Nuclear Fuel for Advanced Burner Reactors

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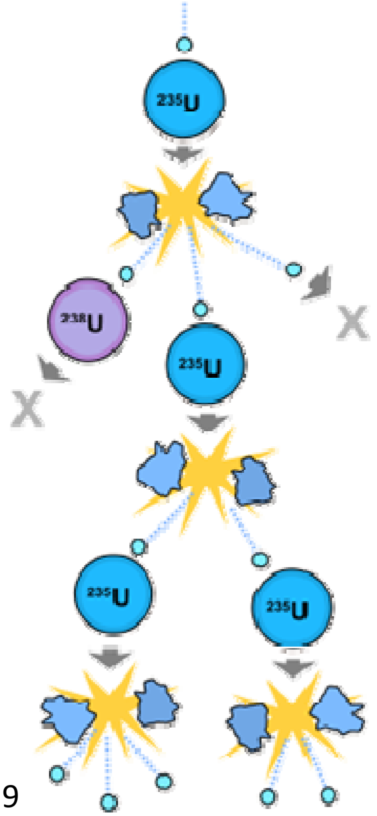
Nuclear Energy: Pros and Cons

- Nuclear power is a viable alternative to carbon-based energy
  - It has been pretty safe
  - It is ~0 CO_2
  - Mature technology
- There are some disadvantages
  - There is no long term solution for dealing with nuclear waste
  - Threat of nuclear proliferation
- Advanced burner reactors will be able to transmute transuranic elements produced in conventional light water reactors, turning waste into fuel

Great Let’s Do it…

- Wait a minute
  - – The behavior of this fuel is not well understood
  - – Failure can occur because noble gases are very insoluble in nuclear fuels such as uranium dioxide
    - These atoms tend to segregate and form bubbles in the fuel, which eventually lead to macroscopic swelling and material degradation
  - Processing of waste to use as fuel is tricky
  - Other design characteristics to worry about
    - – High burnup
  - It is expensive
- Let’s step through the process

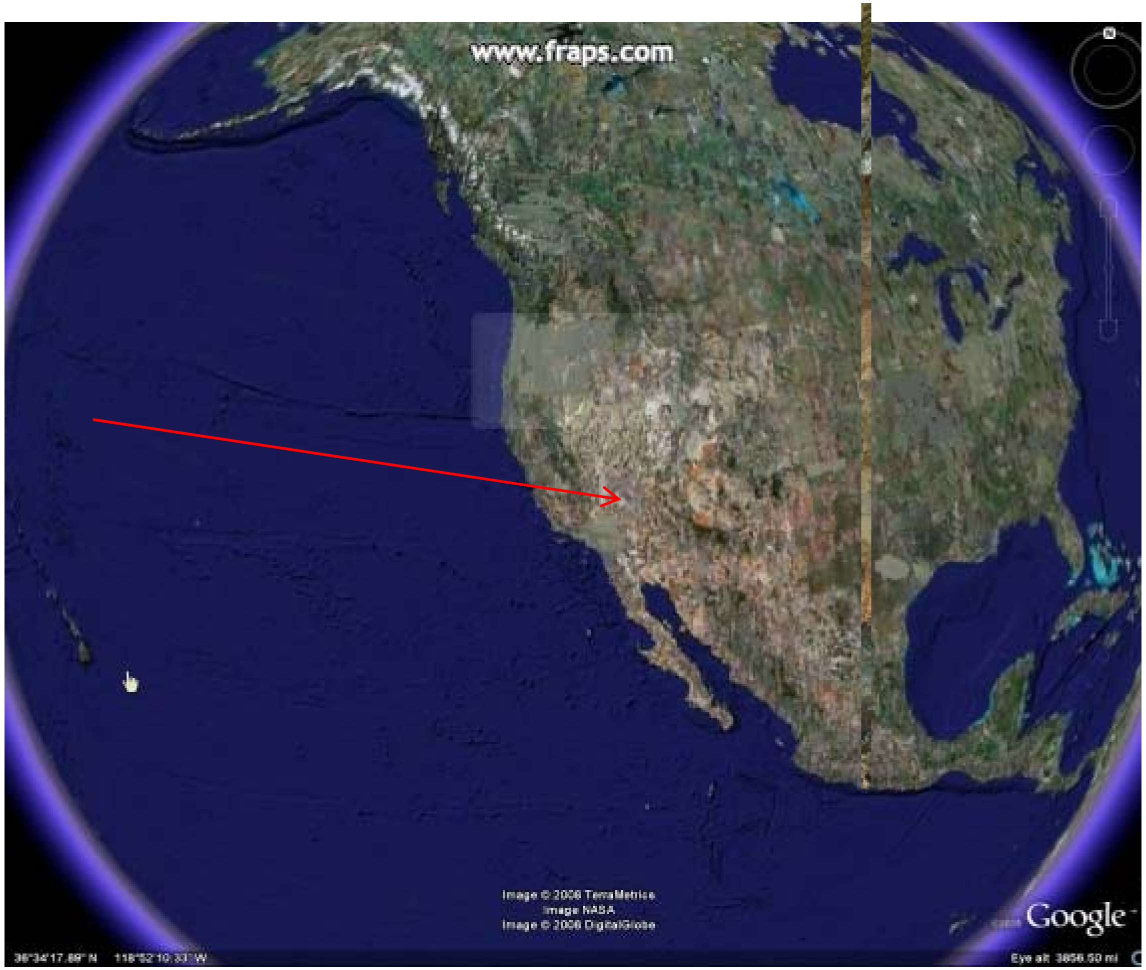
Light Water Reactor- The Open Cycle



- Mine of uranium ore
  - Contains 0.1% U_3O_8
- Converted to uranium hexafluoride
- Gas centrifuge is used to sort the uranium isotopes
  - 0.71% uranium-235
  - LWR operate with 3.5%
- Convert to uranium oxide (UO_2) powder
- Powder is pressed into pellets and sintered
- The pellets are stacked in the reactor core
- A neutron combines with a uranium-235 atom making it a uranium-236
- Uranium-236 spontaneously decays into fissile products and more neutrons
  - Chain reaction!!!
- These neutrons are so called fast neutrons
  - High energy
- If uranium-238 absorbs a neutron, no more the chain reaction
  - The uranium-239 will decay into neptumium-239 and will decay again into plutonium-239
- Using water as a moderator the neutrons are slowed to become so called thermal neutrons
  - Low energy
  - Less likely to interact with uranium-238
- The spent fuel has large amounts of uranium-238, plutonium-239, and even some uranium-235
- The current plan…

Yucca or BUST!

Yucca Mountain

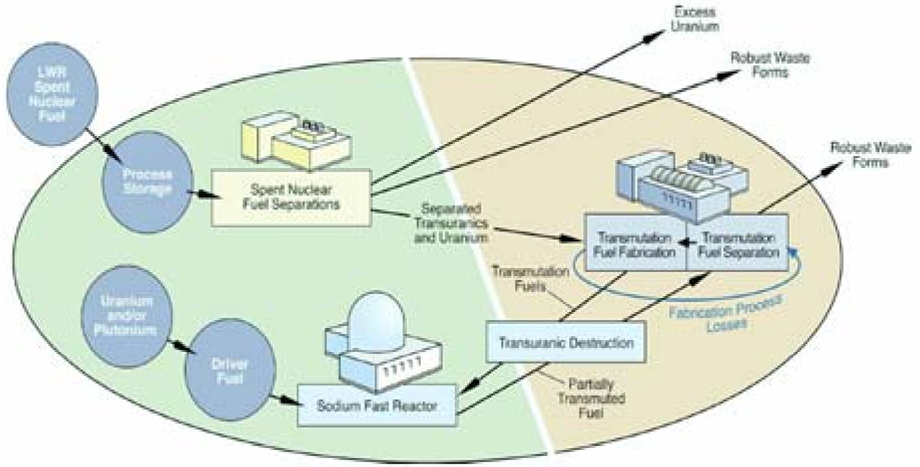


Bust?

- Limited capacity
  - As more nuclear power produced, need more Yuccas
- Yucca delayed
  - No current opening date!
- Store waste for thousands of years
- Lots of opposition
- Large amount of material that could still undergo fission, the spent fuel can be reprocessed in a closed cycle reaction

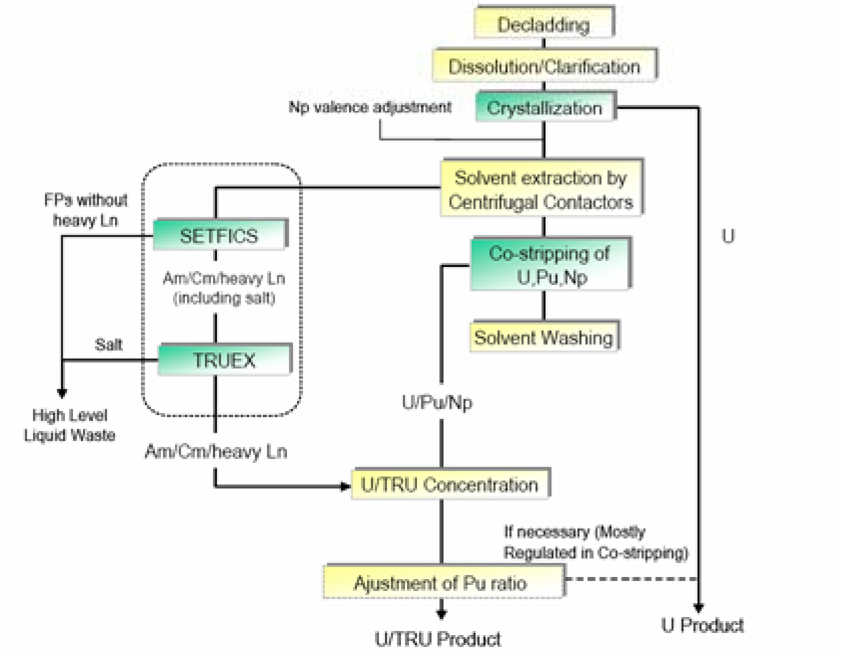
ABR- The Closed Cycle

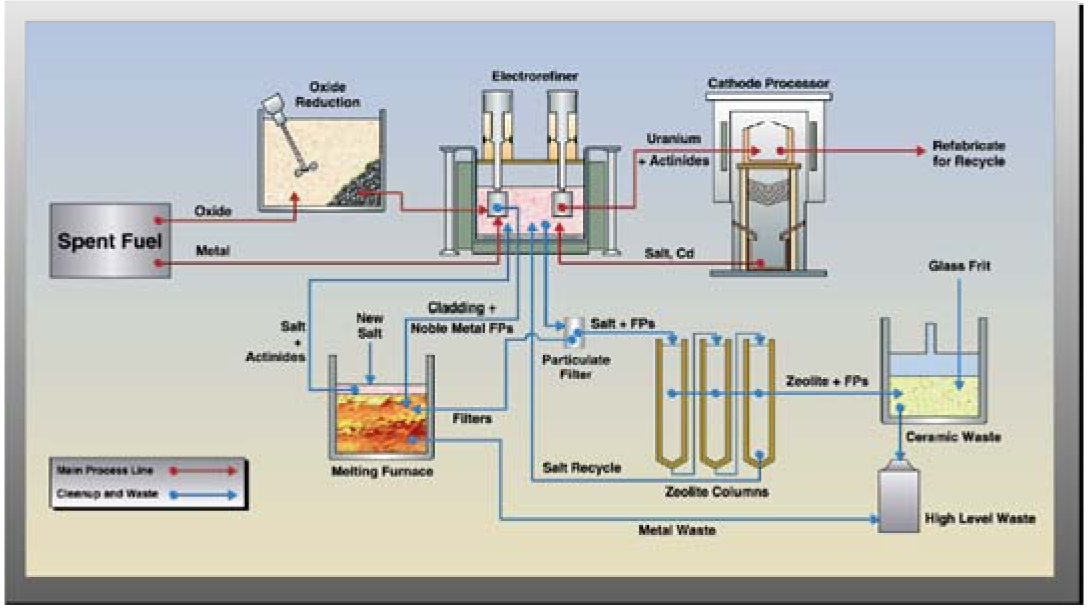
- Global Nuclear Energy Partnership (GNEP) is a program to develop a closed fuel cycle
  - To address the security and environmental concerns about nuclear waste
- First step is to process waste from LWR



Waste Processing

- Aqueous Processing (UREX+, PUREX)
- Pyrochemical Processing
- Iodine and technetium
  - Long half-lives → Yucca Mountain
- Cesium and strontium
  - Short half-lives → low level waste repositories (10s of years)
    - They would have to undergo decay storage until it could technically qualify
- Transuranic elements → reprocessing for advanced burner reactor fuel



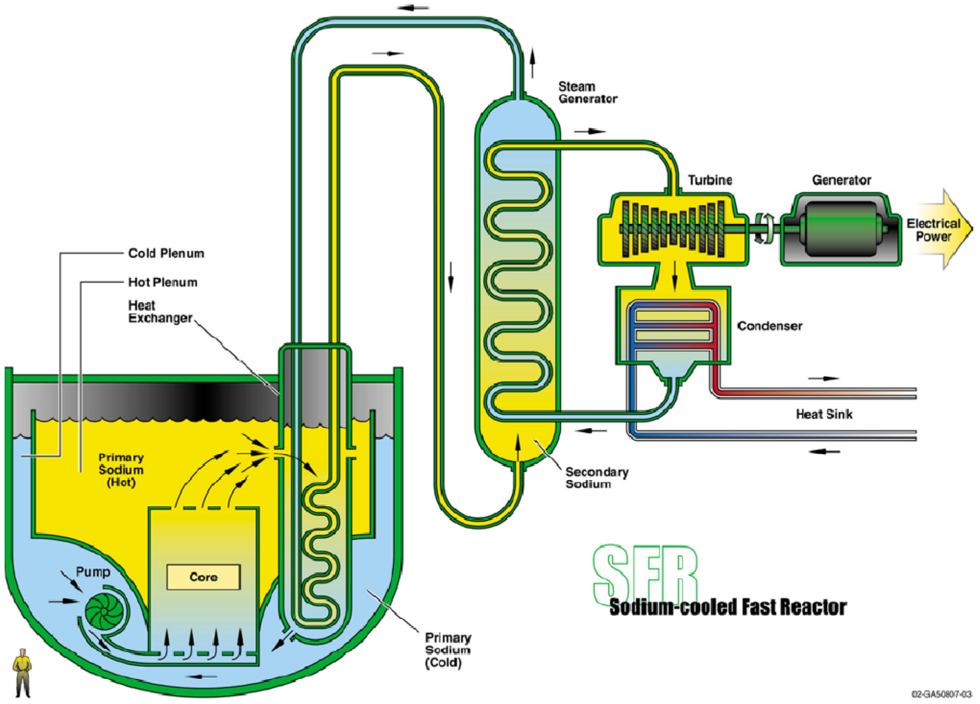


How to Turn Trash to Treasure

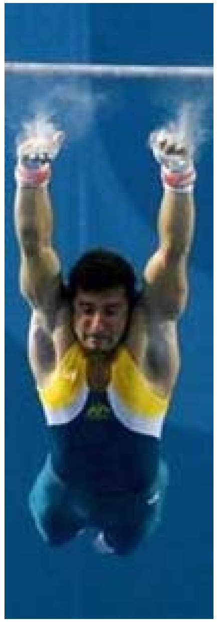
- Obviously need something more than LWR
- LWR used water as coolant and moderator
  - Thermal neutrons
- Need higher energy to transmute Np, Pu, Am, etc.
  - Fast neutrons!
- Can not use a water coolant any more
- Fast neutrons more likely to give nonproductive reactions (U-238)
  - Need higher enrichment

Sodium-Cooled Fast Reactor

- Water around the core replaced with liquid sodium
- Heat exchanger to a sodium loop
- Heat exchanger to a water loop
- Water turned to steam to turn a turbine



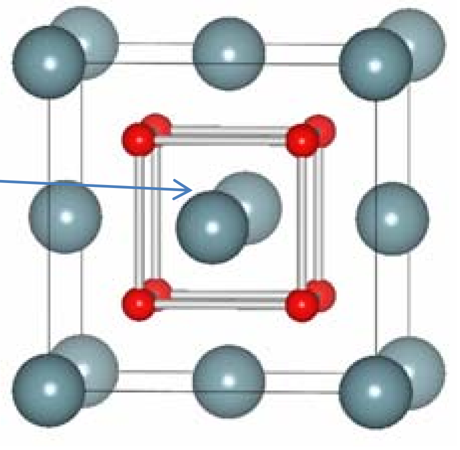
Failure

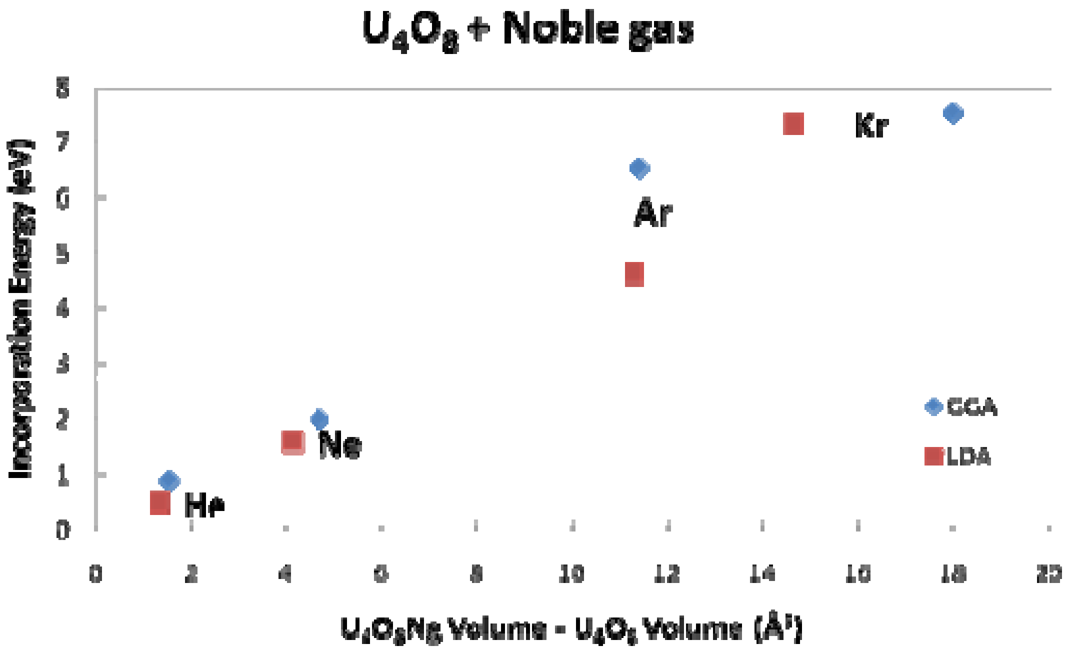


- Fuel-to-cladding chemical interaction (FCCI)
  - Eutectic melting between the fuel and the cladding
- U, Pu, and La (a fission product) interdiffuse with the iron of the cladding
  - The alloy that forms has a low eutectic melting temperature
  - FCCI causes the cladding to reduce in strength
    - Eventual rupture
- Noble gases are very insoluble in nuclear fuels
  - Segregate and form bubbles in the fuel
    - Leads to macroscopic swelling and material degradation

Noble Gas Atoms in U_4O_8

He, Ne, Ar, Kr, placed in the octahedral interstitial site of fluorite urania unit cell





Einc (eV)	He	Xe
UO2	-0.1	11.2
PuO2	0.4	-
AmO2	1.1	-
(Am0.5Pu0.5)O2	0.7	-

No ‘+U’, Nonmagnetic

M. Freyss et al. Journal of Nuclear Materials 352 (2006) 144–150

Very large incorporation energies: consistent with lack of solubility of noble gases, as well as previous calculations of Xe in UO_2

Summary

- Nuclear power is great
  - But serious drawbacks
- Closed cycle fission can alleviate issues of waste
- Processing of waste is difficult
- In the reactor, fast neutrons are used
  - No moderator
- The environment is harsh and materials not well understood
  - Hard to model

References

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[4] D. Hill, "Global Nuclear Energy Partnership Technology Demonstration Program", Nuclear Physics and Related Computational Science R&D for Advanced Fuel Cycles Workshop, 2006

[5] J. J. Laidler, "The Advanced Fuel Cycyle Initiative of the U.S. Department OF Energy: Development of Separations Technologies", WM'04 Conference, 2004

[6] M. J. Lineberry and T. R. Allen, Argonne National Laboratory "The Sodium-Cooled Fast Reactor (SFR)"

[7] George F. Vandegrift et al., "Designing and Demonstration of the UREX+ Process Using Spent Nuclear Fuel", ATATLANTE 2004

[8] S. Bays, M. Pope, B. Forget, R. Ferrer, "Transmutation Target Compositions in Heterogeneous Sodium Fast Reactor Geometries", INL/EXT-07-13643 Rev. 1, 2008

[9] D. R. Olander, Fundamental Aspects of Nuclear Reactor Fuel Elements, National Technical Information Service, Springfield Virginia, 1976

[10] N. G. Jensen, M. D. Asta, C. Wolverton, A. van de Waale, V. Ozolins, "Radiation Damage in Nuclear Fuel for Advanced Burner Reactors: Modeling and Experimental Validation", a Research Proposal

Strain Due to Noble Gases (LDA)

No O displacements

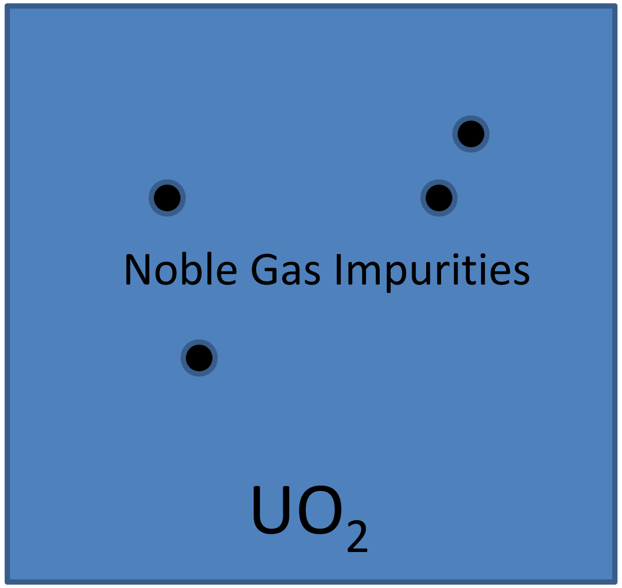
Interstitial Occupancy	$E(U_4O_8Ng)$ –$E(U_4O_8)$ –E(Ng) (eV/unit cell)	E(Distorted U_4O_8)–$E(U_4O_8)$ (eV/unit cell)	ΔE (Strained UO_2) (eV/unit cell)	$\Delta E=K/2V_o*\Delta V^2$ (eV/unit cell)
He	0.487	0.013	0.006	0.007
Ne	1.589	0.075	0.064	0.069
Ar	4.642	0.764	0.469	0.517
Kr	7.366	3.044	0.769	0.867

Still insulating, no longer AFM → NM ⟶

- $E(U_4O_8Ng)$ –$E(U_4O_8)$ –E(Ng) → Energy of urania with interstitial gas atom minus the energy of pure urania minus energy of isolated gas atom
- E(Distorted U_4O_8)–$E(U_4O_8)$ → Energy of injected urania with interstitial gas atom removed and the ions frozen minus the energy of pure urania
- ΔE (Strained UO_2) → Energy of urania strained to equivalent volumes of urania with injected defects
- $\Delta E=K/2V_o*\Delta V^2$ → Calculated energy of hydrostatically strained UO_2 with K=209 GPa

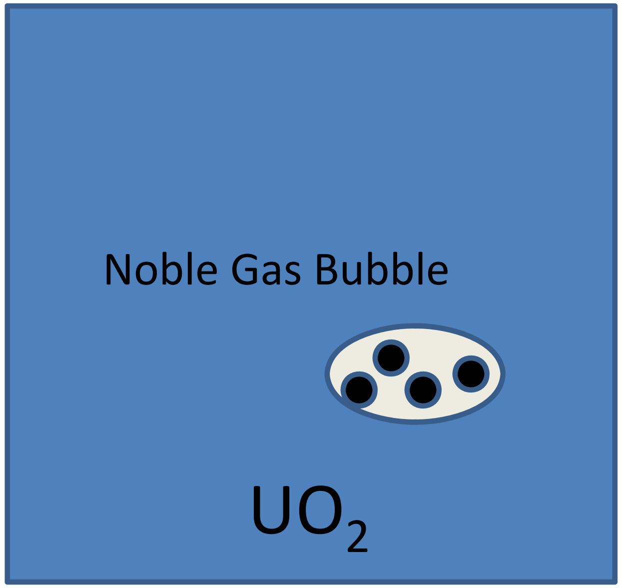
The effect of a gas atom in urania is not purely hydrostatic strain

Simple Model of Noble Gas Bubbles



Noble Gas Impurities

UO_2



Noble Gas Bubble

UO_2

Energetic Cost: Strain induced by Impurities

Energetic Cost: Creation of Bubble
Energetic Gain: Relief of strain energy

At what point does the bubble become energetically favorable?

Energetics of Bubble Formation – Simple Model

Consider a Schottky defect as the smallest possible bubble

Balance energy cost of Schottky vs. Strain relief of impurities

Schottky defect volume ~ 40.4 Å^3

ΔE > Schottky Formation energy (~7.2 eV [+])

	Volume (Å^3)	# Atoms/Schottky D.	$E(U_4O_8Ng) - E(U_4O_8) - E(Ng)$ (eV)
He	1.4	29.8	14.543
Ne	4.1	9.8	15.498
Ar	11.3	3.6	16.569
Kr	14.6	2.8	20.281

- We can do a “thought experiment” calculation to find the energy of a small bubble of gas atoms that fill a Schottky defect
- We take the volume of the Schottky defect as one fourth of the U_4O_8 unit cell
- Take the ΔV between the pure oxide and the defect injected cell
- Calculate how many gas atoms will fit into the Schottky defect strain free (neglecting packing fraction, etc.)
- Multiply the number of atoms with the strain energy of an individual gas atom in UO_2
- This is the amount of energy recovered by allowing the gas atoms to cluster strain free
- The energy to create a Schottky defect is about ~7.2 eV [+]
- The critical number of atoms for nucleation of a bubble is very small
  - It may be more energetically favorable to create a Schottky defect and let the gas atoms cluster in a lower strain environment

[+]Gupta et. al. Philosophical Magazine, 87, No. 17, (2007), 2561–2569