

# Requirements Driven Comprehensive Approach to Fuel Cycle Back-End Optimization

F. Franceschini, M. Carelli, E. Lahoda, M. Wenner, P. Ferroni,  
Y. Arafat, J. Lyons, M. Kretzing

*Research and Technology Unit  
Westinghouse Electric Company, Pittsburgh, PA, USA*

B. Petrovic, L.M. Huang, A. Adeniyi

*Nuclear and Radiological Engineering  
Georgia Institute of Technology, Atlanta, GA, USA*

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**Georgia  
Tech**



# Motivation

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Technical issues

Public acceptability/acceptance of nuclear power ?

Requires adequately addressing:

- Sustainability
- Used/Spent nuclear fuel and high level waste management

# Issues

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- Current Yucca Mountain site (and future “Yucca Mountains”) not deemed acceptable
  - Required time for isolation too long (tens of thousands to millions of years), difficult to demonstrate
  - Need a new repository every N years

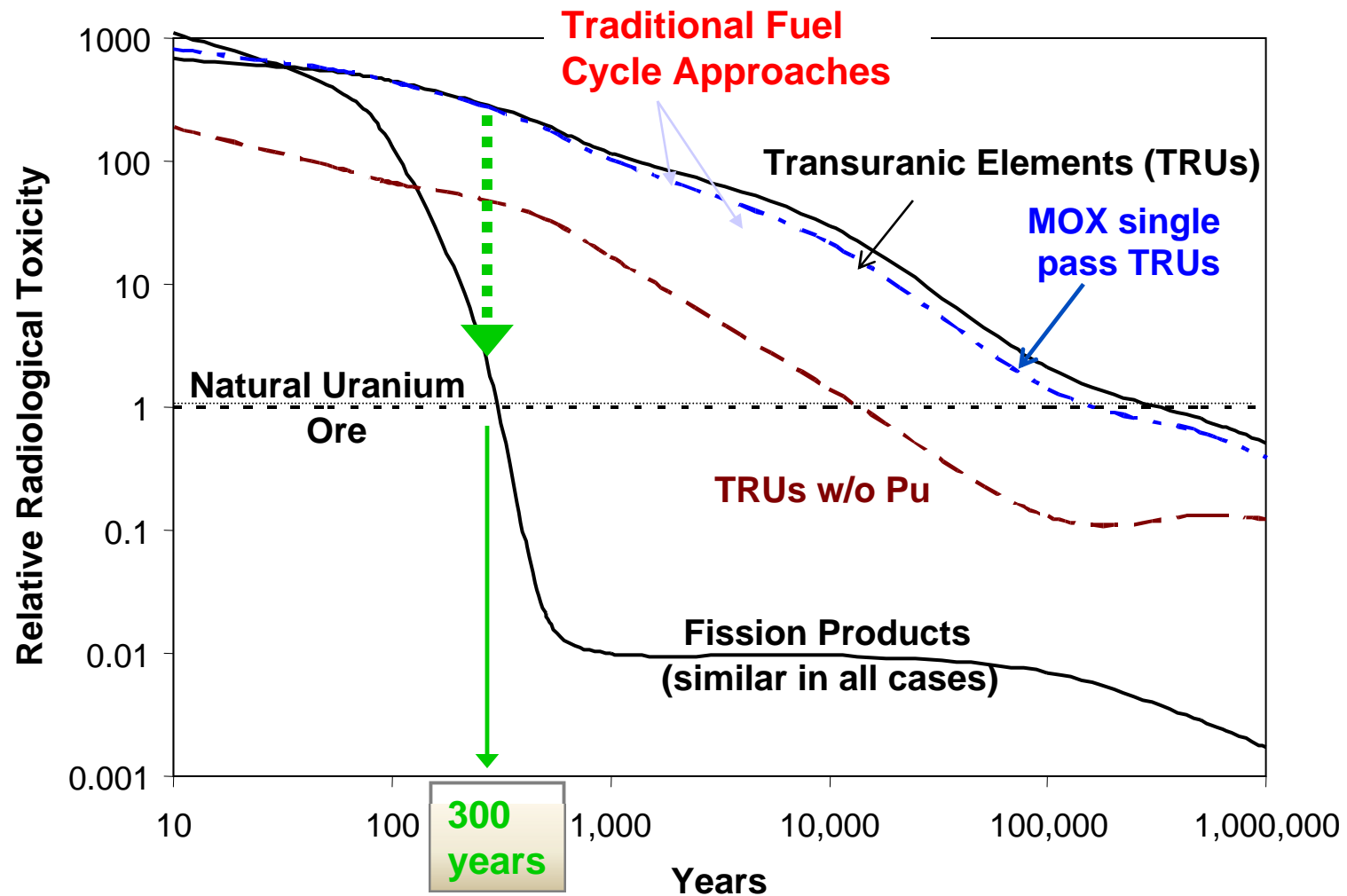
# Objective

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Goal – Generate high level waste with “acceptable” isolation time requirement and radiotoxicity level

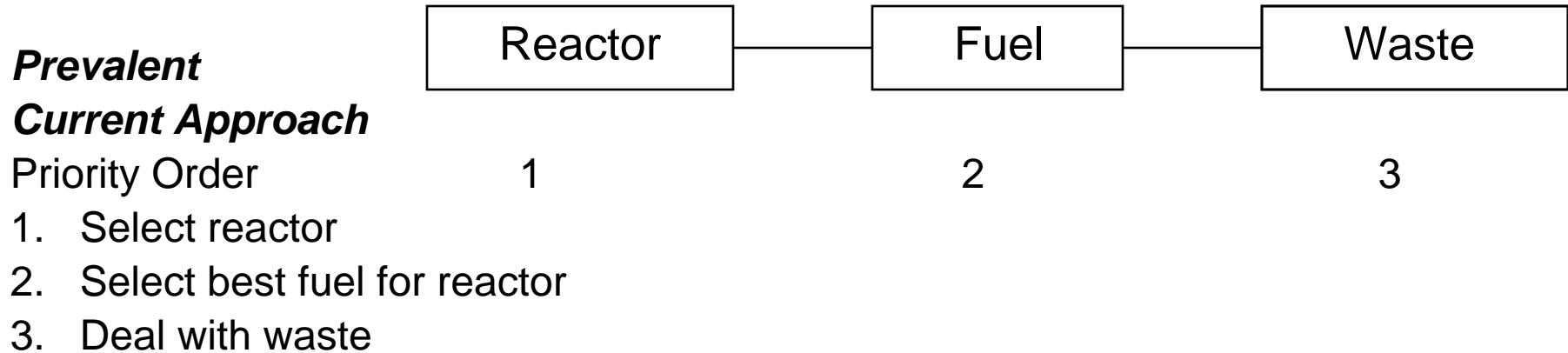
- Assumed acceptable/reasonable time – 300 years
  - Somewhat arbitrary but defensible
- Assumed acceptable/reasonable level – same radiotoxicity as the “equivalent” amount of open-cycle U ore
  - More concentrated, so volume is lower
- “Essentially” no permanent geological repositories

# Transuranic Elements (TRUs) Major Contributors to Long Term Spent Fuel Radiotoxicity



# Approaches to the Nuclear Fuel Cycle

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## **Proposed Approach**

- |                |  |   |   |
|----------------|--|---|---|
| Priority Order | 3  | 2 | 1 |
|                | 1. Define waste objective  |   |   |
|                | 2. Select fuel/cycle best satisfying waste objective                                   |   |   |
|                | 3. Select critical/subcritical system(s) to achieve objective                          |   |   |
|                | 4. Assess potential solutions for technical feasibility and integrated fuel cycle cost |   |   |

**Attempt to avoid pre-conceived solutions, keep all options open longer**

# Key Attributes of Proposed Approach

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- Public acceptance: address concerns on permanent disposal
- Goal-oriented: Reduction of the waste to ore level toxicity in 300 years
- Science-based: The physics of the fuel cycles/transmutations dictates the technology choices
- Solution driven: Each identified technology will be utilized in the most efficient way to reach the ultimate goal
- All present fuel cycle technology (reactors, subcriticals, separation, fabrication) will be utilized to the optimum extent
- New technologies will be developed and introduced as necessary

# Technology Needs

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- Suitable reprocessing technology
- Suitable reactor technology
- Remote fuel fabrication
- Proliferation controls



# Required Capabilities

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- Current and Advanced Reactors (LWR, FR, etc.)
- Interim storage of spent fuel
- Fuel reprocessing
- Interim storage of HLW
- Final disposal of HLW
- Hot and cold fuel fabrication
- Transportation of hot and cold fuel
- Fuel tracking and monitoring
- Transportation of spent fuel

# Path to Evaluation and Implementation

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- Develop capability for extended scenario studies, including sustainability, radiotoxicity, and proliferation characteristics
- Develop models of various reactor and reprocessing options
- Develop isotopic mass balance flow-sheets for various cases
- Determine reactor and reprocessing specifications required to meet the 300-year waste requirement for each case
  - Isotopic composition of fuel at reactor discharge
  - Isotopic composition of streams after reprocessing
- Identify technical, proliferation and cost issues for each case
- Generate specifications for future development programs
  - Fast reactor fuel and fast reactor performance for TRU incineration
  - Reprocessing in terms of elemental recovery ratios

# Progress to date

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- Preliminary approach developed
- Simulation capabilities established/underway
  - Core physics tools (current and advanced systems)
  - Preliminary calculations and benchmarks
  - Devise and evaluate fuel cycle scenarios to scope alternatives
  - Waste radiotoxicity
  - Economics
  - Proliferation resistance
- Specific analyses performed on thorium fuel implementation in multi-tier systems
- Potential role of fission-fusion hybrids and ADS in multi-tier systems
- Open to any technology that could contribute to accomplishing the goals
- Continue to build comprehensive analysis capabilities

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# Fuel Cycle Options

# Open Cycle

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- Dominant fuel cycle
- Poor use of U resources (<1%)
- Generates large volume of radiotoxic material
  
- Typical PWR (UO<sub>2</sub>, up to 5% enriched in <sup>235</sup>U)
- Discharged fuel
  - ~94 wt% U (<1% <sup>235</sup>U)
  - ~5 wt% FP
  - ~1 wt% Pu
  - ~0.1 wt% MAs
  
- Long-term evolution of main radiotoxicity drivers in LWR SNF
  - Pu: 85-90% from 1,000 to 100,000 yrs; ~50% at 1,000,000 yrs
  - Am: varying between ~10% and a few percent over that time

# Radiotoxicity Components of Open Cycle LWR SNF

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- Pu is responsible for ~90% of intermediate/long-term radiotoxicity of LWR SNF
- MAs (Np,Am,Cm) make up the rest
  - Lower concentration than Pu but their specific radiotoxicity higher
- In very long term (>1e6 yrs) U daughters from tails going to secular equilibrium become dominant (~30% of total, but overall level is low at that time)
- Fission products dominate short-term (<300 yrs)
  - Long-lived FP responsible for ~0.1% of long-term residual radiotoxicity
  - Risk may be higher due to migration potential vs. actinides

# Option – Pu recycle in MOX fuel

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- Burn Pu (or Pu+MA) in LWR

## Issues:

- Pu vector degrades, MA content increases
- Need to add more and more fissile to compensate
- Safety parameters deteriorate
- Limited to one, perhaps two passes
  
- Not a long-term solution
- However, this is the only actually implemented alternative to open cycle

# Option – P&T with Full TRU Recycle and Burn

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- Significantly improves utilization of resources (compared to open cycle)
- Reduces volume and radiotoxicity of waste
- Potential to satisfy the 300-year objective
- Requires a number of conditions to be met:
  - Availability of fast spectrum systems (critical, subcritical)
  - Acceptable reprocessing recovery fractions and losses
  - Feasibility of deep burn of TRUs
  - Fuel manufacturability and performance
  - Safe operation of all systems/facilities



# Issues – Full TRU Recycle and Burn via P&T

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- Deep burn of MAs?
- Am and Cm content of recycled fuel tends to increase
- None of the considered approaches is fully satisfactory
  - Homogeneous recycle of Am/Cm (within fuel)
  - Heterogeneous recycle of Am/Cm (in targets)
  - Incineration in ADS/hybrids
- In any case, will need to develop technology to cope with:
  - increasingly hot fuel
  - fuel manufacturability issues
  - recovery ratio challenges  
(need to recover 99.9% Pu and 99% MA from LWR SNF)

# Challenges to Reactor Design and Overall Economics

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- A combination of fast reactors, burners, likely required
- ADS or F-F hybrids likely needed to complete the burning
- More complex design of reactors/systems and their operation (MA content, safety parameters, target design/performance)
- Need to address the “legacy” fuel (that will keep being produced) as well as the “new” fuel(s)

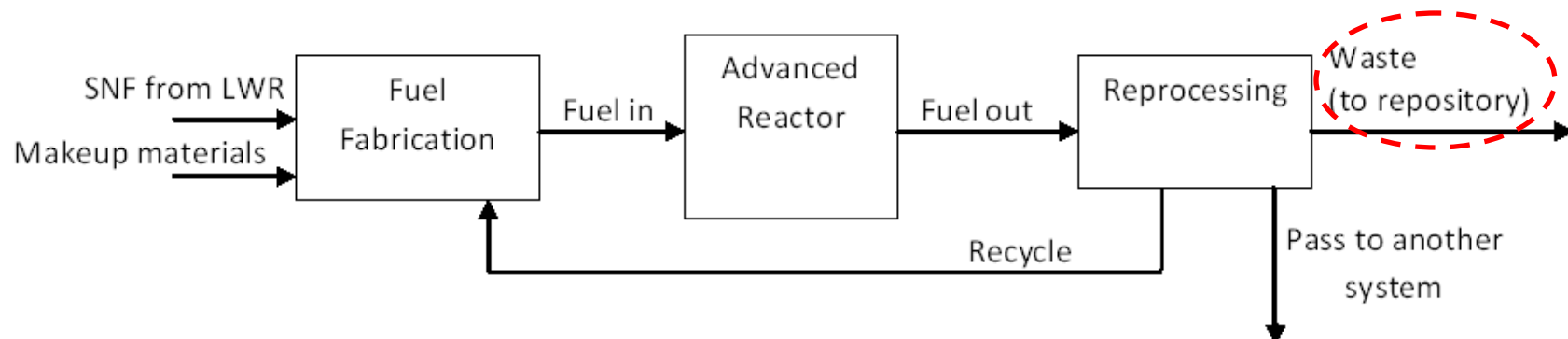
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# Fuel Cycle Modeling



# Methodology

- Material balance and isotopic tracking across the system:
  - Fabrication, irradiation, reprocessing, etc.
  - Transmutation based on or reproduced from in-core neutronic analysis
  - Relevant isotopes tracked across the system
  - Recycle of discharged fuel
  - Different reprocessing options and isotopic recovery ratios
  - Radiotoxicity of wastes for various scenarios
  - Analysis of initial and final state plus transition cycles



# Reprocessing

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- Various reprocessing options are examined to separate and recycle critical elements from discharged fuel with desired efficiency :
  - PUREX, UREX's
  - AIROX
  - Pyroprocessing
  - Fluorex
  - Thorex
- Discriminating Factors:
  - Radiotoxicity
  - Proliferation resistance
  - Cost effectiveness
- Current state-of-the-art barely enables meeting the 300-year objective for LWR SNF. Further challenges for recycled fuel (higher MA content, hotter fuel, higher efficiency needed)

# PUREX & UREX's

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- PUREX – original aqueous reprocessing that separates U and Pu
  - Proliferation concerns – pure Pu stream
- Additional chemical separations added to form derivations of PUREX
  - UREX+1/1a/1b
  - UREX+2/2a
  - UREX+3/3a
  - UREX+4/4a
- Advantages/benefits
  - PUREX is based on industry experience (demonstrated)
  - UREXs widely researched; addresses proliferation resistance

# Pyroprocessing

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- High-temperature dry reprocessing technology that utilizes electrolytic separation (electro-refining)
- Elements targeted: U, TRUs, Rare Earths
- Advantages/benefits
  - More proliferation resistant than aqueous methods – no possibility to recover pure Pu
  - Dry reprocessing technology – low risk of criticality
  - Can be used for oxide or metal fuels
  - Simple process – lower cost
  - Can be located at same facility as fast reactor – no need for transportation

# Fluorex & Thorex

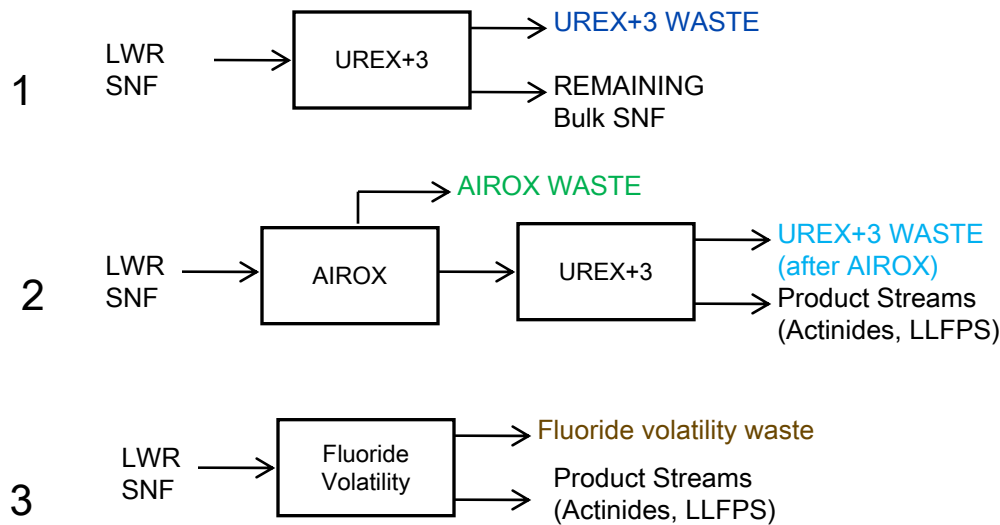
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- Fluorex
  - Combines fluoride volatility and solvent extraction methods
  - Elements targeted: U, Np, Am, Cm, & LL FPs
  - Advantages/benefits
    - » High recovery of U
    - » Greatly decreased criticality problem (absence of neutron moderator)
    - » Reduced proliferation concerns (no ability to separate pure Pu)
- Thorex
  - Similar to PUREX, but for use with Thorium based fuels
  - Separates Thorium and Uranium from the bulk waste
    - » Co-decontamination process
    - » Does not separate any actinides
    - » Would most likely need additional reprocessing to separate out the actinides



# Advanced Reprocessing Technologies

Advanced reprocessing options analyzed to evaluate potential isotopic recovery and impact on the waste radiotoxicity



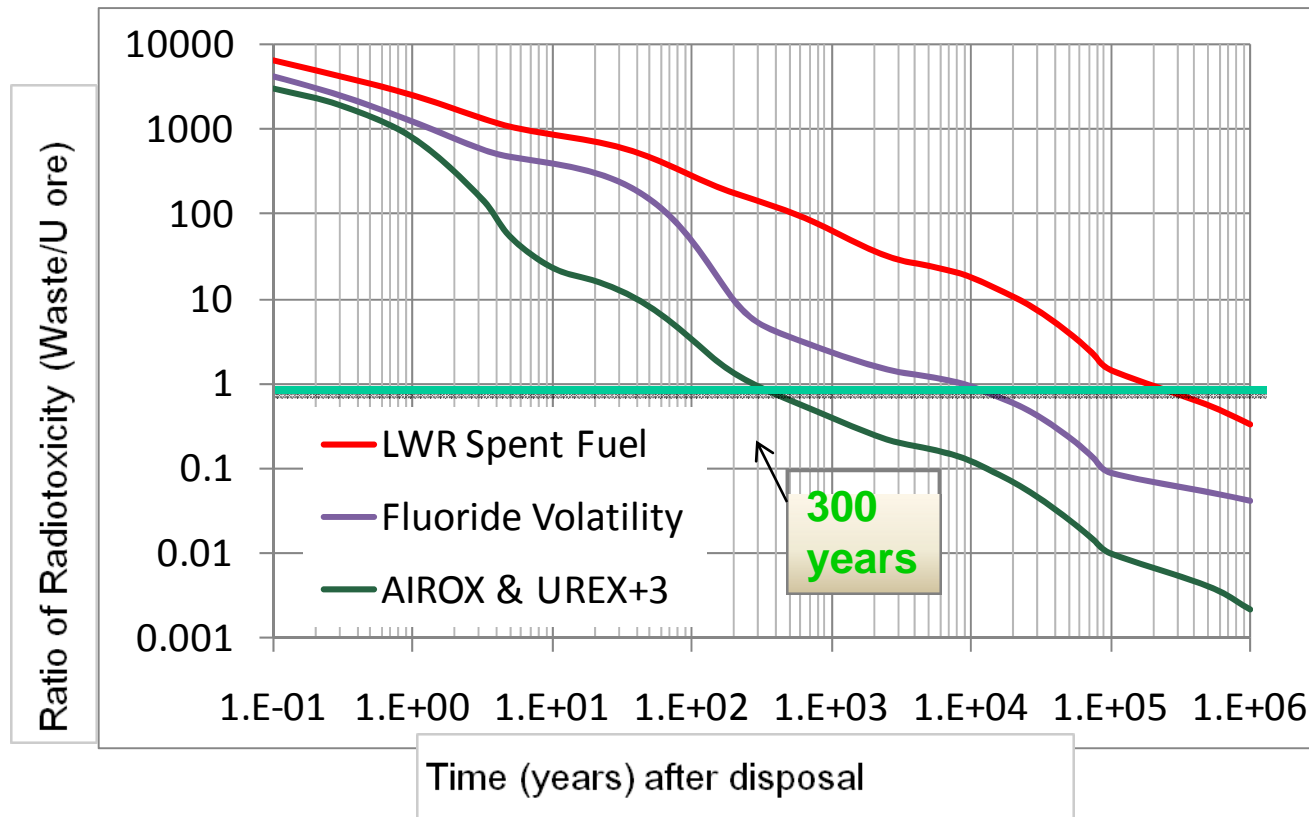
AIROX	
Element	Recovery Ratio
H	1
C	1
Kr	1
I	1
Xe	1
Cs	0.9
Ru	0.9
Cd	0.75
Te	0.75
In	0.75

UREX +3	
Element	Recovery Ratio
U	0.9995
Pu	0.995
Am	0.98
Cm	0.79
Np	0.71
Cs	0.96
Sr	0.99
Tc	0.95
Lanthanides	-
Rare Earths	0.272

FLUORIDE VOLATILITY	
Element	Recovery Ratio
U	0.99
Pu	0.99
H	1
C	1
Kr	1
I	1
Cs	0.98
Tc	1
Ru	1
Rh	0.8
Te	0.9
Mo	0.8

# Radiotoxicity of wastes from LWR SNF

- Advanced reprocessing (AIROX+UREX) applied to LWR SNF barely enables meeting the 300 yrs objective
- Available margin may not be adequate for multi-recycled fuel
- Further advanced reprocessing options will be investigated or developed



# Reprocessing Specifications

- Back-solving from waste to identify elements to be separated and respective reprocessing ratios to achieve the waste target.
- Current reprocessing technologies may not be adequate to fulfill the P&T requirements, either due to low recovery ratios or lack of separation of critical elements/isotopes.
- Collaboration of industry and National Labs would promote achieving those particular reprocessing specifications and evaluate practical feasibility so that the radiotoxicity of the reprocessing waste streams meet 300-year goal.
- Need to scale up and satisfy economical constraints

LWR SNF →

Reprocessing	
Theoretical Specifications	
Element	Recovery Ratio
U	0.9995
Pu	0.995
Am	0.99
Cm	0.999
Np	0.99
Cs	0.98
Sr	0.99
Tc	0.999
Lanthanides	-
Rare Earths	0.272

→ UREX+3 WASTE

→ REMAINING Bulk SNF

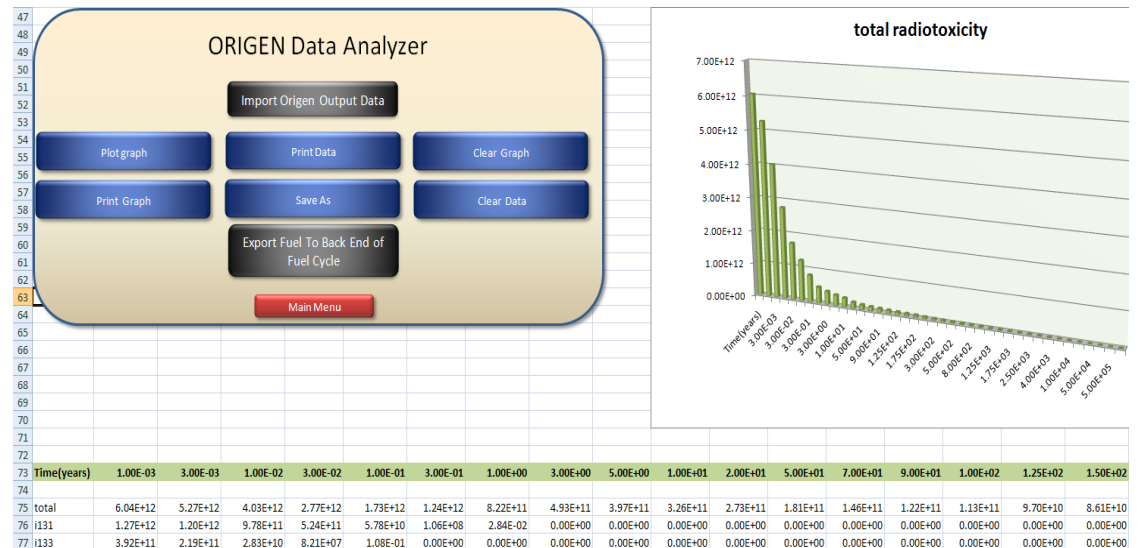
# Approach to integrate reprocessing and reactor options

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- A promising fuel cycle (reactors, fuel technology, fuel composition, discharge burnup etc.) to reach objective is developed
- Scenario calculations for the selected option are performed
- Mass balances and waste streams from “best” reprocessing option (viable, economic, proliferation resistant) calculated
- Waste stream composition used to calculate radiotoxicity
- Optimization feedback on fuel, reactor, reprocessing
  - weak points throughout the system identified
  - potential margins for improvement assessed
  - redesign and reevaluate

# Radiotoxicity Calculation GUI (under development)

- VBA based GUI was developed at Westinghouse to facilitate transferring the ORIGEN output into EXCEL
- It can plot/print/save for individual isotopic values or the total for any ORIGEN output
- It can convert isotopic data from the Waste Calculator and convert them to the respective ZAIDs so that an ORIGEN input can be set up very fast
- Intuitive GUI to help user modify data fast and efficiently



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# Evaluation of Thorium Fuel Cycle Potential toward Meeting the Waste Objectives

## Some advantages of thorium...

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- More abundant than Uranium, more concentrated
- Not fissile, but can breed  $^{233}\text{U}$  as it is irradiated
- No need for enrichment with Th plus better fuel use
  - Reduced mining, no tails, much better front end
  - Sustainability
- Very stable, can withstand high burnup

## ...and some drawbacks of thorium

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- Need fissile to start the cycle
- May need to top-up depending on breeding capability
- Need reprocessing to recover  $^{233}\text{U}$
- General features and infrastructure not as known/established as for U (U-Pu) cycle
- Fuel stability may entail additional difficulty in reprocessing (dissolution)
- Shielding required for gammas from  $^{232}\text{U}$  daughters
- **However Th generates much lower TRU (Pu+MA) than the current U-Pu cycle**
- **Concern for waste management -  $^{231}\text{Pa}$**



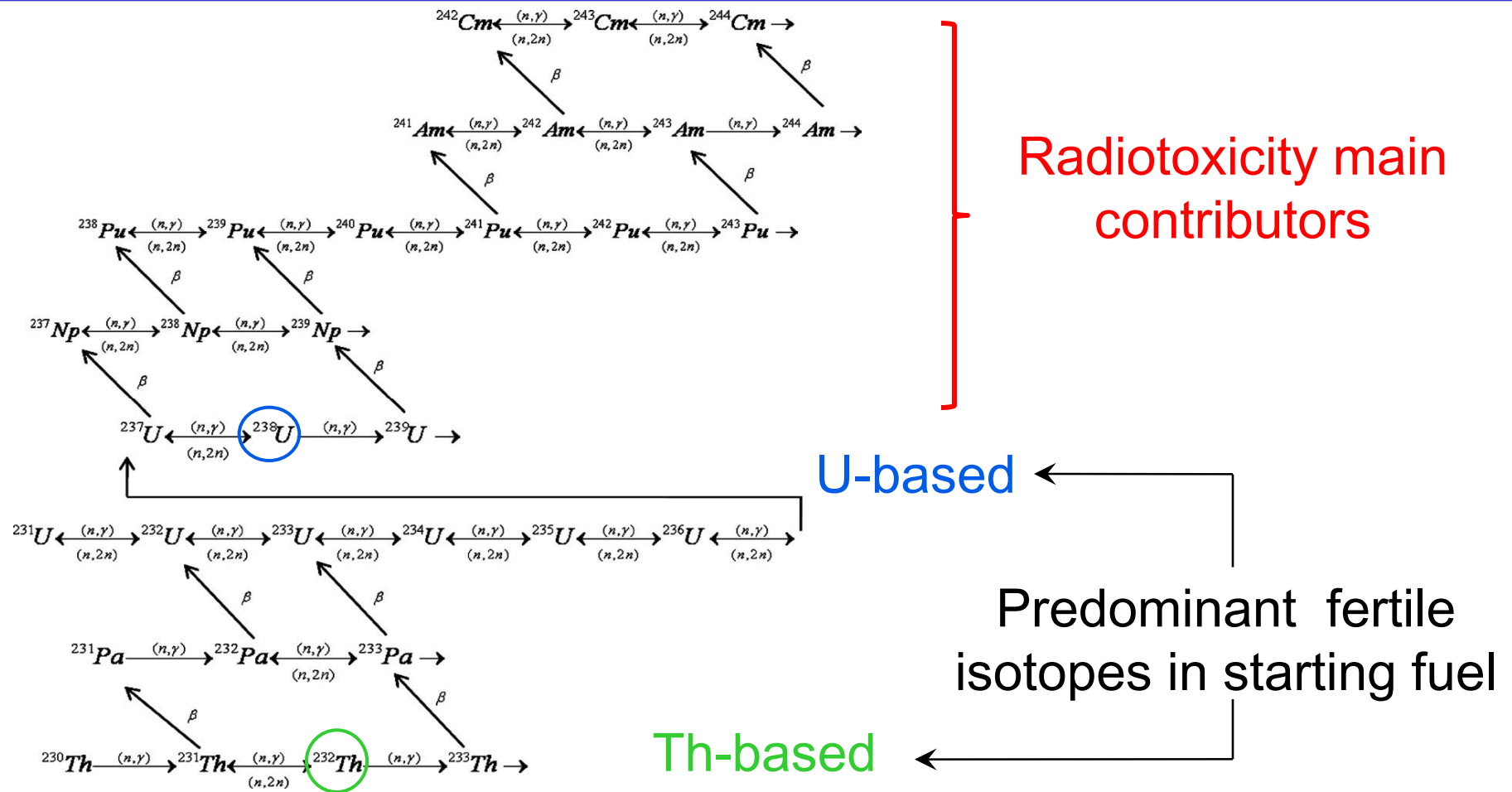
# TRUs generation with pure thorium-U233 fuel

- Thorium reduces TRU generation by orders of magnitude
- Thorium fuel is a viable industrial technology

Element	TRU at discharge (Kg/Assembly)			Ratio (Th/UO2)	Ratio (Th/MOX)
	U-based fuel		Th-based		
	UO2	MOX	Th- <sup>233</sup> UO2		
Total Pu	5.70E+00	5.26E+01	<b>2.91E-03</b>	<b>5.E-04</b>	<b>5.E-05</b>
Total Am	1.25E-01	1.82E+00	<b>2.59E-06</b>	<b>2.E-05</b>	<b>1.E-06</b>
Total Cm	5.47E-02	5.67E-01	<b>6.36E-07</b>	<b>1.E-05</b>	<b>1.E-06</b>

From "Thorium Based Fuel Cycle Options for PWRs", M. Todosow and G. Raits, Proceedings of ICAPP '10 San Diego, CA, USA, June 13-17, 2010

# Th-based fuel reduces TRUs generation because of the longer pathway for their creation



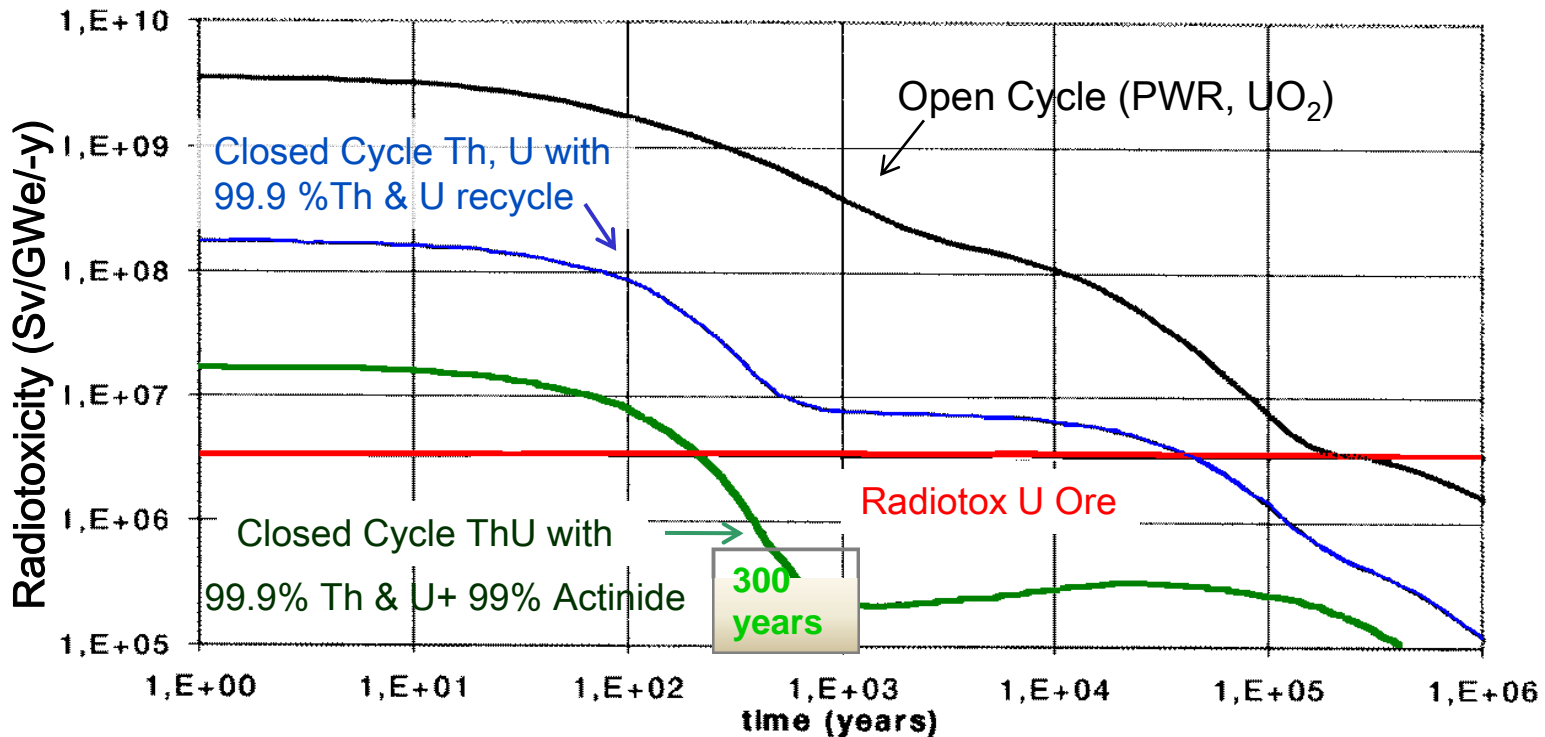
Transmutation chain from "Actinide evolution and equilibrium in fast thorium reactors" David J. Coates, Geoffrey T. Parks Annals of Nuclear Energy 37 (2010) 1076–1088

# Thorium Can Provide a Viable Solution

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- Thorium has been considered for many decades and proposed by various organizations
- Considering the front-end and core residence time only, it had not much chance to economically compete vs. the U-Pu cycle
- The U-Pu cycle can rely on
  - well known and established industrial infrastructure
  - low price and availability
- However, if the criterion is waste reduction/elimination, a Th-<sup>233</sup>U cycle due to its significantly lower TRU/radiotoxicity, may become overall more economical

# The “300-year” goal is achievable with thorium



- Thorium fuel with appropriate reprocessing technology has the potential to close the fuel cycle and bring the waste radiotoxicity below the corresponding open-cycle uranium ore in 300 years

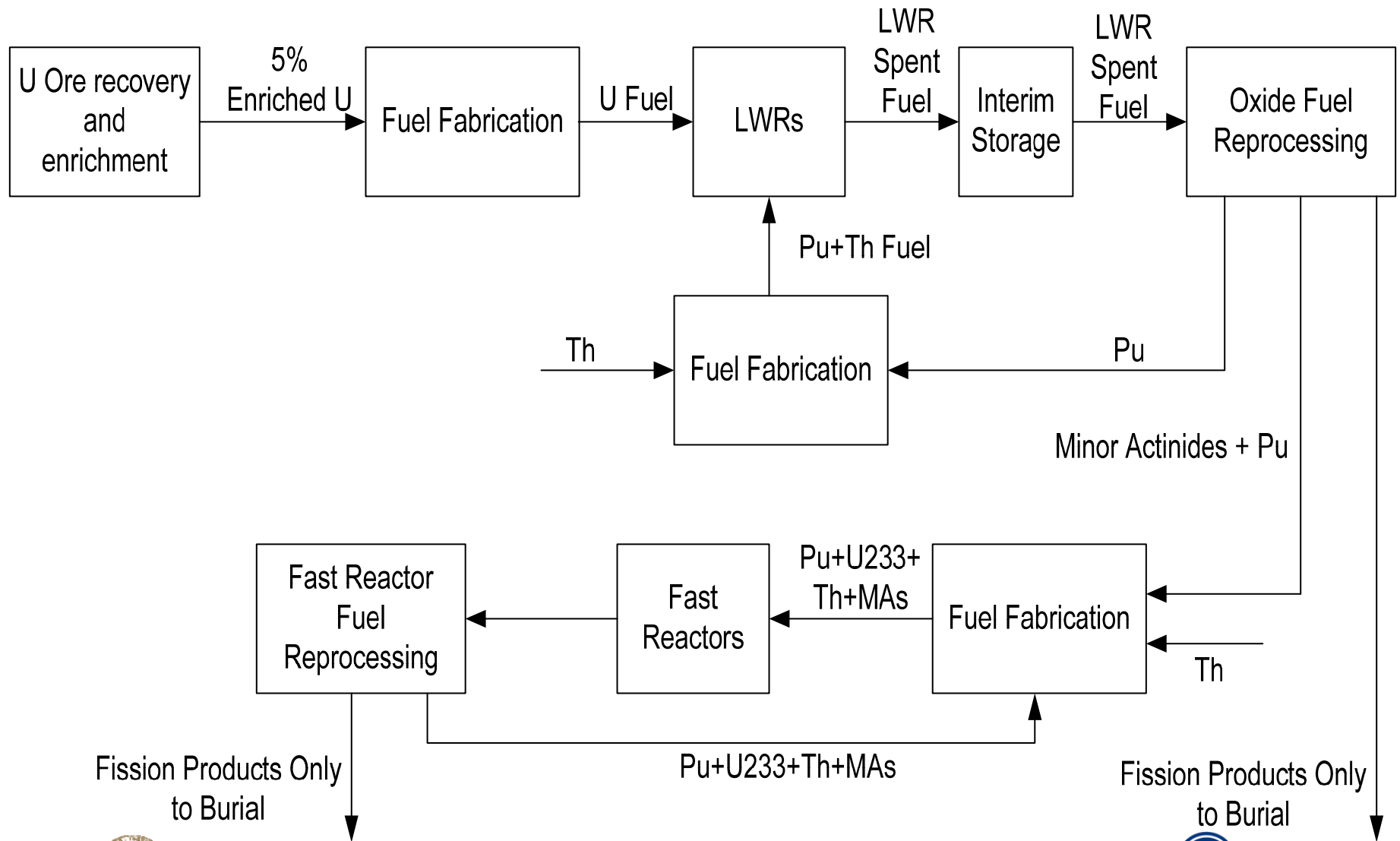
Based on data published in EC report, “Thorium as a waste management option”, H. Gruppelaar (Ed.), 2000

# Approach to Evaluate the Thorium Cycle from the Standpoint of Addressing the Waste Problem

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1. Perform an evaluation of the long-term fuel cycle potential with thorium
  - assume infinite supply of U-233
  - evaluate radiotoxicity in the ideal case
  - assess breeding of U-233 required for sustainability
2. Evaluate the transition cycles:
  - how to make best use of thorium to reduce/eliminate legacy-waste generated in the U-Pu cycle and breed required  $^{233}\text{U}$  to sustain a pure Th-U233 cycle
  - reprocessing technology to meet the radiotoxicity goal

# Possible multi-tier approach to deal with legacy



# Comprehensive Approach

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- Combination of reactors with various fissile requirements and irradiation characteristics to meet the radiotoxicity goal and ensure sustainability
  - Current LWRs
  - Advanced LWRs
  - Fast Reactor with flexible design for breeding/burning
  - ADS and F-F hybrid
  - First generation Pu as seed to LWR (and FR if needed)
  - Full Actinide recycle in FR
  - Need design to breed enough U-233 to transition to pure Th-U-233 cycle in the long-run
  - Current high-specific power LWR may need top-up
  - Need right balance (and design) of breeders/burners

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# Simulation Capabilities for Fuel Cycle Scenario Studies



# Requirements for Scenario Simulations

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- Enable assessment of radiotoxicity and activity
  - Fuel vector and wastes (reprocessing, manufacturing)
  - Track relevant isotopes (ageing, irradiation, contaminants)
  - Inhalation/Ingestion, m<sup>3</sup> air/water, Sv and Ci
- Ensure sustainability
  - Meet energy demand with variety of reactors
  - Reactivity equivalence for various fuel types and discharge BU
  - Track relevant isotopes (ageing, irradiation)
- Proliferation resistance
  - Gamma-heating, spontaneous fission
  - Isolated fissile in stream/repository
- Evaluate other hazards and risks
  - Transport, casks
  - HLW composition and resistance to dissolution

# Requirements for comparison with U-based fuel

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- Comparison of Th-based and corresponding U-based fuel cycle
- Need a “fair” comparison of the two options on the same set of parameters and metrics
  - Radiotoxicity, sustainability, proliferation resistance etc.
- May need to enhance capabilities of the current tools accordingly
  - Isotopes tracked
  - Generate additional libraries (reactors and fuel types)
  - Amount and type of information provided
  - Proliferation resistance
  - Economics
- Possible various metrics within different objectives/constraints
  - Radiotoxicity reduction
  - Accelerated burning of legacy Pu
  - Economics (e.g. LWR vs. FR/ADS)
  - Proliferation resistance
  - Energy demand

# Conclusions

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- Westinghouse with Georgia Tech is developing a requirements driven approach to devising solution(s) for fuel cycle back-end
- Open to collaboration
- Based on using thorium-bearing fuel for HLW radiotoxicity reduction
  - Thorium-based fuel reduces TRU generation
  - Combination of reactors for breeding/burning
  - Sustainability plus legacy-waste reduction
  - Various metrics and constrains account for different scenarios
- No predetermined solution but rather a requirements-driven approach to solving the waste problem
- Utilizes present technologies to the maximum extent possible
- Develops new advanced technologies as necessary
- Devising solutions for spent fuel management will guide the development of the fuel cycle and reactors