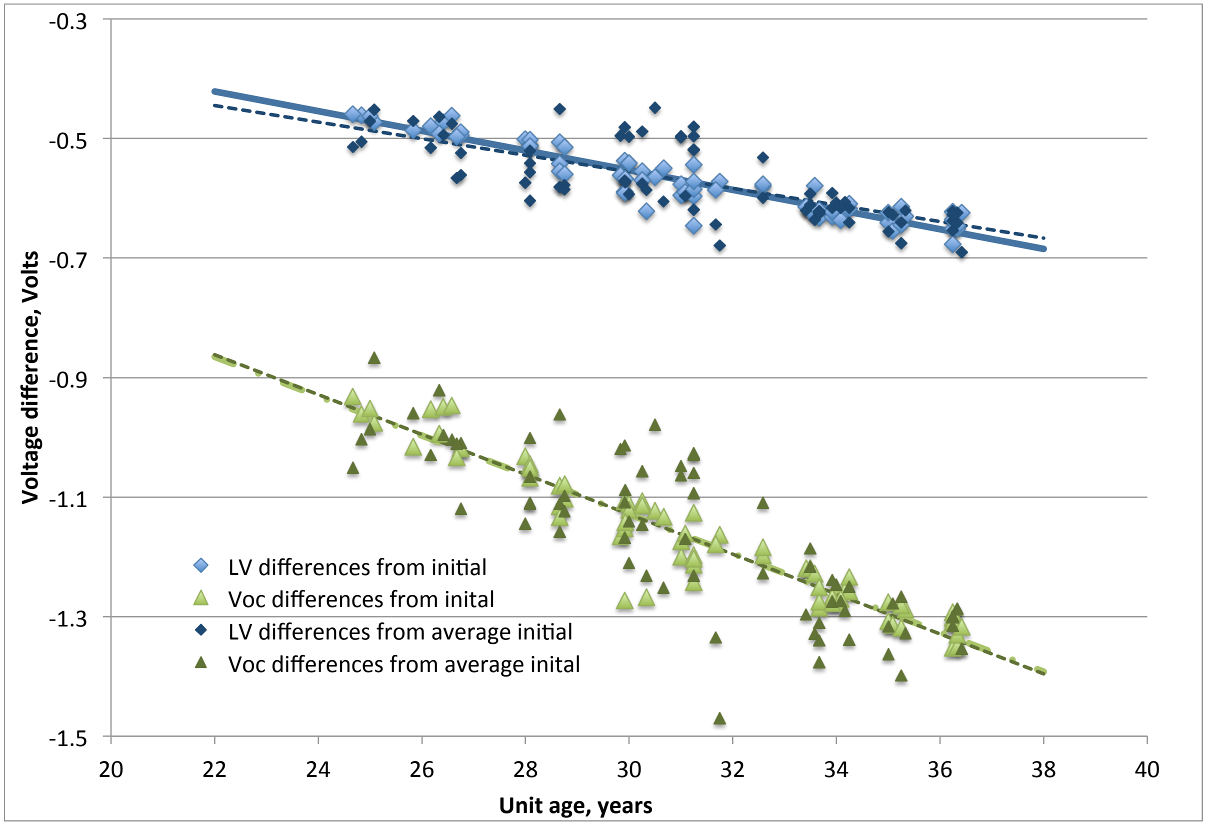
Voltages were recorded at manufacture



Data at 25 C

Scatter in data is reduced by normalization to initial values

Scatter reduced: Scatter due to variable fuel charge removed



LV at 25 C

$R^2 = 0.88$ without subtraction, $R^2 = 0.59$

Voc at 25 C

$R^2 = 0.91$ without subtraction, $R^2 = 0.61$

Slopes of linear fits unchanged for loaded voltage (LV) measurements

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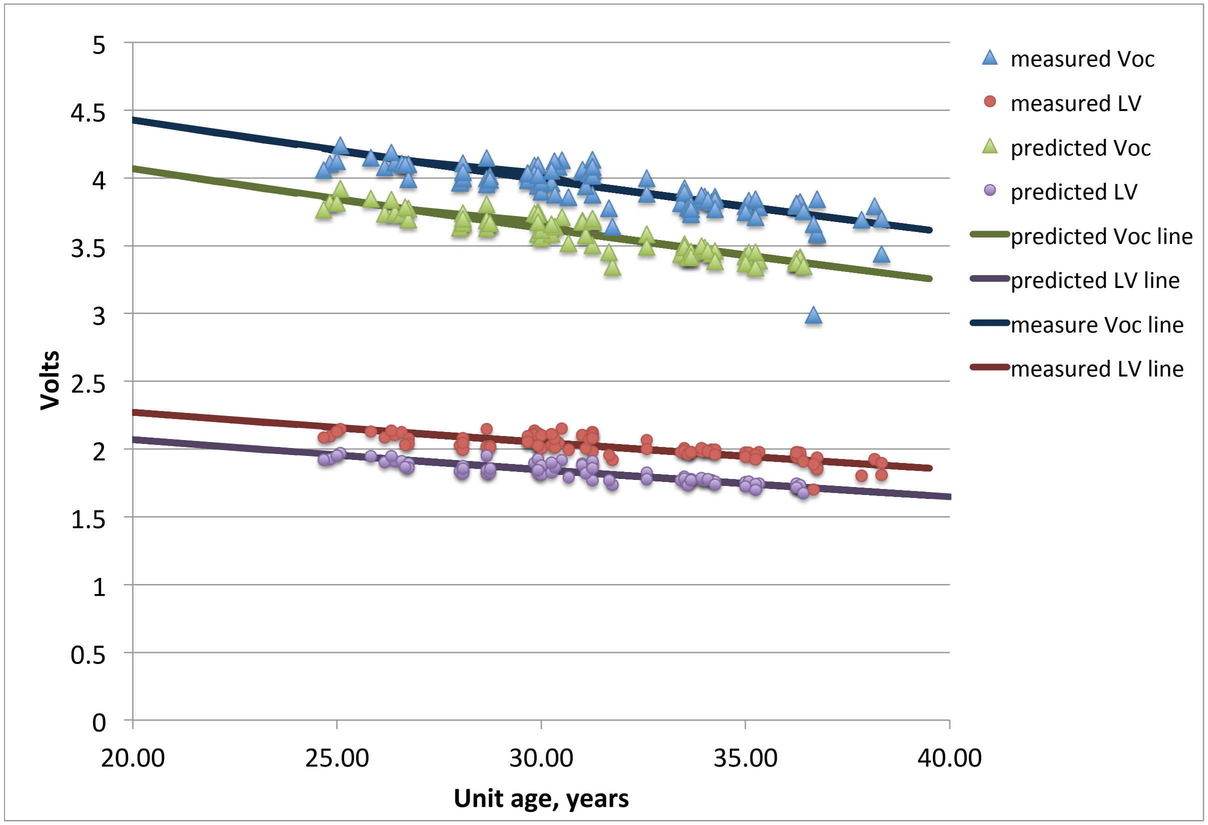
Measured performance is better than predicted performance



Operating temperature 25 C

Initial and measured voltages fit with known decay rate

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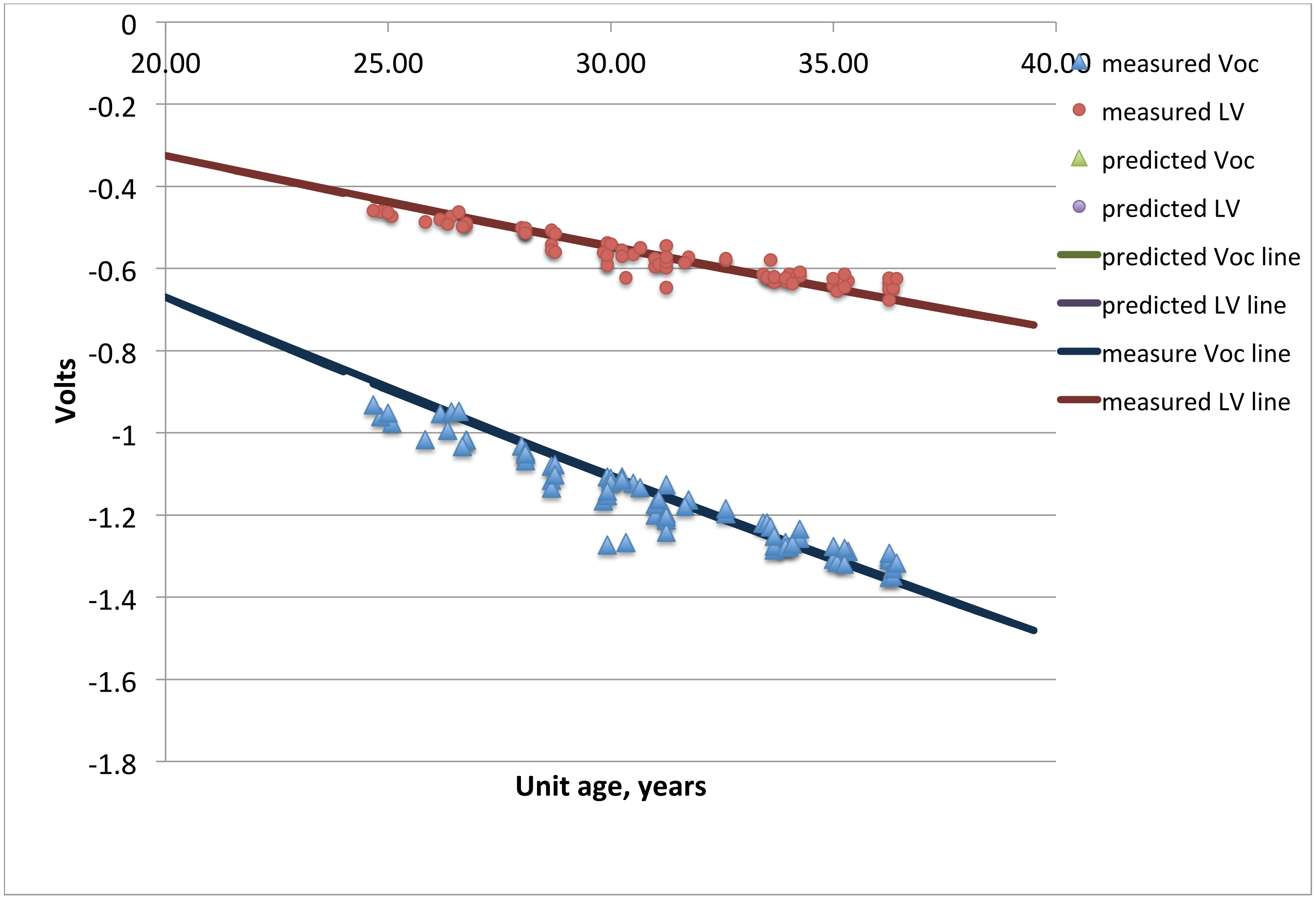
Correcting each data point for initial voltage tightens the distribution



Measured at 25 C

The margin between predicted and measured is indicated

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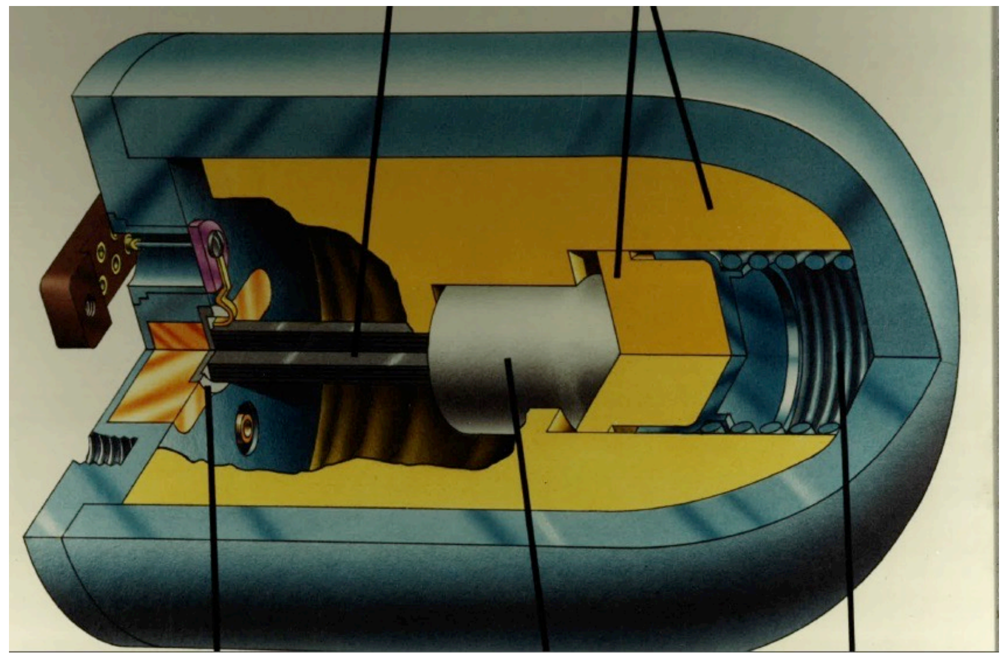
The case is cut in half on a lathe

The parts are embedded in fibrous Min-K thermal insulation

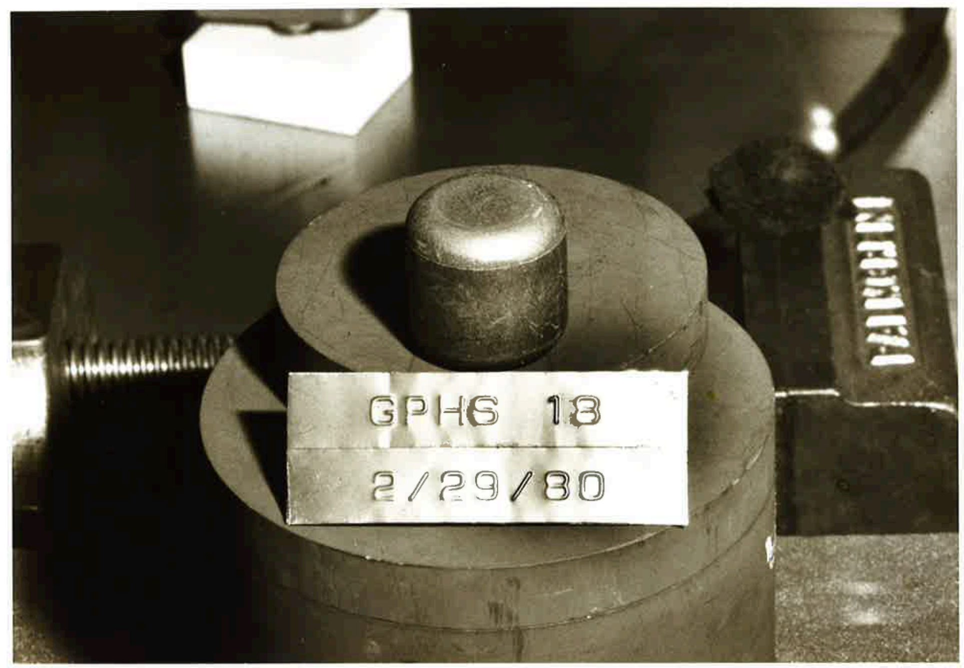


All parts are recovered and examined

Many of the heat sources were made at LANL

Manufacture was originally done at Mound Laboratory

Manufacture moved to Los Alamos in 1989



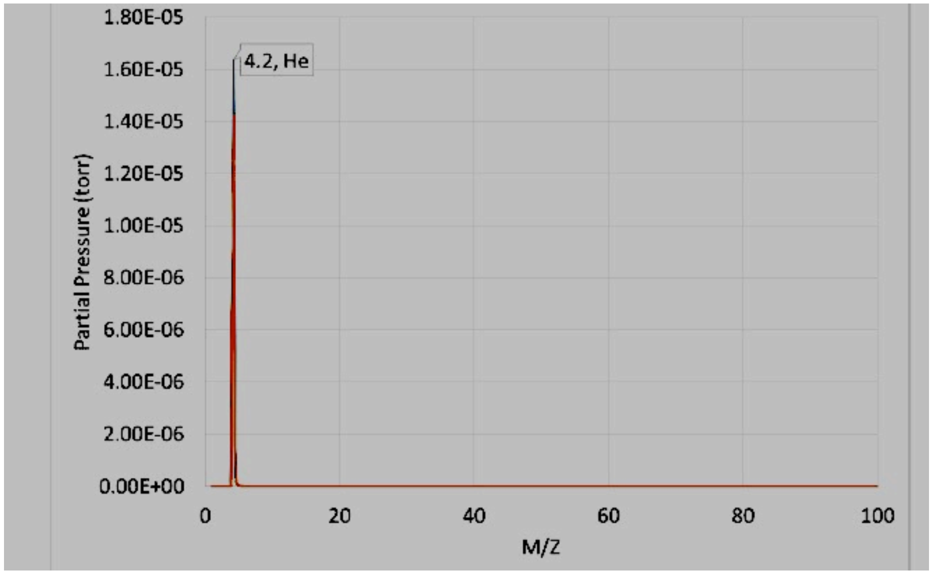
The unit is cut in half and the components measured

The parts are archived so that future questions can be addressed

The heat source is examined further

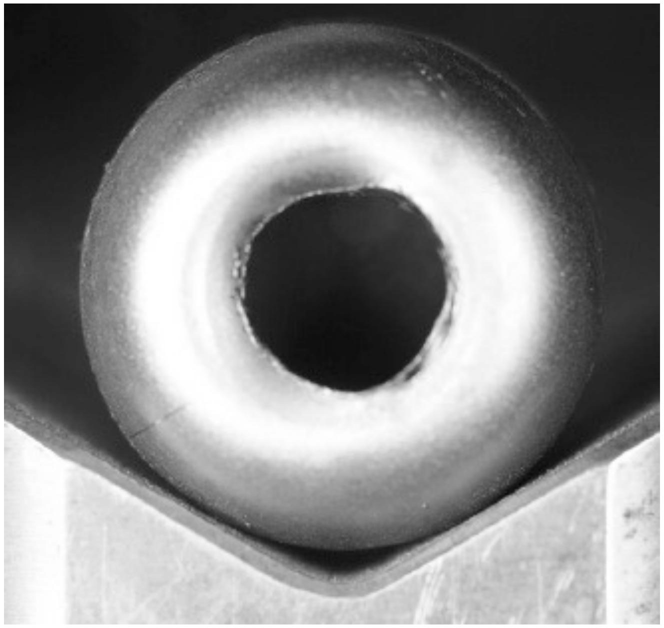
Gas pressure and composition in heat source measured

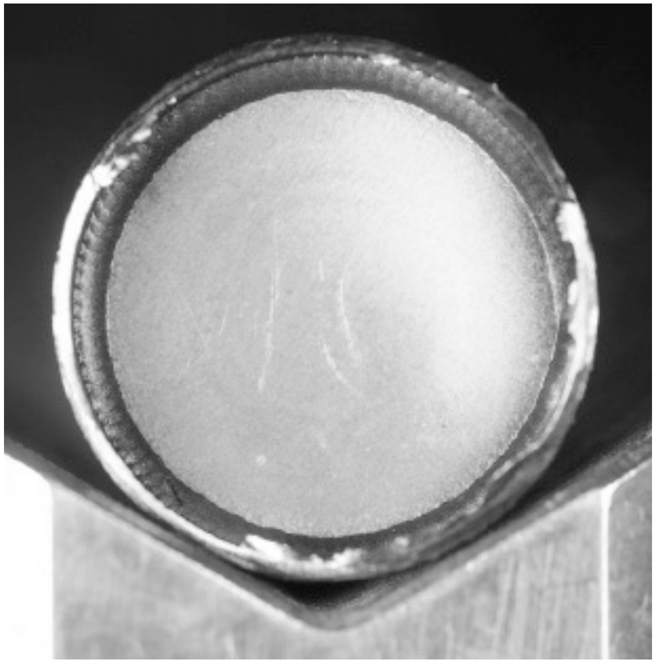
Helium is a product of radioactive decay, measured with mass spectroscopy done on fill gas



The expected pressure can be calculated from the age of the unit

The container is sectioned and welds examined





The granular fuel is emptied out the drilled hole

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Examination of the weld shows pristine sound welds

Welds are examined for evidence of change

Metallography shows grains in the weld

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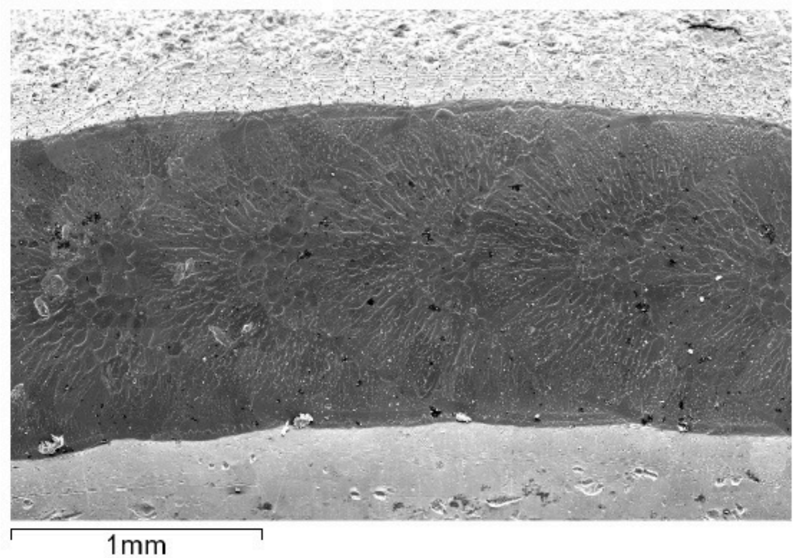
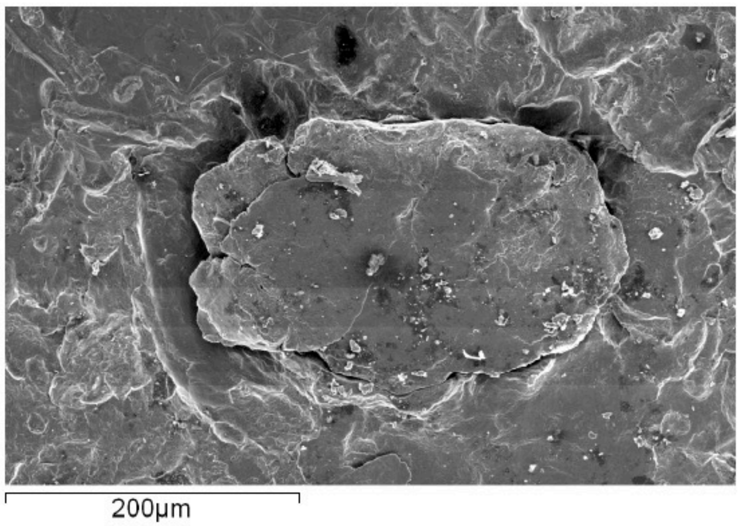
Electron micrographs of the strength member weld

Weld overlap region is beautifully clean

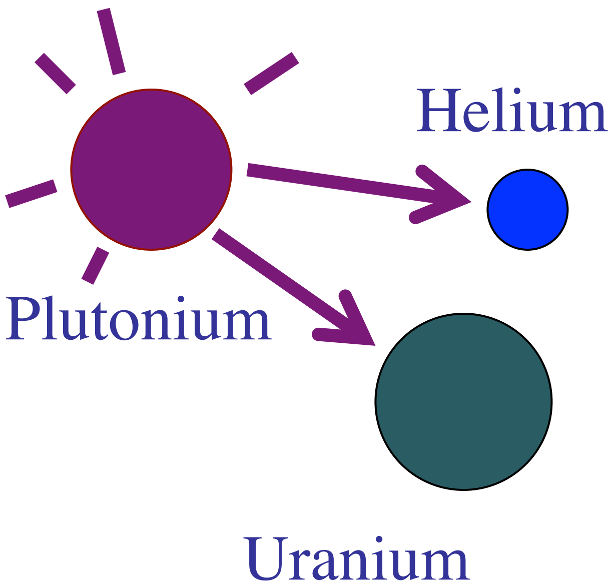


Galling is sometimes seen from metal-metal contact



Pressure data yields interesting science

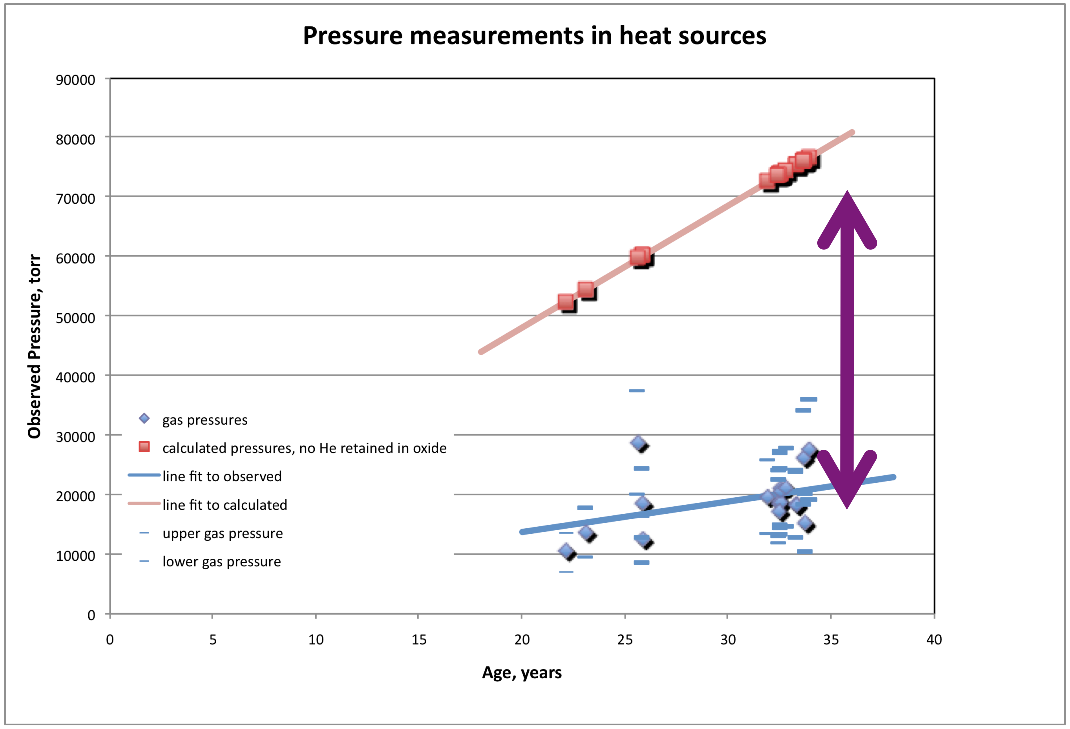
Helium is evolved from Pu-238 oxide–
will there be sufficient gaseous helium at advanced age to deform the capsule?



Helium release from oxide studied in 1970s

Pressure is about 26% of expected

Pressure observed is about ¼ to 1/3 of helium produced over time



Where' s the Helium?

Relatively large error bars from
pressure gauge with error larger than advertised

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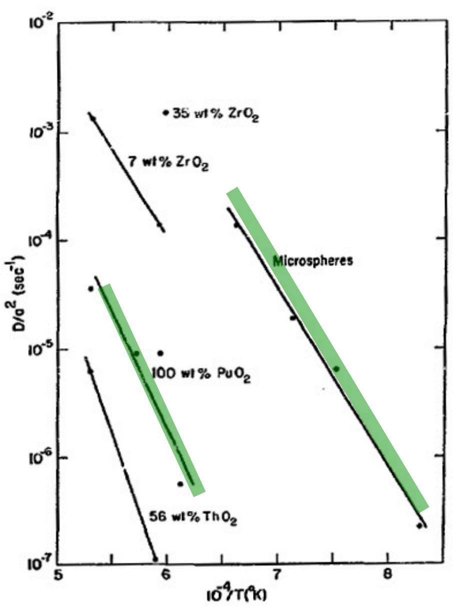
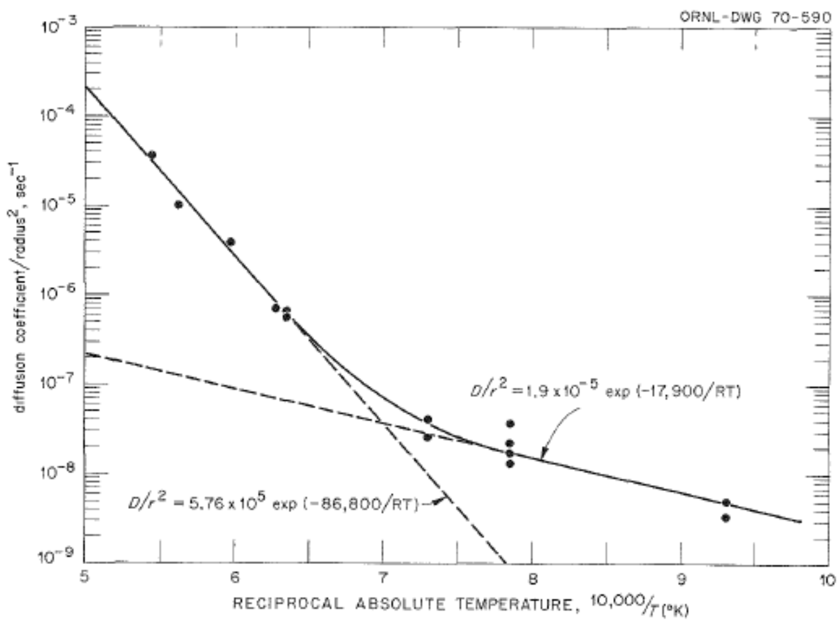
Helium is captured in the oxide matrix

Early work indicated that helium is
captured in the oxide lattice even at high temperatures (900-1400°C)

Arrhenius plots for He diffusion:



RNR Mulford, LASL, 1973



P. Angelini, ORNL, 1970

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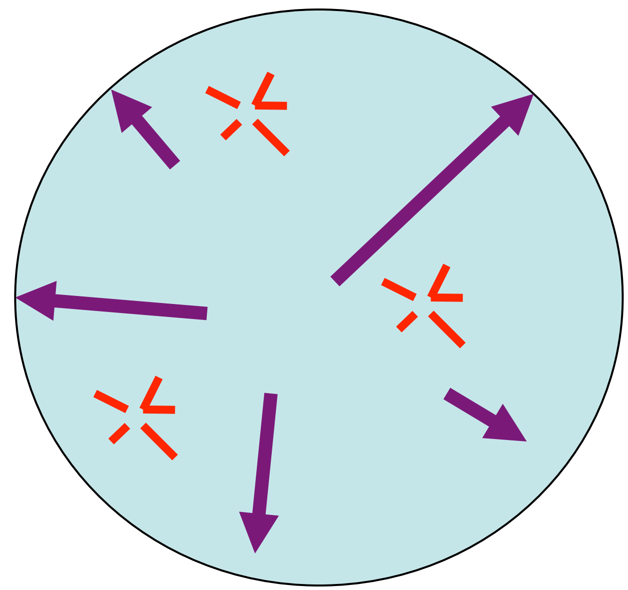
Amount of gas calculated assuming diffusion in radial geometry

Diffusion: Fick's law $P = -D \frac{dC}{dX}$ P is flux and $\frac{dC}{dX}$ concentration

Equation in spherical geometry for radial diffusion

$$F = 1 - \frac{6}{\pi} \sum_n \frac{1}{n^2} \exp(-n^2 \pi^2 D t / a^2)$$

for fraction F of gas escaping



D' is D/a^2 a rate, eliminating radius a

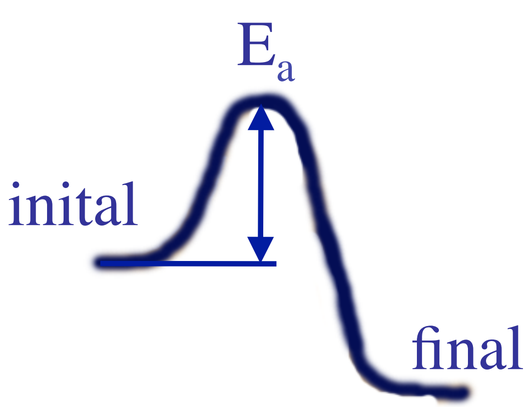
τ is $\int_t D' dt$ to account for continuous decay

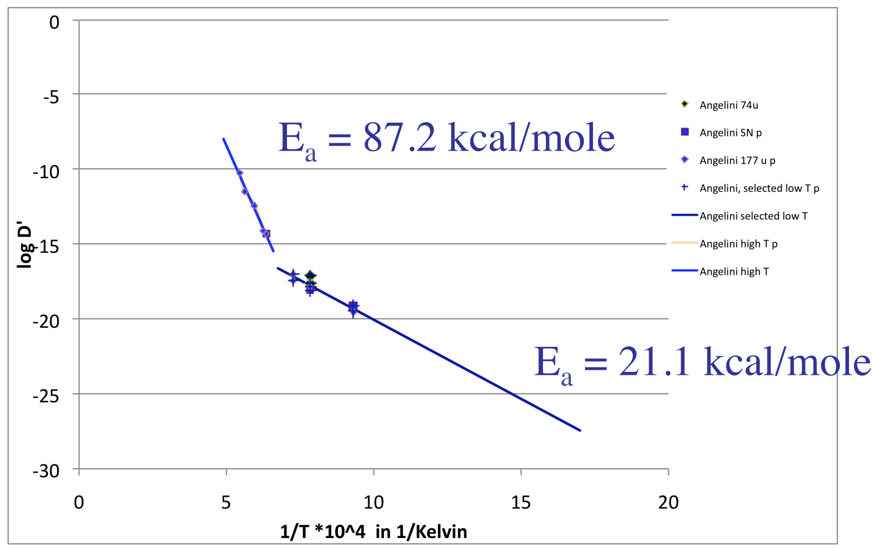
Rate of diffusion of helium assumes Arrhenius kinetics

$$D' = D'_0 \exp(-E_a / RT)$$

$$\text{Log } D' = -E_a / R \left[\frac{1}{T}\right] + \log D'_0$$

Activation energy





Activation Energy E_a derived from slope

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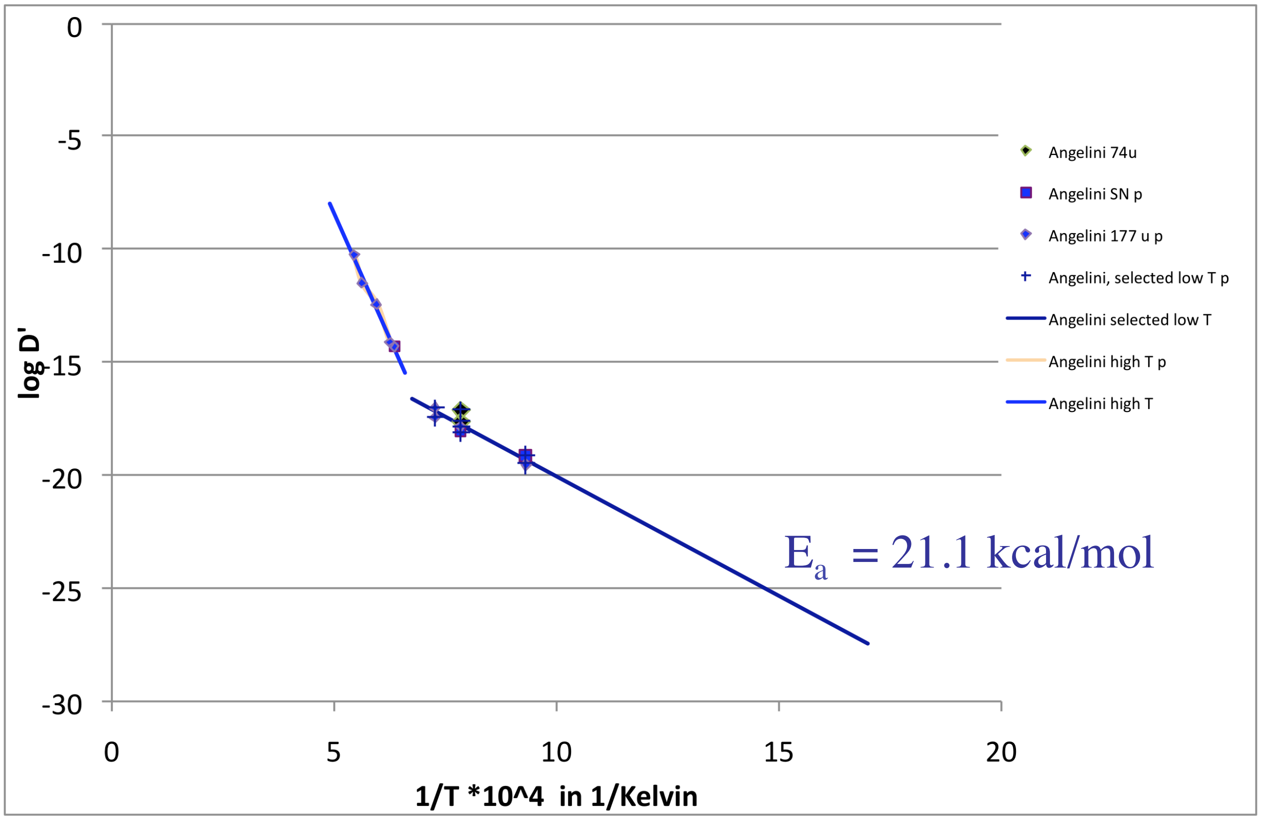
Arrhenius plot obtained in 1970 shows low-temperature diffusion (800-1100°C)



Static data from Mound microspheres 3 years old

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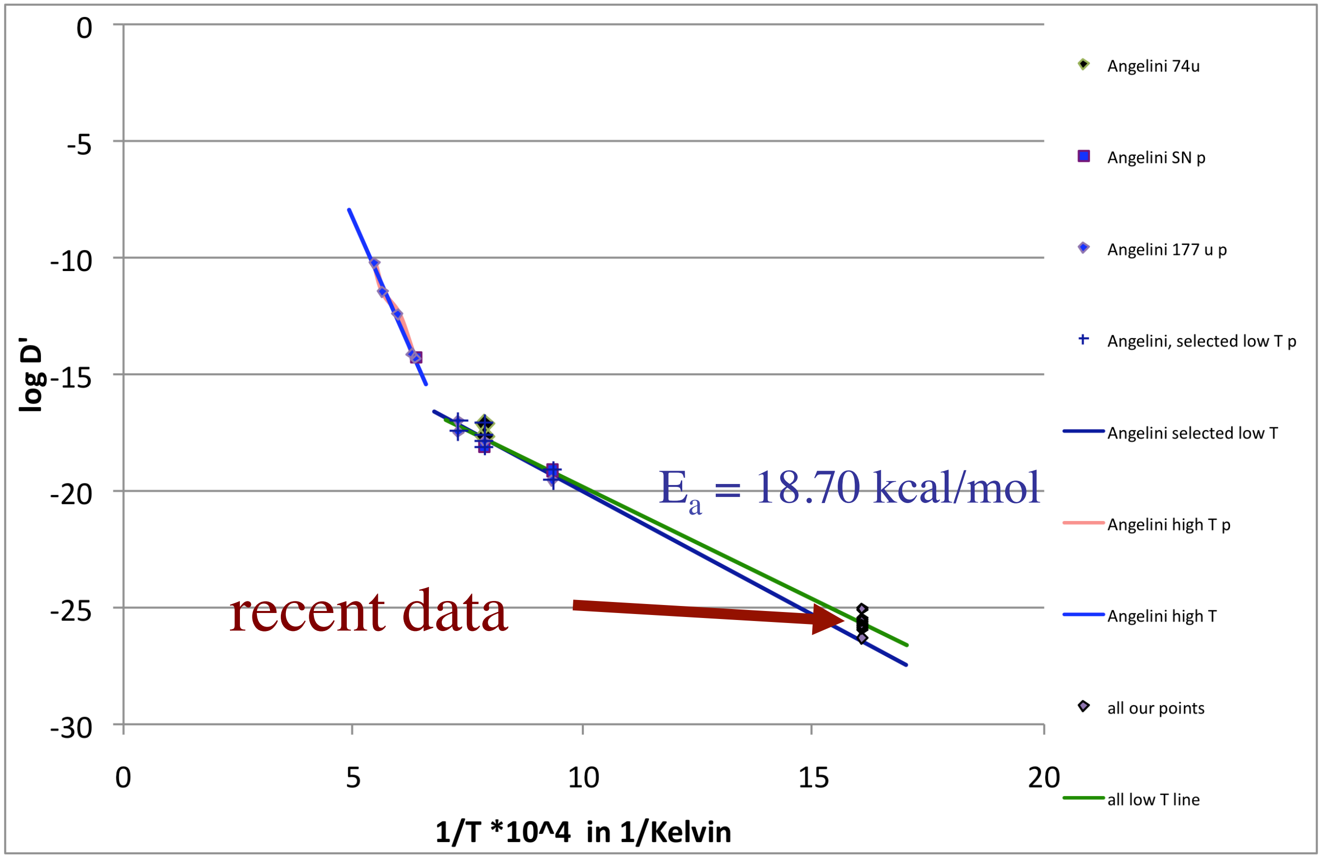
Arrhenius plot with 2011 data shows low-temperature diffusion exactly like 1970



Static data from microspheres 23-34 years old

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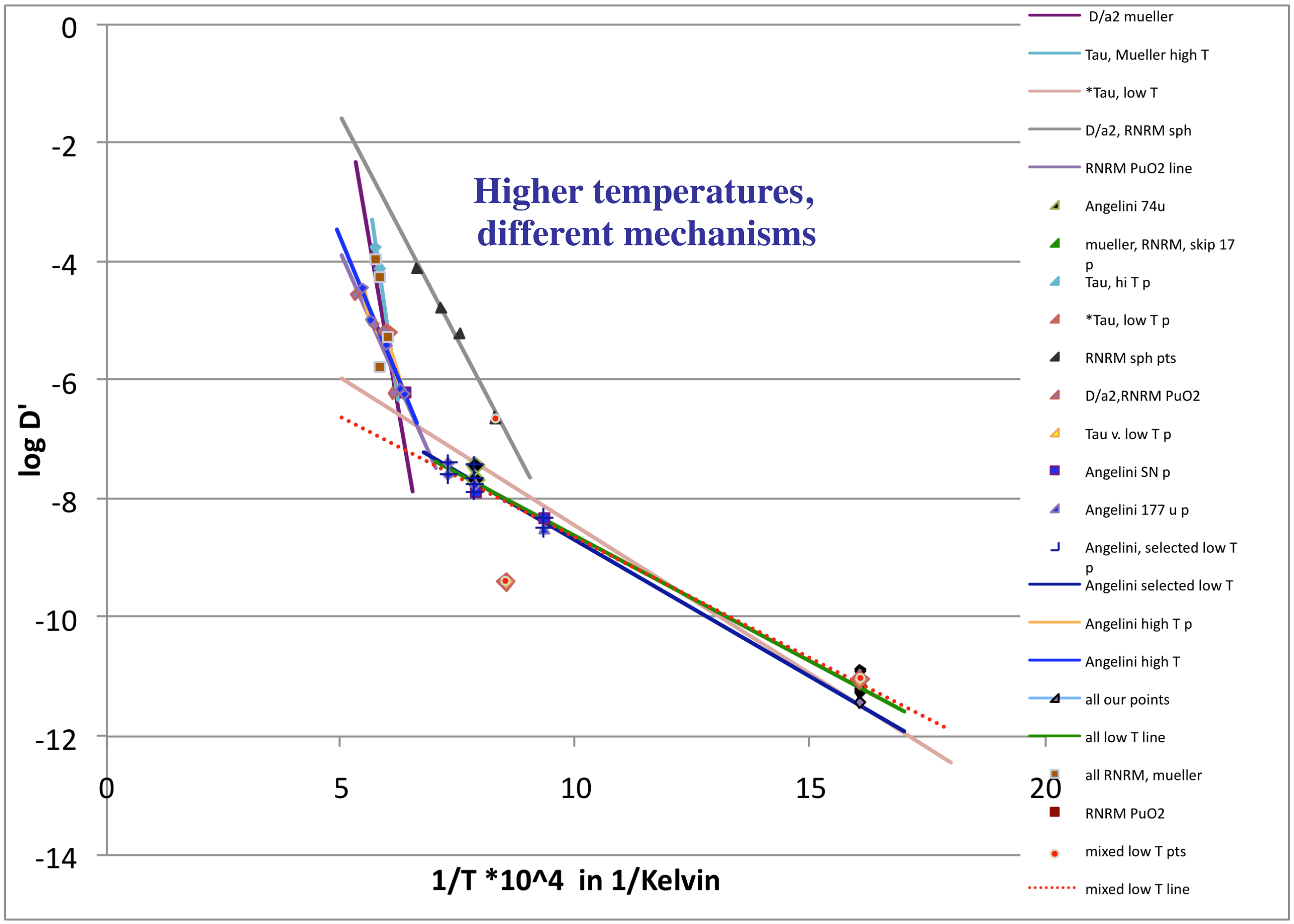
Plot with all available data gives same conclusion



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Arrhenius rates indicate mechanism is similar to previous

kcal/mole

E_a 21.06 for Angelini at low temperature (800°C, 1000°C, 1100°C)

E_a 18.70 for our measurement at low temperature (~350°C) plotted with Angelini's data

E_a 18.49 for our measurement (~350°C) plotted with Mueller 1397°C and Mueller 900°C

Activation energies reasonable for high T diffusion

E_a 123-150 Peterson high T — diffusion of bubbles in grains

E_a 87.09 Mueller Tau at high T

E_a 82.0 RNRM PuO_2

E_a 87.2 Angelini high T

diffusion of bubbles to grain boundaries
high activation energy,
above 80 kcal/mole

E_a 69.2 RNRM microspheres

E_a 49.9 – 52.9 Peterson medium temp

E_a 49.9 movement of oxygen vacancies in ThO_2

Ando, Oishi, et. al. J. Chem. Phys,65(7) 2751(1976)

activation energy consistent with diffusion of point defects

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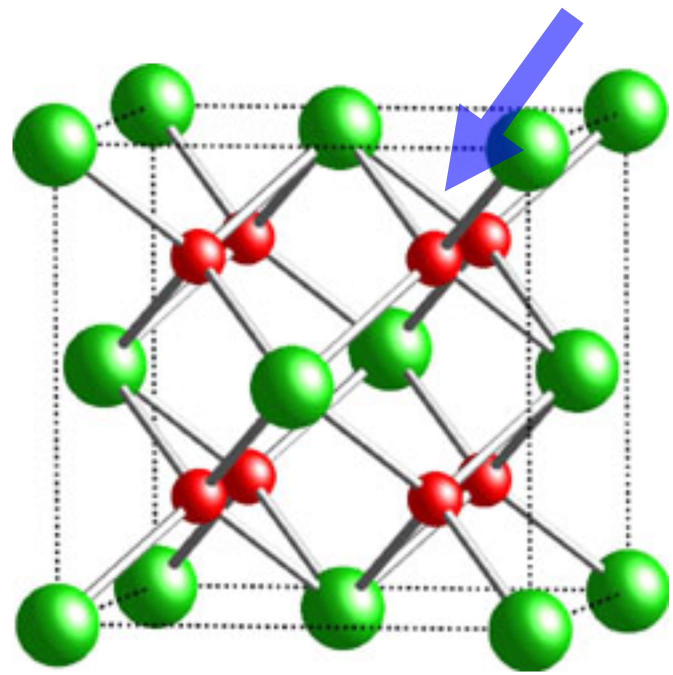
Plutonium matrix can absorb all helium produced

Helium is retained
In holes
in the lattice,

most likely
tetrahedral holes
from geometric
arguments



The fluorite structure has
exactly as many tetrahedral holes as plutonium atoms

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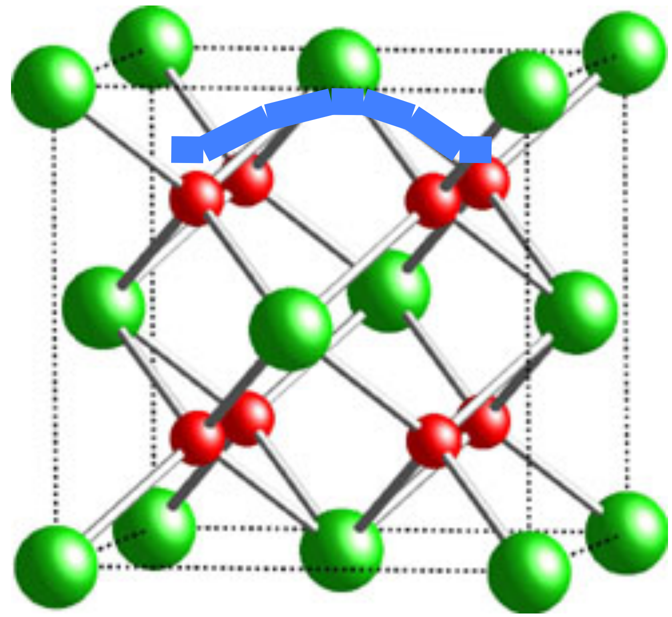
Open lattice allows helium diffusion at low energies



E_a = 18.70 kcal/mol

E_a = 20.3 kcal/mol
concerted
He movement in ErH_2
Wixom, et. al., JAP (2008)

E_a = 50 for point defect

Movement between holes requires only energy of site change
No energy of ion or point defect movement

Concerted He movement minimizes energy

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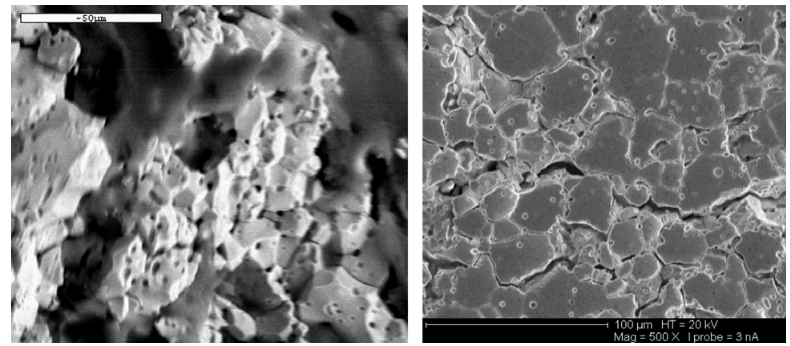
Morphology of the fuel may change

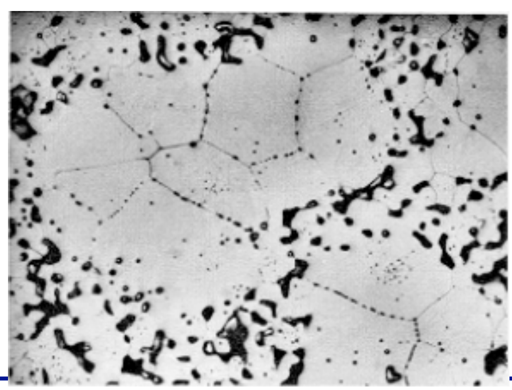
Rapid helium evolution pressurizes grain boundaries
Resulting in fracturing of microspheres

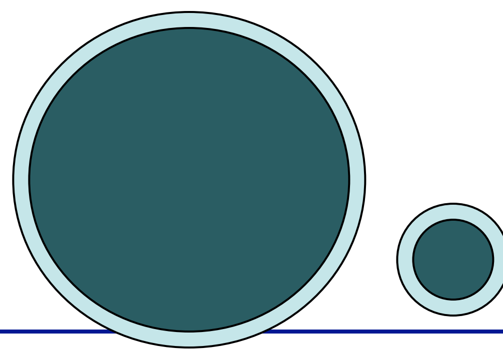
What is our morphology now?
CEA has seen fracturing of oxide:



Roudil (08)
CEA oxide
30 years old

Depleted zone at the edge of each grain:
If the grains get smaller, eventually the He will come out





Conclusion: no change in helium release from fuel

Current data indicates NO CHANGE in the mechanism of Helium release from plutonia at ages of up to 34 years.

The rate has NOT CHANGED appreciably in 34 years.

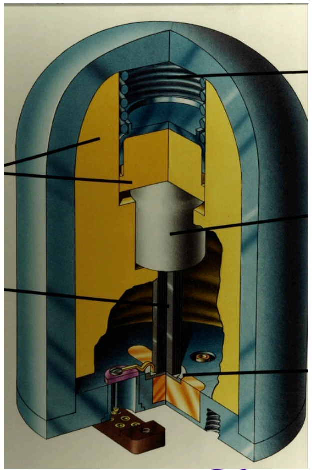
Preservation of chemistry: still all helium

Core surveillance: critical mission and a good reason for doing science

They're well-kept, but will they run?

(and where will you get leaded gas?)

If every part works, it's a reasonable expectation that the entire assembly will work



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DPA: Displacements Per Atom

Calculated atomic ppm helium is about 2000-2950 ppm
depending on unit age,
or about 0.0020 to 0.0029 grams helium per gram oxide.

Assuming 1500 defects per reaction in the ceramic*
the displacements per atom (DPA) in the solid
calculated to be between 3 dpa and 4.42 dpa.

Majority of units about 4 dpa of accumulated damage.

*Roudil et al, J. Nucl. Mats. *378,* pp. 70-78 (2008.)