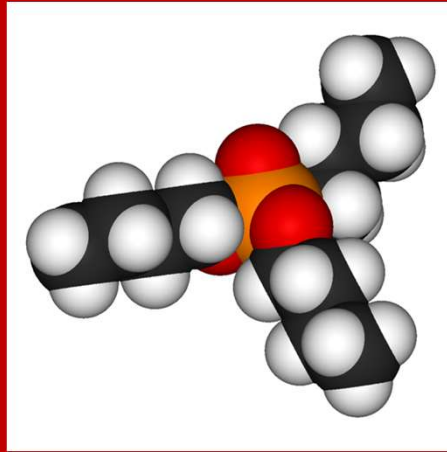


# Chemical Reprocessing of Used Nuclear Fuel



**“At first I saw absolutely nothing. When I was able to open my eyes, I stood still, far more stupefied than astonished.”**

**—Henry Lawson in “Journey to the Center of the Earth”  
by Jules Verne.**

# Used Nuclear Fuel Reprocessing (UNF)

An alternative to the “Once Through” Cycle. Extract U and Pu from UNF, and leave the fission products. Separate the Pu from the U.

Blend the extracted Pu with natural, enriched reprocessed or depleted U to make mixed oxide (MOX) fuel.  $\text{PuO}_2$  can vary from 2 to 30% by weight.

Not new idea; was expected in the 1970s.

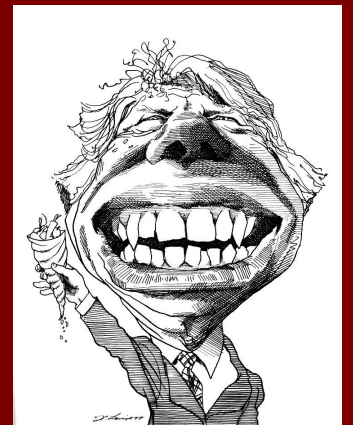
# UNF Reprocessing

Banned by President Carter in 1977 because of Pu accessibility concerns.

The ban was lifted by President Reagan in 1981.

Commercial reprocessing of UNF did not materialize long in the U.S.

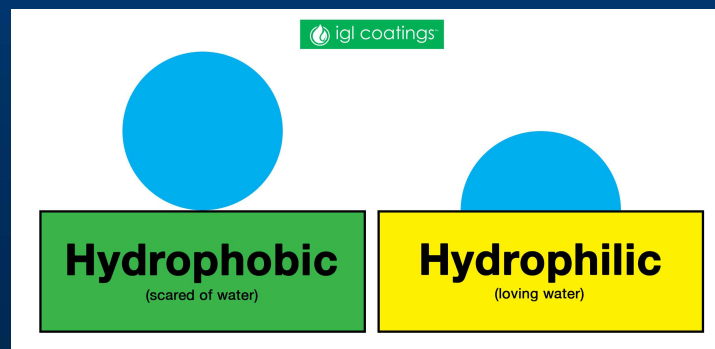
Large-scale reprocessing of UNF developed in the UK, France, Germany, Russia, and Japan.



# Relevant Terms

*Extraction.* The transfer of solute(s) from one liquid phase to another.

Typically the transfer of solutes from an aqueous, hydrophilic (“water loving”) phase to a hydrophobic (“water fearing”) phase composed of an organic solvent.



# Relevant Terms

*Scrubbing.* The purification of an intermediate product such as the removal of fission products from the solvent that were co-extracted with uranium and plutonium.

*Stripping.* The back extraction; the transfer of solute(s) from the hydrophobic phase back into a hydrophilic aqueous phase for additional separations and recovery.

# Early UNF reprocessing

The used fuel was first cooled for only about 40 days. Then the aluminum cladding was dissolved in a hot sodium hydroxide-sodium nitrate solution. The fuel was then dissolved in a 60% nitric acid solution. The plutonium that was dissolved was reduced to  $\text{Pu}^{4+}$  by the addition of sodium nitrite. The nitrite oxidized to nitrate, and the resulting electrons reduced the plutonium.

# Early UNF reprocessing

Then bismuth nitrate and phosphoric acid were added to co-precipitate the plutonium:



The precipitate was collected by centrifugation, then dissolved in nitric acid. The final product was refined to remove impurities such as fission products by numerous dissolution-precipitation steps.

# Modern UNF Reprocessing

The Plutonium Uranium Recovery (also called Redox or Refining) Extraction (Purex) method was invented in about 1954, and has become “the universal choice.”

**Basics of the Purex Process:** It is a liquid-liquid extraction.

UNF is physically broken down and dissolved in concentrated nitric acid yielding a dissolution liquor that contains U, Pu, and all the fission products.

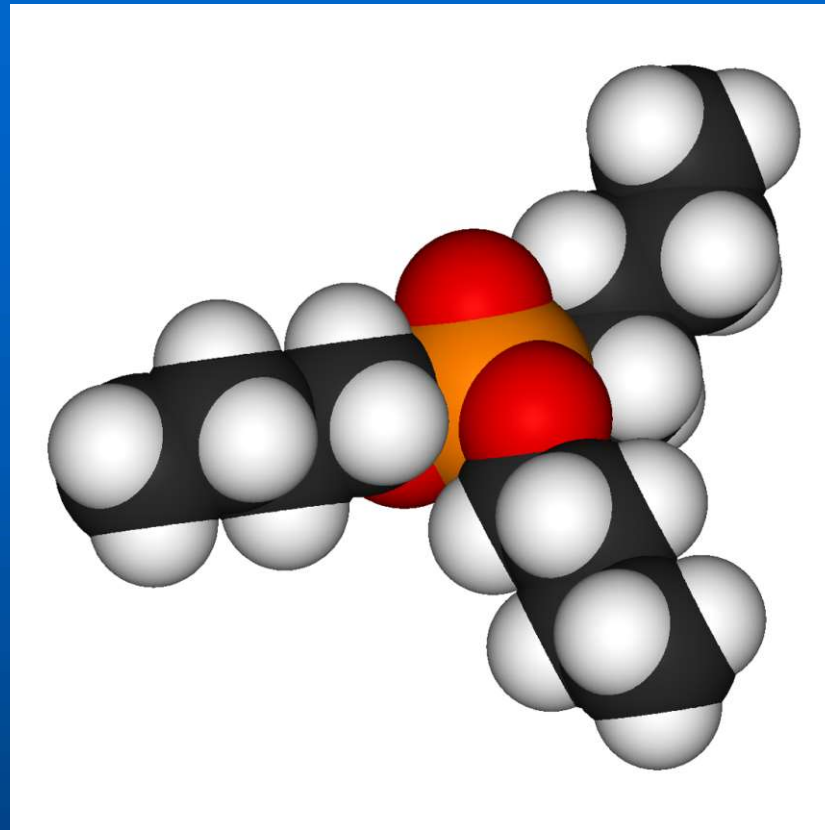


# The Purex Reaction

The acidic ( $\sim 3\text{ N HNO}_3$ ) solution is reacted with a solution containing 30% tributyl phosphate (TBP) dissolved in kerosene or dodecane.

TBP, is an organophosphorus compound with the formula  $\text{CH}_3\text{CH}_2\text{CH}_2\text{CH}_2\text{O})_3\text{PO}$ .

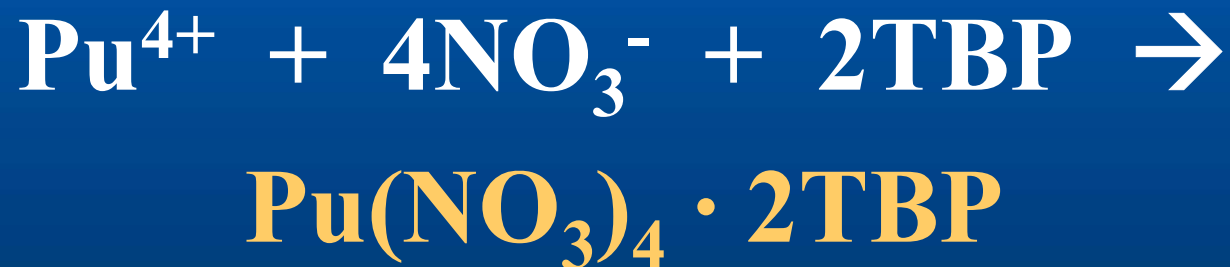
**TBP**



**We are mixing a hydrophilic solution (“water loving”) with a hydrophobic (“water fearing”) solution.**

# The Purex Reaction

$\text{U}^{4+}$  and  $\text{Pu}^{4+}$ , which are hydrophilic, react with TBP in the presence of nitrate to form organic-soluble complexes which are more hydrophobic.



Fission products such as  $\text{Cs}^+$  do not react with TBP as strongly and remain in solution.

# Purex Reaction

Liquor →

30% TBP in Kerosene  
Hydrophobic liquid

Extract →  
U, Pu, some  
Th, Np

← Raffinate  
Fission  
products  
(a new  
Waste)

3 N  $\text{HNO}_3$   
Hydrophilic liquid

# Purex Reaction

Pu (IV) is then chemically reduced to Pu (III) by adding hydroxylamine ( $\text{NH}_2\text{OH}$ ), and then extracted with 6-M nitric acid.

$\text{UO}_2^{2+}$  remains in the organic phase.

Multiple extraction cycles are required. Extracts can be further refined by the removal of Np and Th.

# **Commercial UNF reprocessing in the US**

**West Valley Plant. New York.**

**Operated by Nuclear Fuels Services  
(NFS) from 1966 to 1972.**

**Used a three-cycle Purex process.**

**U and Pu-nitrate solutions were the  
end product.**

# West Valley Plant. New York.



# **West Valley Plant**

**Nuclear Fuels Services operated the site as a nuclear fuel reprocessing center from 1966 to 1972, and accepted radioactive waste for disposal until 1975.**

**During the operation of the plant, 640 tonnes of used reactor fuel were processed, resulting in 660,000 gallons (2,500 kL) of radioactive liquid waste.**

**The liquid waste is stored in underground waste tanks.**



# West Valley Plant

NFS also used a 15-acre (6 ha) area for the disposal of radioactive waste from commercial waste generators, and another 7-acre (2.8 ha) landfill was used to dispose of radioactive waste generated from reprocessing.

In 1976, NFS decided the costs and regulatory requirements of reprocessing made the venture impractical. Left lease to State of New York in 1980.

# Contaminated soil and groundwater issues remain.



# **Commercial Reprocessing (almost)**

**The Nuclear Fuel Services facility was the first and only private plant in the United States to reprocess UNF.**

**General Electric completed its Midwest Fuel Recovery Plant at Morris, Illinois, in 1974, but immediately declared it “inoperable” and never used it.**

# **Commercial Reprocessing (almost)**

**In 1970, Allied General Nuclear Services began constructing a 1,500 ton/year (1,360 tonnes/a) reprocessing plant at Barnwell, South Carolina. Because of the 1977 ban, the plant was not completed.**

# **Commercial Reprocessing (almost)**

**In the future, we may reconsider reprocessing of UNF, but what about the wastes such as waste acids containing fission and TRU products? These are the High-Level Wastes.**

# Recycling used nuclear fuel - Orano la Hague - English

<https://www.youtube.com/watch?v=V0UJSIKIy8g> (9:57)

# Questions?

