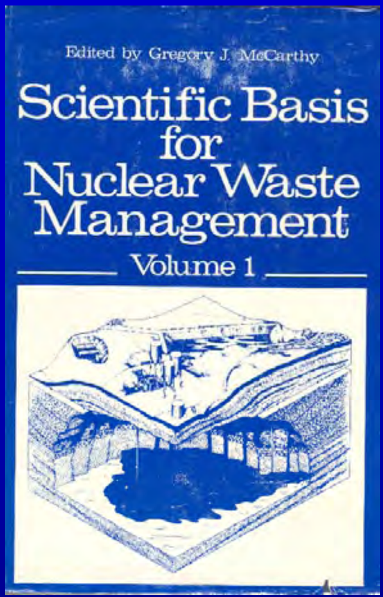
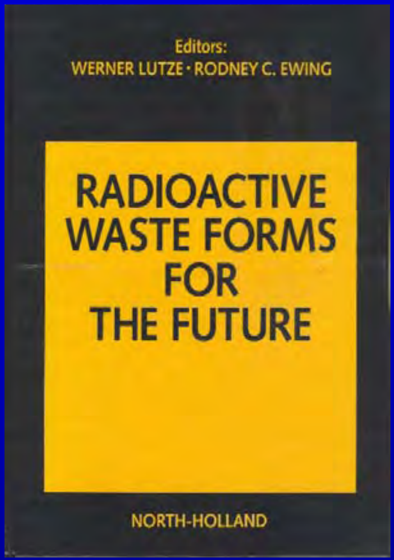
Waste Forms & Repositories





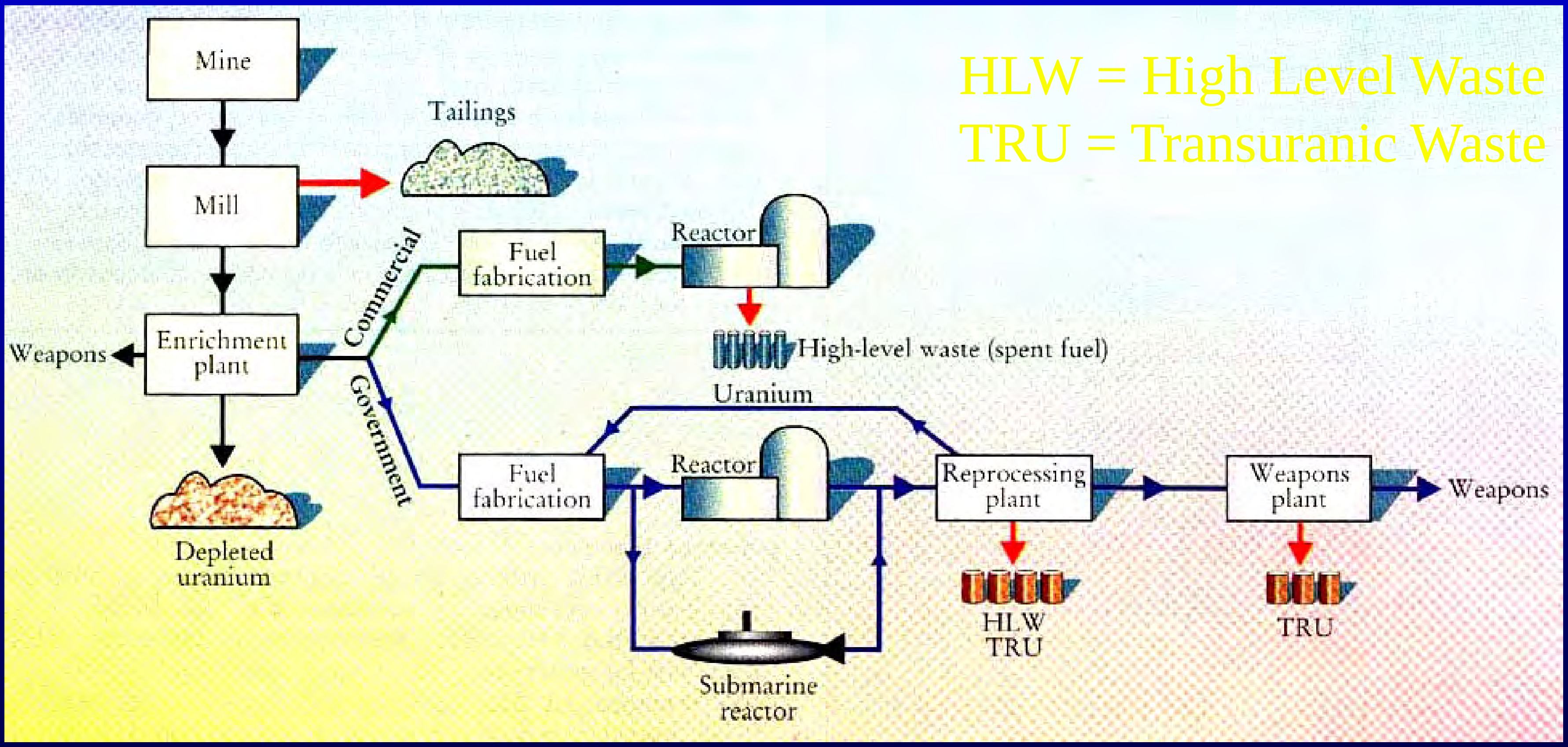
Rod Ewing

Geological Sciences
Materials Science & Engineering
Nuclear Engineering & Radiological Sciences

University of Michigan

Nuclear Integration Project Workshop:
"The Back-end: Healing the Achilles Heel of the Nuclear Renaissance?"
Vanderbilt University
March 3, 2008

The Nuclear Fuel Cycle



John Ahearne (1997) *Physics Today*

Nuclear Wastes in U.S.: 2010

Uranium mine & mill tailings	438 million m^3 3,000 MCi
Depleted uranium (UF_6)	500,000 metric tonnes
High-level Waste (defense reprocessing)	380,000m^3 2,400 MCi
Buried waste (TRU, LLW, hazardous)	6.2 million m^3
Spent Nuclear Fuel (commercial)	61, 800 mTHM 39,800 MCi
Contaminated soil	79 million m^3
Contaminated water	1,800 to 4,700 million m^3

Clean-up and disposal cost = 300 billion US dollars

Plutonium Inventories in 2000

in spent nuclear fuel (70-100 tons/year) 1,000 mT

in operating reactors 80 mT

separated by reprocessing of 'civil" SNF 210 mT

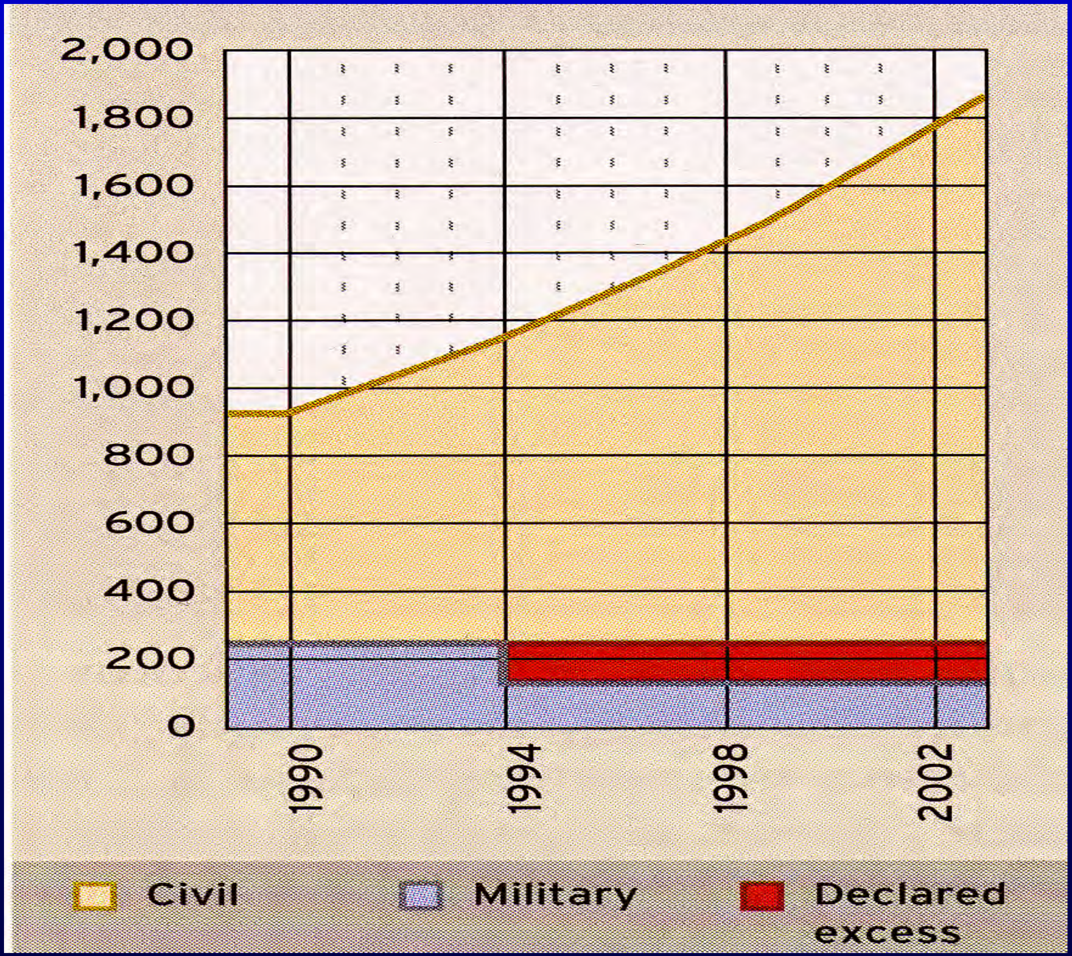
(France, 73 MT; UK, 60 MT; Russia, 30 MT; Japan 24 MT)

military inventories:

Russia	140 mT
USA	100 mT
others	15 mT

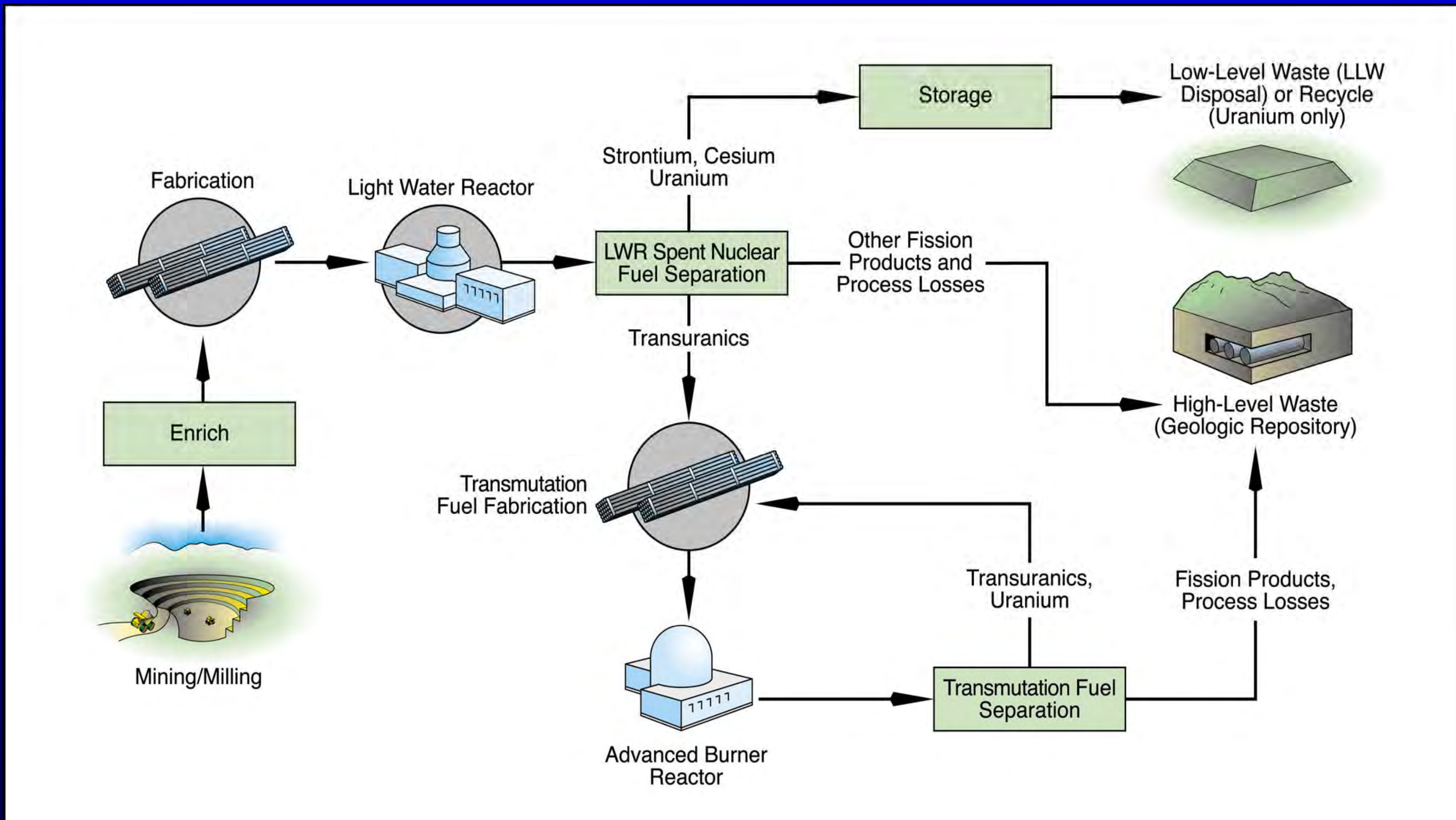
Estimated World Total 1,545 metric tons

Growth of "Civil" Pu-Inventory

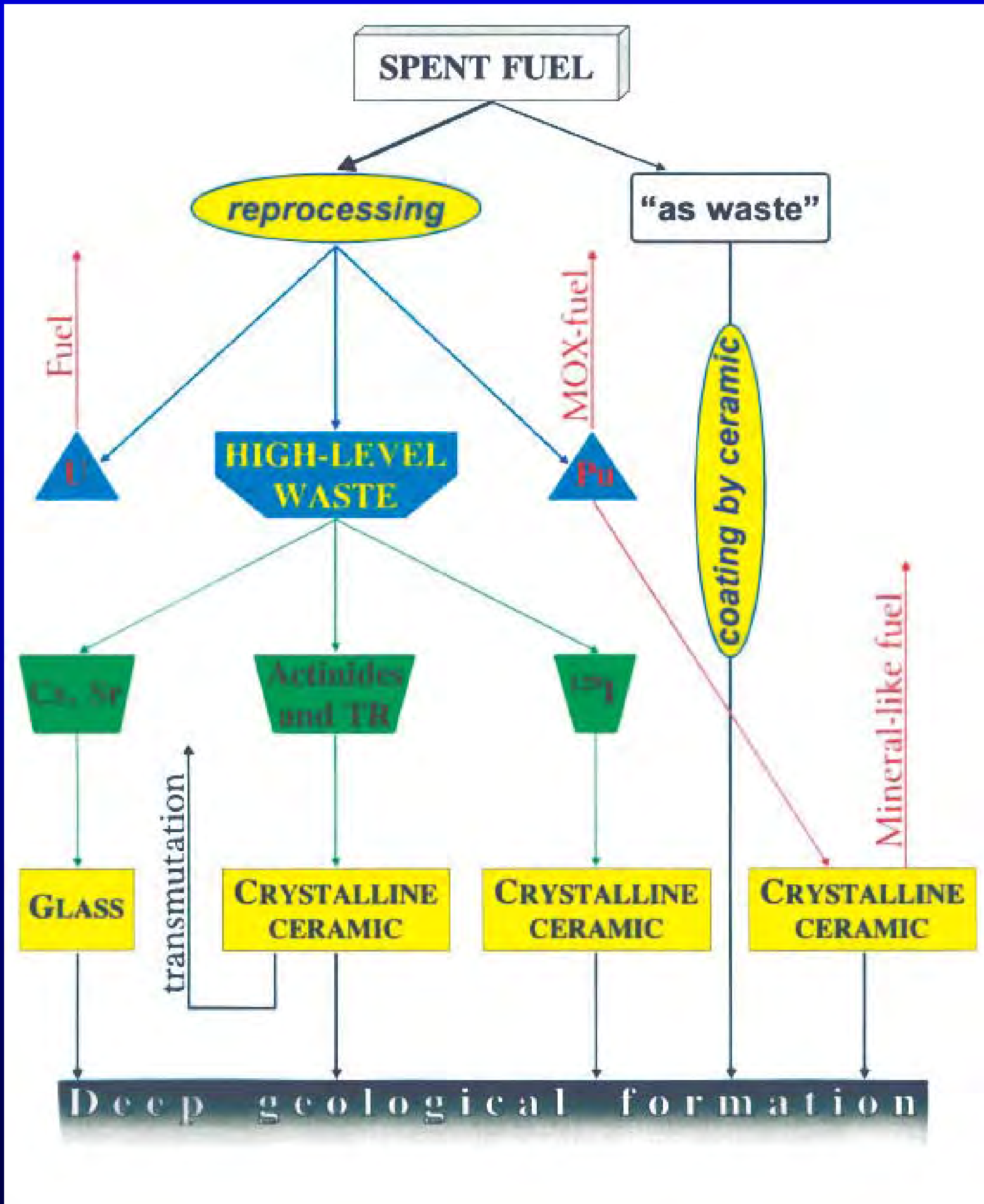


Albright and Kramer (2004) *Bulletin of the Atomic Scientists*

GNEP: Closed Fuel Cycle Strategy



courtesy Department of Energy



courtesy of Boris Burakov

Characteristics of the Waste Stream

radionuclide
radiotoxicity
half-life
crystal chemistry
geochemistry
characteristics of the repository

<u>Why is the waste form important?</u>

- **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 <u>designed</u> to match waste stream***

Challenges for Waste Forms

- **design**
- **development**
- **synthesis**
- *evaluate performance*

Designing Nuclear Waste Forms

- ✓ Radionuclide should guide the selection of the solid phase.
- ✓ If long-term durability is important, let Nature guide.
- ✓ Chemical durability is not only a matter of experimental leach rate, but also depends on the corrosion mechanism and the specific geologic setting.
- ✓ For crystalline waste forms, radiation-induced transformations may have a profound effect on chemical and mechanical properties. This requires:
  - ✓ the study of natural U- and Th-samples of great age;
  - ✓ well controlled experiments using ion beam irradiations;
  - ✓ accelerated experiments, e.g., ^{238}Pu and ^{244}Cm

Typical Requirements for Waste Forms

- match the waste stream composition
- high waste loading
- easy processing
- chemical flexibility
- durability
- radiation stability
- natural analogues

Science & Technology Plan: Three Tiers

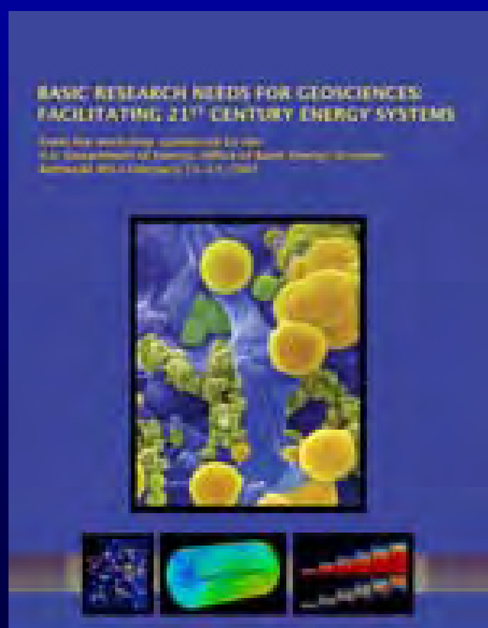
- Immediate Research Needs
- Long-term Research Plan
- “Integration by Simulation”:
  Properties and Performance

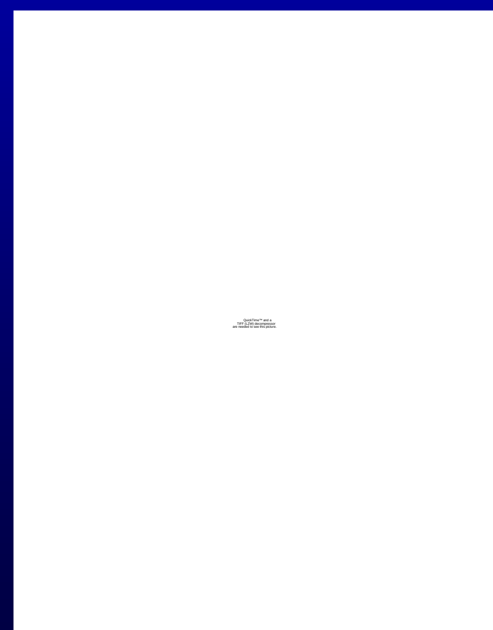
Immediate Research Needs

- short-lived fission products
- long-lived fission products
- complex waste streams
- off-gas radionuclides
- grouts for low level waste streams
- low activity waste streams containing mixed and hazardous chemicals

Long-Term Research Plan

- "Use-inspired" research of specific materials
- Cross-cutting fundamental research





“Use-inspired” Research of Specific Materials

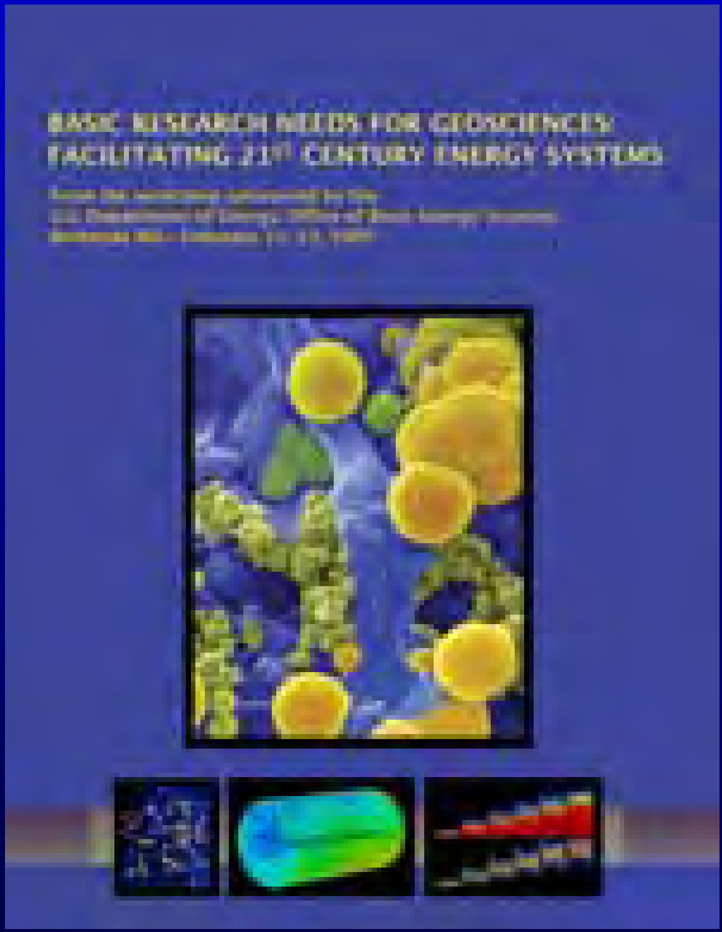
- structure and chemistry
- phase transitions as a function of temperature and radiation field
- chemical durability over a range of conditions (thermodynamic and kinetic studies)
- corrosion mechanisms and rates
- mechanical properties
- radiation response to different types of radiation
- natural “analogue” studies
- synthesis technologies

Basic Research Needs for Advanced Nuclear Energy Systems

QuickTime™ and a TIFF (LZW) decompressor are needed to see this picture.

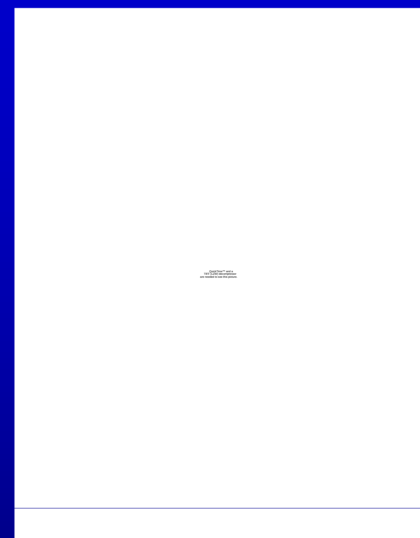
- Materials under extreme conditions
- Chemistry under extreme conditions
- Separations science
- Advanced actinide fue
- Advanced waste forms
- Predictive modeling and simulation

Basic Research Needs for Geosciences: Facilitating 21st Century Energy Systems



- Multiphase fluid transport in geologic media
- Chemical migration processes in geologic media
- Subsurface characterization
- Modeling and simulation of geologic systems

“Cross-cutting” Fundamental Research

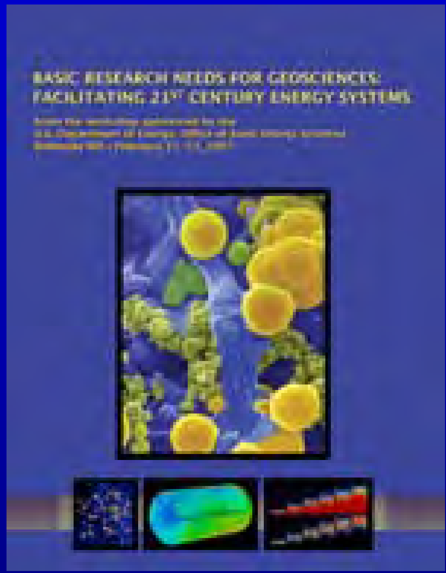


- novel materials
- interfaces
- thermodynamics of complex systems
- radiation & radiolysis effects
- predictions of long-term performance

Grand Challenges

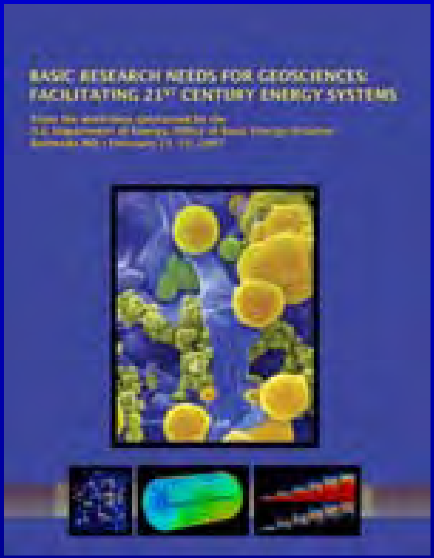
- f-electron challenge for the chemistry and physics of actinide-bearing materials
- First principles multiscale description of material properties under extreme conditions
- Understanding and designing new molecular systems to gain unprecedented control on chemical selectivity during processing

“Cross-cutting” Fundamental Research



- **Microscopic basis of macroscopic complexity**
- **Highly reactive subsurface materials and environments**
- **Thermodynamics of the solute-to-solid continuum**

Grand Challenges



- Computational thermo-dynamics of complex fluids and solids
- Integrated characterization, modeling, and monitoring of geologic systems
- Simulation of multi-scale systems for ultra-long times

The Seam Between “Immediate” and “Fundamental” Research Needs

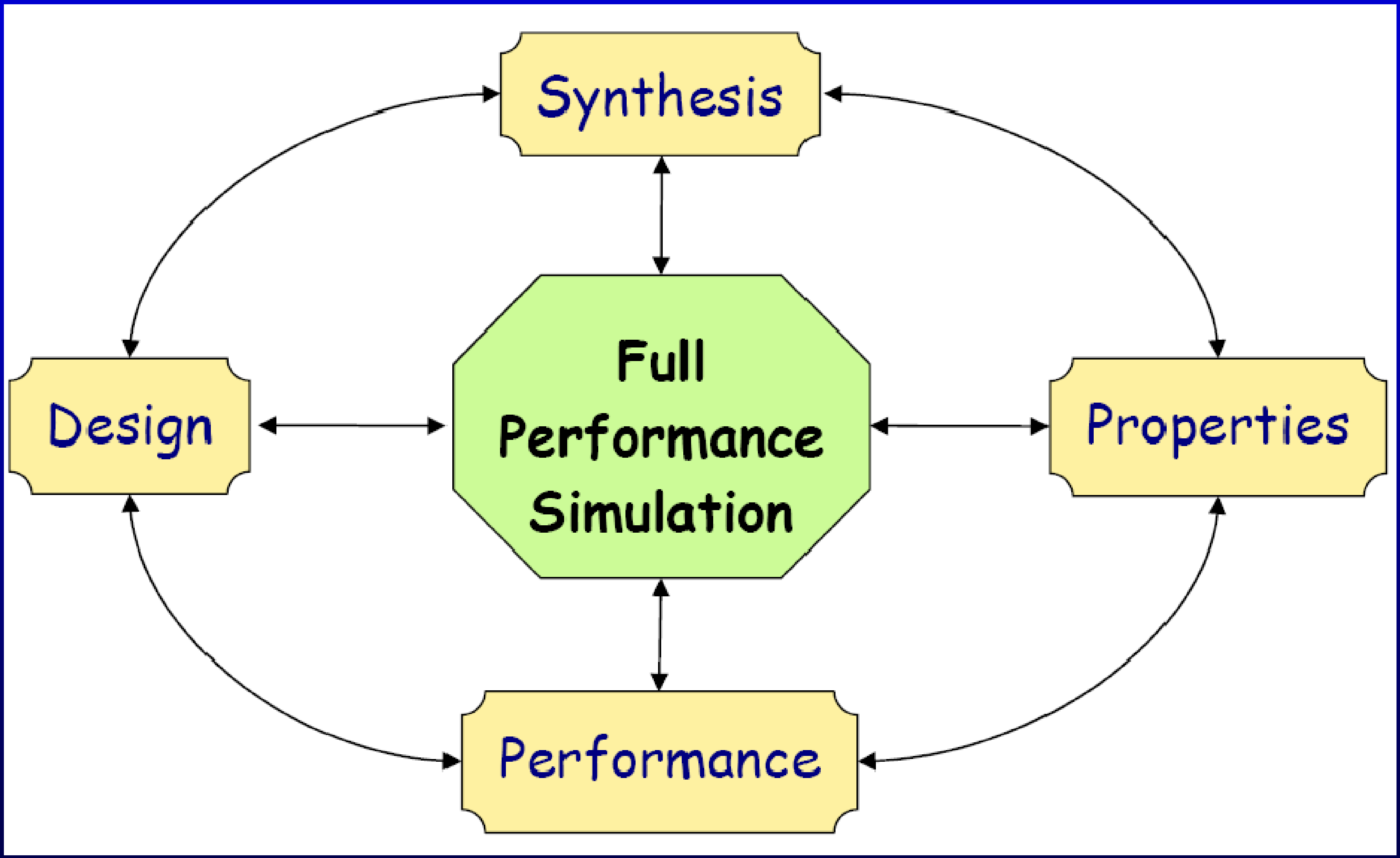
- Novel materials (mesoporous)
- New approaches to the design of materials
- Computational simulation of waste form properties
- Waste forms that are part of the processing technology
- Waste forms that use the natural environment to advantage

“Integration by Simulation” Waste Form Properties & Performance

Computational simulation of:

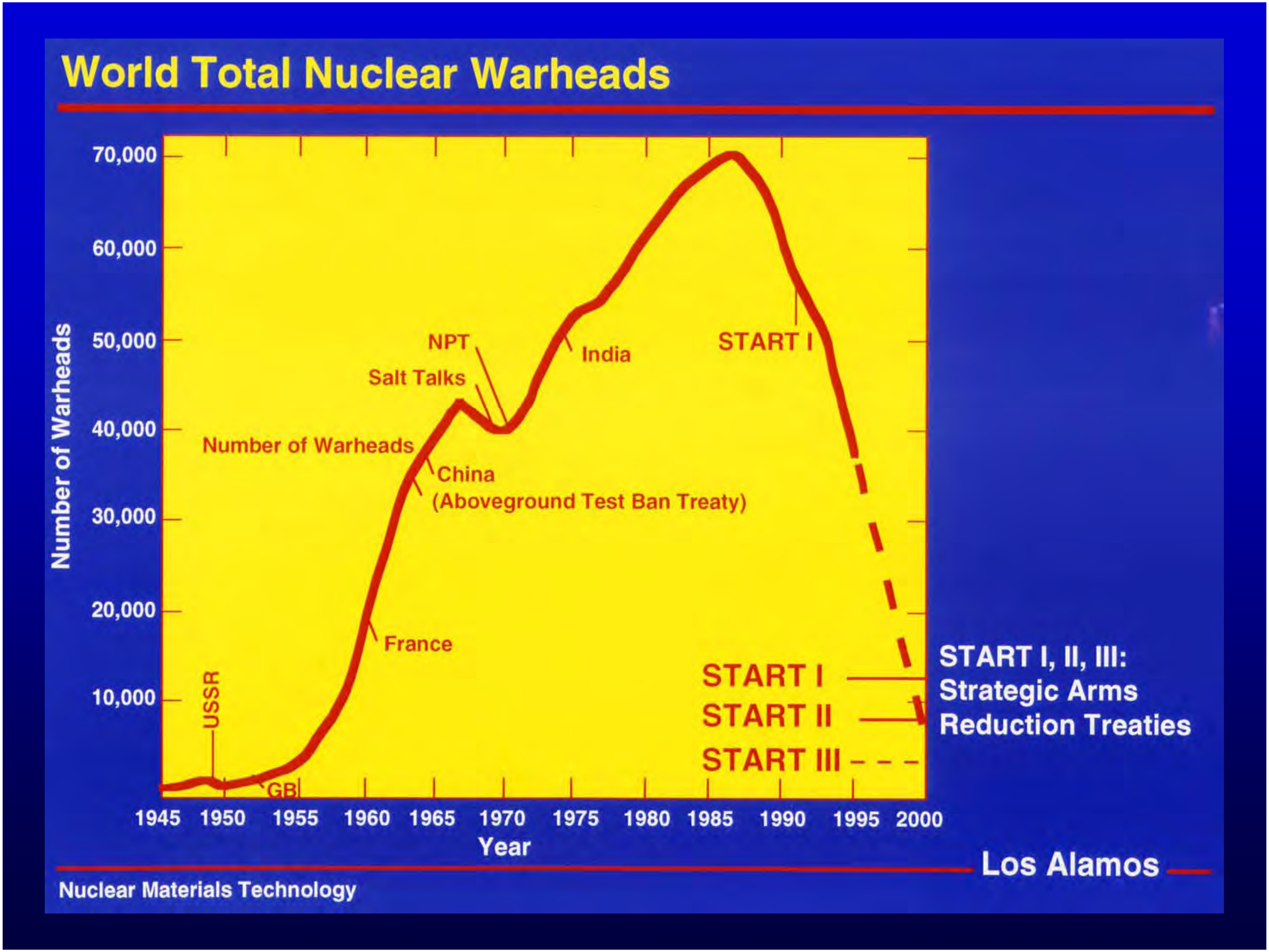
- ✓ waste form properties
- ✓ waste form behavior in the environment

Full Performance Simulation



Tomas de la Rubia (2007) *Basic Research Needs for Materials in Extreme Environments*

Example


World Total Nuclear Warheads
Number of Warheads
70,000
60,000
50,000
40,000
30,000
20,000
10,000
USSR
GB
France
China
(Aboveground Test Ban Treaty)
Number of Warheads
Salt Talks
NPT
India
START I
START I
START II
START III
1945
1950
1955
1960
1965
1970
1975
1980
1985
1990
1995
2000
Year
START I, II, III:
Strategic Arms
Reduction Treaties
Los Alamos
Nuclear Materials Technology

Actinide Matrices: *Mineralogical Solution*

simple oxides:	*zirconia*	ZrO_2
complex oxides:	*pyrochlore*	$(Na,Ca,U)_2(Nb,Ti,Ta)_2O_6$
	murataite	$(Na,Y)_4(Zn,Fe)_3(Ti,Nb)_6O_{18}(F,OH)_4$
	zirconolite	$CaZrTi_2O_7$
	perovskite	$CaTiO_3$
silicates:	*zircon**	$ZrSiO_4$
	*thorite**	$ThSiO_4$
	*garnet**	$(Ca,Mg,Fe^{2+})_3(Al,Fe^{3+},Cr^{3+})_2(SiO_4)_3$
	britholite	$(Ca,Ce)_5(SiO_4)_3(OH,F)$
	titanite	$CaTiSiO_5$
phosphates:	*monazite**	$LnPO_4$
	*apatite**	$Ca_{4-x}Ln_{6+x}(PO_4)_y(O,F)_2$
	*xenotime**	YPO_4

*durable heavy minerals

Pyrochlore

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

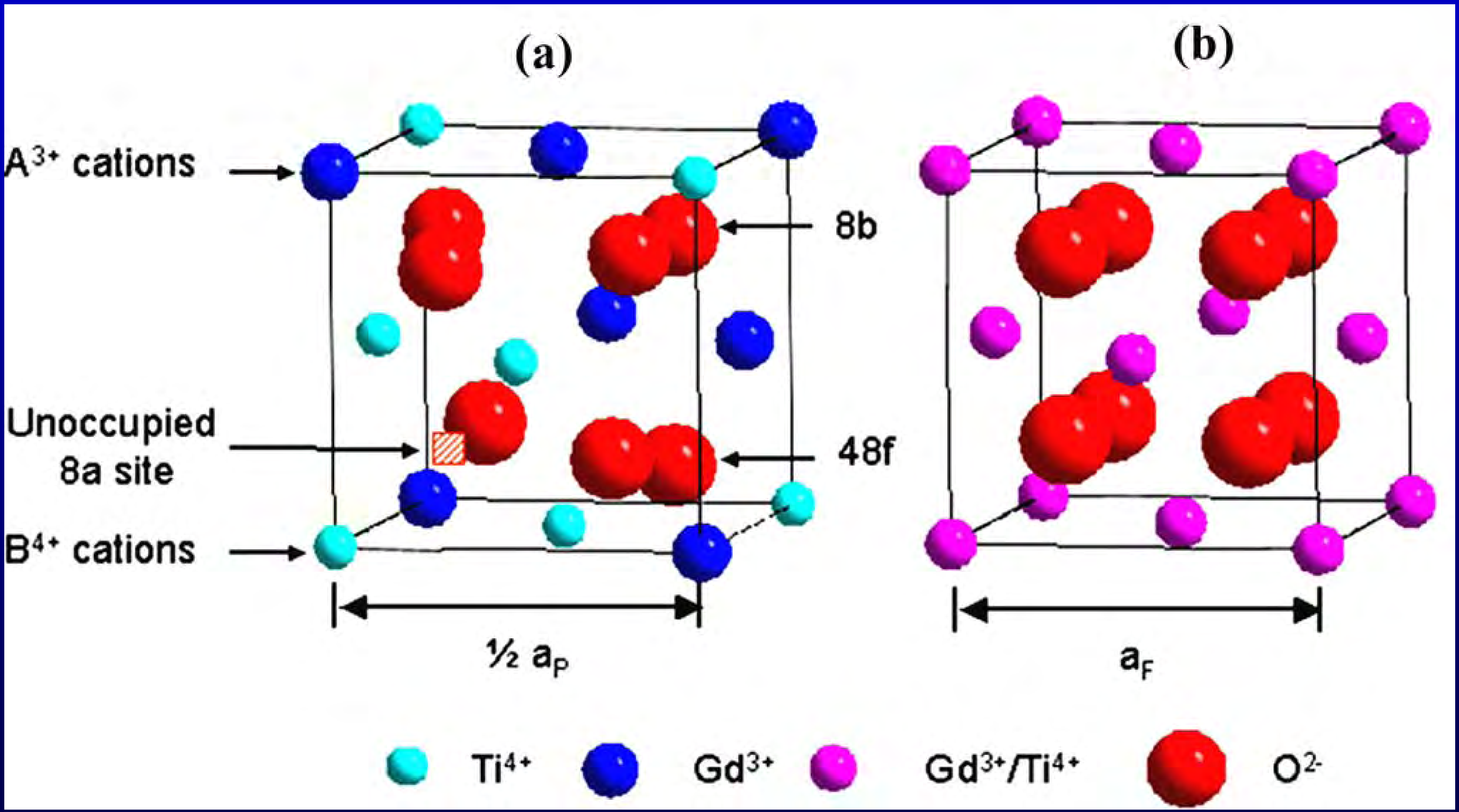
$(Ca,Na,U,$
$REE)_2(Nb,Ta,Ti)_2O_6(OH,F)$

$(Ca,Gd,Pu,U,Hf)_2Ti_2O_7$

$CaZrTi_2O_7$ (monoclinic)

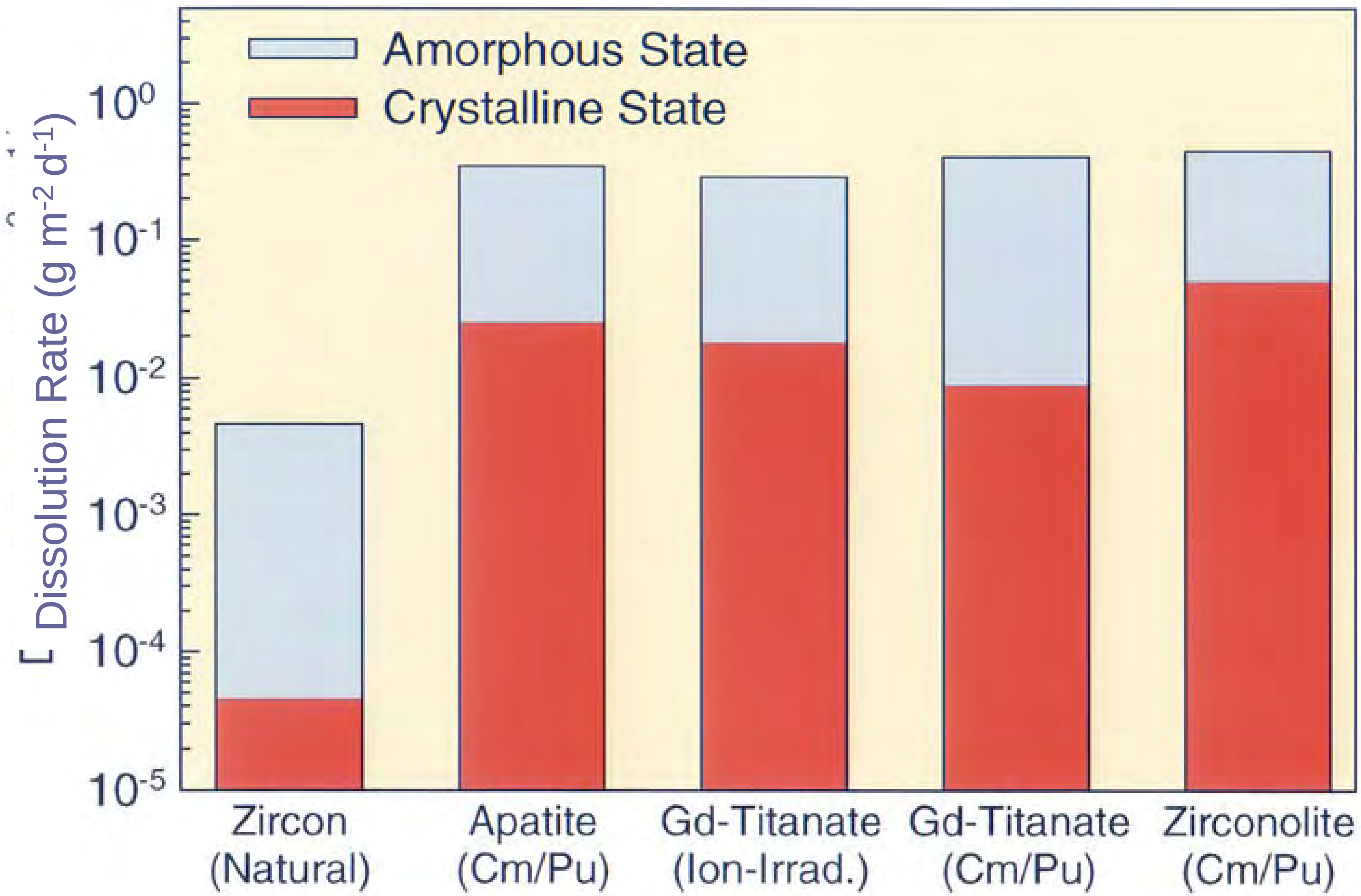
$CaPuTi_2O_7$ (monoclinic to cubic)

Pyrochlore to Fluorite



Lian, Wang, Wang, Chen, Boatner and Ewing (2001) *Phys. Rev. Lett.*

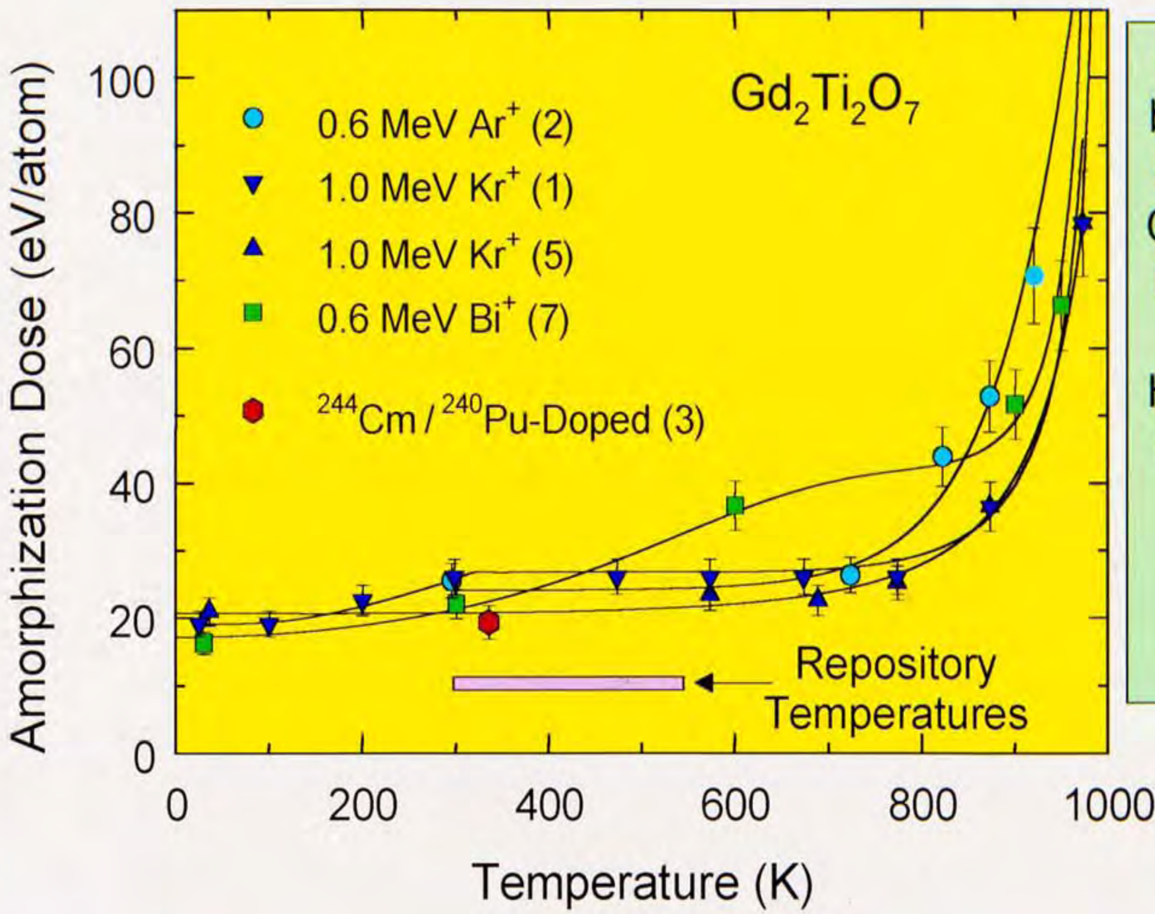
Amorphization: Effect on Dissolution Rate



Based on Literature Data: Ewing et al. 1982; Wald & Weber 1984; Helean et al. 1999; Begg et al. 2000

Radiation-Induced Amorphization in Gadolinium Titanate

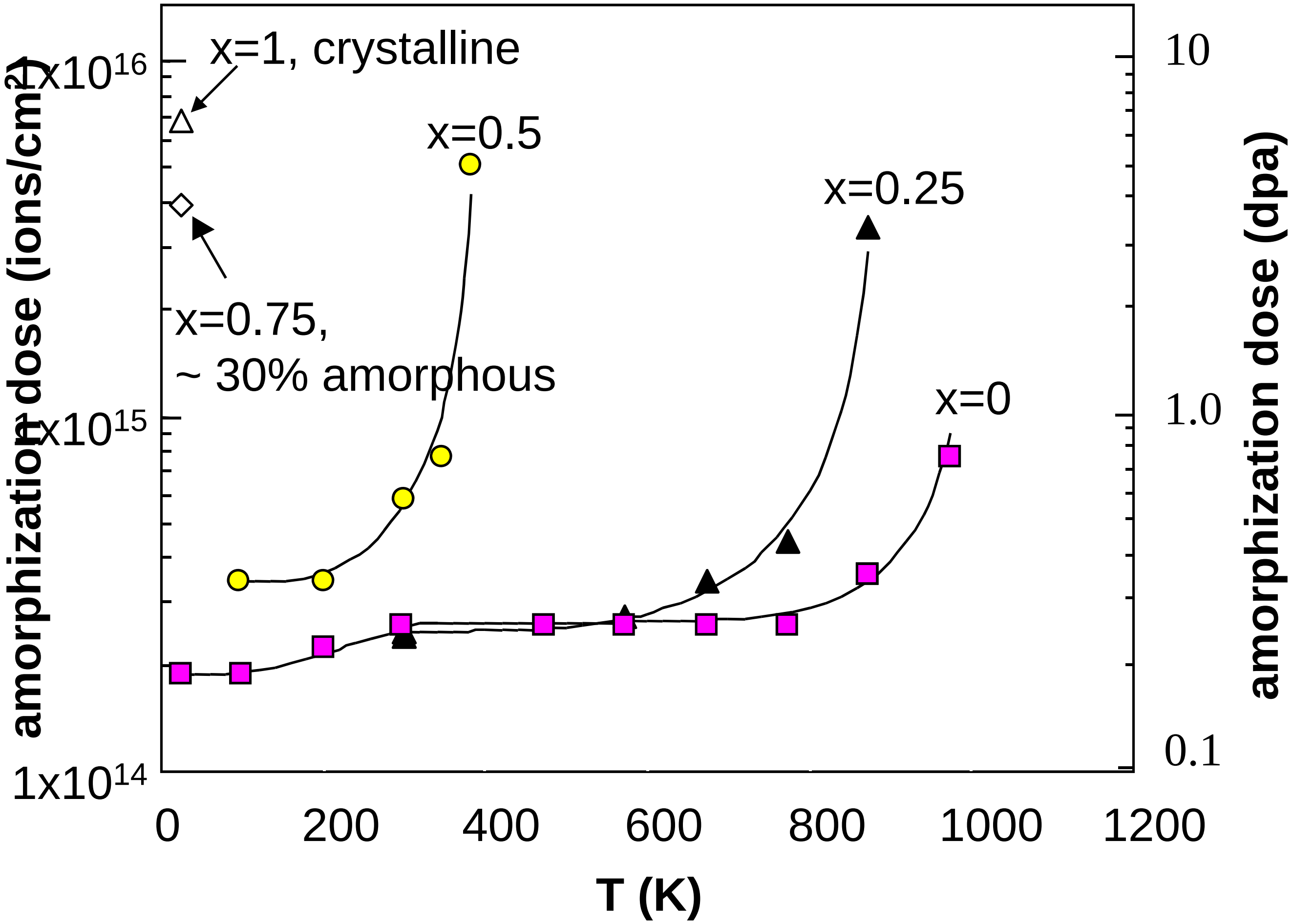
(Primary Candidate Phase for Plutonium Immobilization)



Good agreement between results of alpha-decay (i.e., Cm/Pu decay) and heavy-ion studies at 300 K.
High temperatures required to inhibit amorphization compared to repository temperatures.

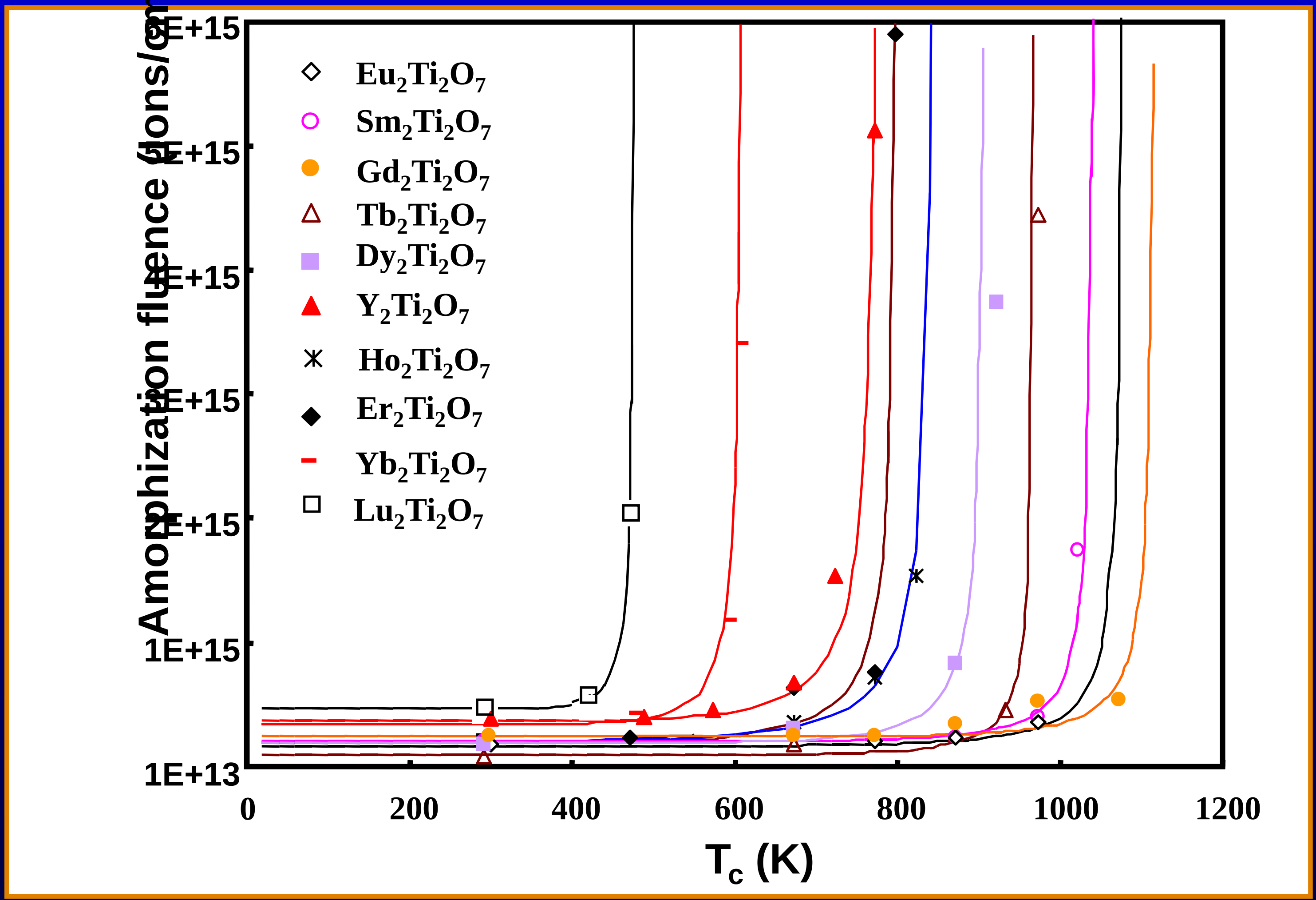
From WJ Weber and RC Ewing *Science* **289** (2000) 2051-2052

Radiation-Induced Amorphization (1 MeV Kr^+): $Gd_2(Zr_xTi_{1-x})_2O_7$



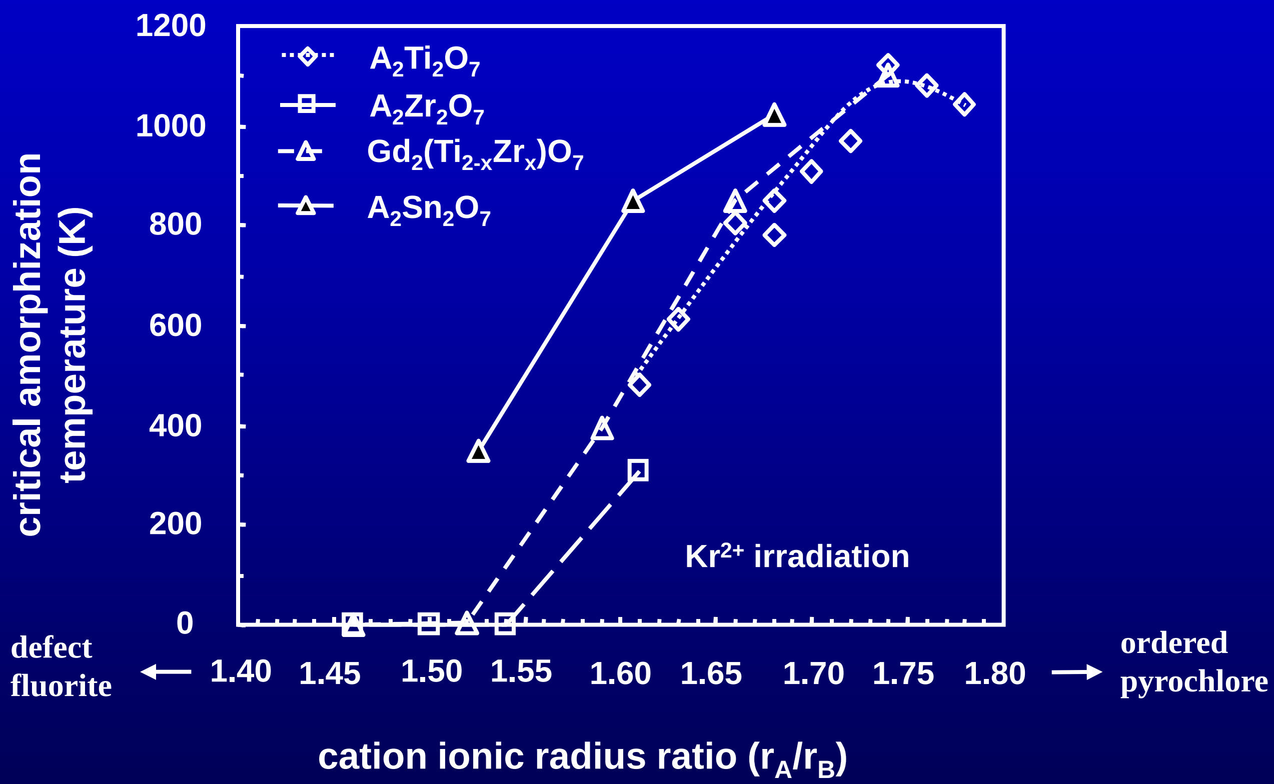
Wang, Begg, Wang, Ewing, Weber and Kutty (1999) *J. Mater. Res.*

REE$_2$Ti$_2$O$_7$ Irradiated with 1 MeV Kr$^+$



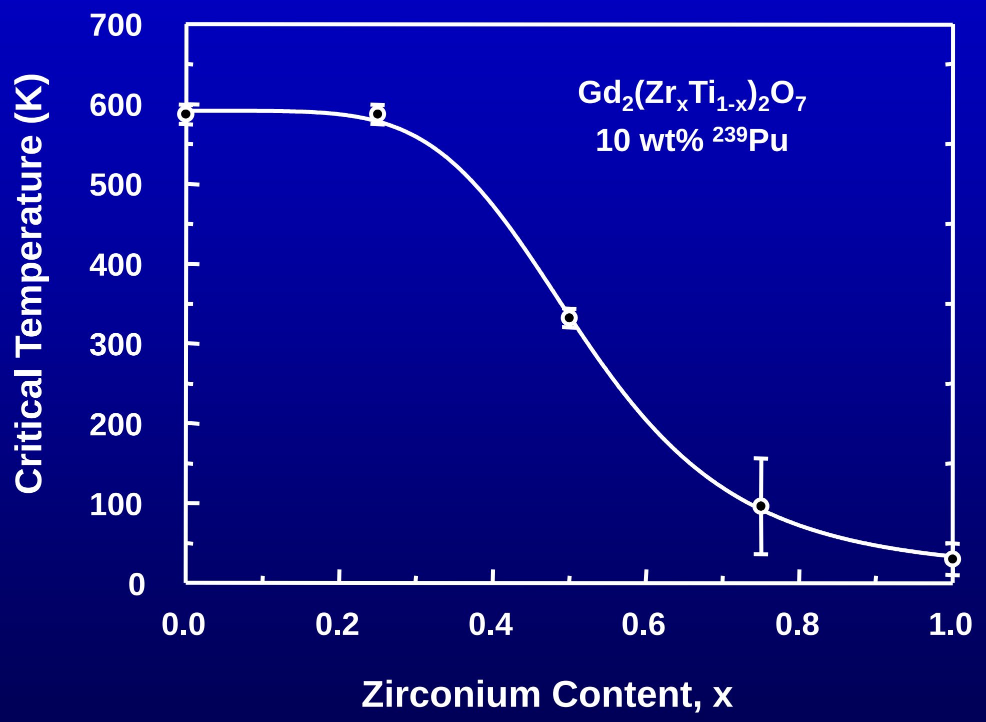
Lian, Chen, Wang, Ewing, Farmer, Boatner and Helean (2003) *Phys. Rev. B*

Critical Temperature, T_c, *vs.* A-site:B-site Radius Ratio



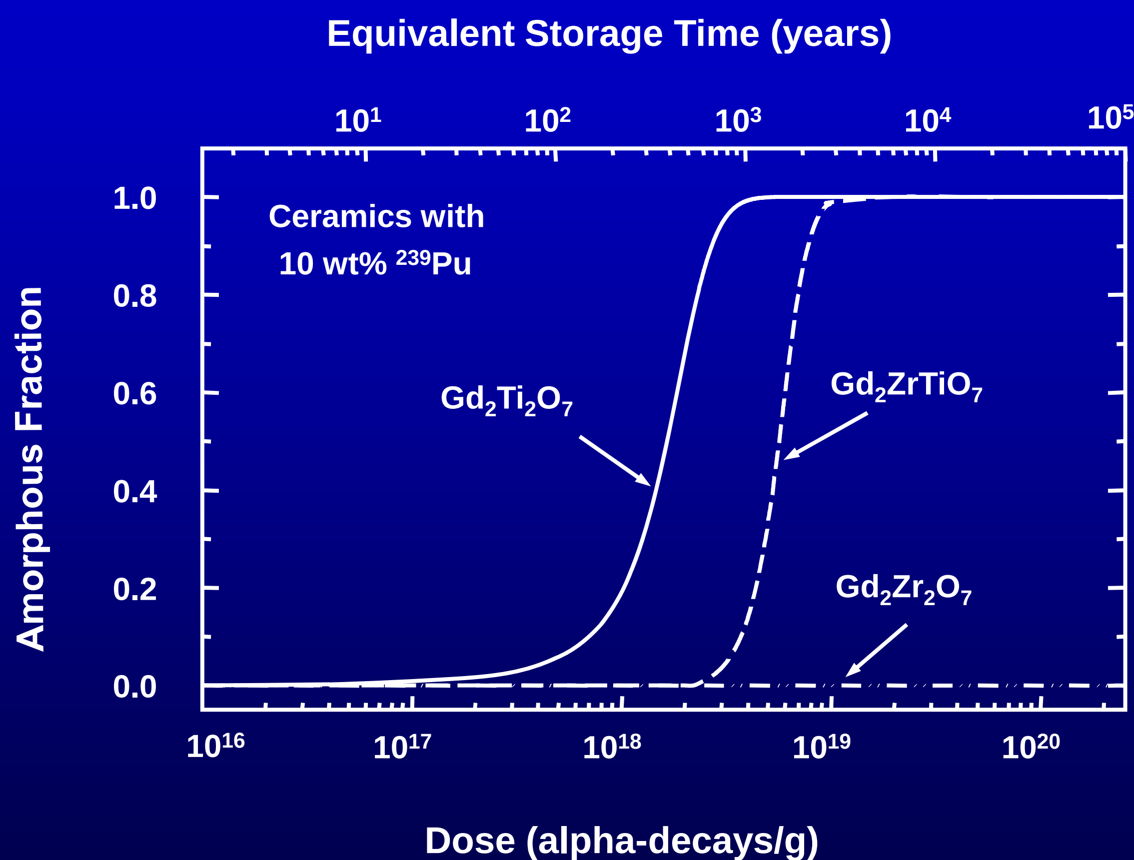
Ewing, Weber and Lian (2004) *J. Applied Physics*

T_c *vs.* $Gd_2(Zr_xTi_{1-x})_2O_7$



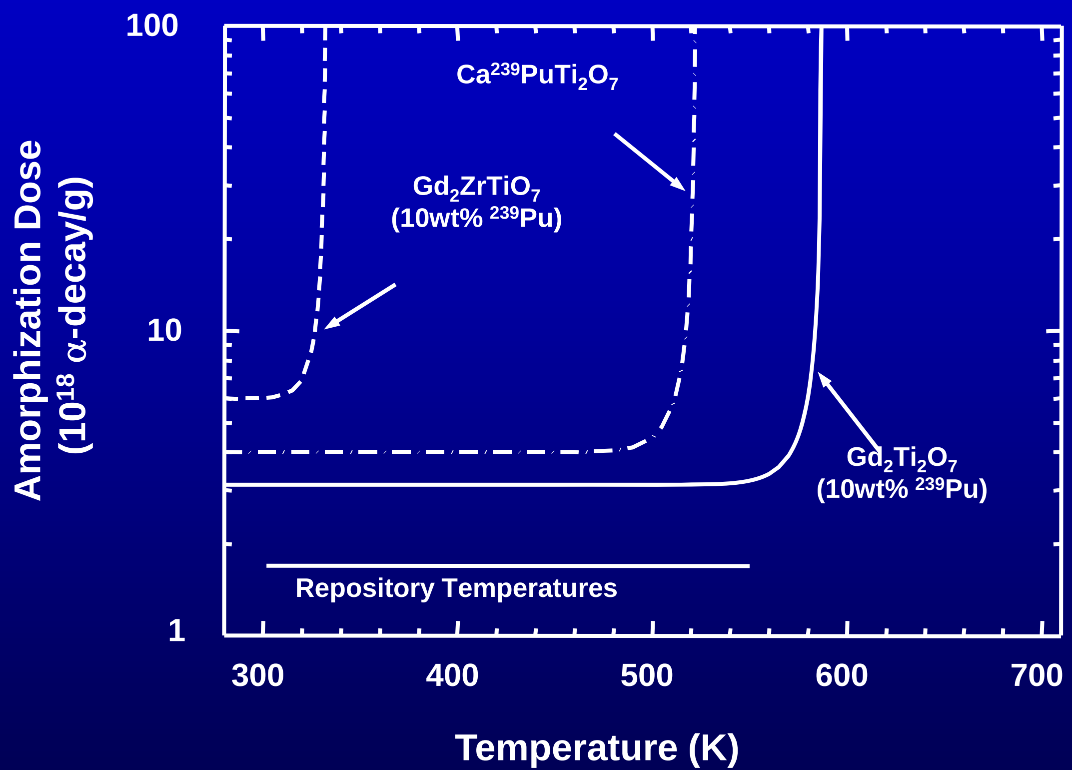
Ewing, Weber & Lian (2004) J. Applied Physics

Storage Time *vs.* Amorphization



Ewing, Weber & Lian (2004) *J. Applied Physics*

Temperature Dependence of Amorphization for Pu-pyrochlore



Ewing, Weber & Lian (2004) J. Applied Physics

State of the Science

We now have *the <u>beginnings</u>* of the fundamental understanding of radiation-induced structural transformations required to design nuclear materials, such as waste forms and nuclear fuels, to specific performance requirements, such as chemical durability and radiation "resistance".

We should use this knowledge as a foundation for designing waste forms to specific geologic repository conditions.


Thanks