Hybrid Processing of Spent Fuel

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GLOBAL 2005 International Conference

October 9–13, 2005,
Tsukuba International Congress Center
Tsukuba, Japan

OAK RIDGE NATIONAL LABORATORY
U. S. DEPARTMENT OF ENERGY

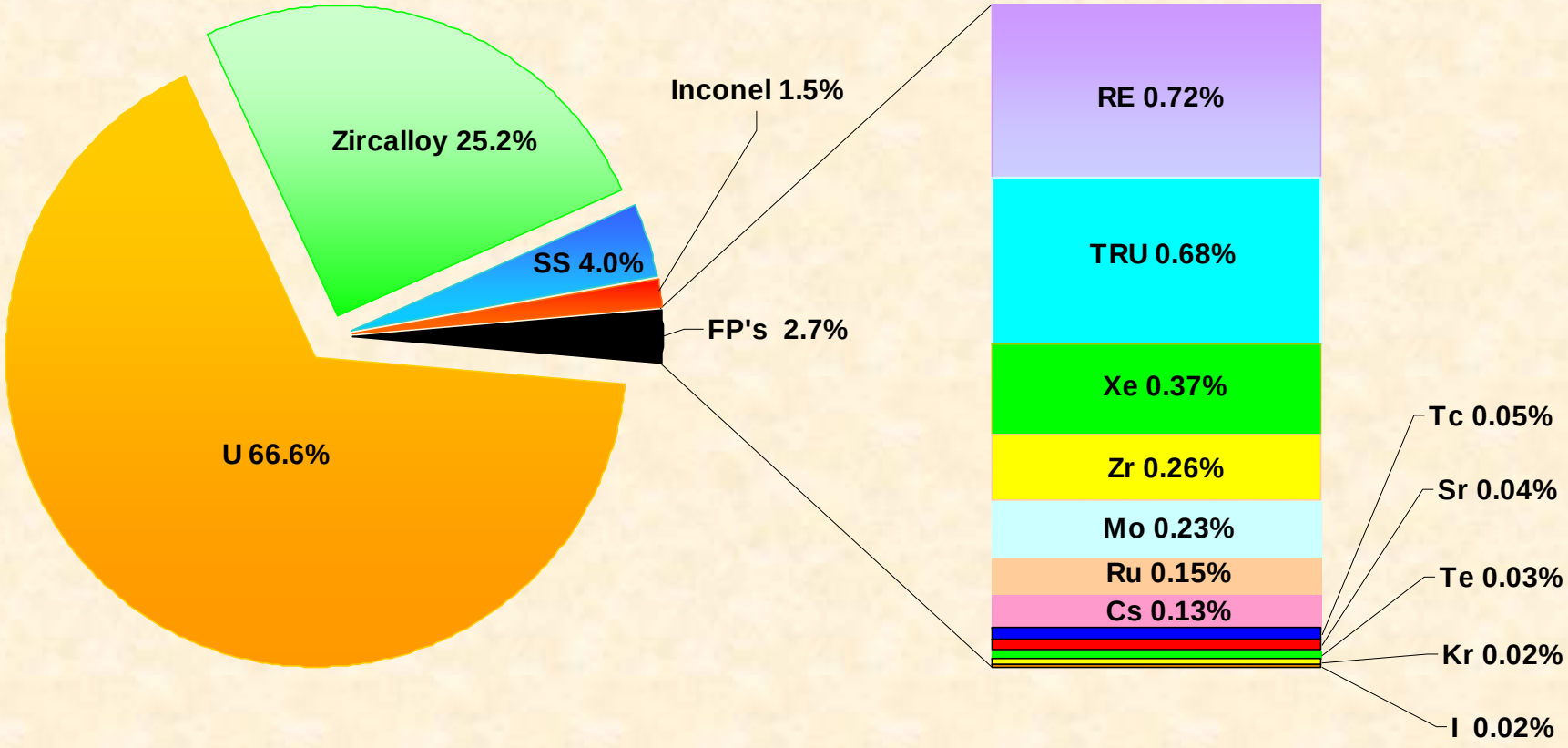
Energy System for Future Generations and Global Sustainability

- **A responsible and sustainable deployment of nuclear energy should be based on a life fuel cycle that maximizes the use and reuse of resources and minimizes the amount of wastes, particularly those requiring geological disposition**
- **The U.S. Advanced Fuel Cycle Initiative (AFCI) is developing an advanced spent-fuel treatment process that will separate the major heat sources, volatile species, long-lived fission products, actinides, and cladding and should be able to recover more than 95% of the spent fuel mass**
- **If a plant were to start-up in the United States by 2025 with a capacity matching the present annual U.S. demand for new fuel (2000 to 2500 MTU), on an oldest-fuel-first basis, the plant would never process fuel that had been cooled less than 40 years**

Why Process Spent Fuel?

- **The statutory capacity of Yucca Mountain (YM) (63 KT) will be reached around 2009-2010**
- **Afterward a capacity expansion or a 2nd repository will be needed**
- **Nuclear growth in the future will require many geological repositories**
- **Early deployment of a nuclear waste processing plant fully integrated with YM could extend the life of the repository and significantly delay the need for a second one**
- **Prolonged storage can be part of the solution**
  - **But without actual processing of fuel, storage will be regarded as "not knowing what to do" and "passing the buck to future generations"**
- **The excellent operational and safety record of nuclear reactors fostered public acceptance of nuclear energy**
- **A credible answer to the issue of spent nuclear waste is key for a "renaissance" of nuclear energy in our country**
- **To be successful, we must seek the widest possible consensus**

Composition of U.S. Spent Nuclear Fuel



Average composition, pressurized water reactor and boiling water reactor, accumulated so far (1968 to 2002) and calculated on the basis of 40 years of cooling

Spent Fuel Stored at Reactor Sites

- **During this prolonged cooling period, a significant decay of activation products, fission products, and heavy actinides—notably ^{241}Pu and ^{244}Cm—would occur, facilitating the processing and unlimited recycle of major constituents back to the existing fleet of reactors at a lower cost**

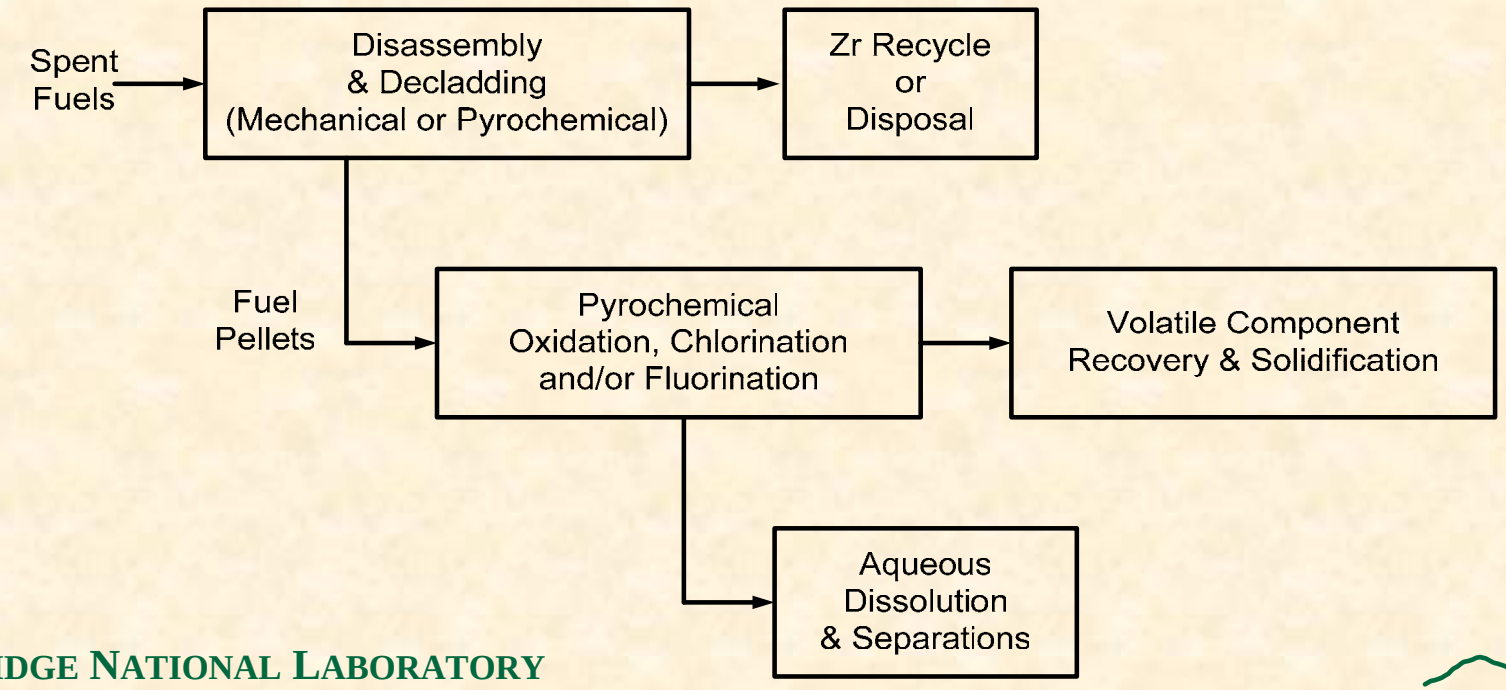
- **This combination of relatively inexpensive interim storage (e.g., 40 to 60 years) followed by processing for recycle should minimize the life cost of the fuel cycle by limiting expensive processing, maximizing the recycle, and minimizing the need for geological disposal**

As Part of This Effort, an Advanced Hybrid Process Is Being Studied

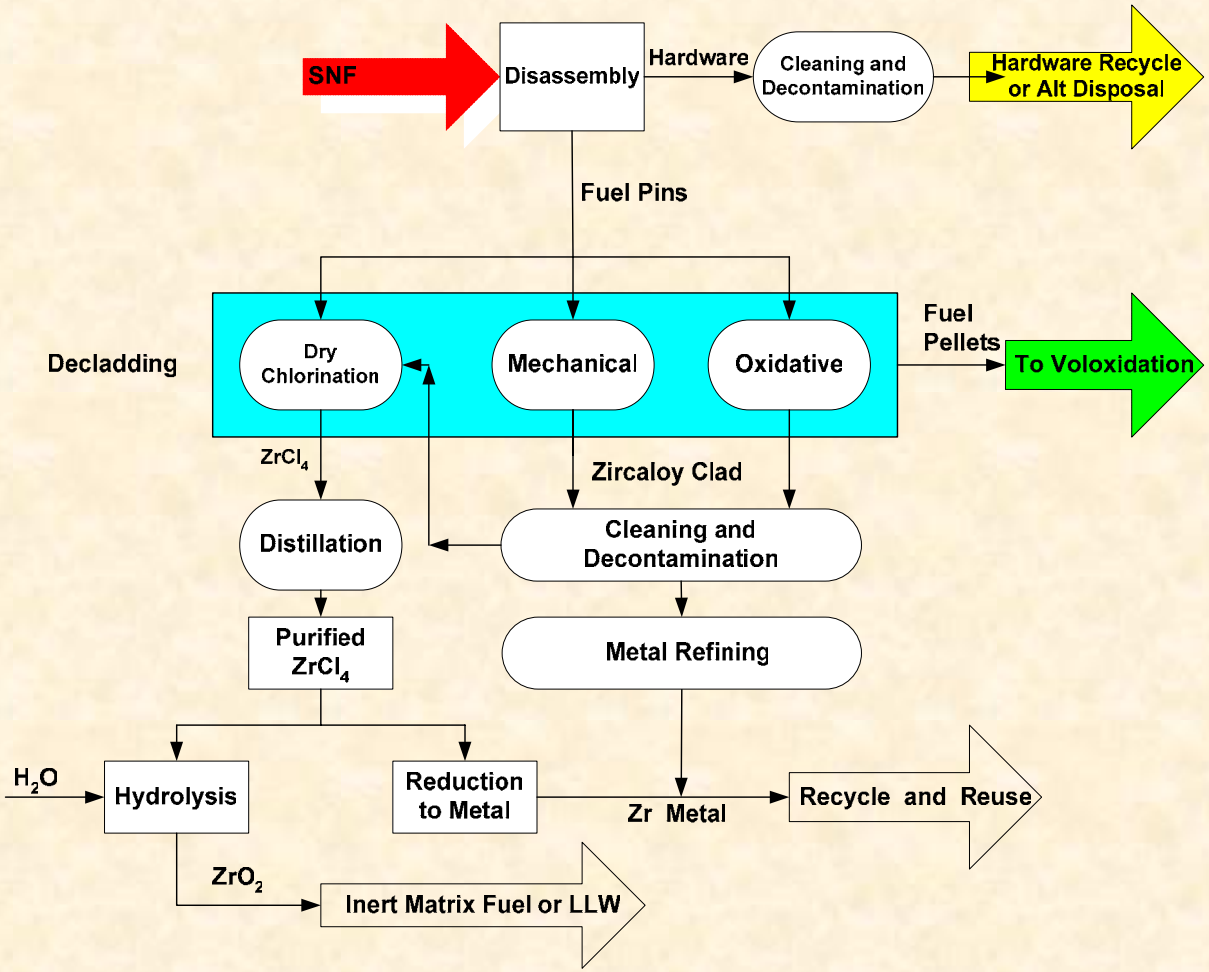
- **This scheme includes a pyrochemical head-end followed by aqueous-based separations, which may provide significant simplifications, improvements, and reduced costs**
- **The process of disassembly and decladding removes the second-largest mass and volume fraction of spent fuel components**
  - **Processes might include mechanical methods to remove the fuel pellets and produce a metal waste stream suitable for either recycle or low-level waste disposal**
  - **Alternatively, a chemical decladding method might be used to remove and recover the zirconium with similar or better results**

As Part of This Effort, an Advanced Hybrid Process Is Being Studied (continued)

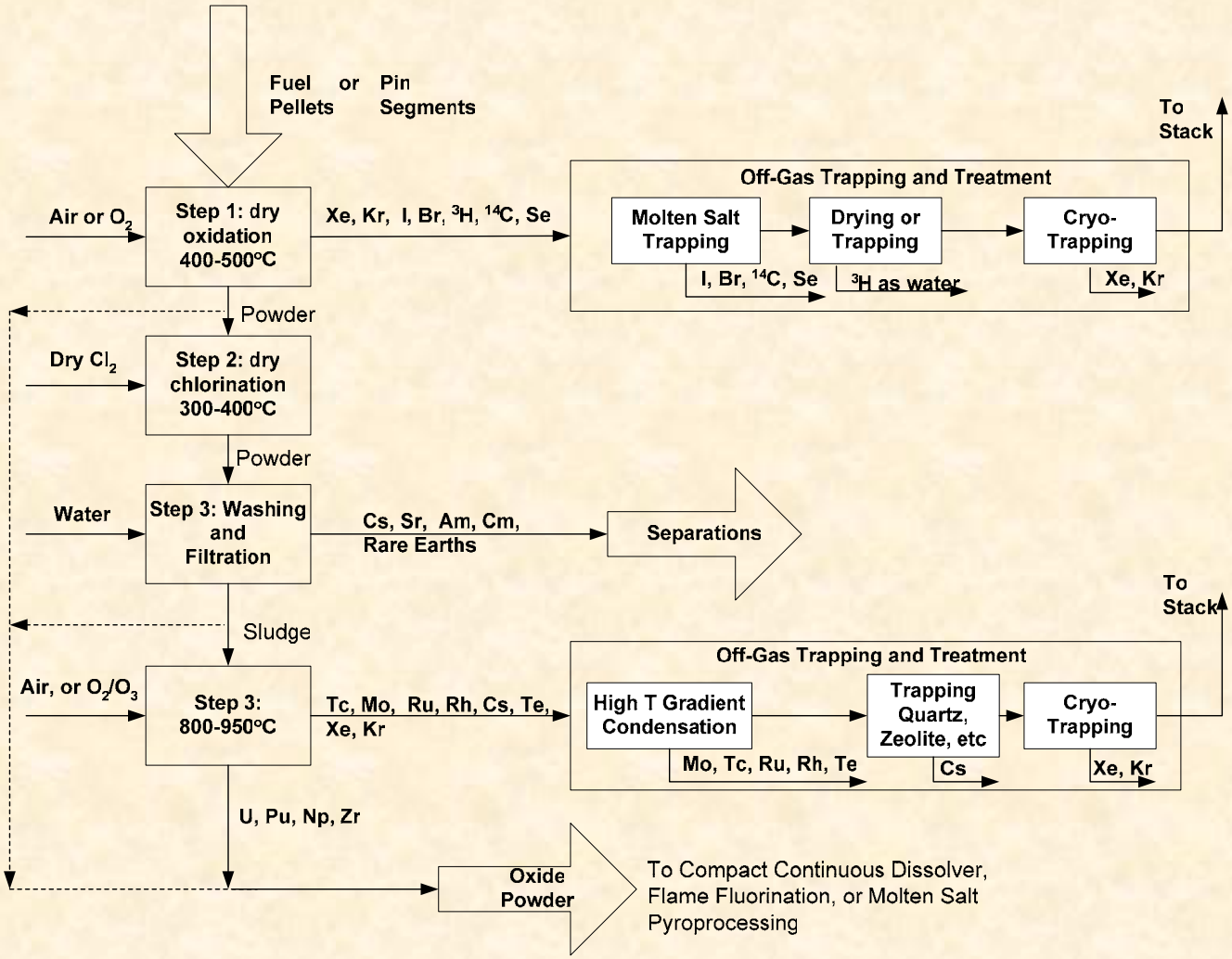
- **The proposed Hybrid Processing includes the up-front pyro-processing recovery and reuse of cladding, hardware, and many volatile and semivolatile species**
  - **The resulting fine oxide powder can be continuously interfaced with aqueous processing or fluoride volatility**



Treatment Alternatives for Hardware and Cladding



Pyrochemical Head-End



Recycle of Uranium

- **Disposal of ruthenium is problematic. Even purified as class C-LLW, there is no clear disposal path**
- **Recycling of uranium for re-enrichment has been practiced in past years in the United States and is currently being done in France (COMURHEX, subsidiary of Cogema) at ~350 MT/y**
- **At least one cycle is possible within standard ASTM specs**
  - **Assuming long fuel cycle and a second cycle late this century, then probably:**
    - **Fast reactors**
    - **New enrichment technologies such as Atomic Vapor Laser Isotope Separation (AVLIS)**
- **Informal contacts with Converdyn (Fluorination) and USEC (Enrichment) indicated recycle is quite feasible under a growing demand scenario**
  - **New independent fluorination line**
  - **An independent enrichment line (centrifuges)**
  - **Private investment will require iron-clad guarantees of steady supply**

Potential Advantages

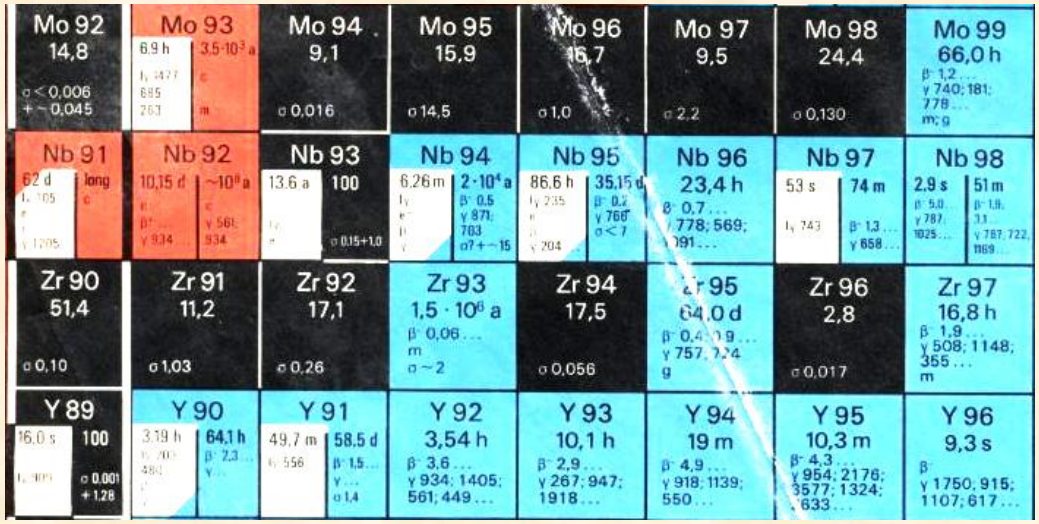
- **Most spent nuclear fuel management scenarios include separation of long-lived fission products, including technetium. If not removed up front, a significant amount of technetium follows the dissolver insolubles, making it very difficult to isolate**
- **Molybdenum and technetium interfere with several downstream separation processes**
- **Unless ^{3}H is separated up front, it is not practical to isolate it during the aqueous processing**
- **Iodine is a source of corrosion and complicates the treatment of dissolver off-gas**
- **Cesium is a major source of radiation and heat. Up-front removal reduces the need for shielding and limits the radiolytic degradation of the solvents downstream. Its early removal facilitates tracking of fissile materials**

Mechanical Decladding

- **Several design alternatives were considered for this front-end process**
- **The selected system provides for mechanical disassembly of the fuel elements and extraction of the fuel rods, followed by removal of the fuel pellets from the individual fuel rods by electro-mechanical means using a laser to split and clean the cladding**

Dry Chlorination Could Be a Practical Way of Treating Cladding from Spent Fuel for Purification and Recovery

- **Dry Cl_2 will not significantly affect the steel components**
  - **i.e., decladding with minimal or no disassembly**
- **Zirconium (Zr) can be purified from most contaminants**
  - **$ZrCl_4$ purification is a conventional step for making Zr and Zircalloy**
  - **Because of its long half-life and very weak emissions, ^{93}Zr is not a significant radiological problem**



Enhanced Voloxidation Option

- **Repeated cycling between UO_2 and U_3O_8 using air at ~500°C and H_2 at ~800°C enhances the release of fission products (e.g., OREOX process) by breaking the particles**
- **Our tests show that black U_3O_8 (prepared by voloxidation of UO_2 at 500°C) readily reacts with ozone to form a red colored UO_3 at temperatures below 200°C that decomposes back to U_3O_8 at temperatures above 300°C**
  - **This fact opens the possibility of a lower-temperature cycle without the potential danger of cycling O_2 and H_2 atmospheres**

XRD:
Monoclinic UO_3

Conclusions

- **A conceptual design by Scientech for a mechanical disassembly and decladding process that includes decontamination appears very promising**
- **In addition, proof-of-principle tests showed that dry chlorination could be used for chemical decladding**
- **"Standard" voloxidation can be used for decladding and has been shown to remove volatile species. A second step, using a combination of oxygen and steam and/or ozone, can remove semivolatile species**
  - **The volatilized species can be directly sorbed and trapped, thus generating a waste form suitable for final disposal (e.g., cesium trapped by an aluminosilicate mineral), condensed and separated for future transmutation (e.g., iodine and technetium); or recovered for alternative uses (e.g., xenon, ruthenium, cladding, and hardware)**
- **Cold surrogate tests have shown that chlorination (Cl_2 or HCl) of the oxide powder generated by voloxidation, followed by water washing, could remove the soluble chlorinated products (minor actinides, rare earths, cesium, and strontium) while leaving behind the insoluble uranium, neptunium, plutonium, and zirconium oxides**
  - **This solid residue will require only minimal treatment, such as the removal of some of the uranium, to allow for the recycling of the material as nuclear fuel. In some instances, the washed residue could be recycled as fuel without further separation**