Nuclear Fuel Cycle and Proliferation Monitoring

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CONSORTIUM FOR VERIFICATION TECHNOLOGY

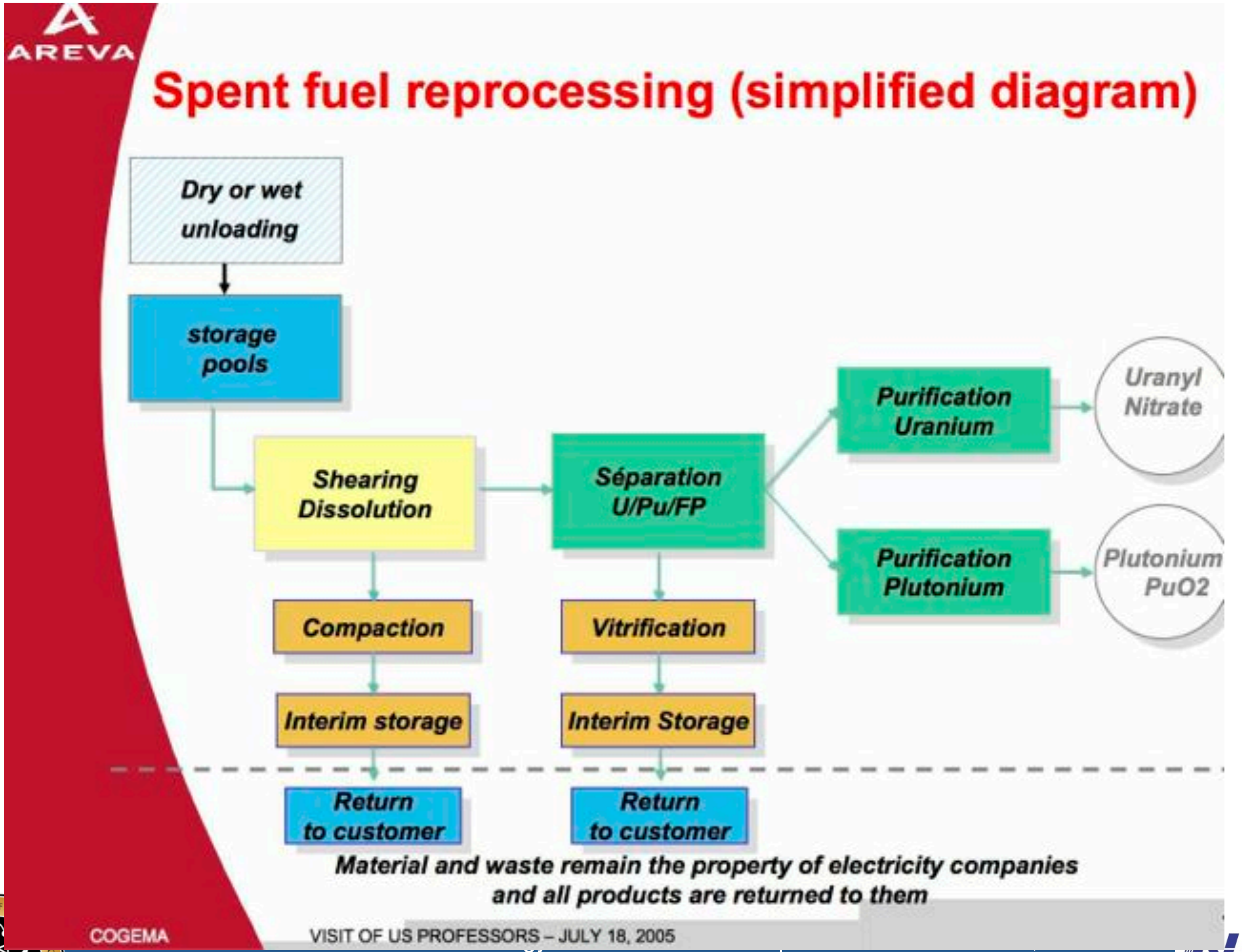
National Nuclear Security Administration

Outline

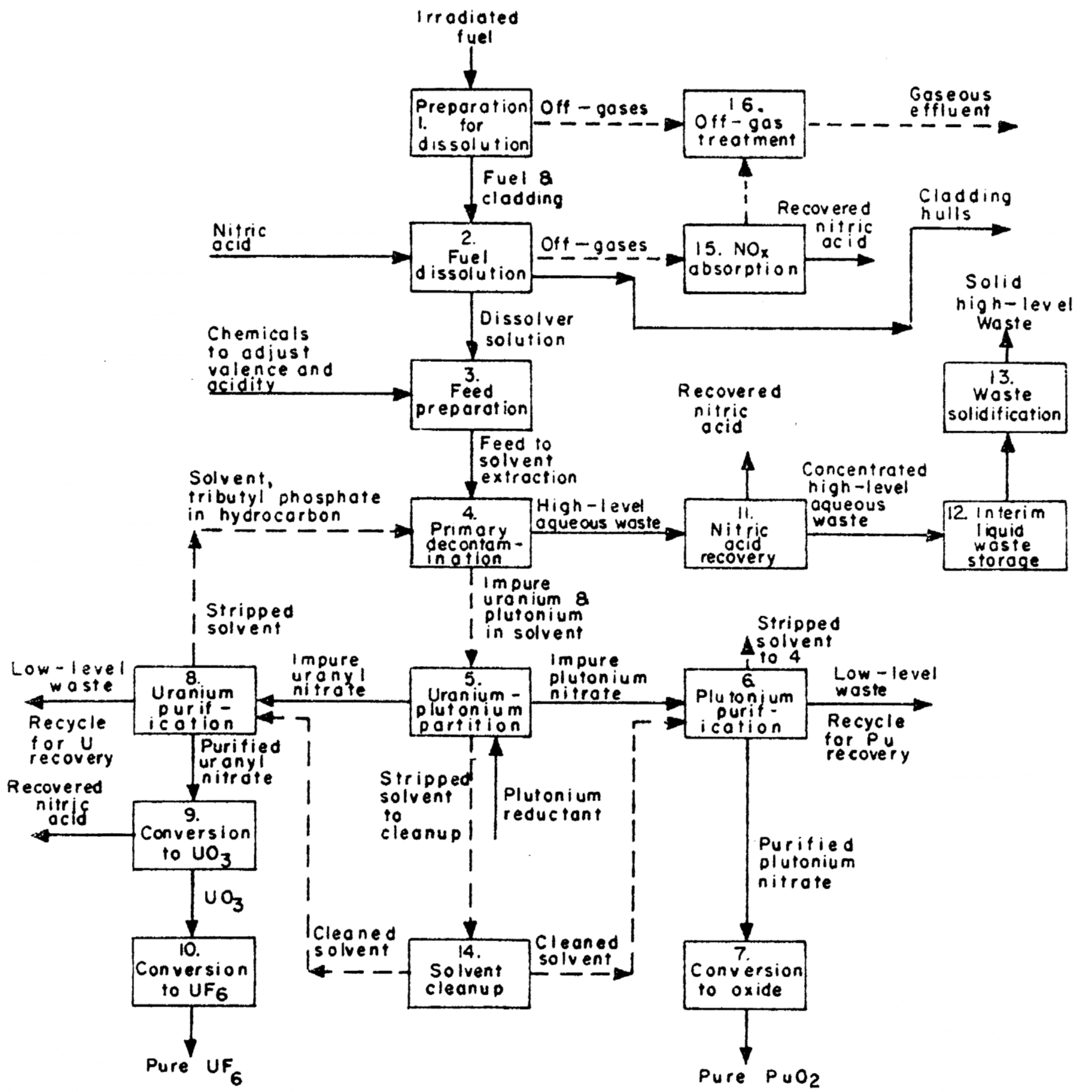
- Fuel cycle overview
- Used fuel reprocessing and management
- Proliferation risk of separations technology
- Examples of fuel cycle analysis
- Lattice physics modeling of fission products
- FP transport monitoring

Overview on Nuclear Fuel Cycle

- LWRs operate with once-through closed cycles using UO_2 with <5.0 wt% enrichment and discharge burnup of 40~50 MWd/kgHM.
- LWR used fuel composition:
  - 3~5% fission products
  - 1% transuranics, 0.1% minor actinides [Np, Am, Cm]
  - 94~95% U.
- Current French recycling strategy:
  - Separate MA + FP + assembly metal and store vitrified logs above ground
  - Recycle $(Pu+U)O_2$ [MOX] once only in PWRs, ¼ core
  - Store once-recycled MOX assemblies above ground
  - PUREX process used for reprocessing and vitrification.
- Pyroprocessing technology used for U-Pu-Zr metallic fuel from sodium-cooled fast reactor and under development for LWR fuel.

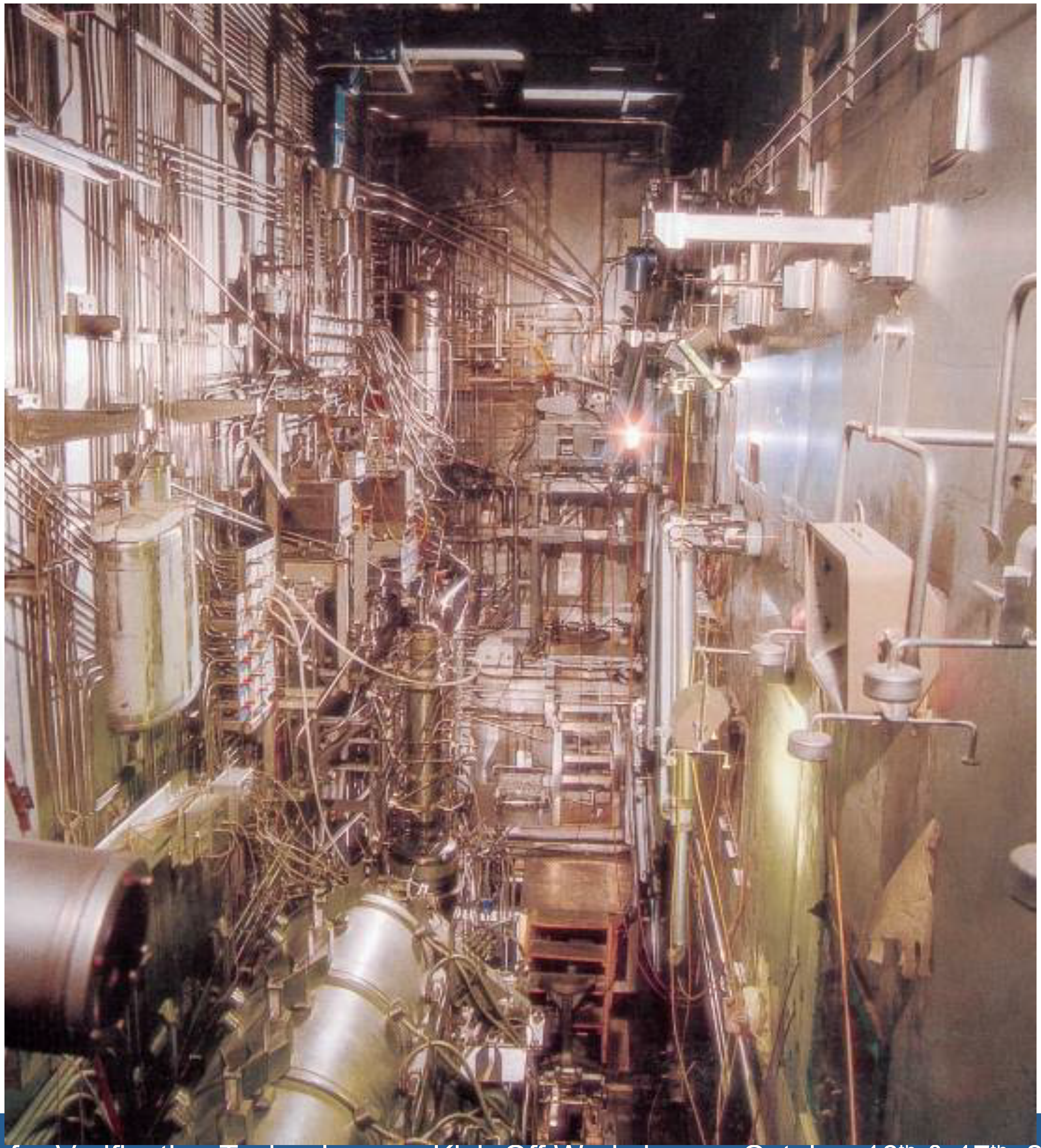

AREVA
Spent fuel reprocessing (simplified diagram)
Dry or wet unloading
storage pools
Shearing Dissolution
Séparation U/Pu/FP
Purification Uranium
Uranyl Nitrate
Purification Plutonium
Plutonium PuO2
Compaction
Vitrification
Interim storage
Interim Storage
Return to customer
Return to customer
Material and waste remain the property of electricity companies and all products are returned to them
COGEMA
VISIT OF US PROFESSORS – JULY 18, 2005

Principal Steps of PUREX Process



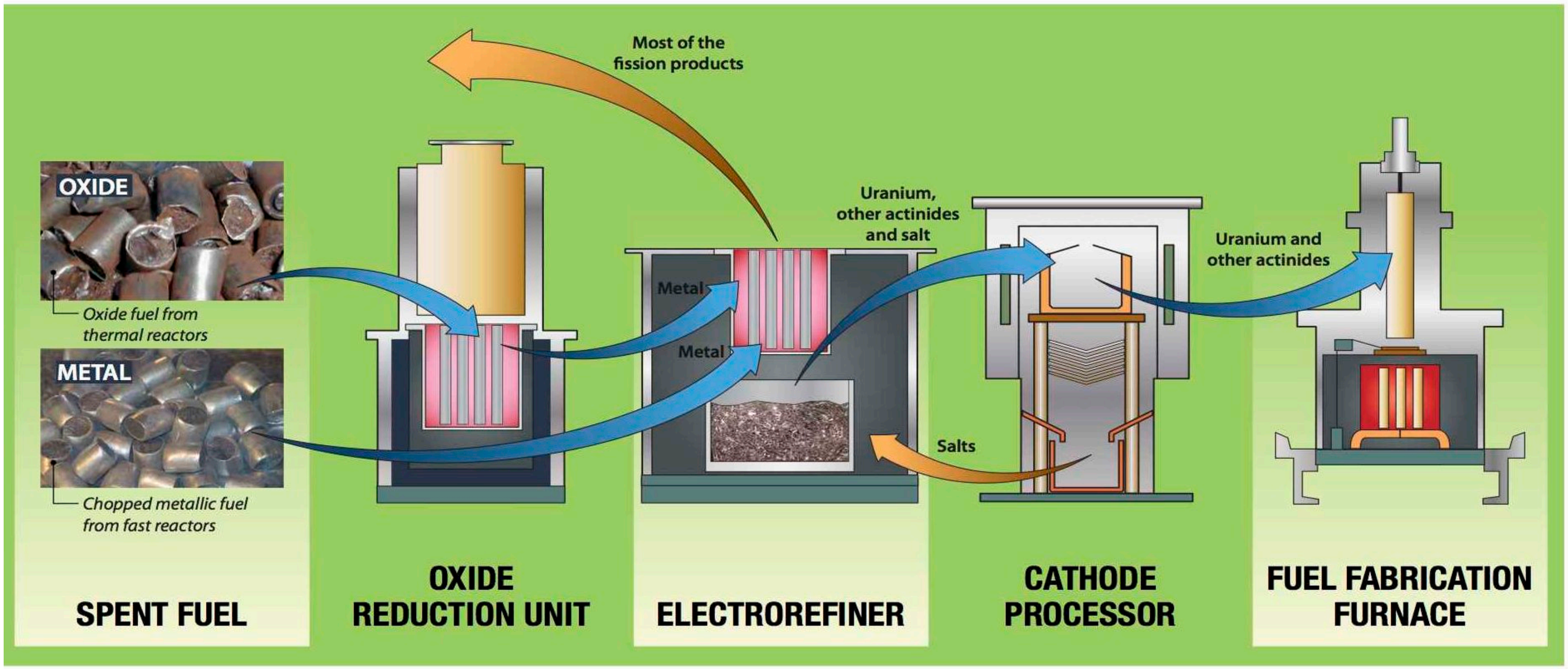
Storage pool at La Hague

Vitrification cell at La Hague



Pyroprocessing Technology

- U-Pu metallic fuel is loaded at an anode in a molten salt electro-refiner.
- Electric current dissolves used fuel and plates out U-Pu on the cathode.



http://www.ne.anl.gov/About/headlines/20120723.shtml

Proliferation Potential of Separations Technology

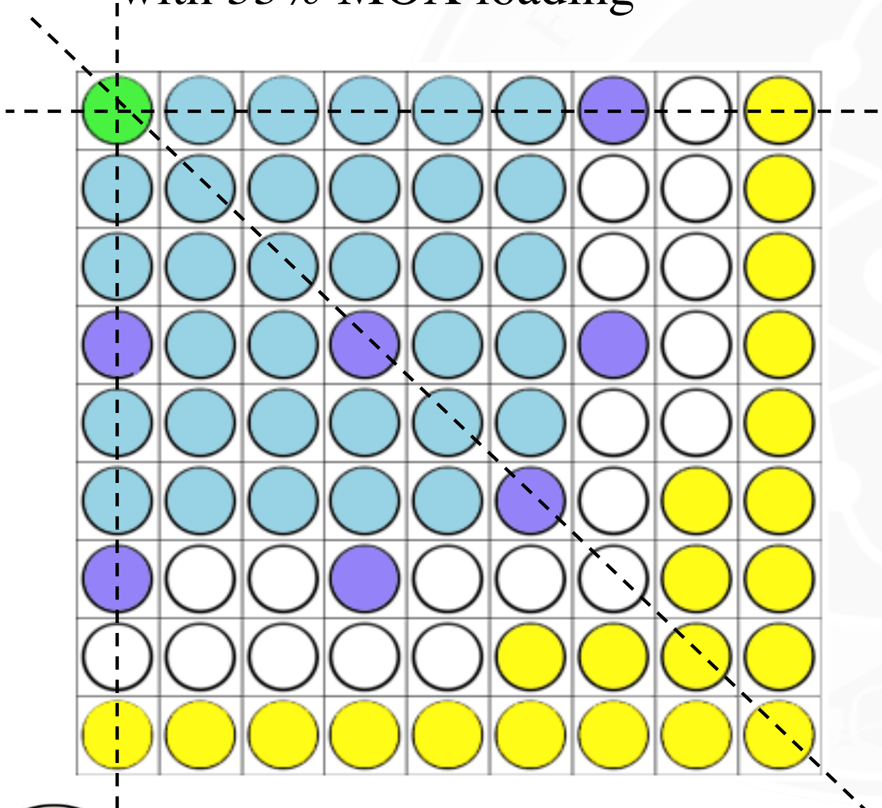
- Aqueous Process (PUREX)
  - Separates SNM but the material is protected via safeguards steps and physical protection in production facilities.
  - Modifying extractants and/or reductants could yield a separated stream of SNM.
- Non-aqueous Process (Pyroprocessing)
  - Batch process involving electro-refining technology in molten salt.
  - FPs (lanthanides) are lumped with SNM providing proliferation barrier.
  - Chemical composition of the salt may be adjusted to collect SNM.
  - Primarily developed for reprocessing of U-Pu-Zr metallic fuel.

- No separations technology is proliferation proof.

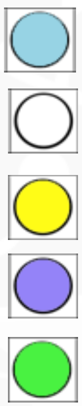
Alternate LWR Cycle: Th-Pu MOX

Thorium-Based **M**ixed-**O**xide (**TMOX**) Assembly

Standard 17x17 PWR assembly with 33% MOX loading

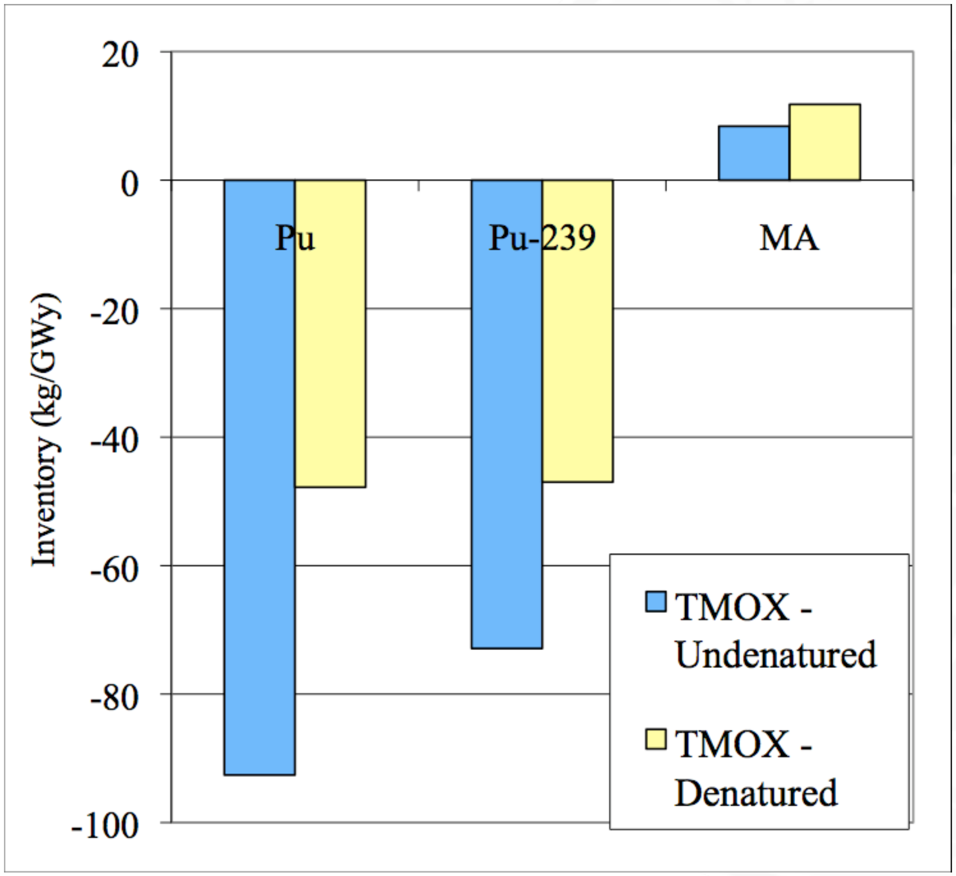


- Natural Th serves as the host for Pu in the MOX.
- TMOX not only stabilizes Pu inventory, but consumes Pu.
- Denaturing Th with ^{238}U reduces ^{233}U proliferation risk.



TMOX Performance

Net Change in Inventory



- With **zero** ^{239}Pu production, once-through TMOX allows for a deep burn of the initial Pu loading:
  - 95% ^{239}Pu destruction
  - 70 % total Pu destruction
- Denaturing U via adding natural U deteriorates the Pu depletion capability.
- Natural U also leads to a larger MA production.

Sacrifice Pu depletion and waste reduction for proliferation resistance

Anti-neutrino Monitoring of Reactor Fuel

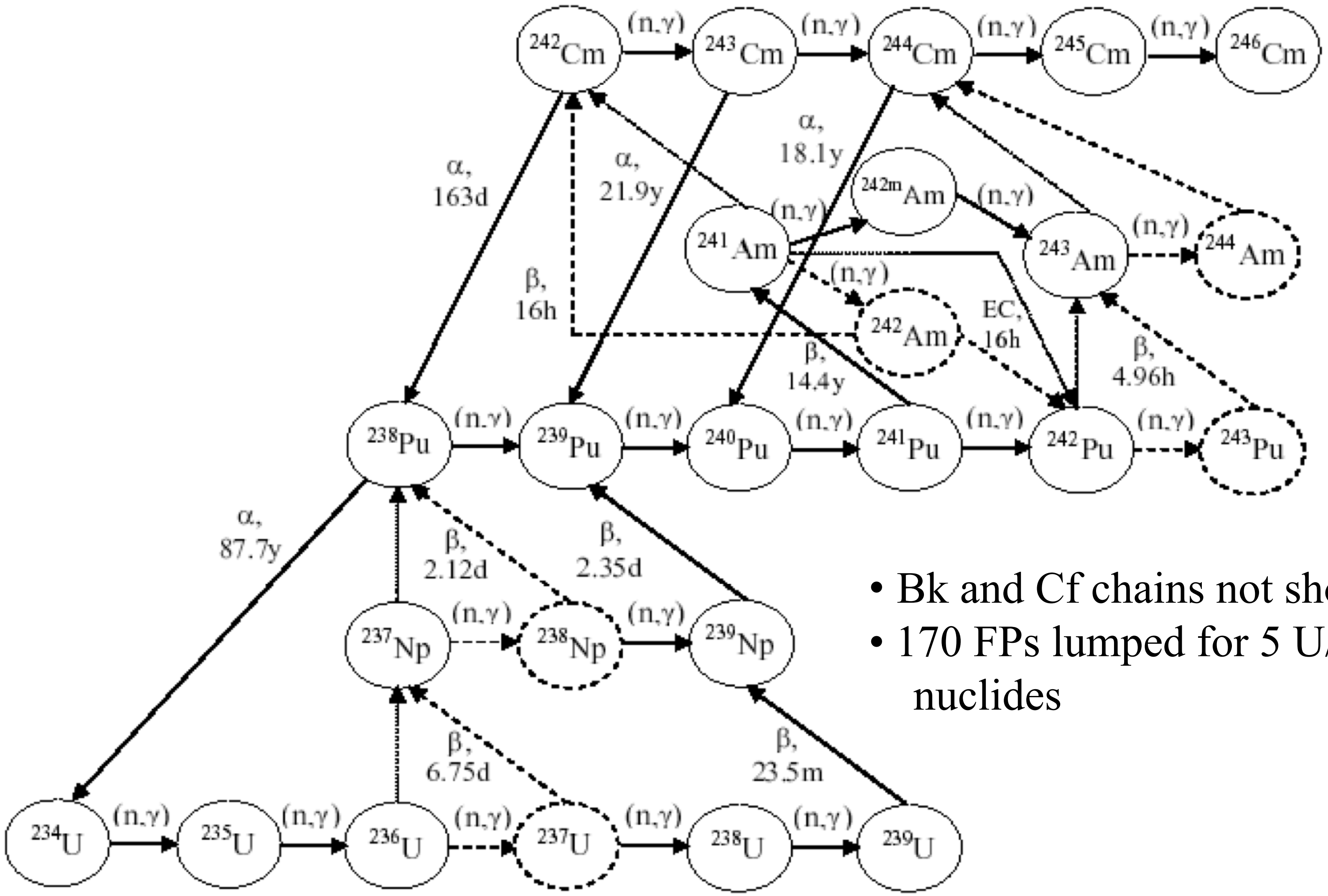
- Number of anti-neutrino produced per fission depends significantly on fissionable nuclides:

  ^{235}U: 1.9
  ^{238}U: 2.4
  ^{239}Pu: 1.5
  ^{231}Pu: 1.8

- Total anti-neutrino production rate in LWR core could be 10% different between UO_2 and MOX fuel.
- Total anti-neutrino production rate in SFR core could be 20% different between UO_2 and WG-Pu fuel.

- Need accurate data for anti-neutrino production rates for fertile Pu and higher actinides to be able to monitor fuel swaps accurately.

Typical Decay Chain for U-Pu Fuel Cycle



- Bk and Cf chains not shown
- 170 FPs lumped for 5 U/Pu nuclides

Fission Products for Radionuclide Monitoring

- NAS Report (2012) suggests increased development of radionuclide transport monitoring with the International Monitoring System.
- Lattice physics codes developed for reactor design and fuel depletion calculations focus on nuclides with large neutron absorption cross section:

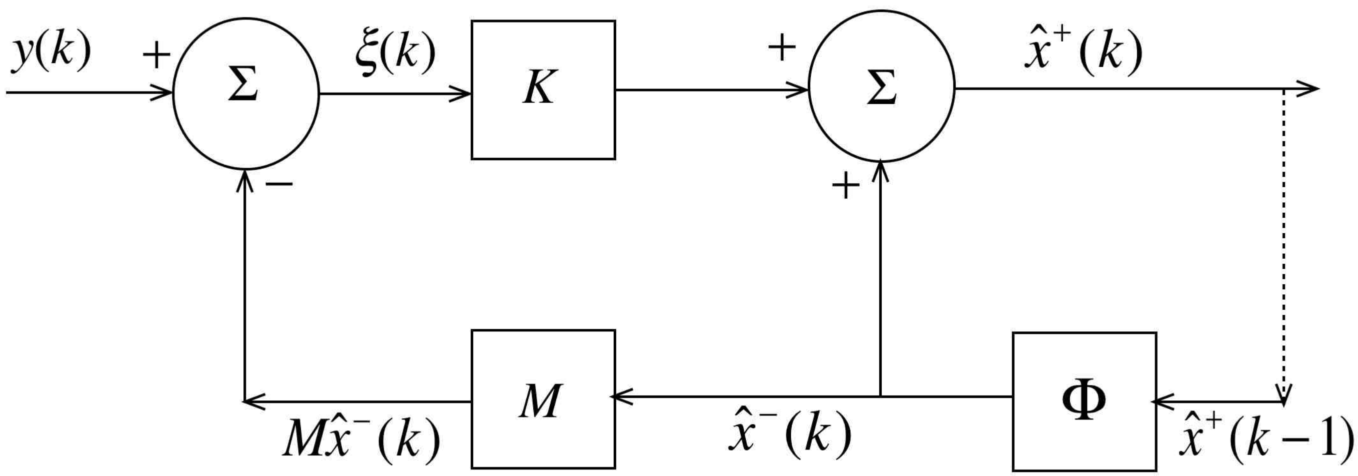
$$^{135}\text{Xe}: \sigma_a \simeq 2.7\times 10^6 \text{ b}, t_{1/2} = 9.1 \text{ h}$$
$$^{133}\text{Xe}: \sigma_a \simeq 190 \text{ b}, t_{1/2} = 5.2 \text{ d}$$
$$^{133m}\text{Xe}: \sigma_a \text{ (?)}, t_{1/2} = 2.2 \text{ d}$$

- Remaining 170 FP nuclides are lumped for 5 major U and Pu nuclides.
- ENDF-VII offers nuclear data for ~400 nuclides and TENDL-2013 includes >2000 nuclides.
- Isotopic depletion code ORIGEN represents 1119 FPs but performs one-group ENDF-VI calculations for a few pre-calculated neutron spectra.
- Need to develop accurate FP generation and transmutation modeling capability for representative fuel designs.

Time-dependent Radionuclide Transport Modeling

- Kalman filter generates minimum-variance (optimal) estimates for time-dependent system behavior combining uncertain system predictions with noisy observations.
- The filter algorithm naturally accounts for multi-modal observations.
- Recent developments of unscented Kalman filter overcomes the limitation of traditional algorithm requiring linear system representation.

$$\xi(k) = y(k) - M\hat{x}^{-}(k)$$



Nuclear Fuel Cycle Analysis and FP Monitoring

- Fuel cycle simulation code VISION represents fuel inventories, material flows, transmutations, economics, and other system interactions for nuclear reactor systems.
- VISION comprises several Excel I/O files built around Powersim system dynamics simulation software.
- Cyclus performs fuel cycle simulations using agent-based algorithms with agent interactions represented via dynamic resource exchange.
- Agent structure allows flexibility in implementing various fuel cycle scenarios and modes of interactions between systems.

- Proliferation markers, e.g., ^{85}Kr, ^{129}I, and ^{3}H in the off-gas stream of the PUREX process or ^{133}Xe, ^{131}I, or ^{137}Cs emission from clandestine tests could be tracked via Kalman filter and incorporated as a Cyclus agent.
- Ability to model and track FPs accurately will be useful for cross-calibration and benchmarking of IMS data.