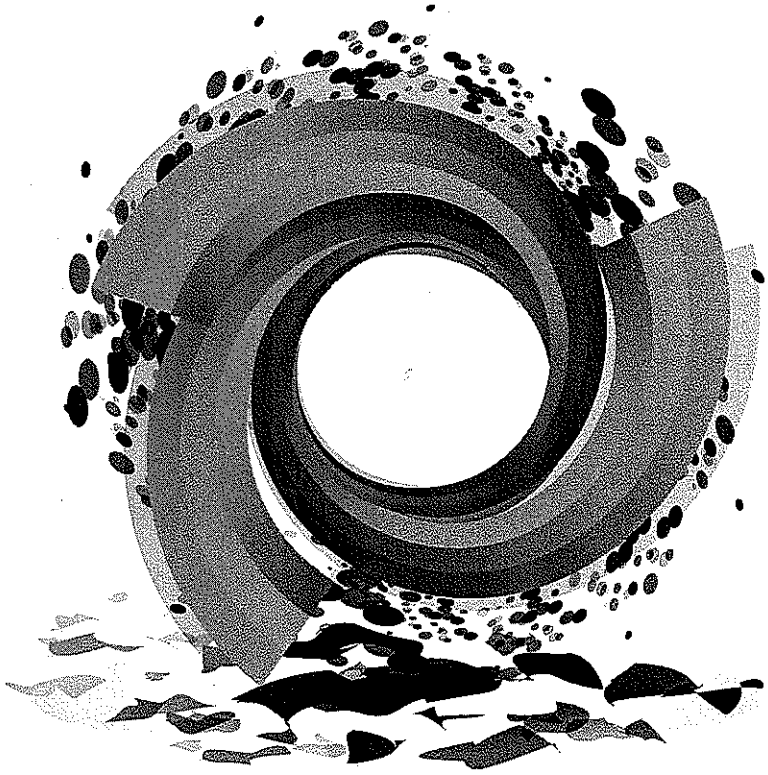
Argonne NATIONAL LABORATORY



Pyroprocessing Technologies

RECYCLING USED NUCLEAR FUEL FOR A SUSTAINABLE ENERGY FUTURE

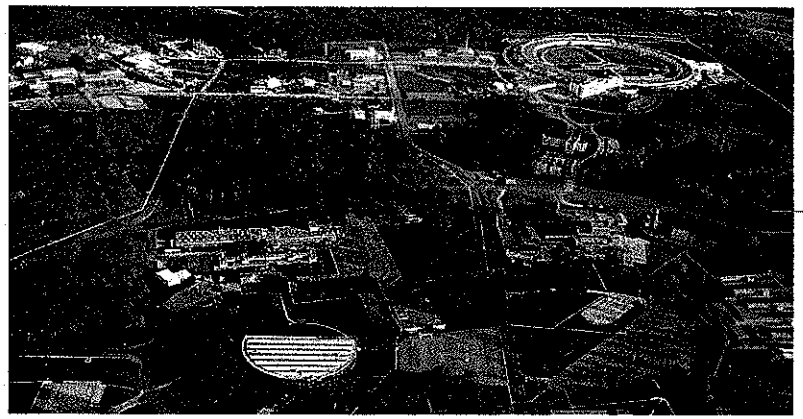
Storing Used Nuclear Fuel is a Real Waste

Nuclear power is the most environmentally friendly way of generating large amounts of electricity on demand. Through the development and application of advanced technologies for recycling "spent," or used nuclear fuel, nuclear power could also become truly sustainable and essentially inexhaustible.

Argonne National Laboratory pioneered the development of pyrochemical processing, or pyroprocessing, a high-temperature method of recycling reactor waste into fuel. When used in conjunction with nuclear fast reactors, pyroprocessing would:

- *Allow 100 times more of the energy in uranium ore to be used to produce electricity compared to current commercial reactors.*
- *Ensure almost inexhaustible supplies of low-cost uranium resources.*
- *Minimize the risk that used fuel would be used for weapons production by recycling the uranium and transuranics to fast reactors for energy production.*
- *Markedly reduce both the amount of waste and the time it must be isolated—from approximately 300,000 to approximately 300 years—by recycling all actinides.*

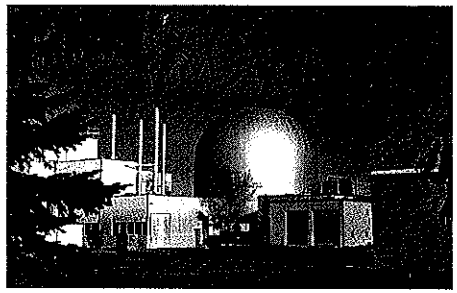
Today, Argonne National Laboratory researchers are developing and refining several pyroprocessing technologies for both light water and fast reactors, working to improve the technologies' commercial viability by increasing their process efficiency and scalability.



Building on its Historic Past, Argonne is a World Leader in Developing Today's Advanced Pyroprocessing Technologies

EXPERIMENTAL BREEDER REACTOR II

Argonne's pioneering development of pyrometallurgical processes led to the design and implementation of the first-ever recycle process for spent metallic reactor fuel from Argonne's Experimental Breeder Reactor II (EBR-II), a fast reactor. Starting in 1964, Argonne researchers treated more than four metric tonnes of used fuel with the pyroprocess to recover and recycle uranium and plutonium to EBR-II. This successful fuel treatment effort led to continued research and development of pyrochemical processes for the recycle of oxide, carbide and other advanced fuels and laid the foundation for Argonne's work today.



INTEGRAL FAST REACTOR PROGRAM

Argonne's development of the Integral Fast Reactor (IFR), a groundbreaking liquid metal-cooled fast reactor designed for energy production and waste consumption, ushered in the use of another pyroprocess for recycling used fuel. Researchers found that an electrochemical process separated the desired actinides from the fission products for recycling into new fuel and addressed the problems associated with increasing requirements for disposal of high-level waste.

The Basics of Pyroprocessing

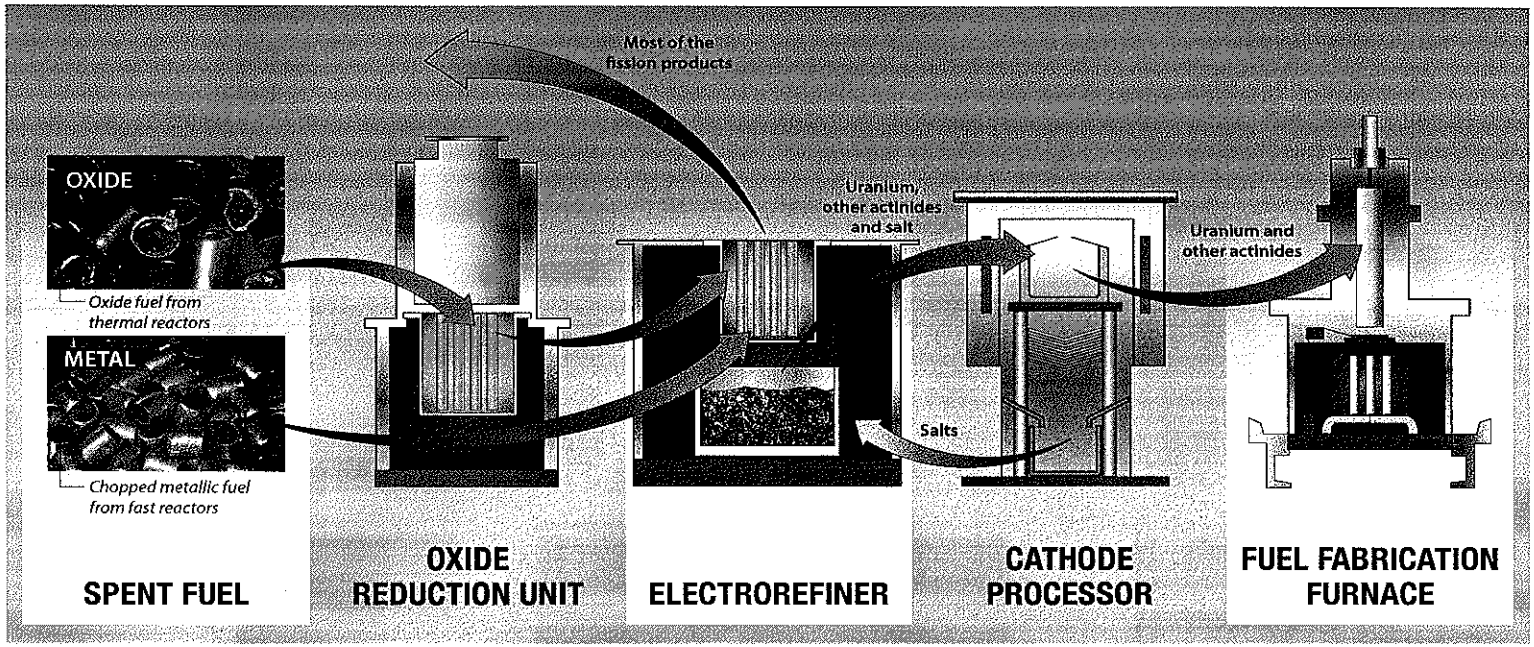
The electrorefining procedure is key to pyrochemical recycling of used nuclear fuel. This process removes the waste fission products from the uranium and other actinides (heavy radioactive elements) in the used fuel. The unfissioned uranium and actinides are then recycled to fast reactors.

Used fuel from today's light water, or thermal, reactors—uranium oxide with a small amount of plutonium and other actinide oxides—is first converted to metal through oxide reduction. While used metallic fuel—uranium, plutonium, and other actinides—from fast reactors goes straight to the refiner.

Electrorefining is very similar to electroplating. Used fuel attached to an anode is suspended in a chemical bath; electric current then dissolves the used fuel and plates out the uranium and other actinides on the cathode. These extracted elements are then sent to the cathode processor where the residual salt from the refining process is removed.

Finally, the remaining actinides and uranium are cast into fresh fuel rods and the salt is recycled back into the electrorefiner.

Through pyroprocessing and the much more efficient fast reactor fuel cycle, vastly more of the energy in the uranium ore can be used to produce electricity.



WHAT IS FISSION (AND OTHER DEFINITIONS)?

Fission: the splitting of the nuclei of heavy-metal atoms (mainly uranium and other actinides) resulting in the release of large amounts of energy.

Fission products: the true waste of fission, a mixture of lighter elements created when the heavy atom splits.

Spent fuel: unfissioned uranium and other actinides (including plutonium and several other heavy radioactive elements). The actinides are the main source of today's nuclear waste problem.

Actinides: heavy radioactive elements, including neptunium, plutonium, americium, and curium, that are separated out during recycling, and remain with the unfissioned uranium as they are recycled back into new fuel. The actinides are then destroyed in fission, turning them into short-lived fission products.

If the reprocessing and refueling steps are repeated enough times, nearly all the actinides will have been fissioned, leaving only short-lived waste fission products containing very little actinides. This process not only reduces the amount of waste created, but also the time it must be isolated—from approximately 300,000 years to approximately 300 years.

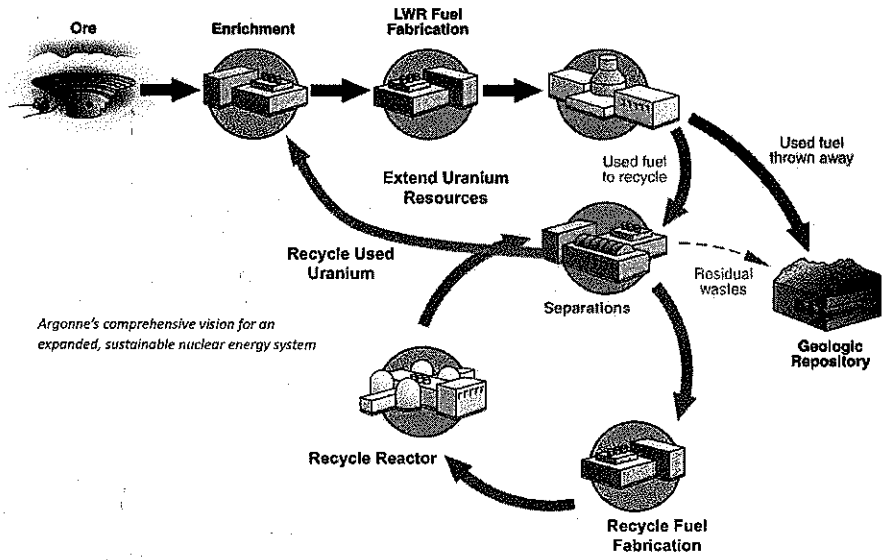
Turning Nuclear Waste into a "Wonderfuel"

Argonne's pyrochemical process research is opening the doors to a sustainable nuclear energy future for the nation. The Laboratory's goals are to:

- *Optimize energy production and use of resources*
- *Manage the fission waste in an environmentally responsible manner*
- *Provide advanced pyroprocessing technologies that are economical to use*

Argonne scientists and engineers are developing commercially viable technologies with the following characteristics:

- *Robust process chemistry and engineering*
- *High product quality*
- *Scalability*
- *Minimal secondary waste production*
- *In line with U.S. non-proliferation objectives*



Argonne's comprehensive vision for an expanded, sustainable nuclear energy system

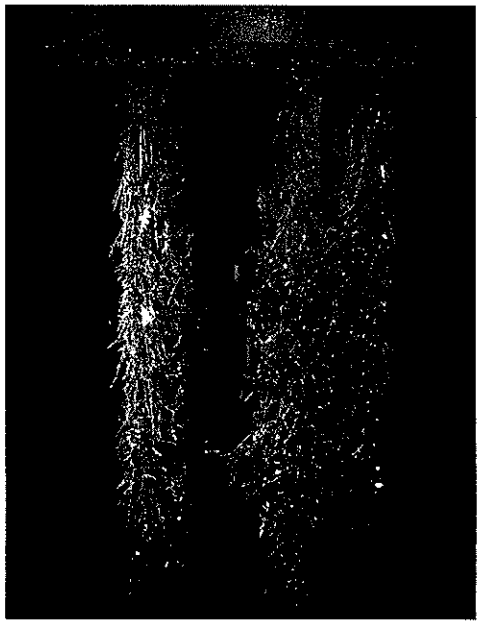
ELECTROREFINING RESEARCH

Electrorefining is the key to pyrochemical processing: an electrometallurgical treatment of spent nuclear fuel that uses molten salt to recover the uranium and other actinides for recycling into new fuel. Electrorefining enables:

- Fission product and actinide partitioning
- Electrodeposition of actinides for recycle
- Fission products to be recovered in a subsequent process and encapsulated in durable waste forms

Argonne's current electrorefining R&D focuses on process efficiency and scalability. Research activities include:

- Increasing throughput (i.e., batch size) to enable the treatment of used light water reactor fuel
- Incorporating automated product recovery to enhance and increase throughput
- Developing intermittent actinide removal from cathodes to enhance process efficiency
- Conceiving and evaluating prototype test electrorefining module designs to establish data essential to the design of commercial systems



Uranium product collected during advanced refiner testing.

Turning Nuclear Waste into a "Wonderfuel"

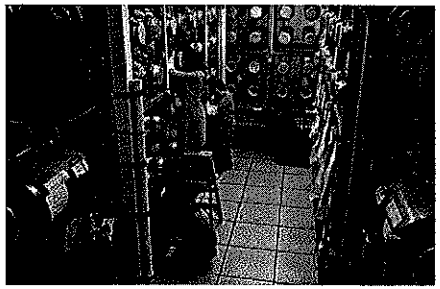
ELECTROREDUCTION TECHNOLOGY

Argonne researchers also pioneered an electrolytic reduction process that has become the standard head-end treatment for converting oxide fuel to base metals for subsequent treatment by electrorefining. This approach enables the treatment of used light water reactor and fast reactor oxide fuel.

Recovering samples from pyroprocessing testing for analysis.

The viability of the electroreduction process chemistry was demonstrated in laboratory-scale and high-capacity testing many years ago. Argonne researchers designed a flexible engineering test-bed for equipment engineering and integration with other unit operations. This includes:

- Modular components with integrated heat shielding
- Multiple cathode slots to test staggered reduction process operation
- An anode and anode shroud that have the flexibility to evaluate alternative materials, geometries, and immersion depths
- A flexible cathode design to evaluate variable bed thicknesses, inter-electrode spacing, and immersion depths
- Multiple fuel baskets compatible with the planar electrorefiner prototype module



Researchers prepare gloveboxes for pyroprocessing tests.

ADVANCED ELECTROCHEMICAL ACTINIDE CO-DEPOSITION

Argonne researchers recently patented an innovative technology for depositing both the uranium and transuranic (other actinides such as neptunium, plutonium, and americium) metal product onto a solid cathode during electrorefining for recycling into fast reactor fuel. During their work, a more detailed understanding of the fundamental electrochemistry behind co-deposition is being gained. The technology:

- Features robust process chemistry that limits the impurities being carried over into the fuel. Fewer impurities in the fuel could increase fuel burn-up in the reactor, resulting in fewer recycle steps
- Reduces the complexity of the process when compared to older technologies
- Maintains the non-proliferation features critical to U.S. fuel cycles

IN SITU PROCESS MONITORING FOR PYROCHEMICAL SYSTEMS

A powerful process monitoring and safeguards technology for the electrorefining systems used in actinide recovery is being developed by Argonne's research team, which includes experts in process research and nuclear safeguards. Reliable process monitoring and control technologies are essential for operating a commercial fuel treatment facility.

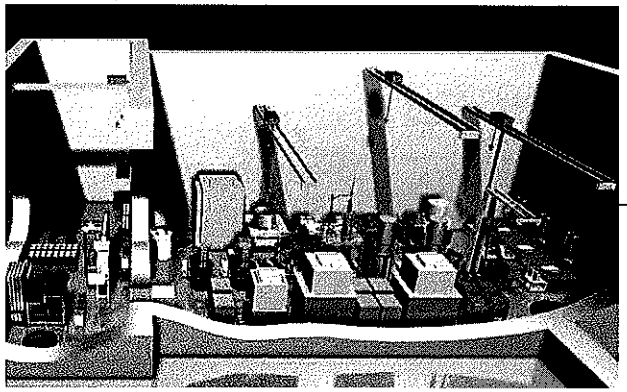
A variety of electroanalytical methods including cyclic and square-wave voltammetry, and spectroscopic techniques are being developed and evaluated to determine the quantity of actinide in molten salt. This research includes developing:

- Methods that achieve representative and reproducible conditions at the sensing electrode/molten salt solution interface
- Methods to determine the sensing electrode's effective area, which is vital to accurate concentration measurements

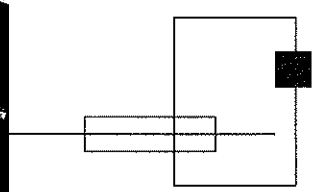
Scientists review data from in situ process monitoring for pyrochemical systems.

PYROCHEMICAL PROCESSING FACILITY CONCEPT

To further advance Argonne's pyroprocessing work and the potential for recycling used nuclear fuel, researchers developed a conceptual 100 metric tonne per year pyroprocessing facility. This work includes the development of processes, equipment concepts, an operations model, and the identification of materials handling issues.



Conceptual pyroprocessing facility



Key Argonne Breakthroughs in Pyroprocessing Technology

URANIUM ELECTROREFINING – Eliminated the need for adding chemicals during the fuel treatment process. Instead, uranium and other actinides are separated from the fission products by electrochemical methods. This approach eliminates the need for extensive solvent recycle and simplifies the design of process facilities.

PURIFIED URANIUM METAL – The first deposit of purified uranium metal greater than a kilogram in weight was produced during the early electrorefiner development.

MARK IV ELECTROREFINER – The first pilot-scale uranium electrorefiner successfully recovered uranium from the spent driver fuel of Experimental Breeder Reactor-II (EBR-II).

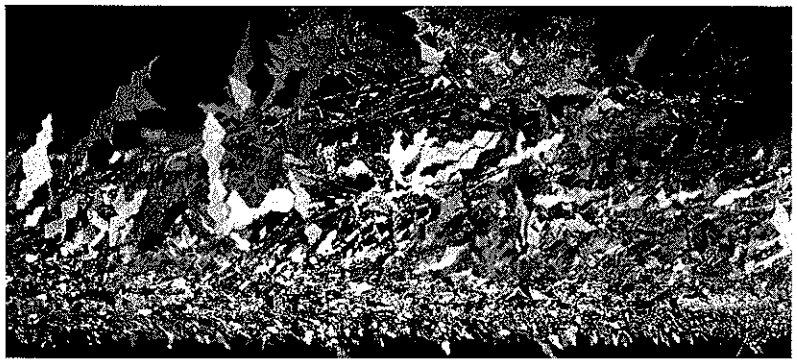
MARK V ELECTROREFINER – The first high-throughput system (see image at right) successfully demonstrated the treatment of EBR-II used fuel.

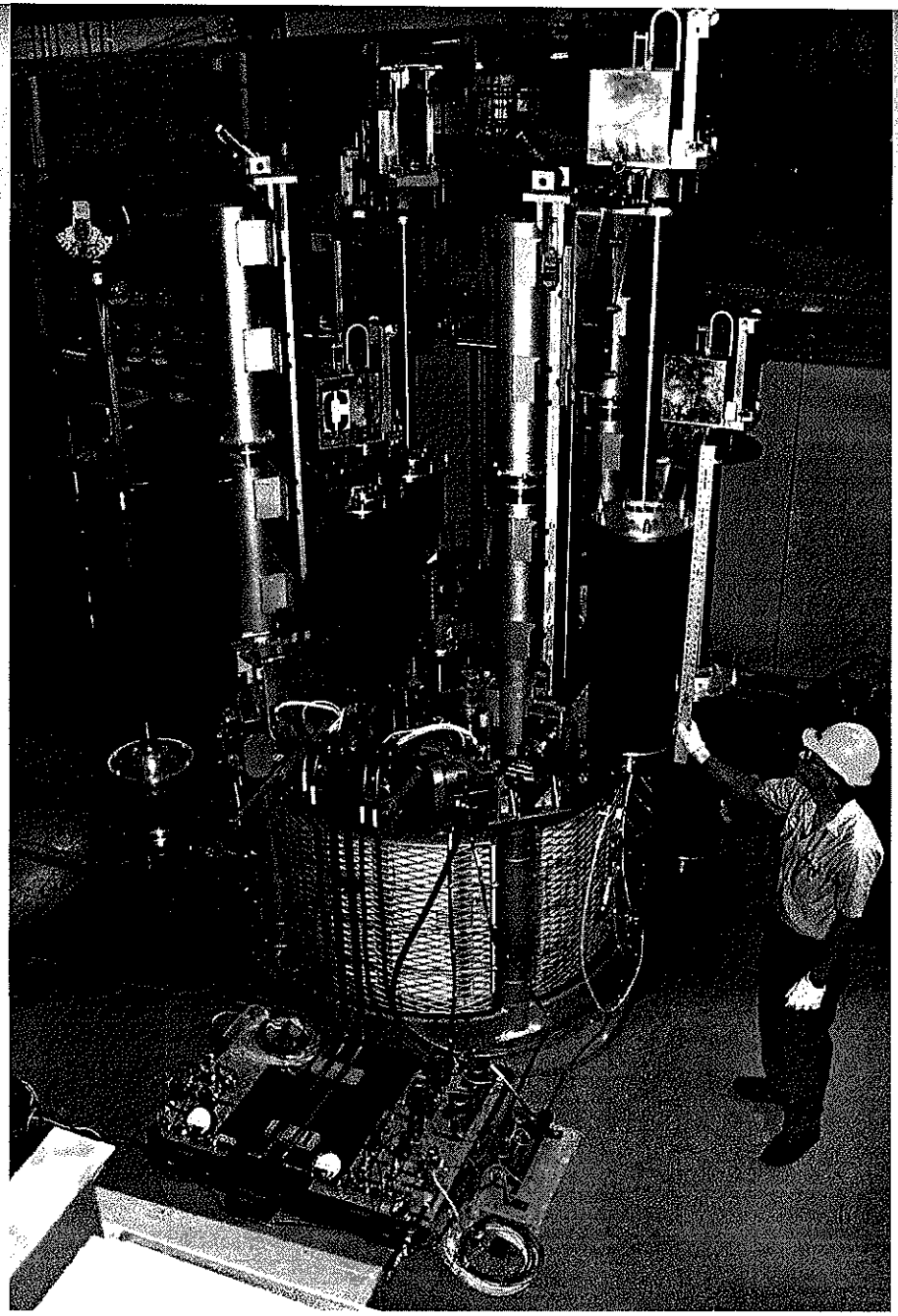
ADVANCED PROTOTYPE ELECTROREFINER – The highly scalable design is essential for treating used fuel from light water reactors because of its substantial—almost 95 percent—uranium content.

ELECTROCHEMICAL REDUCTION OF OXIDE FUEL – Enables the closure of the nuclear fuel cycle by recovering uranium and other actinides from used light water reactor fuel for use in fast reactor fuel.

ACTINIDE CO-DEPOSITION – Enables recovery of uranium and other actinides from the refiner; this limits the impurities being carried over into the fuel and meets U.S. non-proliferation objectives.

Uranium dendrites collected during the evaluation of Argonne's advanced electrorefining technology.





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See the video on Argonne's pyroprocessing research:

http://youtu.be/MlMDDhQ9-pE

Without recycling, most of the energy that could be extracted from the original uranium ore will be "tossed away," and stored as waste.

Argonne's groundbreaking pyroprocessing technologies enables 100 times more of the energy in uranium ore to be used, and by recycling all actinides it significantly reduces the amount of nuclear waste and the time it must be isolated.

U.S. DEPARTMENT OF ENERGY