FUKUSHIMA

THE STORY
OF A NUCLEAR
DISASTER

DAVID LOCHBAUM, EDWIN LYMAN, SUSAN Q. STRANAHAN
AND THE UNION OF CONCERNED SCIENTISTS
FUKUSHIMA
THE STORY OF A NUCLEAR DISASTER

David Lochbaum, Edwin Lyman, Susan Q. C. Ng, and the Union of Concerned Scientists

CLIMATE CHANGE 2013
The Physical Science Basis

WORKING GROUP I CONTRIBUTION TO THE FIFTH ASSESSMENT REPORT OF THE INTERGOVERNMENTAL PANEL ON CLIMATE CHANGE
e = mc²
Nuclear Fuel Cycle vs. Carbon Cycle
Pu vs. C

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Center for International Security and Cooperation
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Energy Frontier Research Center
Materials Science of Actinides

*This presentation is not an official position of the NWTRB.
Views expressed are my own.

Precourt Energy Seminar
Stanford, CA
June 2, 2014
Carbon Cycle
Carbon Cycle

Nuclear Fuel Cycles
Fission

fission ($^{235}\text{U}$ and $^{239}\text{Pu}$):

$^{235}\text{U} + {}^1\text{n}_o \rightarrow \text{fission fragments} + \text{extra neutrons} + \text{energy!}$

neutron capture and $\beta$-decay:

$^{238}\text{U} + {}^1\text{n}_o \rightarrow ^{239}\text{U} \rightarrow ^{239}\text{Np} \rightarrow ^{239}\text{Pu} + \text{minor actinides}$
Nuclear Fuel Cycles
Nuclear Fuel Cycles vs. Geochemical Cycles

- Reactor
  - Reprocessing
  - Enrichment
  - Mining
    - Storage
      - Release
        - Transport
          - Dilution or Concentration

- Repository

- $^{129}\text{I}$, $^{99}\text{Tc}$
- $^{237}\text{Np}$, $^{239}\text{Pu}$
- $^{238}\text{U}$, $^{226}\text{Ra}$, $^{222}\text{Rn}$, $^{40}\text{K}$

- <3 mrem
- <300 mrem
“Symbiotic” Fuel Cycle Strategy

courtesy Department of Energy
Carbon Cycle

Geochemical Cycles

Nuclear Fuel Cycles

Nuclear Weapons
Uranium/Plutonium Fuel Cycle

courtesy Ron Fleming
Plutonium Inventories in 2010

in spent nuclear fuel (70-100 mt/yr) 1,700 mt
In operating reactors 80 mt
separated by reprocessing of “civil” SNF 250 mt
(France, 73 mt; UK, 60 mt; Russia, 30 mt; Japan, 24 mt)

military inventories:
  Russia 140 mt
  USA 100 mt
  others 15 mt

Estimated world total: ~ 2,300 metric tonnes
Plutonium vs. Carbon
plutonium vs. carbon

Plutonium “... is a clear and present danger to national and international security.”

Plutonium vs. Carbon

“Warming of the climate system is unequivocal, and since the 1900s, many of the observed changes are unprecedented over decades to millennia.”

WG1 IPCC Fifth Assessment Report (2013)
Nuclear Power & Carbon Reduction in 2000

number of NPP (U.S. = 103)  433
power generation (GW-year/yr)  350
fraction of electricity production (%)  17

carbon reduction (Gt/yr)  0.5
Energy Production: 2050

population 9 billion

total primary energy (EJ/yr)* 900
contribution from NPP (EJ/yr)** 300
fraction of electricity production (%) >50

*50% increase in per capita energy consumption
**3,300 GW-year of output annually (GW-yr = single NPP)

Energy Production: 2050

3,300
power plants

~ 70,000 metric tonnes
spent nuclear fuel per year

~ 700 metric tonnes
Pu per year
Strategies for the Disposition of Pu

- **Nuclear:** Use nuclear reactors to “burn” or reduce inventories of plutonium and “minor” actinides in MOX or inert matrix fuels.

- **Geologic:** Direct disposal of spent nuclear fuel and/or separated actinides in durable solids.
Actinides: Geologic Disposal Issues

- radioactivity
- radiotoxicity
- decay heat
- geochemical mobility
  - radiolysis and redox conditions
  - solution chemistry
  - solid-state chemistry
  - sorption and colloids
Radioactivity over Time

Radiotoxicity over Time

Redox Effects on Geochemical Mobility

Sorption & Colloid-Facilitated Transport

Novikov et al. (2006) Science
100,000-year Dose (at ~20 km)

Yucca Mountain Viability Assessment (1998)
Geologic Disposal of Actinides in Clay

Multi-Barrier System
Defense-in-Depth
Actinide Waste Forms

- Near-field containment.
- Long-term behavior can be modeled.
- Natural “analogues” can be used to verify extrapolated behavior.
- With chemical processing waste form can be designed to match the waste stream and the geochemical conditions of disposal.
# Actinide Waste Forms

<table>
<thead>
<tr>
<th>Category</th>
<th>Example</th>
<th>Formula</th>
</tr>
</thead>
<tbody>
<tr>
<td>Simple oxides</td>
<td>Zirconia</td>
<td>$\text{ZrO}_2$</td>
</tr>
<tr>
<td>Complex oxides</td>
<td>Pyrochlore</td>
<td>$(\text{Na, Ca, U})_2(\text{Nb, Ti, Ta})_2\text{O}_6$</td>
</tr>
<tr>
<td></td>
<td>Murataite</td>
<td>$(\text{Na, Y})_4(\text{Zn, Fe})_3(\text{Ti, Nb})<em>6\text{O}</em>{18}(\text{F, OH})_4$</td>
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<tr>
<td></td>
<td>Zirconolite</td>
<td>$\text{CaZrTi}_2\text{O}_7$</td>
</tr>
<tr>
<td></td>
<td>Perovskite</td>
<td>$\text{CaTiO}_3$</td>
</tr>
<tr>
<td>Silicates</td>
<td>Zircon*</td>
<td>$\text{ZrSiO}_4$</td>
</tr>
<tr>
<td></td>
<td>Thorite*</td>
<td>$\text{ThSiO}_4$</td>
</tr>
<tr>
<td></td>
<td>Garnet*</td>
<td>$(\text{Ca, Mg, Fe}^{2+})_3(\text{Al, Fe}^{3+}, \text{Cr}^{3+})_2(\text{SiO}_4)$</td>
</tr>
<tr>
<td></td>
<td>Britholite</td>
<td>$(\text{Ca, Ce})_5(\text{SiO}_4)_3(\text{OH, F})$</td>
</tr>
<tr>
<td></td>
<td>Titanite</td>
<td>$\text{CaTiSiO}_5$</td>
</tr>
<tr>
<td>Phosphates</td>
<td>Monazite*</td>
<td>$\text{LnPO}_4$</td>
</tr>
<tr>
<td></td>
<td>Apatite*</td>
<td>$\text{Ca}<em>{4-x}\text{Ln}</em>{6+x}(\text{PO}_4)_y(\text{O, F})_2$</td>
</tr>
<tr>
<td></td>
<td>Xenotime*</td>
<td>$\text{YPO}_4$</td>
</tr>
</tbody>
</table>

* indicates durable heavy minerals

Radiation Damage

**Alpha-Decay Event**

Recoil Nucleus

0.07 – 0.09 MeV
10 nm

boom!

Alpha-Particle

4.5 – 5 MeV
10,000 nm

Ewing et al. (2000) RIMG, vol. 39
Radiation Damage

Ewing et al. (2000) RIMG, vol. 39
Alpha-decay Event Radiation Damage

Ewing et al. (2000) RIMG, vol. 39
Alpha-decay Event Radiation Damage

Zircon: ZrSiO$_4$

Chakoumakos, Murakami, Lumpkin and Ewing (1987) *Science*
Zircon: $\text{ZrSiO}_4$

Zircon: \( \text{ZrSiO}_4 \)


zircon photographs courtesy of John Hanchar
Pyrochlore

\[ A_{1-2}B_2O_6(O,OH,F)_{0-1} \, pH_2O \]

\[ (Ca,Na,U,REE)_2(Nb,Ta,Ti)_2O_6(OH,F) \]

\[ (Gd,Pu,U,Hf)_2Ti_2O_7 \]

Damage Accumulation

Damage Accumulation

Ion Beam Irradiation
IVEM-Tandem Facility at Argonne National Lab.

Radiation-induced Amorphization:
\( \text{Gd}_2(\text{Zr}_x\text{Ti}_{1-x})_2\text{O}_7 \) (1 Mev Kr+)

\( x=0, \) crystalline
\( x=0.25 \)
\( x=0.5 \)
\( x=0.75, \sim 30\% \) amorphous
\( x=1, \) crystalline

Radiation-induced Amorphization: 
\( \text{REE}_2\text{Ti}_2\text{O}_7 \) (1 Mev Kr+)

![Graph showing the radiation-induced amorphization of different REE$_2$Ti$_2$O$_7$ compounds.](image)

Radiation-induced Amorphization:

$\text{REE}_2\text{Ti}_2\text{O}_7$ (1 Mev Kr+)


Lumpkin (2006) *Elements*
Radiation-Resistant Materials for Plutonium Immobilization
(From Current Gadolinium Titanate to Stable Gadolinium Zirconate)

Equivalent Storage Time (years)

Gd₂Ti₂O₇ (10 wt% Pu)
Amorphous

Predicted Behavior From Systematic Studies

Gd₂Zr₂O₇ (10 wt% Pu)

Plutonium Release Rate Increased by 50x in Amorphous Gd₂Ti₂O₇

Plutonium Immobilized for Millennia in Stable Structure of Gd₂Zr₂O₇

RC Ewing, SX Wang, LM Wang, KV Govidan Kutty (IGCAR) University of Michigan
WJ Weber, BD Begg (ANSTO) Pacific Northwest National Laboratory
World Total Nuclear Warheads

- NPT
- India
- START I
- START II
- START III
- START I, II, III: Strategic Arms Reduction Treaties

Number of Warheads:
- USSR
- GB
- France
- China (Aboveground Test Ban Treaty)

Year:
- 1945
- 1950
- 1955
- 1960
- 1965
- 1970
- 1975
- 1980
- 1985
- 1990
- 1995
- 2000

Los Alamos

Nuclear Materials Technology
Excess Weapons Pu in the United States

1993 – President Clinton issues Nonproliferation and Export Control Policy.
1994 – NAS review and the “spent fuel standard”.
1995 – U.S. declared 38.2 MT of weapons grade Pu and 14.3 MT of non-weapons grade Pu as no longer needed.
1997 – DOE reviews 37 options; adopts a high-bred, dual-path strategy of MOX fuel in LWR and immobilization by can-in-can configuration.
2000 – United States-Russia Plutonium Management and Disposition Agreement confirming the use MOX in LWR; some US Pu would be disposed of by immobilization.
2002 – Bush administration canceled the immobilization option; life-cycle cost estimated at $3.8 billion dollars over 20 years.
2007 – Consolidation of Pu-holdings across the DOE complex; additional 9 MT declared as surplus; construction of MOX plant at Savannah River begins.
2011 – PMDA renegotiated and approved by Russian Duma and U.S.
2014 – Life-time cost of MOX plant at Savannah River (~60% complete) estimated at ~$30 billion; MOX plant construction may be placed on “cold standby, although construction continues during 2014.
2014 – DOE reviews other options.
Excess Weapons Plutonium: Five Options

- Irradiation of MOX fuel in Light Water Reactors
- Irradiation of Plutonium Fuel in Fast Reactors
- Immobilization (ceramic or glass form) with High-Level Waste
- Down-blending and Disposal
- Deep Borehole Disposal

Report on the Plutonium Disposition Working Group, April, 2014
Excess Weapons Plutonium: Five Options

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Excess Weapons Plutonium: Criteria for Options

- meeting international commitments
- cost
- duration to begin disposition and complete 35 MT mission
- technical viability
- legal, regulatory and other issues

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Excess Weapons Plutonium:
Criteria for Options

- meeting international commitments
- cost
- duration to begin disposition and complete 35 MT mission
- technical viability
- legal, regulatory and other issues [e.g. materials science and geochemistry]

Report on the Plutonium Disposition Working Group, April, 2014
THANKS