Simulation and Modeling at the Exascale for Energy, Ecological Sustainability and Global Security

Town Hall Meeting: Energy Breakout

Reprocessing perspective

May 17, 2007

Oak Ridge National Laboratory

A Central Challenge: Efficient Partitioning of Spent Nuclear Fuel

"Reprocessing is one of the most complicated chemical processes ever endeavored on an industrial scale"

Choppin, Rydberg, Liljenzin, "Radiochemistry and Nuclear Chemistry", 2001

Contents of 1 metric ton of PWR fuel (~ 2 fuel assemblies) from 50 MWd/kg burn-up after 10 years cooling:

936.0 kg U

- **9.1 kg Pu (5.0 kg Pu-239)**
- **1.1 kg Np-237**
- **1.3 kg Am**
- **0.2 kg Cm**

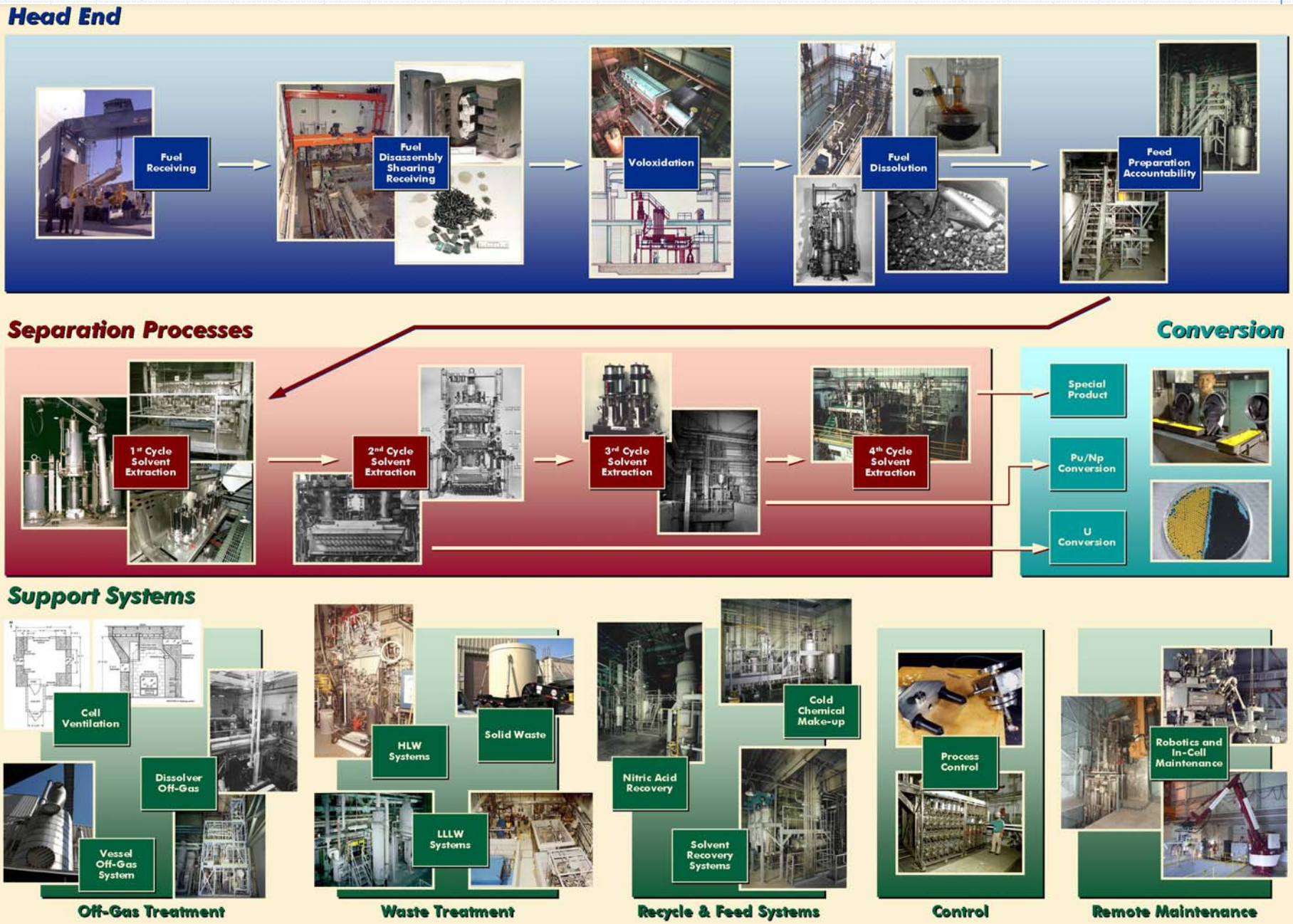
52.7 kg fission products:

- **0.53 kg Kr (0.19 kg Kr-85)**
- **0.54 kg Rb**
- **1.2 kg Sr (0.65 kg Sr-90)**
- **0.70 kg Y**
- **5.6 kg Zr**
- **5.2 kg Mo**
- **1.2 kg Tc-99**
- **3.4 kg Ru**
- **0.5 kg Rh**
- **2.4 kg Pd**
- **0.71 kg Te**
- **0.36 kg I (0.29 kg I-129)**
- **8.2 kg Xe**
- **3.3 kg Cs (1.5 kg Cs-137)**
- **3.1 kg Ba**
- **15.3 kg lanthanides**

10^9 Sv/TWhe

From Carol J. Burns, "Frontiers in Chemical Research for Advanced Nuclear Energy Systems," Workshop on Basic Research Needs for Advanced Nuclear Energy Systems, *Bethesda, MD, July 31, 2006.*

Reprocessing is more than solvent extraction



Separations science: basic-science challenges, opportunities, and needs

- **Fundamental research on development of separating agents. Functionalities specified by**
  - **Affinity for target species**
  - **Ease of synthesis**
  - **Interaction with solvents/solubility**
  - **Interaction with other materials**
  - **Chemical stability**
  - **Radiolysis**
  - **Chemical and physical properties**
- **Applied science research for improved processes**
  - **Improved models and computational tools for**
    - **Equilibrium partitioning in complex mixtures**
      - **Solution thermodynamics at high ionic strength**
    - **Transport and kinetics in non-equilibrium multiphase systems**
      - **Multiple time/size scales**
      - **Complex interactions of reactions, mass transfer, interfacial phenomena, turbulent fluid flow**

Separations science: basic-science challenges, opportunities, and needs

Closely coupled computations and experiments can improve and accelerate development of selective separating agents

Opportunities

Multi-scale modeling

- **Discrete-event**
  - Plant-level models
- **Macroscopic process**
  - Unit operations level
- **First-principles models**
  - Small-scale phenomena as necessary

Implementation

- **Assessment**
  - Evaluate existing codes
  - Prioritize development
- **Integration**
  - Common component architecture
- **Advanced Modeling**
  - Develop specialized codes

Multiple component systems

Engineering Tests
Multiple component systems

System Design, Pilot Tests

Simulation of transport and kinetics in non-equilibrium multiphase systems, including mass transfer, reaction kinetics, fluid dynamics

Prediction of physical and transport properties (viscosity, vapor pressure, etc.), interfacial effects

Models of partitioning based on first-principles thermodynamics that incorporate multicomponent effects at high ionic strength

Plant Implementation

Full process simulations allowing process optimization and sensitivity analysis, structured to incorporate advanced chemical models

2015 Vision - Nuclear energy simulation is "on par with experiment"

- Is the vision correct and attainable?
  - Vision is Correct
    - The alternative - "we don't need computing" - isn't acceptable
    - Simulation will not replace experiment
- KEY QUESTION is Attainability
  - Are programs embracing revolutionary changes through M&S to achieve mission goals?
  - Will campaigns say they cannot accomplish goals without M&S?
  - Need to establish credible goals, capabilities embraced by development programs
    - How accurate do chemical models have to be/can they do it?
    - Can M&S reduce the time for research and development?

Solvent Extraction Scales and Models

Contactors at realistic operational conditions

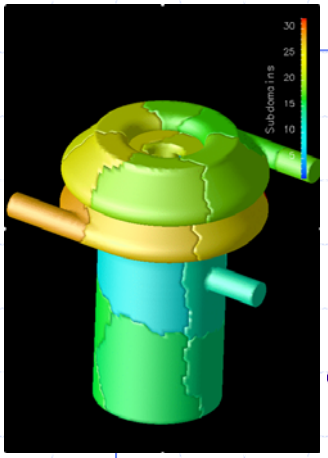
- **Quantum**
  - Solvent structure design
  - Thermo-chemistry
- **Classic atomistic**
  - Solvent-diluent interaction
  - Solvent-solvate complexation
  - Interfacial transport
- **Continuum**
  - Multiphase sharp interface
  - Multispecies
  - Mass-action reactive
- **Averaged Continuum**
  - Classic multiphase
  - Turbulence modeling
  - Flow regime dependent
    - Mixing zone
    - Separation zone
    - Transition zone
- **Lumped models**
  - Contactor dependent
  - Plant scale input

Å nm µm mm cm m

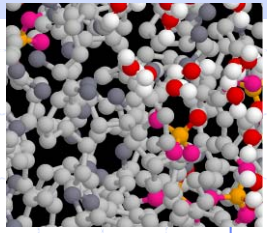
- Exascale computing significant impact

Separations Simulation

Solvent Extraction



Centrifugal contactor



Tri-butyl-phosphate / Dodecane / Water

- **Drivers**
  - Current exps do not allow for scale-up to plant operations
  - Predict extraction efficiency of contactors
  - Predict foot-print of extraction facility
- **Terascale**
  - Accurate 3-D geometry and meshing of contactors
  - Limited two-phase (volume-average based), few-species, reactive, modeled turbulent flow simulation
  - No flow-regime transition (interfacial area) prediction (dependent on experimental calibration)
  - Nano-second molecular dynamics on tens-of-thousands-molecule aqueous-organic systems with few species
- **Petascale**
  - Accurate simulation of volume-average-based multi-phase, multi-species, modeled turbulence
  - Improved prediction of extraction but dependent on experimental calibration
  - MD on realistic aqueous-organic systems on pico-seconds time scale
- **Exascale**
  - Simulation of sharp-interface, multi-phase, multi-species, modeled turbulence
  - Flow regime transition prediction (interfacial area)
  - MD on realistic aqueous-organic systems on nano-second time scale

2015 Assumptions - Policy

- There is a national commitment to a closed fuel cycle solution
- Regulatory agency role has changed to accommodate new simulation capabilities

2015 Assumptions – simulation capabilities

- An exaflops platform is successfully designed and deployed
- Next-generation nuclear energy simulation tools have had 8 full years of DOE program support for sustained research, development, deployment, V&V, and application
- Nuclear energy simulation capability requirements, roadmaps, and milestones are established and integrated with a validation experimental plan

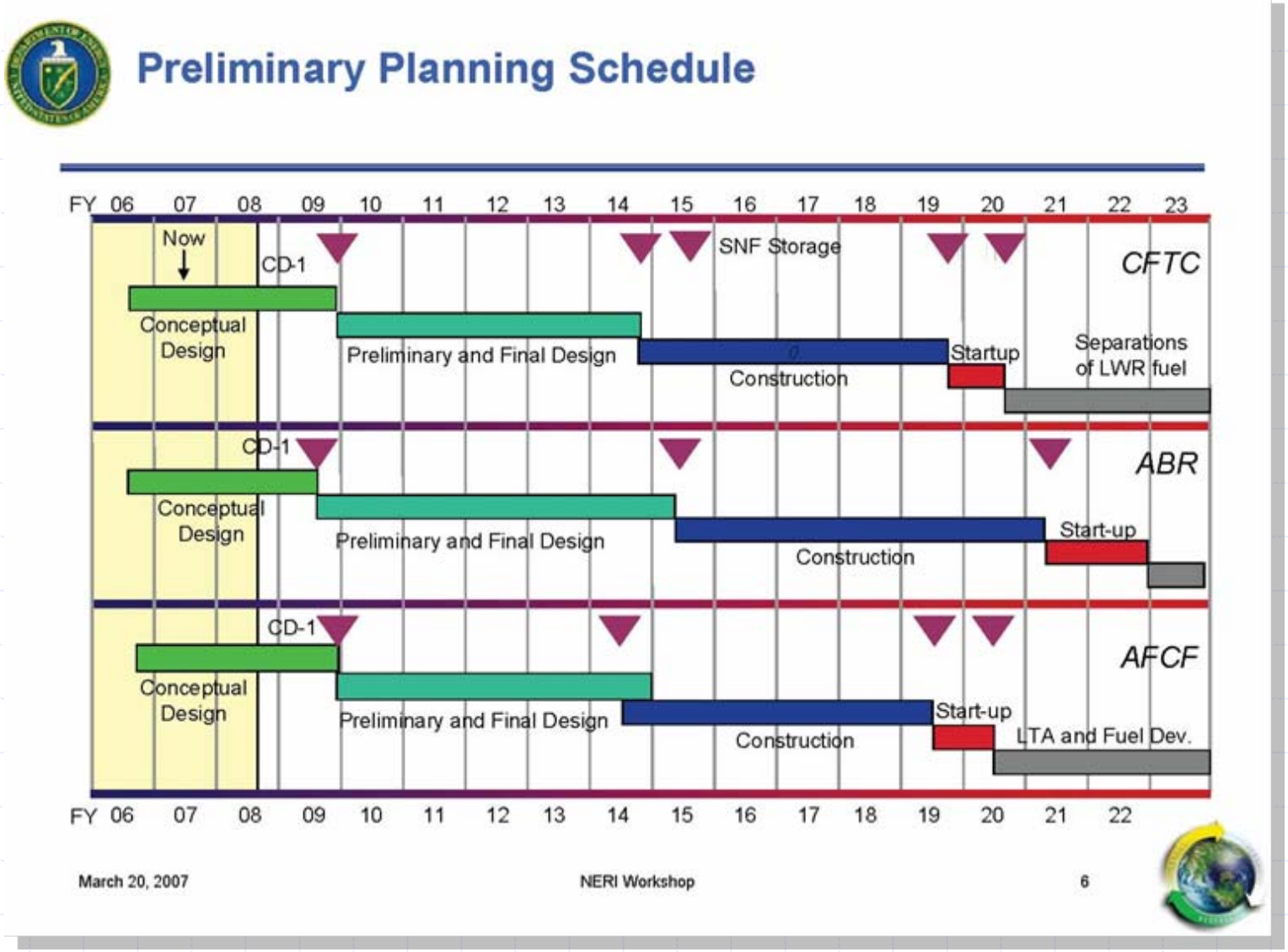
What are the potential payoffs?

- Reduced R&D cost – focus experimental programs
- Improved/accelerated design
  - Process scale-up
  - Reduced facility cost
- Opportunity for major change?
  - Shorten path to XXX by X years, or bypass the need for a generation of facilities
  - One possibility: AFCF
    - large pilot plant for long-term R&D
    - purpose to do development work on reprocessing
    - Model that simulates every function inside AFCF would allow prediction of changes.
    - PAYOFF: smarter about tests done in AFCF. Focus testing and accelerate progress in very expensive testing.

Utility: linkage with GNEP facilities

- Advanced Fuel Cycle Facility (AFCF):
  - an R&D center of excellence for developing transmutation fuels and improving fuel cycle technology.
    - provide experience needed to design and operate the commercial scale fuel fabrication and separations facilities.
    - develop and demonstrate:
      - advanced aqueous and pyroprocessing separations technologies,
      - transmutation fuel fabrication technologies, and
      - state-of-the-art safeguards instrumentation and monitoring systems.
- Consolidated Fuel Treatment Center (CFTC):
  - provide proof of the feasibility, reliability, and cost of an integrated separations process at prototype scale using commercial reactor spent nuclear fuel.

M&S Capability can impact value of CFTC and AFCF



From Paul Lisowsky, "The Global Nuclear Energy Partnership Program Overview/University Programs," NERI Workshop, March 20, 2007. http://www.ne.doe.gov/neri/07MarchWorkshop/GNEP%20(Lisowski).pdf

What can simulation do to facilitate the overarching GNEP vision?

- Can you make reprocessing cheaper than once-through?
- Waste issues
- Solvent design:
  - Reduced degradation in radiation field
  - Improved selectivity
  - Cheaper

Some technical questions

- Can quantum and molecular modeling make enough progress to accurately predict solution thermodynamics?
- Is ORIGEN (used for input data) correct?
  - Do we know all cross sections are correct
  - Output is many isotopes - How do you measure every one to validate?
  - Variability/uncertainty?
- Waste forms are a major issue
  - How do you design better waste forms? Need prediction of long-term behavior of waste forms (can't wait to rely solely on experiment!)
  - Example: Cs decays to Ba in waste form, with heat generation. What are effects of change in atomic size, radiation damage, etc. on waste form?
  - Need atomic-level materials models!

Can M&S Offset Criticism?

Costs too much
Waste management issues

INSIDE ENERGY | MAY 14, 2007

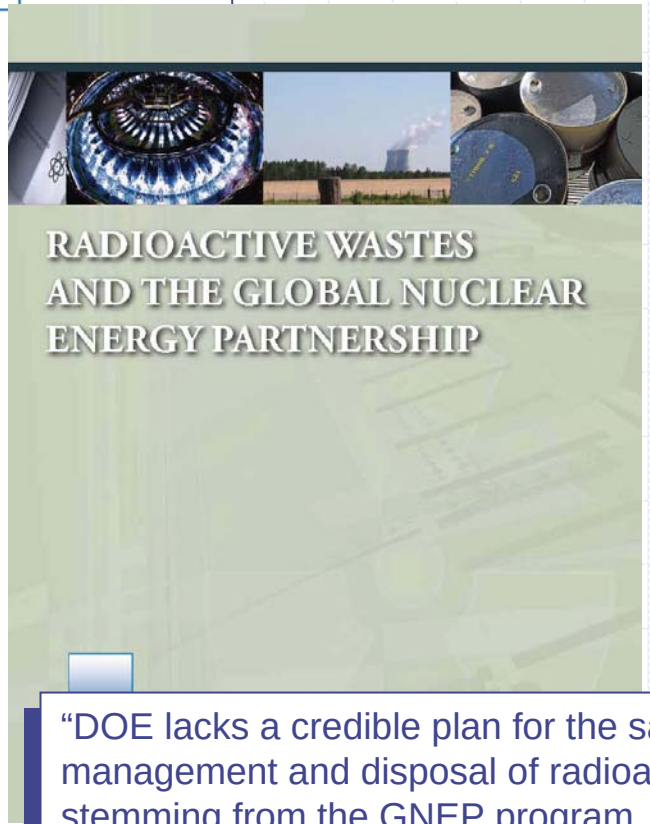
Nuclear reprocessing called futile

Opponents of reprocessing spent nuclear fuel say experiences in Britain, France and Japan indicate any such U.S. effort would be doomed because of cost, technical complication and administrative problems.

At a Monday forum sponsored by Public Citizen, which opposes nuclear energy, three experts said the Bush administration's Global Nuclear Energy Partnership would cost the United States many billions of dollars and create almost nothing in the way of new energy. "It can't be done," Aileen Mioko Smith, director of Green Action of Kyoto, Japan, said of the prospect of obtaining "any reasonable amount of energy" from reprocessing. She said Japan has spent $26 billion over 50 years working on reprocessing with little to show for it. The country has 11,000 metric tons of nuclear waste in pools at nuclear plants and generates 1,000 additional tons a year, she said. A plant due to come online in Japan in late 2007 would reprocess 800 tons a year, but Smith said it would not produce useable nuclear fuel.

Shaun Burnie, a consultant and former Greenpeace staffer who has studied France's reprocessing efforts, said that country recycles a very small proportion of the nuclear waste it generates and has spent $25 billion on its program. He maintained that reprocessing was inherently unsafe and unprofitable.

William Walker, a professor at St. Andrews University in Scotland, said U.S efforts to reprocess will likely lead to more complexity and difficulty in coordination between the government and industry on policy.



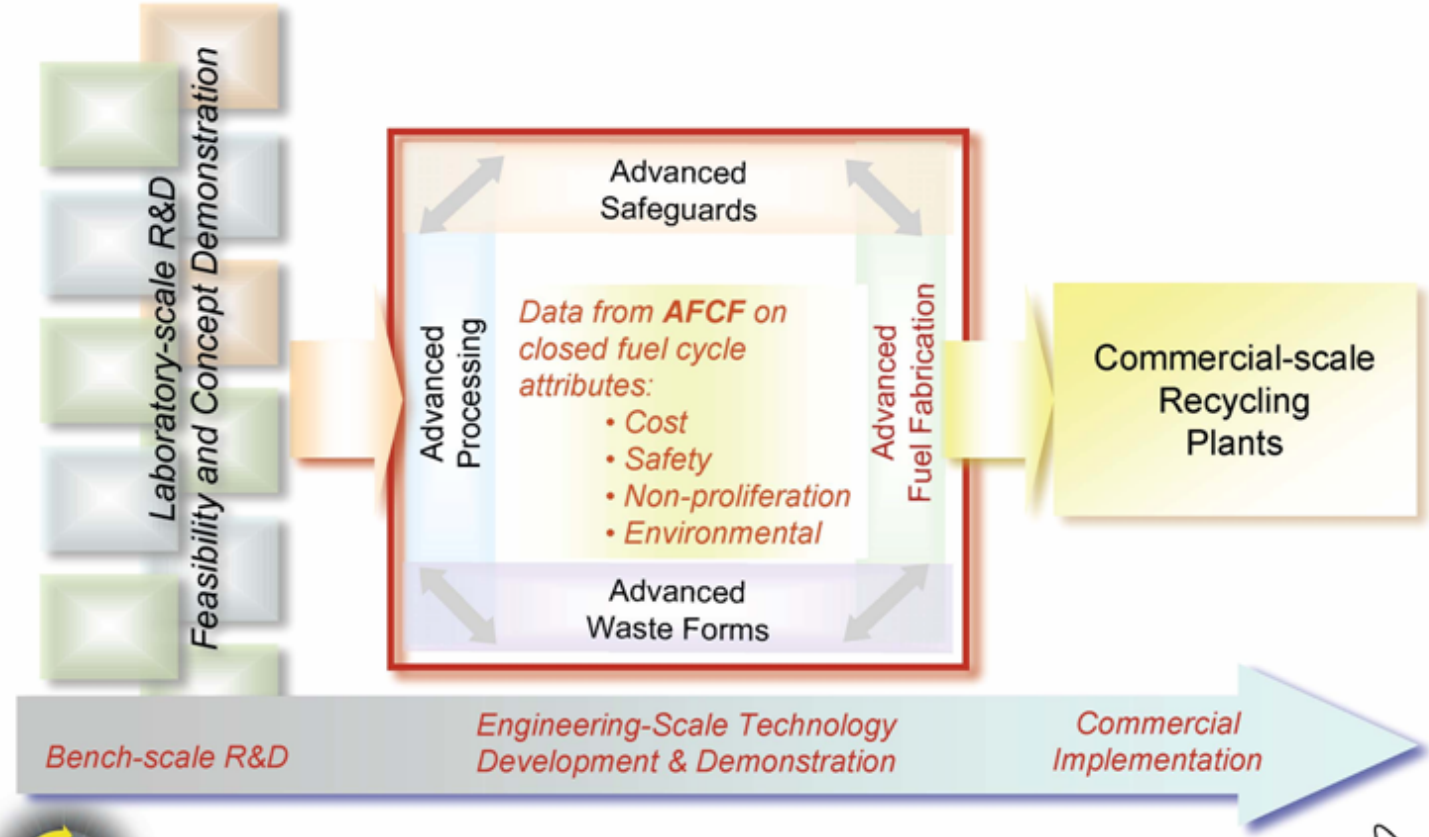
"DOE lacks a credible plan for the safe management and disposal of radioactive wastes stemming from the GNEP program. This plan should address waste volumes, disposition paths, site specific impacts, regulatory requirements and life-cycle costs. *Given past failures to address waste problems before they were created, DOE's rush to invest major public funds for deployment of reprocessing should be suspended."*

Other questions

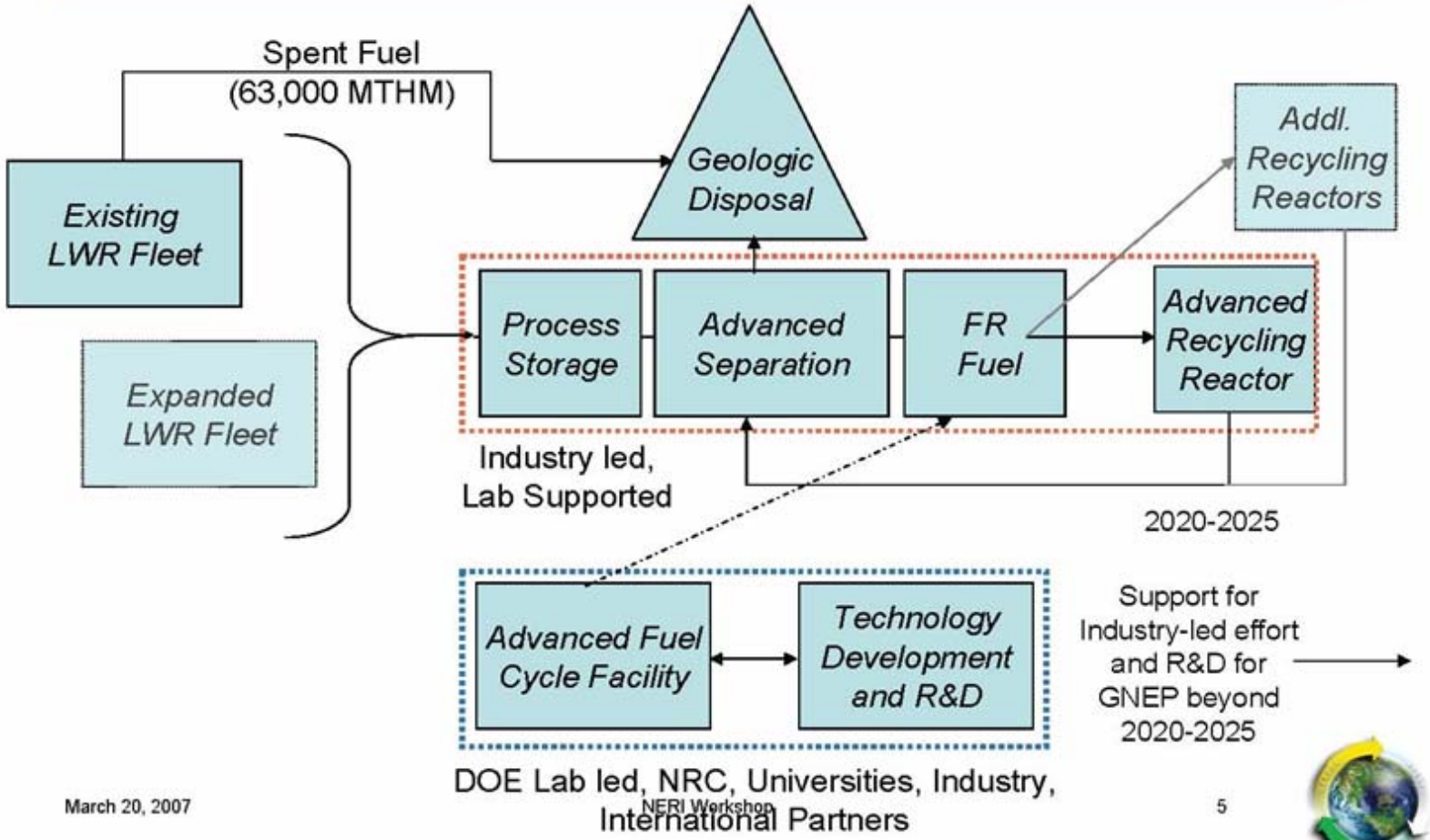
- Who are the stakeholders?
- What are the enabling technologies and facilities?
- What are the technology barriers?
- What is the technology roadmap?
- What are the major annual program gates?
- What are the management and technological risks?
- What are the budget estimates?

BACKUP

The over-arching objective of the AFCF is to perform fuel recycling technology development and demonstration of at a scale sufficient to provide input for subsequent commercialization decision.



Supporting the GNEP Strategy Requires New Facilities, Technology Development and R&D



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