Physical Chemistry for Energy Engineering
(24th: 2018/12/10)

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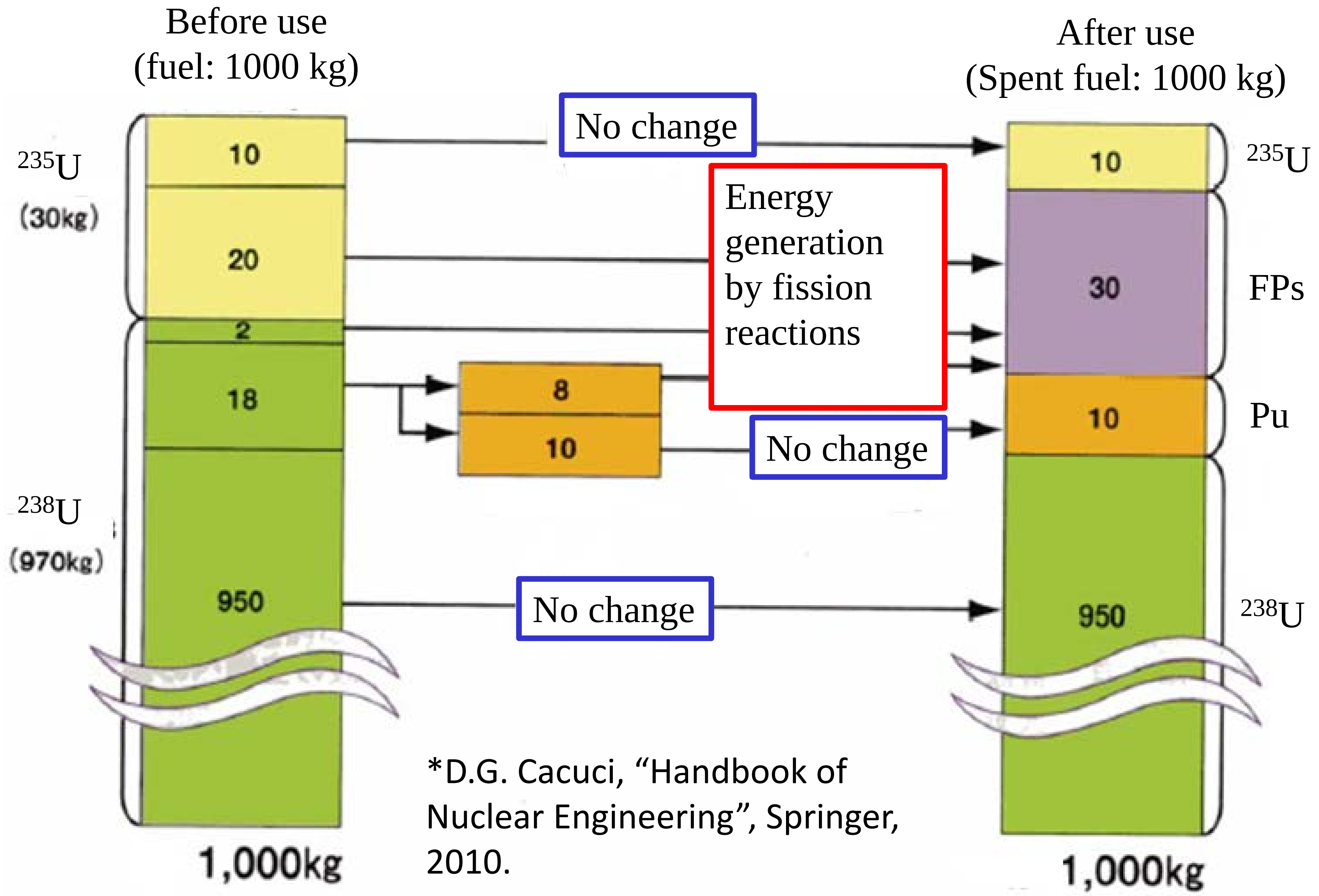
*The class follows the text book: D.A. McQuarrie, J.D. Simon, “Physical Chemistry: A Molecular Approach", University Science Books (1997).

5. Chemistry in reprocessing of spent nuclear fuels
-composition of fuels before and after the use-



Before use
(fuel: 1000 kg)

After use
(Spent fuel: 1000 kg)

^{235}U

^{238}U

No change

Energy generation by fission reactions

No change

No change

^{235}U

FPs

Pu

^{238}U

*D.G. Cacuci, “Handbook of Nuclear Engineering”, Springer, 2010.

5. Chemistry in reprocessing of spent nuclear fuels
-composition of fuels before and after the use-

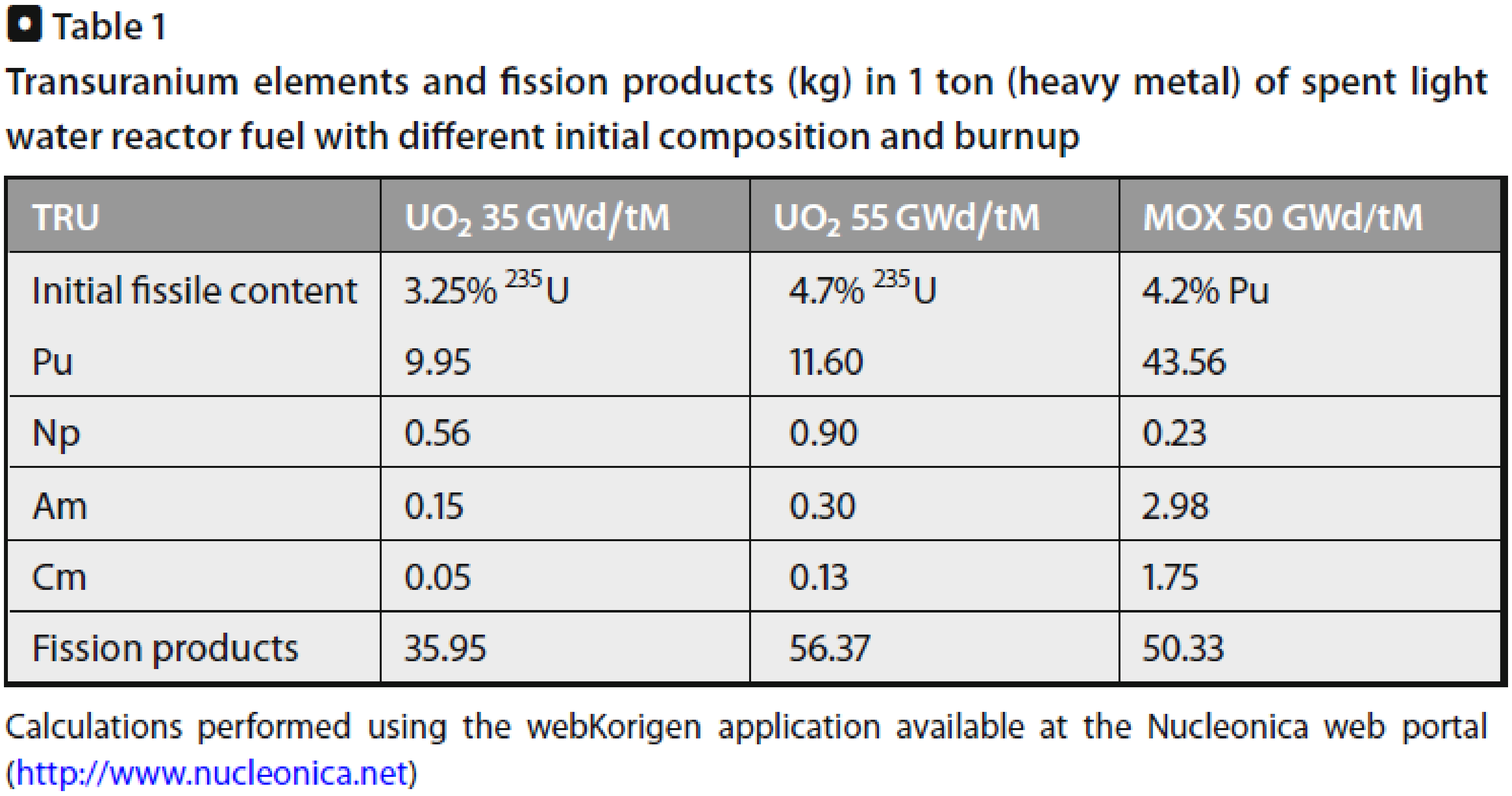


Table 1

Transuranium elements and fission products (kg) in 1 ton (heavy metal) of spent light water reactor fuel with different initial composition and burnup

TRU	UO_2 35 GWd/tM	UO_2 55 GWd/tM	MOX 50 GWd/tM
Initial fissile content	3.25% ^{235}U	4.7% ^{235}U	4.2% Pu
Pu	9.95	11.60	43.56
Np	0.56	0.90	0.23
Am	0.15	0.30	2.98
Cm	0.05	0.13	1.75
Fission products	35.95	56.37	50.33

Calculations performed using the webKorigen application available at the Nucleonica web portal (http://www.nucleonica.net)

Minor actinides

*D.G. Cacuci, “Handbook of Nuclear Engineering”, Springer, 2010.

5. Chemistry in reprocessing of spent nuclear fuels
-composition of fuels before and after the use-

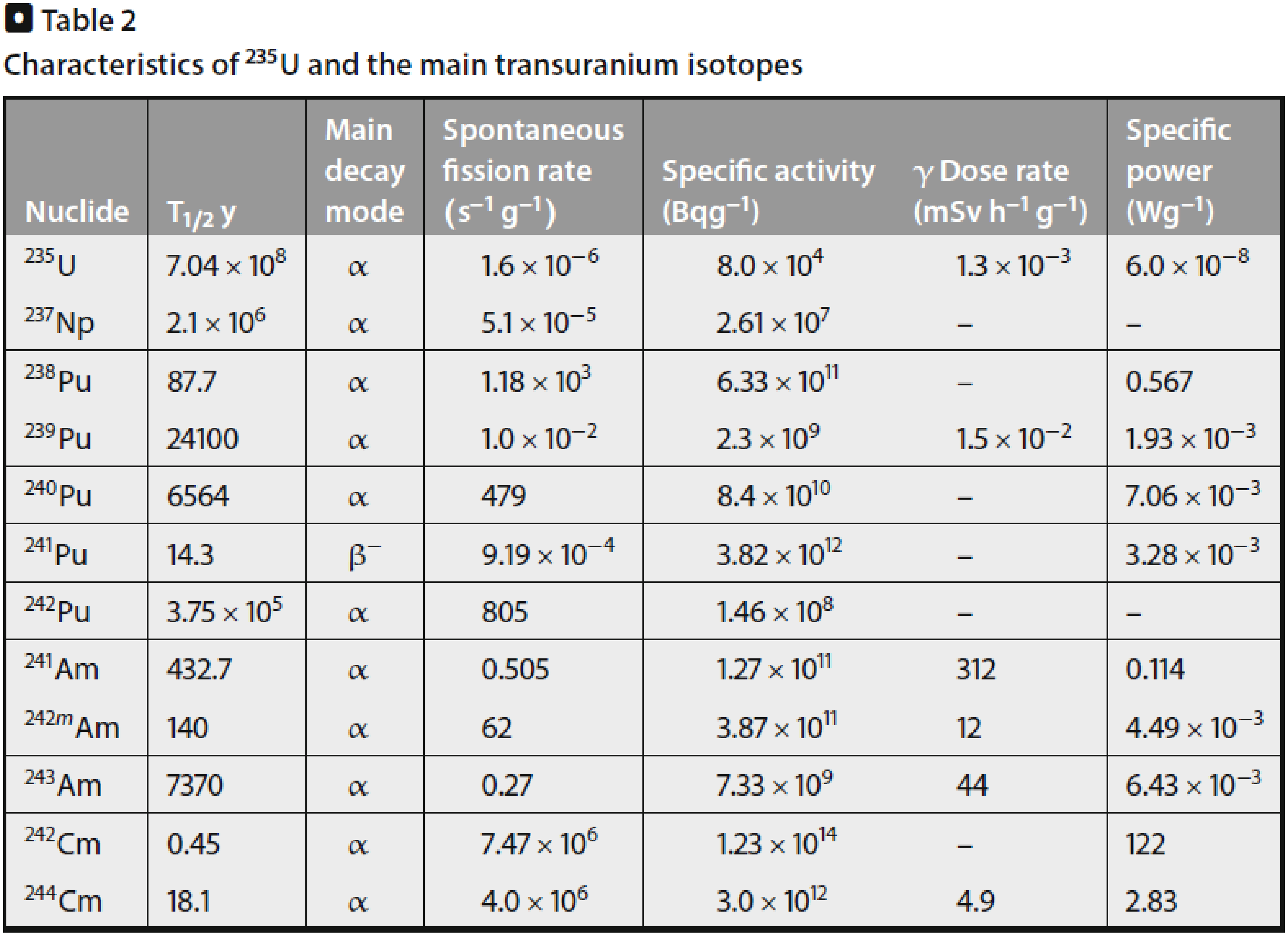


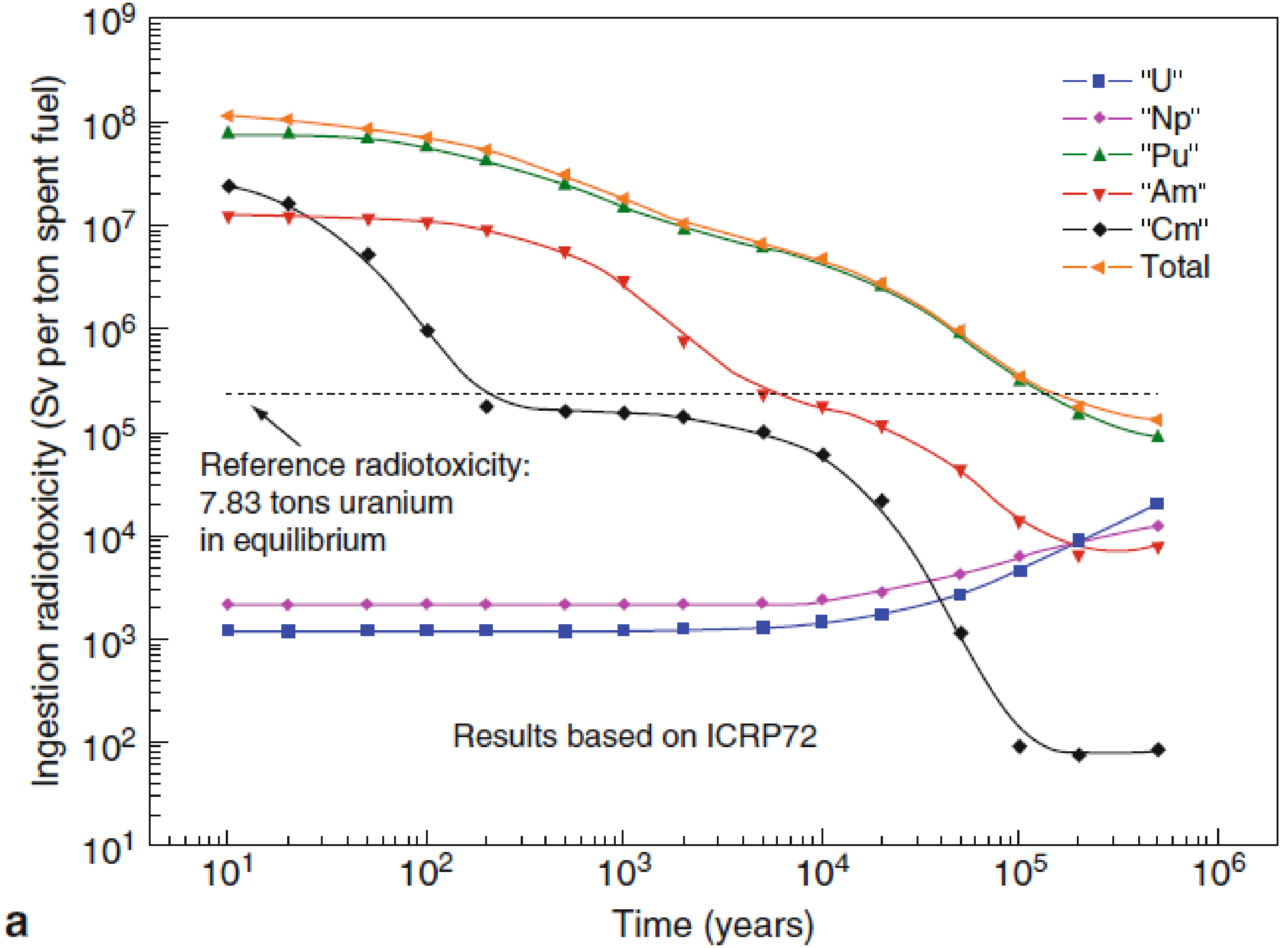
Table 2

Characteristics of ^{235}U and the main transuranium isotopes

Nuclide	$T_{1/2}$ y	Main decay mode	Spontaneous fission rate ($s^{-1}\,g^{-1}$)	Specific activity ($Bq g^{-1}$)	γ Dose rate ($mSv\,h^{-1}\,g^{-1}$)	Specific power ($W g^{-1}$)
^{235}U	7.04×10^{8}	α	1.6×10^{-6}	8.0×10^{4}	1.3×10^{-3}	6.0×10^{-8}
^{237}Np	2.1×10^{6}	α	5.1×10^{-5}	2.61×10^{7}	–	–
^{238}Pu	87.7	α	1.18×10^{3}	6.33×10^{11}	–	0.567
^{239}Pu	24100	α	1.0×10^{-2}	2.3×10^{9}	1.5×10^{-2}	1.93×10^{-3}
^{240}Pu	6564	α	479	8.4×10^{10}	–	7.06×10^{-3}
^{241}Pu	14.3	β^-	9.19×10^{-4}	3.82×10^{12}	–	3.28×10^{-3}
^{242}Pu	3.75×10^{5}	α	805	1.46×10^{8}	–	–
^{241}Am	432.7	α	0.505	1.27×10^{11}	312	0.114
^{242m}Am	140	α	62	3.87×10^{11}	12	4.49×10^{-3}
^{243}Am	7370	α	0.27	7.33×10^{9}	44	6.43×10^{-3}
^{242}Cm	0.45	α	7.47×10^{6}	1.23×10^{14}	–	122
^{244}Cm	18.1	α	4.0×10^{6}	3.0×10^{12}	4.9	2.83

*D.G. Cacuci, “Handbook of Nuclear Engineering”, Springer, 2010.

5. Chemistry in reprocessing of spent nuclear fuels
-why reprocessing is attractive?-



(a) The radiotoxic inventory of the U, Pu, and MA in spent nuclear fuel as a function of storage time.

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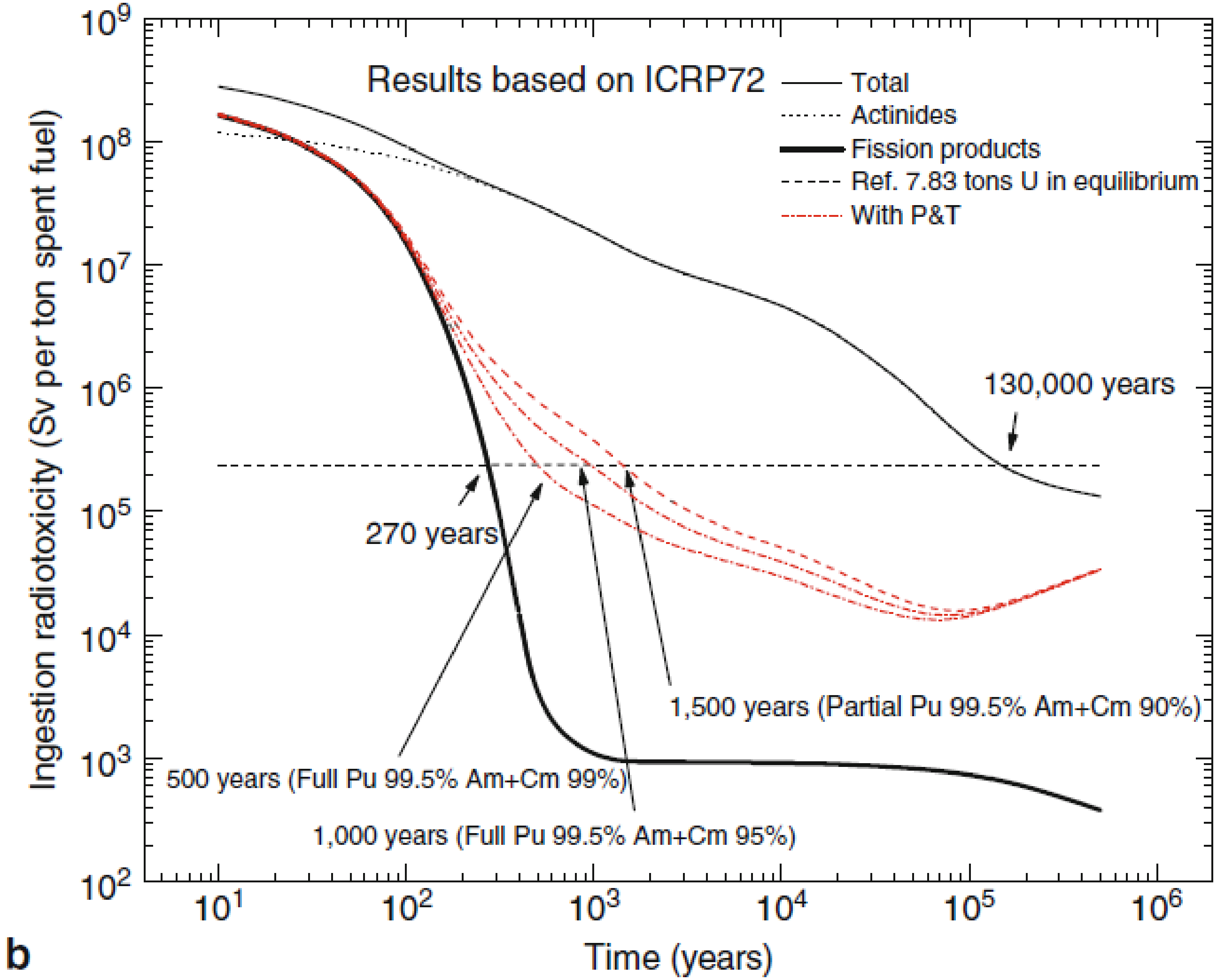
5. Chemistry in reprocessing of spent nuclear fuels
-why reprocessing is attractive?-



(b) The radiotoxic inventory of spent fuel as a function of storage time separated into the contributions of the actinides and the fission products

*D.G. Cacuci, “Handbook of Nuclear Engineering”, Springer, 2010.

5. Chemistry in reprocessing of spent nuclear fuels
-Partitioning/reprocessing goals -

- ✓ The main radionuclides targeted for separation for partitioning-transmutation (P&T) and partitioning-conditioning (P&C) are the actinides neptunium, americium and curium (Np, Am, Cm; along with U & Pu), and the fission products iodine-129, technetium-99, caesium-135 and strontium-90.
  - ✓ Removal of the latter two significantly reduces the heat load of residual conditioned wastes. Of course. any chemical process will not separate different isotopes of any particular element.

- ✓ Conceptually reprocessing can take several courses, separating certain elements from the remainder, which becomes high-level waste. Reprocessing options include:
  - ✓ Separate U, Pu, (as today; PUREX, Plutonium Uranium Redox EXtraction).
  - ✓ Separate U, Pu+U (small amount of U).
  - ✓ Separate U, Pu, minor actinides[f].
  - ✓ Separate U, Pu+Np, Am+Cm.
  - ✓ Separate U+Pu all together.
  - ✓ Separate U, Pu+actinides, certain fission products.

*D.G. Cacuci, “Handbook of Nuclear Engineering”, Springer, 2010.

5. Chemistry in reprocessing of spent nuclear fuels
-Preparation needed for PUREX process-

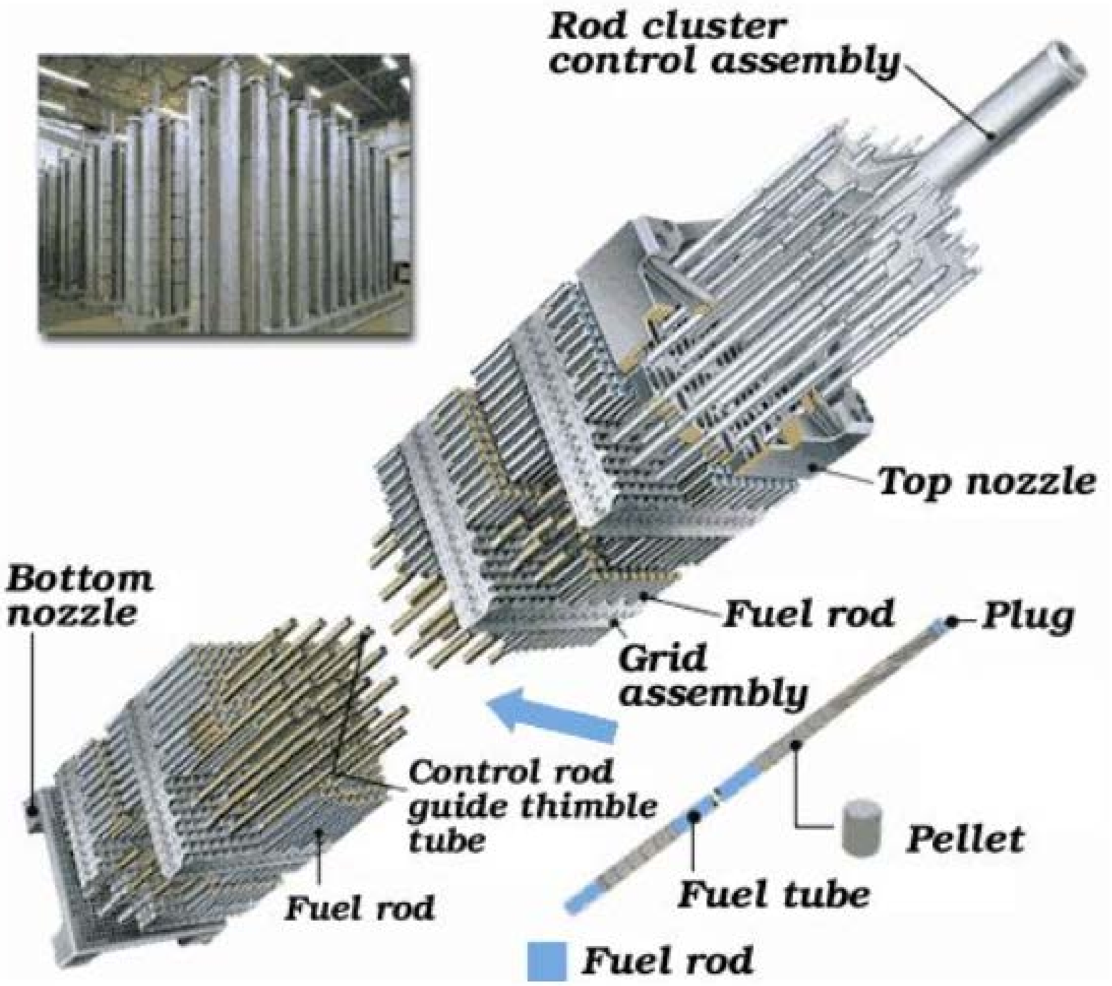


Figure 4: A PWR fuel assembly

*http://www.world-nuclear.org/info/Nuclear-Fuel-Cycle/Conversion-Enrichment-and-Fabrication/Fuel-Fabrication/

5. Chemistry in reprocessing of spent nuclear fuels
-PUREX: Fuel Dissolution-

✓ Preparation for dissolution

- Cladding is opened to permit subsequent dissolution of oxide by mechanical shearing or sawing for steel or zircaloy.
- Off-gases from decladding contain up to 10% of Kr and Xe in the fuel and some of the $^{14}CO_2$, ^{3}H, and other volatile fission products.

✓

✓ Fuel dissolution:

- Fuel and cladding are reacted with hot nitric acid to convert U, Pu, and fission products into chemical states most favorable for their subsequent separation (+6 for U and +4 for Pu).
- The acidity of dissolver solution is adjusted to pH of around 2.5 and Pu is brought into its most extractable valence of 4, by controlled addition of N_2O_4.

$3UO_2 + 8HNO_3 \rightarrow 3UO_2(NO_3)_2 + 2NO + 4H_2O$ (dominant at acid concentration <10M)

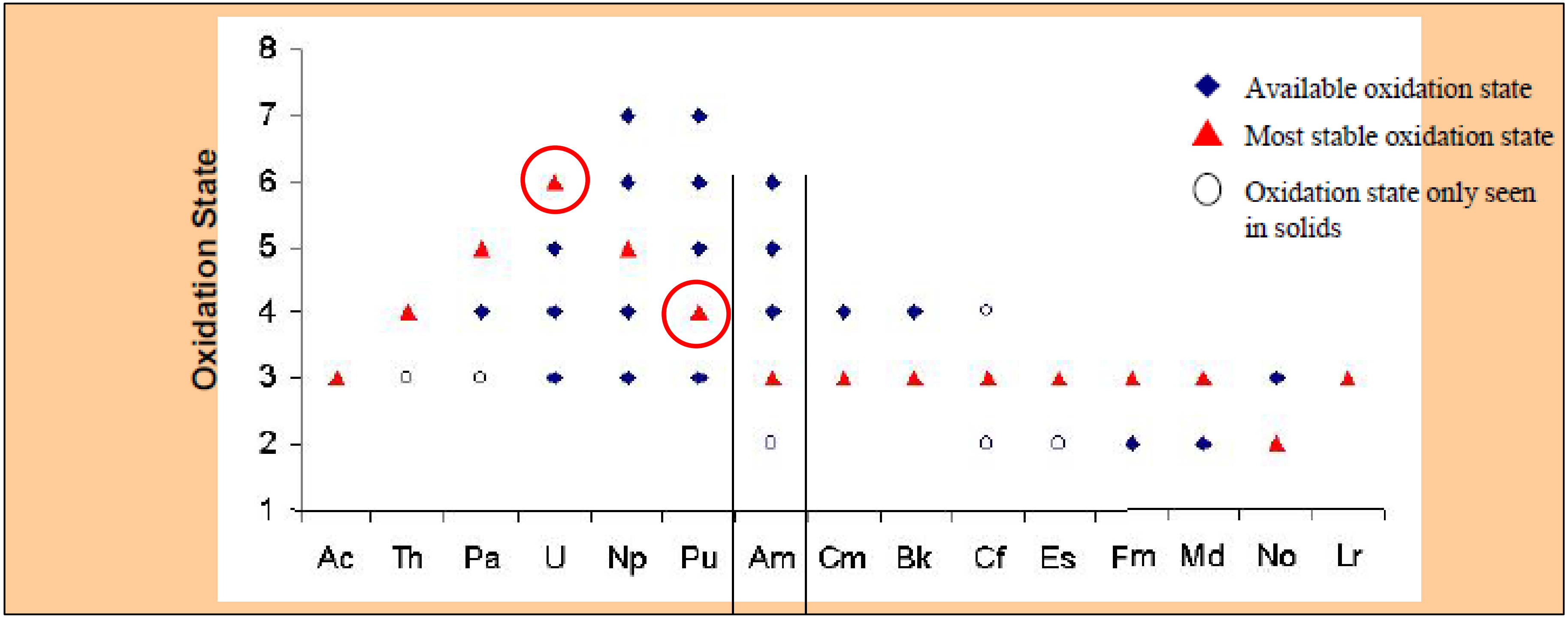
$UO_2 + 4HNO_3 \rightarrow UO_2(NO_3)_2 + 2NO_2 + 2H_2O$ (dominant at higher concentration)

$Pu^{(VI)}O_2^{2+} + N_2O_4 + 2H^+ \rightarrow Pu^{4+} + 2HNO_3$

$4Pu^{3+} + N_2O_4 + 4H^+ \rightarrow 4Pu^{4+} + 2NO + 2H_2O$

*https://notes.engineeringonline.ncsu.edu/courses/ne/NE531/NE531_s09_08.pdf

5. Chemistry in reprocessing of spent nuclear fuels
-PUREX: Fuel Dissolution-



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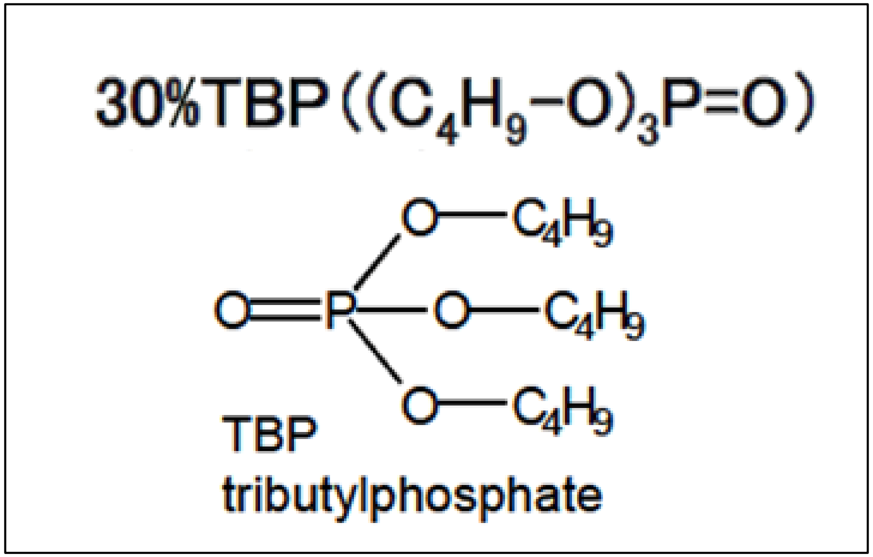
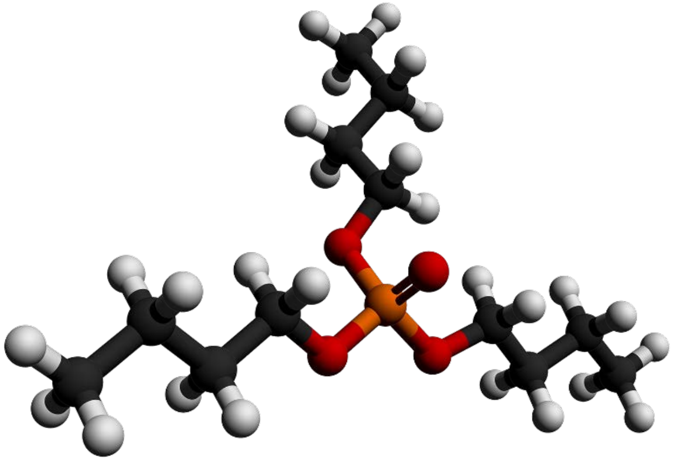
5. Chemistry in reprocessing of spent nuclear fuels
-PUREX: extraction of Pu and U with TBP-

- An organic solvent composed of 30% tributyl phosphate (TBP) in a hydrocarbon solvent (e.g. $C_{12}H_{26}$) is used to extract the uranium as $UO_2(NO_3)_2 \cdot 2TBP$ complexes, and plutonium as $Pu(NO_3)_4 \cdot 2TBP$, from other fission products, which remain in the aqueous phase.
  - The transuranium elements americium and curium also remain in the aqueous phase.
- TBP is also radiation resistant which is an important consideration in the processing of spent fuel or HLW.


30%TBP $((C_4H_9-O)_3P=O)$

TBP
tributylphosphate

+ 70% $C_{12}H_{26}$



*http://www.wikiwand.com/en/Tributyl_phosphate

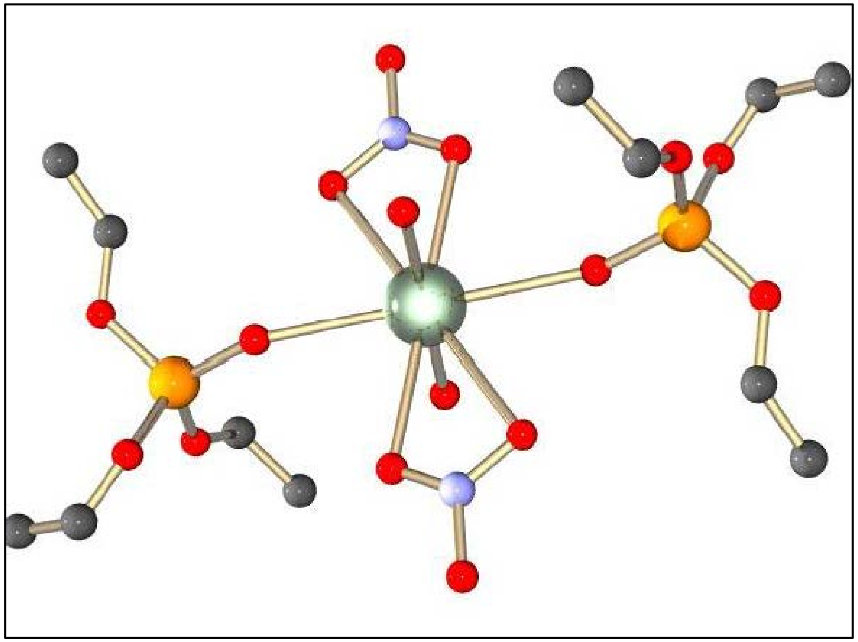
5. Chemistry in reprocessing of spent nuclear fuels
-PUREX: extraction of Pu and U with TBP-

The nature of the organic soluble uranium complex has been the subject of some research. A series of complexes of uranium with nitrate and trialkyl phosphates and phosphine oxides have been characterized.

$$HNO_3\ (aq) + TBP\ (org) \Leftrightarrow HNO_3 \cdot TBP\ (org)$$

$$UO_2(NO_3)_2(aq) + 2TBP\ (org) \Leftrightarrow UO_2(NO_3)_2 \cdot 2TBP\ (org)$$

$$Pu(NO_3)_4\ (aq) + 2TBP\ (org) \Leftrightarrow Pu(NO_3)_4 \cdot 2TBP\ (org)$$



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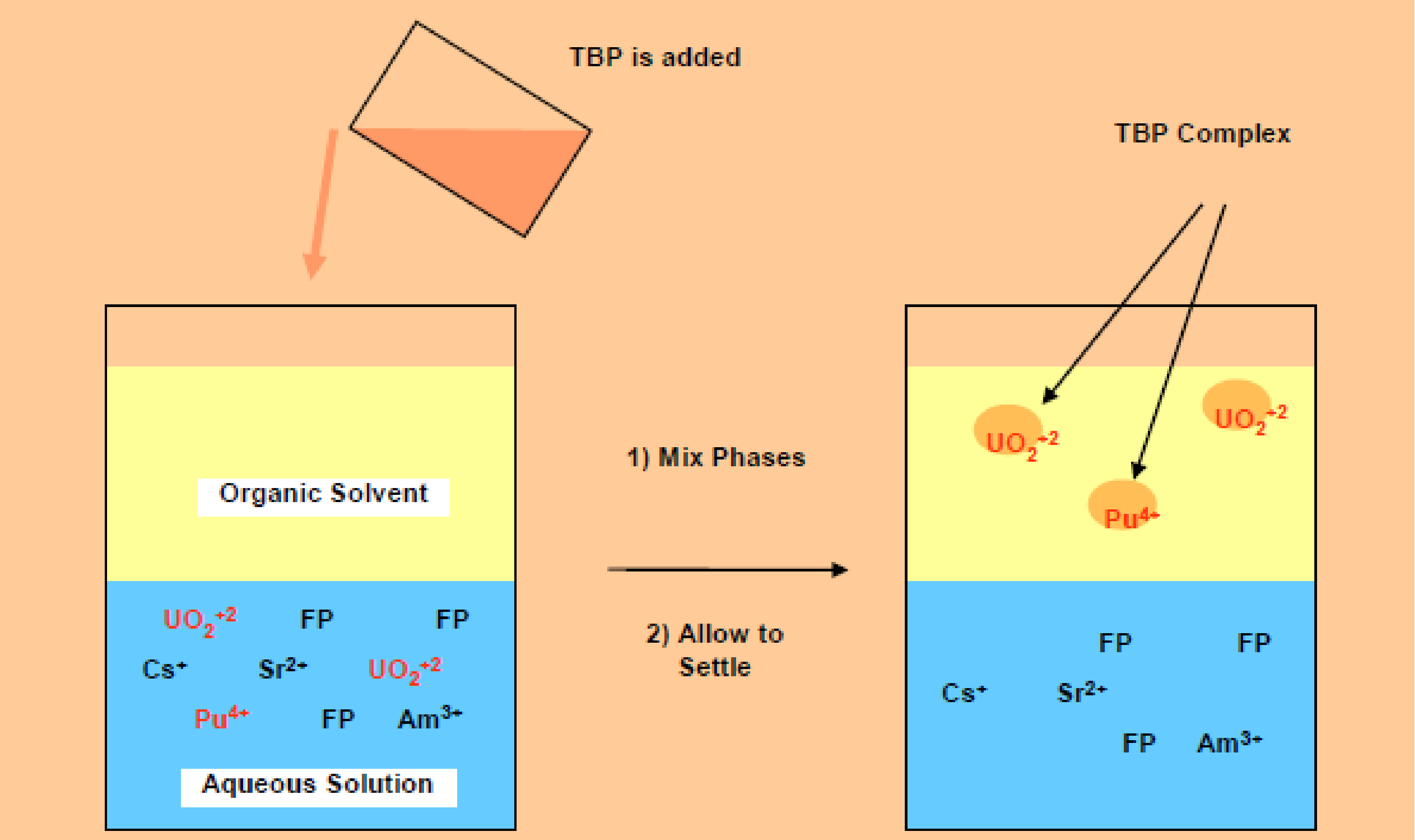
5. Chemistry in reprocessing of spent nuclear fuels
-PUREX: extraction of Pu and U with TBP-



$$UO_2^{2+} + 2NO_3^- + 2TBP \rightleftarrows UO_2(NO_3)_2 \bullet 2TBP$$

$$Pu^{4+} + 4NO_3^- + 2TBP \rightleftarrows Pu(NO_3)_4 \bullet 2TBP$$

*https://notes.engineeringonline.ncsu.edu/courses/ne/NE531/NE531_s09_08.pdf

5. Chemistry in reprocessing of spent nuclear fuels

-distribution coefficient, distribution ratio, decontamination factor-

$$\boxed{UO_2(NO_3)_2(aq) + 2TBP\ (org) \Leftrightarrow UO_2(NO_3)_2 \cdot 2TBP\ (org)}$$

$$\boxed{UO_2^{2+}{}_{(aq)} + 2NO_3^-{}_{(aq)} + 2TBP_{(org)} \Leftrightarrow UO_2(NO_3)_2 \cdot 2TBP_{(org)}}$$

distribution coefficient (KD, which is equilibrium constant), distribution ratio (DR), decontamination factor (DF)

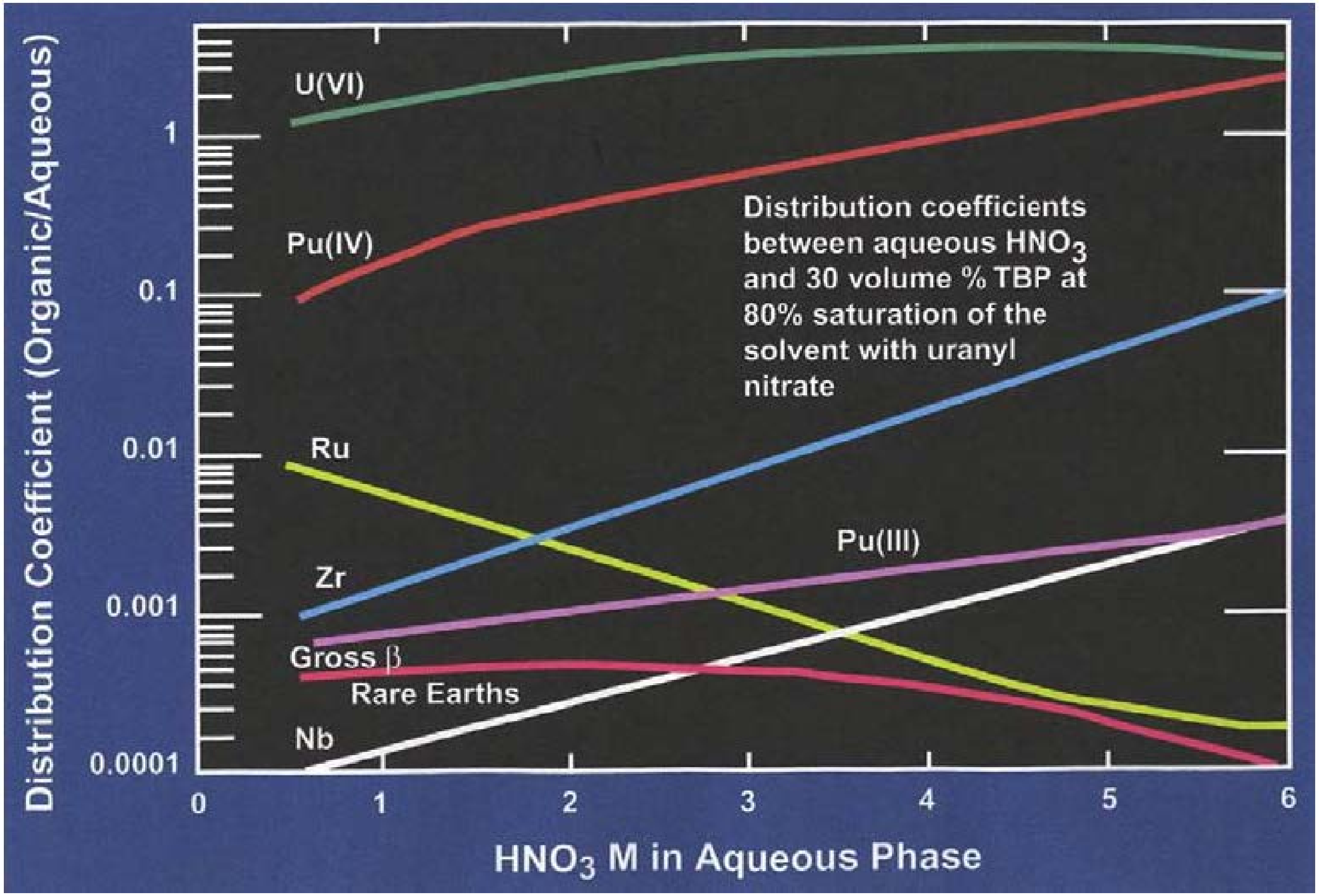
$$KD_U = \frac{\left[UO_2\left(NO_3\right)_2\text{-}2TBP_{(org)}\right]}{\left[UO_2{}^{2+}{}_{(aq)}\right]\left[NO_3{}^{-}{}_{(aq)}\right]^2\left[TBP_{(org)}\right]^2}$$

$$DR_U = \frac{\left[UO_2\left(NO_3\right)_2\text{-}2TBP_{(org)}\right]}{\left[UO_2{}^{2+}{}_{(aq)}\right]} = KD_U\left[NO_3{}^{-}{}_{(aq)}\right]^2\left[TBP_{(org)}\right]^2$$

$$DF_U = \frac{\left(\left[FP_{feed}\right]/\left[UO_2{}^{2+}{}_{feed}\right]\right)}{\left(\left[FP_{product}\right]/\left[UO_2{}^{2+}{}_{product}\right]\right)}$$

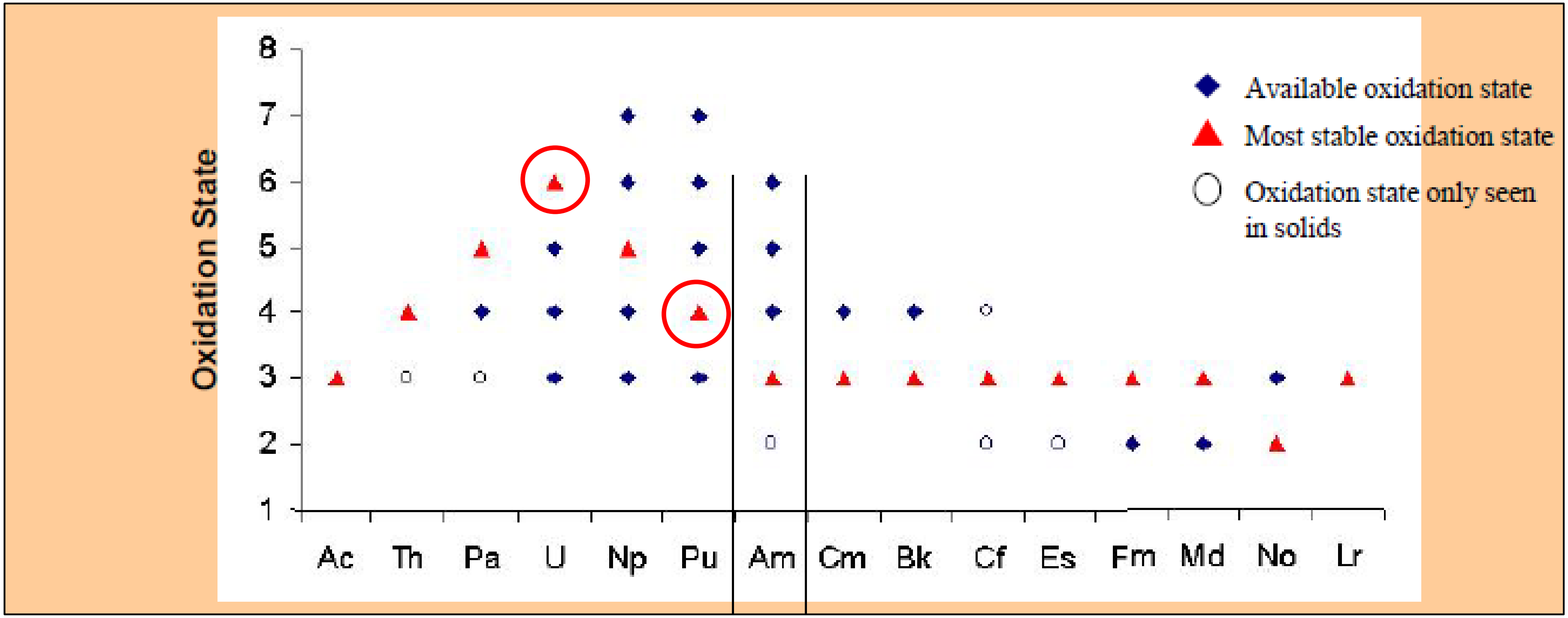
5. Chemistry in reprocessing of spent nuclear fuels
-comparison of distribution coefficients (KD_M) -

$$KD_U = \frac{\left[UO_2(NO_3)_2\text{-}2TBP_{(org)}\right]}{\left[UO_2{}^{2+}{}_{(aq)}\right]\left[NO_3{}^{-}{}_{(aq)}\right]^2\left[TBP_{(org)}\right]^2}$$



*http://www.cresp.org/NuclearChemCourse/monographs/07_Jubin_Introduction

5. Chemistry in reprocessing of spent nuclear fuels -comparison of distribution coefficients (KD_M) -



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1.2.1. Engineering processes for fuel cycles: reprocessing (wet) -PUREX: extraction of Pu and U with TBP-

$$UO_2^{2+}{}_{(aq)} + 2NO_3^-{}_{(aq)} + 2\,TBP_{(o)} \Leftrightarrow UO_2(NO_3)_2 \cdot 2\,TBP_{(o)}$$

$$Pu^{4+}{}_{(aq)} + 4NO_3^-{}_{(aq)} + 2\,TBP_{(o)} \Leftrightarrow Pu(NO_3)_4 \cdot 2\,TBP_{(o)}$$

$$KD_U = \frac{\left[\mathrm{UO_2(NO_3)_2\text{-}2TBP_{(org)}}\right]}{\left[\mathrm{UO_2^{2+}}_{(aq)}\right]\left[\mathrm{NO_3^-}_{(aq)}\right]^2\left[\mathrm{TBP_{(org)}}\right]^2}$$

$$DR_U = \frac{\left[\mathrm{UO_2(NO_3)_2\text{-}2TBP_{(org)}}\right]}{\left[\mathrm{UO_2^{2+}}_{(aq)}\right]} = KD_U\left[\mathrm{NO_3^-}_{(aq)}\right]^2\left[\mathrm{TBP_{(org)}}\right]^2$$

$$KD_{Pu^{4+}} = \frac{\left[Pu\mathrm{(NO_3)_4\text{-}2TBP_{(org)}}\right]}{\left[Pu^{4+}{}_{(aq)}\right]\left[\mathrm{NO_3^-}_{(aq)}\right]^4\left[\mathrm{TBP_{(org)}}\right]^2} \qquad DR_{Pu} = KD_{Pu}\left[\mathrm{NO_3^-}_{(aq)}\right]^4\left[\mathrm{TBP_{(org)}}\right]^2$$

- ✓ By increasing the concentration of uncombined TBP in the organic phase and by increasing the concentration of aqueous nitrate ion, the reactions are shifted to the right: e.g. 99 to 99.9% of fission products are separated.

*https://notes.engineeringonline.ncsu.edu/courses/ne/NE531/NE531_s09_08.pdf

5. Chemistry in reprocessing of spent nuclear fuels
-PUREX: U and Pu partition-

- Pu (specifically, Pu^{4+}) is separated from U by reducing Pu to the organic-insoluble, trivalent state (Pu^{3+}) with a reductant strong enough to act on plutonium but not so strong as also to reduce U.
  - U is left in the extractable hexavalent condition.
- Reductants used for this purpose include Fe^{2+}, U^{4+}, or cathodic reaction.

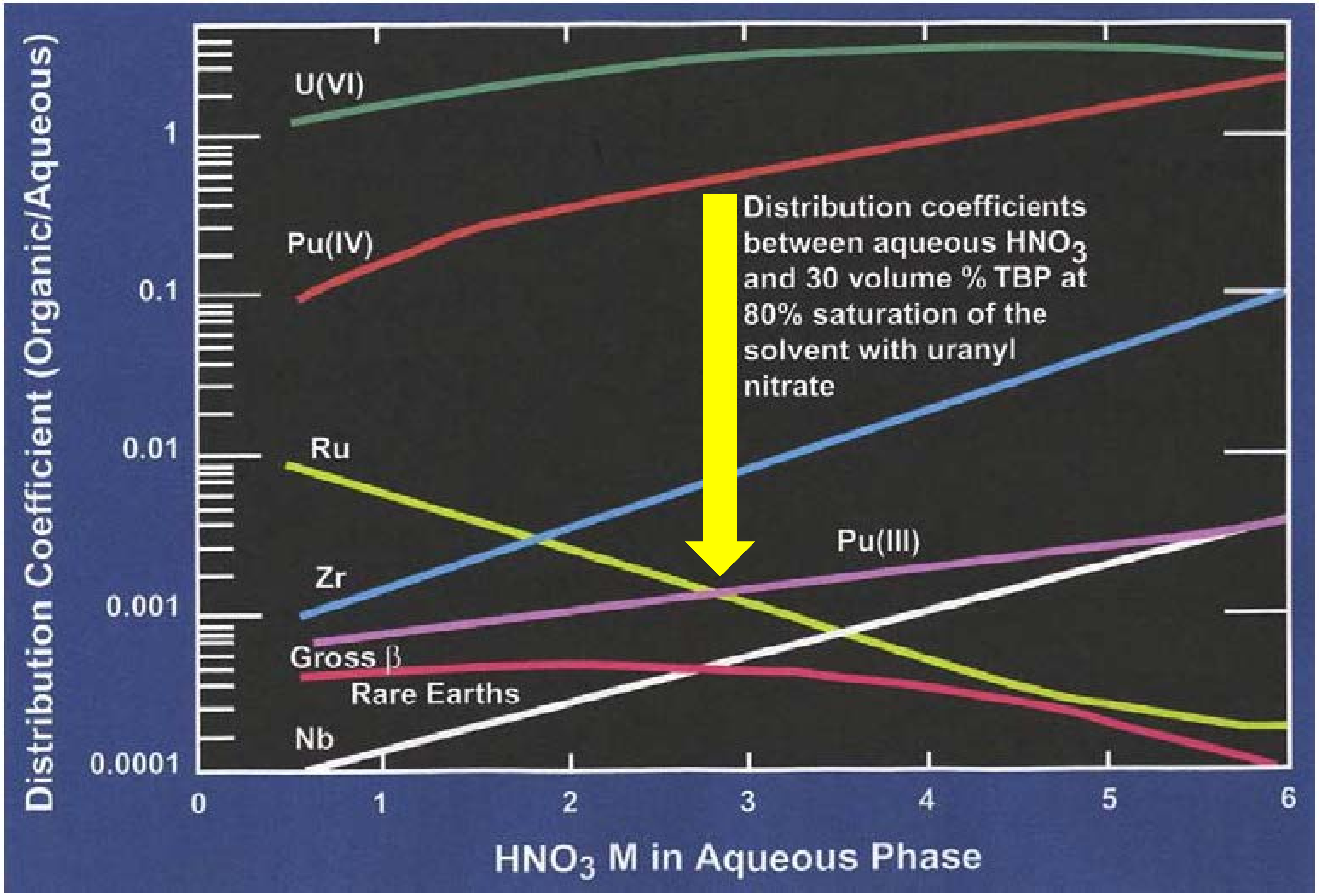
$$U^{4+} + 2Pu^{4+} + 2H_2O \longrightarrow UO_2^{2+} + 2Pu^{3+} + 4H^{+}$$

*https://notes.engineeringonline.ncsu.edu/courses/ne/NE531/NE531_s09_08.pdf

5. Chemistry in reprocessing of spent nuclear fuels
-PUREX: U and Pu partition-

$$KD_U = \frac{\left[UO_2\left(NO_3\right)_2\text{-}2TBP_{(org)}\right]}{\left[UO_2{}^{2+}{}_{(aq)}\right]\left[NO_3{}^{-}{}_{(aq)}\right]^2\left[TBP_{(org)}\right]^2}$$

$$KD_{Pu^{3+}} = \frac{\left[Pu\left(NO_3\right)_3\text{-}2TBP_{(org)}\right]}{\left[Pu^{3+}{}_{(aq)}\right]\left[NO_3{}^{-}{}_{(aq)}\right]^3\left[TBP_{(org)}\right]^2}$$

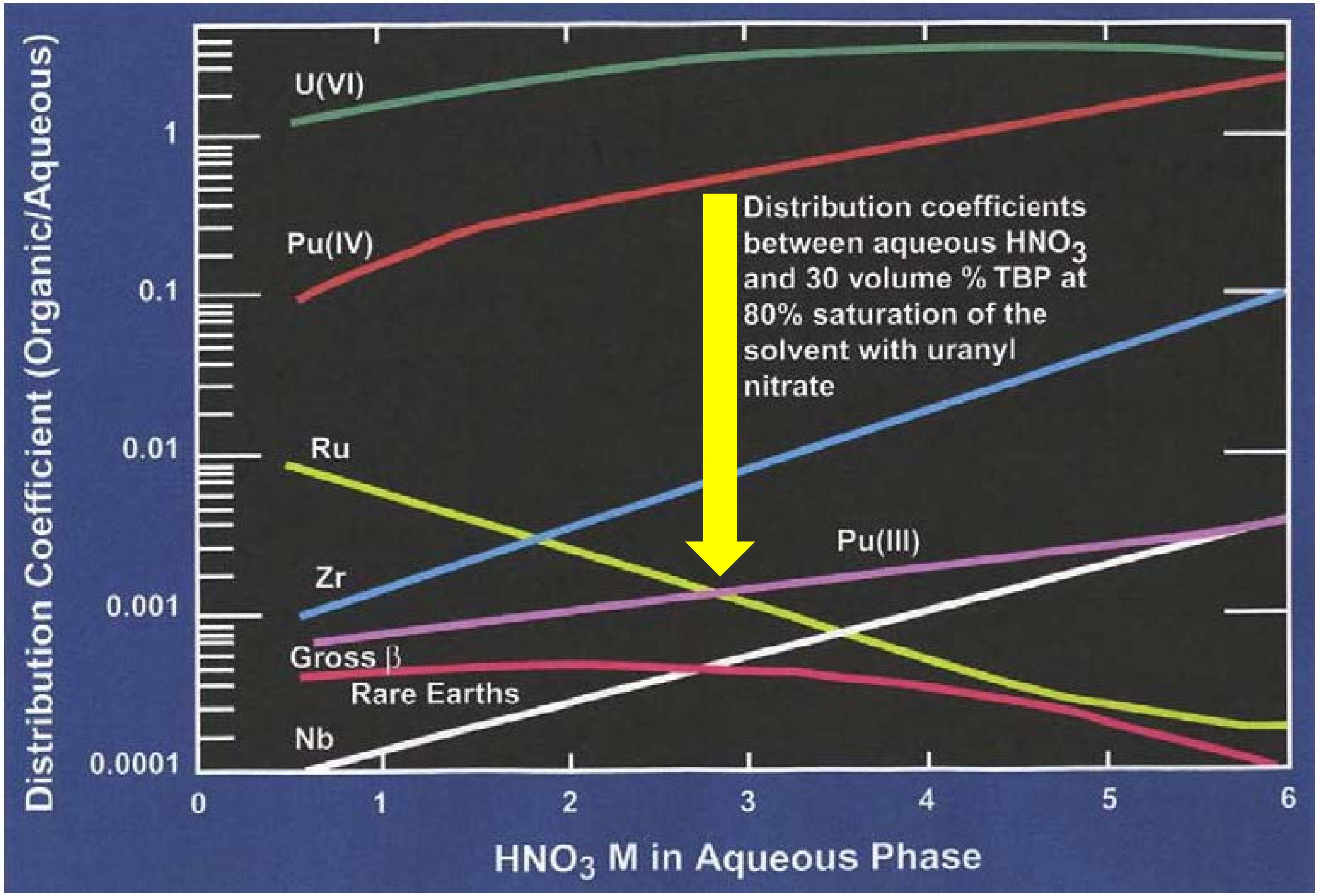


Distribution coefficients between aqueous HNO$_3$ and 30 volume % TBP at 80% saturation of the solvent with uranyl nitrate

*http://www.cresp.org/NuclearChemCourse/monographs/07_Jubin_Introduction

5. Chemistry in reprocessing of spent nuclear fuels
-PUREX: U and Pu partition-

$$\frac{KD_{Pu}}{KD_{U}} = \frac{\left[Pu\left(\mathrm{NO_3}\right)_3\text{-}2\mathrm{TBP}_{(org)}\right] / \left[Pu^{3+}{}_{(aq)}\right]}{\left[\mathrm{UO_2}\left(\mathrm{NO_3}\right)_2\text{-}2\mathrm{TBP}_{(org)}\right] / \left[\mathrm{UO_2}^{2+}{}_{(aq)}\right]} \times \frac{1}{\left[\mathrm{NO_3}^{-}{}_{(aq)}\right]} = \frac{DR_{Pu}}{DR_{U}} \frac{1}{\left[\mathrm{NO_3}^{-}{}_{(aq)}\right]} \sim 0.001$$



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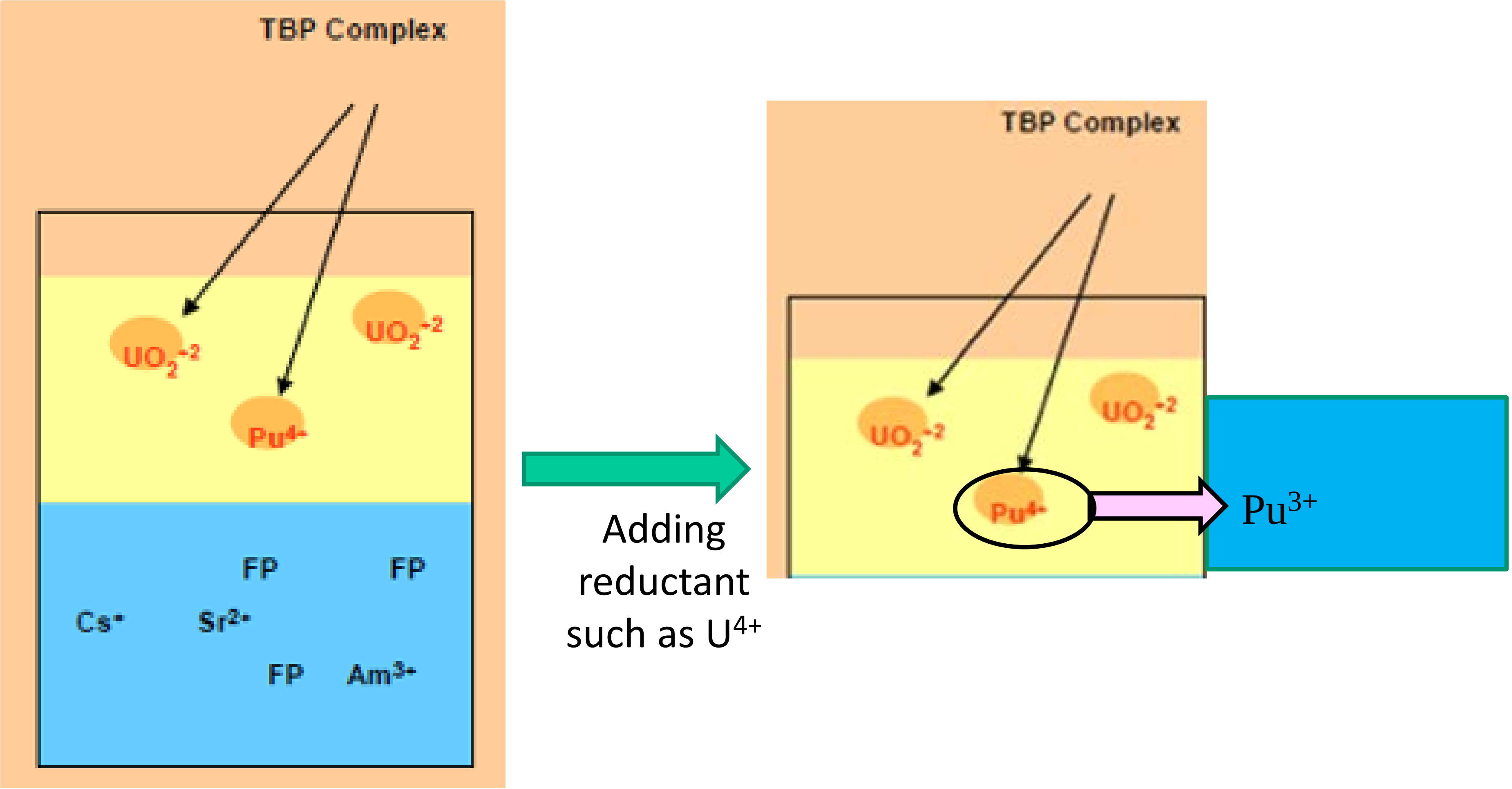
5. Chemistry in reprocessing of spent nuclear fuels
-PUREX: U and Pu partition-



Adding reductant such as U^{4+}

Pu^{3+}

*http://www.cresp.org/NuclearChemCourse/monographs/07_Jubin_Introduction

5. Chemistry in reprocessing of spent nuclear fuels
-PUREX: purification-

- ✓ Impure Pu nitrate is purified by one or more additional solvent extraction cycles.
  - ✓ Purified Pu nitrate is converted to the preferred product from PuO2 either by evaporation to dryness and calcinations or by precipitation as the oxalate or peroxide and calcinations.

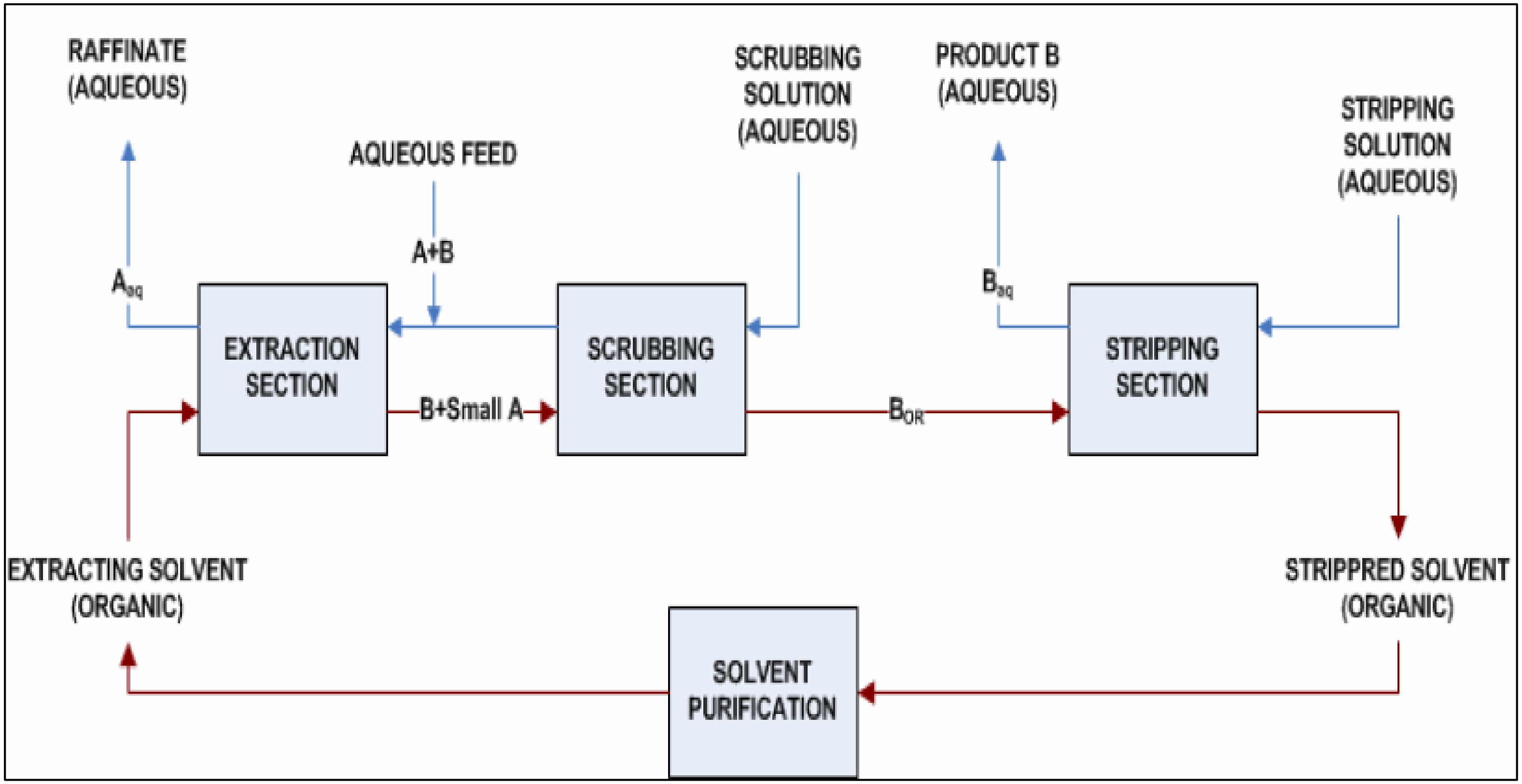
- ✓ Impure uranyl nitrate is purified by one or more additional cycles of solvent extraction.
  - ✓ Purified uranyl nitrate solution is evaporated to dryness and calcined to UO_3. Nitric acid vapors are condensed and recycled.

- ✓ If the UO_3 product is to be re-enriched, it is converted to UF_6.
  - ✓ Note that the isotope ratio of U in the spent fuel is around 1% ^{235}U.
  - ✓ According to Areva, about eight fuel assemblies reprocessed can yield one MOX fuel assembly, two-thirds of an enriched uranium fuel assembly, and about three tonnes of depleted uranium (enrichment tails) plus about 150 kg of wastes

*https://notes.engineeringonline.ncsu.edu/courses/ne/NE531/NE531_s09_08.pdf

*D.G. Cacuci, “Handbook of Nuclear Engineering”, Springer, 2010.

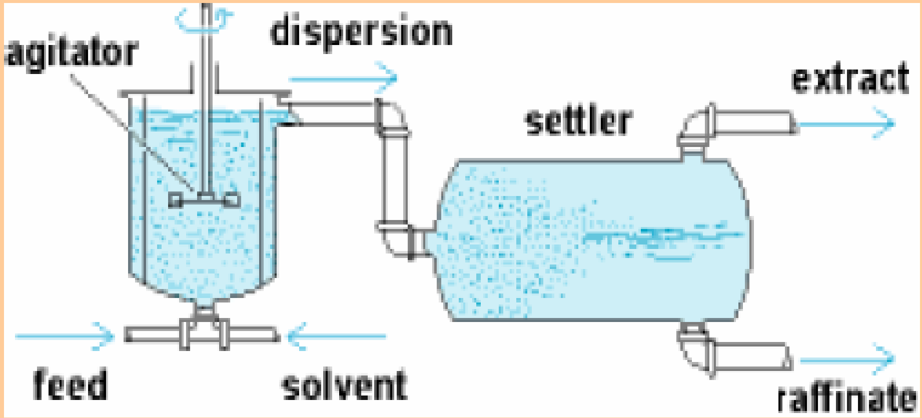
5. Chemistry in reprocessing of spent nuclear fuels
-PUREX: flow chart-



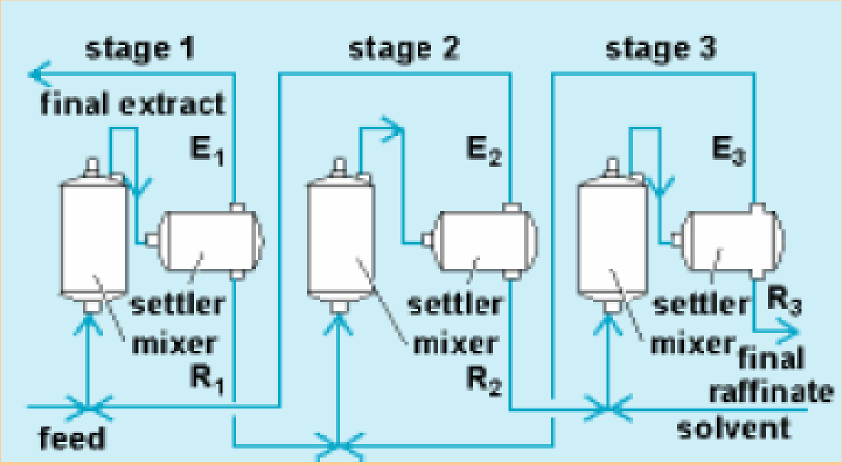
- ✓ **Extraction:** the initial transfer step whereby the main solute, often together with impurities, is transferred from feed to solvent.
- ✓ **Scrubbing:** the process of selectively removing contaminating solutes (impurities) from an extract that contains these as well as the main extractable solute by treatment with a new immiscible liquid phase.
- ✓ **Stripping**: The process of removing solute(s) from a loaded solvent or extract.
- ✓ **Raffinate**: the phase remaining after extraction of some specified solute(s).

*D.G. Cacuci, “Handbook of Nuclear Engineering”, Springer, 2010.

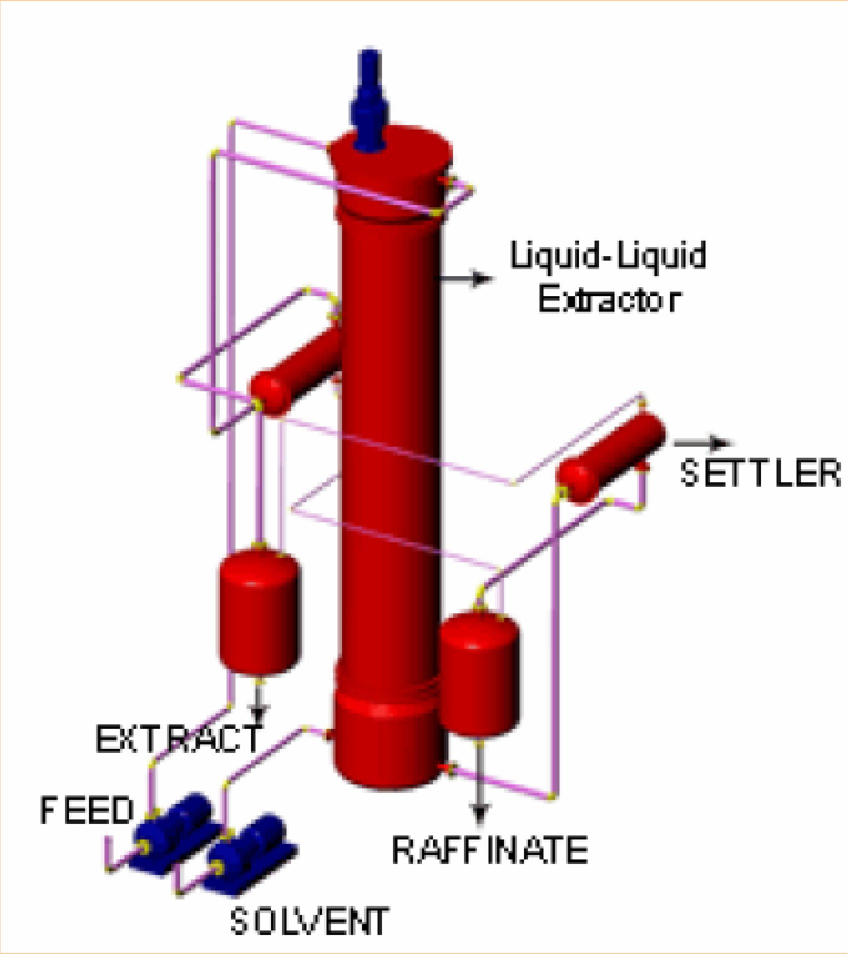
5. Chemistry in reprocessing of spent nuclear fuels
-PUREX: Mixer settler-



Single-Stage Mixer-settler



3-Stage Mixer-settler

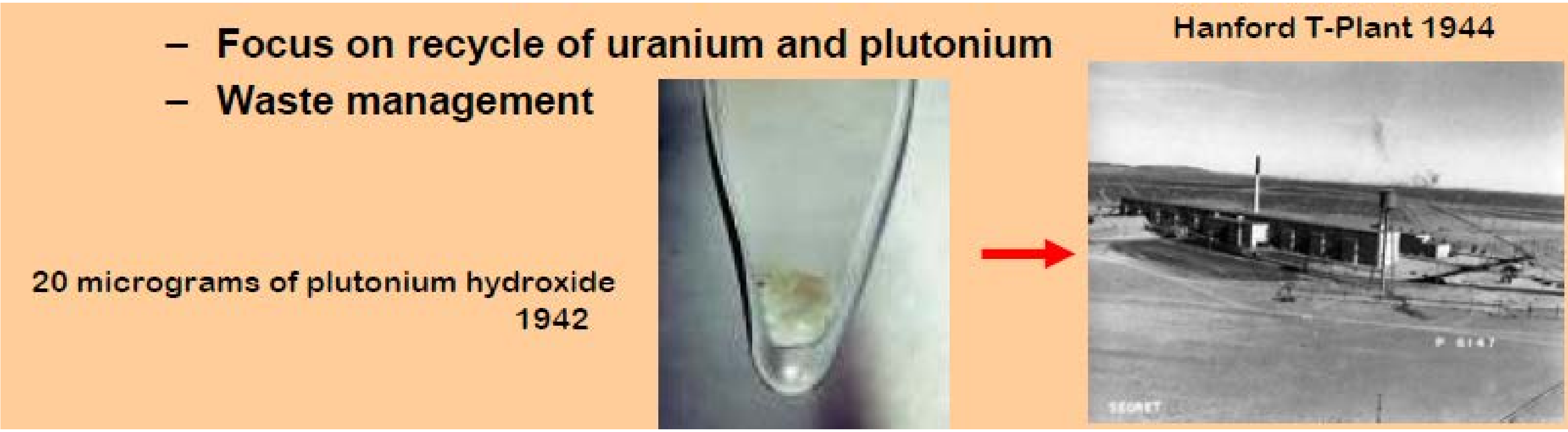


Commercial Multi-Stage Mixer-settler

*https://notes.engineeringonline.ncsu.edu/courses/ne/NE531/NE531_s09_08.pdf

5. Chemistry in reprocessing of spent nuclear fuels
-History of reprocessing

- Reprocessing began during Manhattan project to recover Pu-239.
  - Seaborg first separated microgram quantities of Pu in 1942 using "Bismuth-Phosphate precipitation process"
  - Process scaled to kilogram quantity production at Hanford in 1944.
- Solvent extraction processes followed to allow concurrent separation and recovery of both U and Pu.
- Reprocessing transitioned from defense to commercial use



- Focus on recycle of uranium and plutonium
- Waste management

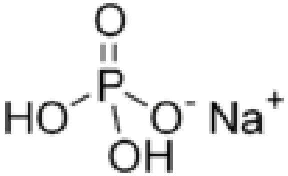
20 micrograms of plutonium hydroxide
1942

Hanford T-Plant 1944

*https://notes.engineeringonline.ncsu.edu/courses/ne/NE531/NE531_s09_08.pdf

5. Chemistry in reprocessing of spent nuclear fuels
-History of reprocessing: Bismuth Phosphate process-

- ✓ Pu is co-precipitated as $Pu_3(PO_4)_4$ by the addition of bismuth nitrate, $Bi(NO_3)_3$, and sodium phosphate.



*wikipedia

<Advantages of Bismuth phosphate processes>

- ✓ Recovery of >95% of Pu
- ✓ Decontamination factors: 10^7 for fissile products

$$DF_{Pu} = \frac{\left([FP_{\text{feed}}]/[Pu^{4+}{}_{\text{feed}}]\right)}{\left([FP_{\text{product}}]/[Pu^{4+}{}_{\text{product}}]\right)}$$

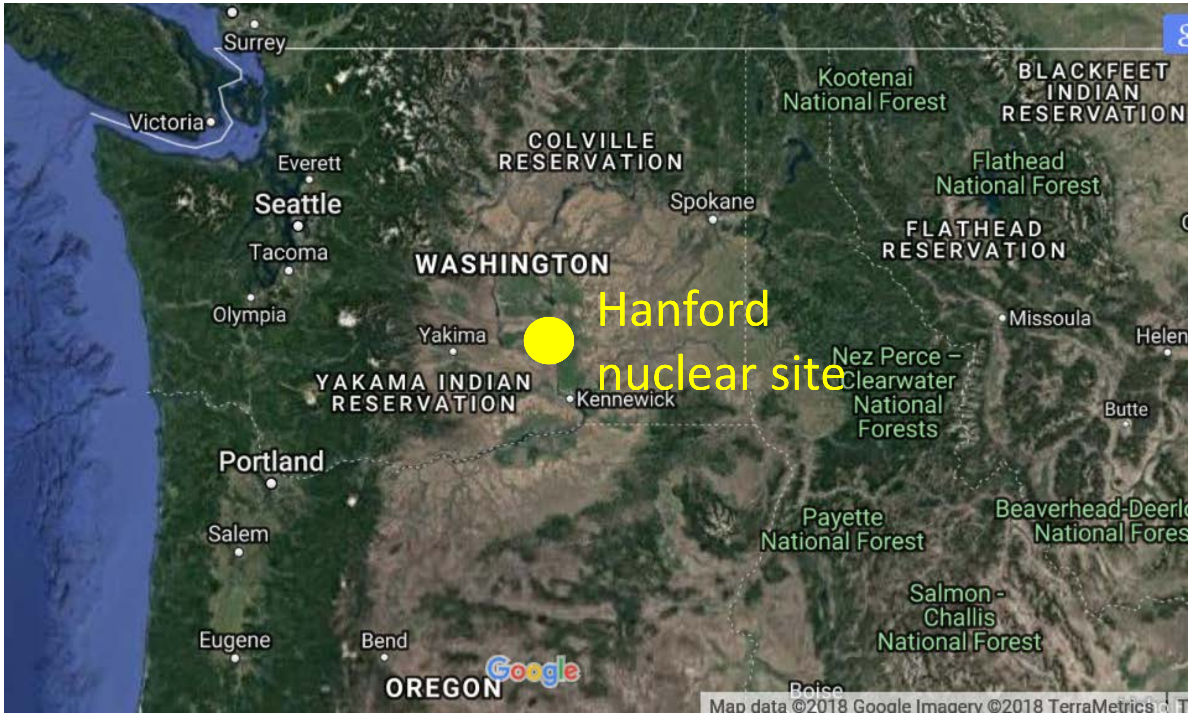
<Disadvantages of Bismuth phosphate processes>

- ✓ Batch operations
- ✓ Inability to recover uranium
- ✓ Required numerous cycles and chemicals
  - ✓ Produced large volume of high-level waste

*https://notes.engineeringonline.ncsu.edu/courses/ne/NE531/NE531_s09_08.pdf

5. Chemistry in reprocessing of spent nuclear fuels
-History of reprocessing: REDOX process-

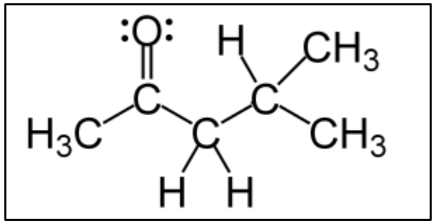
- ✓ First solvent extraction process used in reprocessing
  - ✓ Continuous process
  - ✓ Recovers both U and Pu with high yield and high decontamination factors from fission products.
- ✓ Developed at Argonne National Laboratory (ANL).
- ✓ Tested in pilot plant at Oak Ridge National Laboratory (ORNL) in 1948-49.
- ✓ REDOX plant built in Hanford in 1951.


Hanford nuclear site


Hanford REDOX -Plant (1951)

*https://notes.engineeringonline.ncsu.edu/courses/ne/NE531/NE531_s09_08.pdf

5. Chemistry in reprocessing of spent nuclear fuels
-History of reprocessing: REDOX process-



- ✓ Hexone (methyl isobutyl ketone) used as the extractant.
  - ✓ Immiscible with water
  - ✓ Used to purify uranium ore concentrates
  - ✓ Extracts both uranyl and plutonyl nitrates selectively from fission products
  - ✓ Highly flammable and volatile

- ✓ Plutonium is oxidized to Pu (VI) for highest recovery.
- ✓ U(VI) and Pu(VI) co-extracted, then Pu is reduced to Pu(III) by ferrous sulfamate, $Fe(H_2NO_3S)_2$, and scrubbed from the solvent.

- ✓ Large amounts of salt reagent (aluminum nitrate), to increase the nitrate concentration in the aqueous phase to obtain a reasonable distribution ratio.

$$DR_U = \frac{\left[UO_2\left(NO_3\right)_2\text{-}2TBP_{(org)}\right]}{\left[UO_2{}^{2+}{}_{(aq)}\right]} = KD_U\left[NO_3{}^{-}{}_{(aq)}\right]^2\left[TBP_{(org)}\right]^2$$

*https://notes.engineeringonline.ncsu.edu/courses/ne/NE531/NE531_s09_08.pdf
*https://en.wikipedia.org/wiki/Nuclear_reprocessing

5. Chemistry in reprocessing of spent nuclear fuels
-History of reprocessing: PUREX process-

- ✓ Tributyl phosphate is stated to be used as the extraction in a hydrocarbon diluent (dodecane or kerosene)
  - ✓ Suggested by Warf in 1949 for the recovery of Ce (IV) from rare earth nitrates.
  - ✓ Developed by Knolls Atomic Power Lab. And tested at Oak Ridge Natinoal Lab. In 1950-52
  - ✓ Used for Pu production plant at Savannah River in 1954 (H-canyon facility still operational in 2008)
  - ✓ Replaced REDOX process at Hanford in 1956.
  - ✓ Modified PUREX used in Idaho National Laboratory began in 1953 (first cycle)

<Advantages of PUREX over REDOX process>

- ✓ Nitric acid is used as salting and scrubbing agent and can be evaporated, which results in less HLW.
- ✓ TBP is less volatile and flammable than hexone.
- ✓ TBP is more chemically stable in a nitric acid environment.
- ✓ Consequently, the operating cost is lower.

*https://notes.engineeringonline.ncsu.edu/courses/ne/NE531/NE531_s09_08.pdf

5. Chemistry in reprocessing of spent nuclear fuels
-History of reprocessing: PUREX process-

<Advantages>

- ✓ Continuous operation, high throughput
- ✓ Countercurrent operation, high purity and selectivity
- ✓ Recycle solvent, minimizing waste

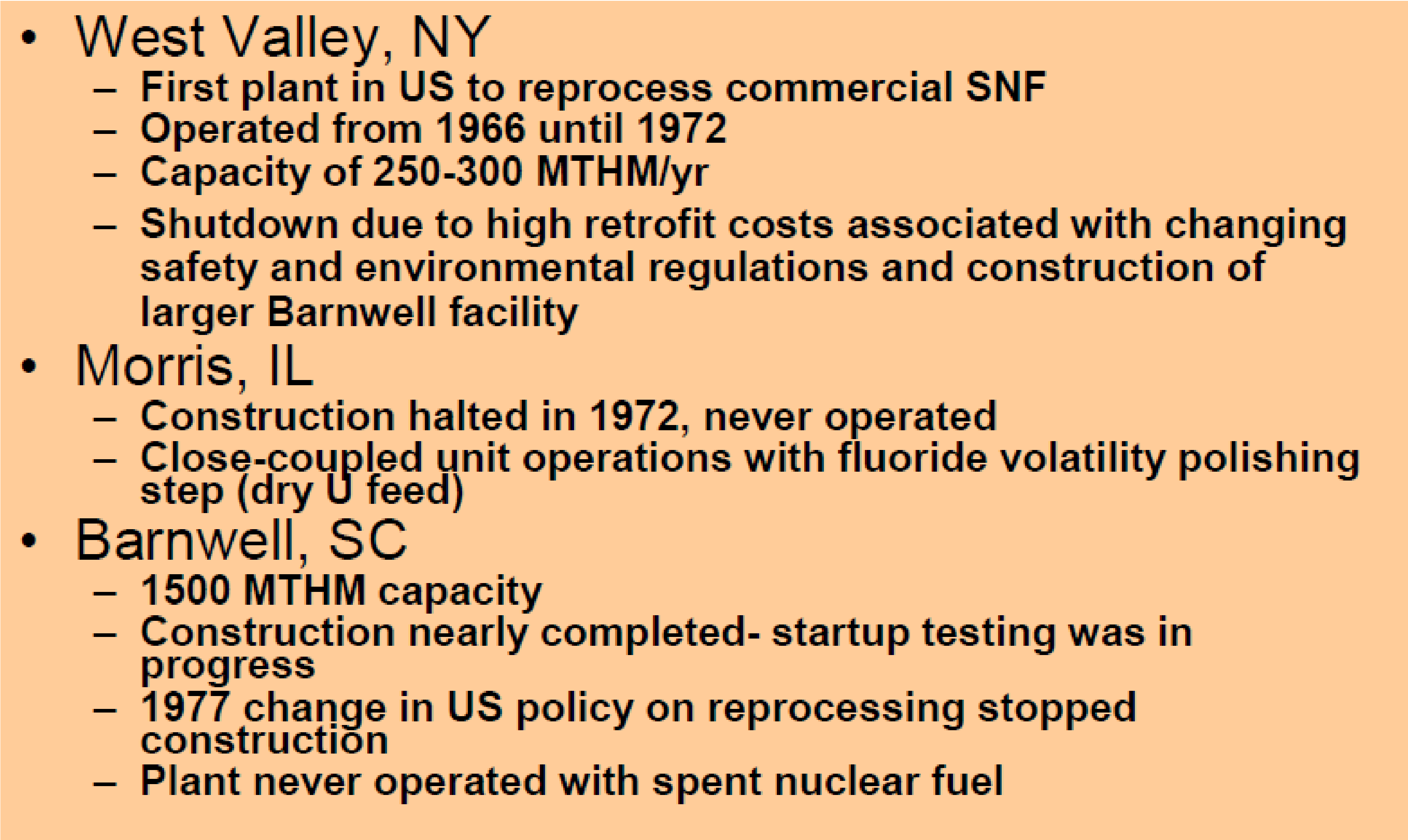
<Disadvantages>

- ✓ Solvent degradation due to hydrolysis and radiolysis
- ✓ Requires substantial tankage and reagents
- ✓ Proliferation risk as Pu is isolated.

- ✓ Initial use was for defense purposes, to recover plutonium from spent nuclear fuel
  - ✓ Hanford, Savannah River and Idaho all utilized the PUREX process for the recovery of Pu and/or highly enriched U.
  - ✓ UK, France, China, India (and North Korea) used the PUREX process to recover plutonium for weapons.

*https://notes.engineeringonline.ncsu.edu/courses/ne/NE531/NE531_s09_08.pdf

5. Chemistry in reprocessing of spent nuclear fuels
-History of reprocessing, commercial use in the US-



- West Valley, NY
  - First plant in US to reprocess commercial SNF
  - Operated from 1966 until 1972
  - Capacity of 250-300 MTHM/yr
  - Shutdown due to high retrofit costs associated with changing safety and environmental regulations and construction of larger Barnwell facility
- Morris, IL
  - Construction halted in 1972, never operated
  - Close-coupled unit operations with fluoride volatility polishing step (dry U feed)
- Barnwell, SC
  - 1500 MTHM capacity
  - Construction nearly completed- startup testing was in progress
  - 1977 change in US policy on reprocessing stopped construction
  - Plant never operated with spent nuclear fuel

*https://notes.engineeringonline.ncsu.edu/courses/ne/NE531/NE531_s09_08.pdf

5. Chemistry in reprocessing of spent nuclear fuels
-History of reprocessing, commercial use in other nations-

- ✓ **France**
    - o Magnox plant in Marcoule began operation in 1958 (~400 MT/yr)
    - o Magnox plant in La Hague began operation in 1967 (~400 MT/yr)
    - o LWR oxide plant (UP2) began in La Hague in 1976 (800 MT/yr)
    - o LWR oxide plant (UP3) began in La Hague in 1990 (800 MT/yr)

- ✓ **United Kingdom**
    - o Windscale plant for Magnox fuel began in 1964 (1200-1500 MT/yr)
    - o THORP LWR oxide plant began in 1994 (~1200 MT/yr)

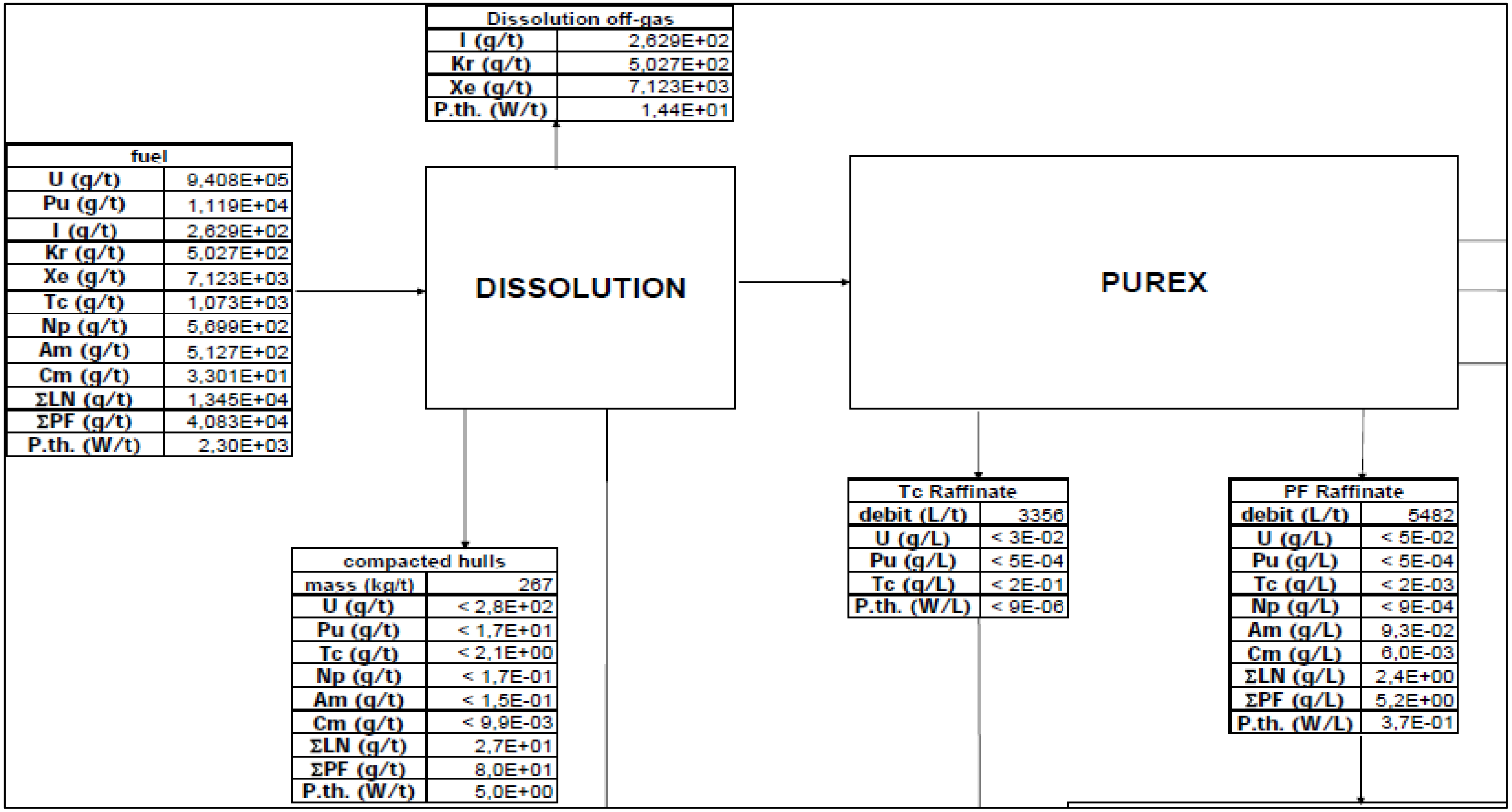
- ✓ **Japan**
    - o Tokai-Mura plant began in 1975 (~200 MT/yr)
    - o Rokkasho plant currently undergoing hot commissioning (800 MT/yr)

- ✓ **Russia**
    - o Plant RT-1
    - o Began operation in 1976, 400 MT capacity

*https://notes.engineeringonline.ncsu.edu/courses/ne/NE531/NE531_s09_08.pdf

5. Chemistry in reprocessing of spent nuclear fuels

-Flowsheet for UOX fuel 45 GWd/t 5-year cooled-



Dissolution off-gas	
I (g/t)	2,629E+02
Kr (g/t)	5,027E+02
Xe (g/t)	7,123E+03
P.th. (W/t)	1,44E+01

fuel	
U (g/t)	9,408E+05
Pu (g/t)	1,119E+04
I (g/t)	2,629E+02
Kr (g/t)	5,027E+02
Xe (g/t)	7,123E+03
Tc (g/t)	1,073E+03
Np (g/t)	5,699E+02
Am (g/t)	5,127E+02
Cm (g/t)	3,301E+01
ΣLN (g/t)	1,345E+04
ΣPF (g/t)	4,083E+04
P.th. (W/t)	2,30E+03

Tc Raffinate	
debit (L/t)	3356
U (g/L)	< 3E-02
Pu (g/L)	< 5E-04
Tc (g/L)	< 2E-01
P.th. (W/L)	< 9E-06

PF Raffinate	
debit (L/t)	5482
U (g/L)	< 5E-02
Pu (g/L)	< 5E-04
Tc (g/L)	< 2E-03
Np (g/L)	< 9E-04
Am (g/L)	9,3E-02
Cm (g/L)	6,0E-03
ΣLN (g/L)	2,4E+00
ΣPF (g/L)	5,2E+00
P.th. (W/L)	3,7E-01

compacted hulls	
mass (kg/t)	267
U (g/t)	< 2,8E+02
Pu (g/t)	< 1,7E+01
Tc (g/t)	< 2,1E+00
Np (g/t)	< 1,7E-01
Am (g/t)	< 1,5E-01
Cm (g/t)	< 9,9E-03
ΣLN (g/t)	2,7E+01
ΣPF (g/t)	8,0E+01
P.th. (W/t)	5,0E+00

- ✓ [Raffinate] In chemical separation terminology, raffinate is a product which has had a component or components removed.
- ✓ [Sludge] thick, soft, wet mud or a similar viscous mixture of liquid and solid components, especially the product of an industrial or refining process.
- ✓ [Hull] the main body of a ship or other vessel, including the bottom, sides, and deck but not the masts, superstructure, rigging, engines, and other fittings.

*"Spent Nuclear Fuel Reprocessing Flowsheet", OECD-NEA, 2012.

5. Chemistry in reprocessing of spent nuclear fuels
-Advanced method: additional separation after PUREX-

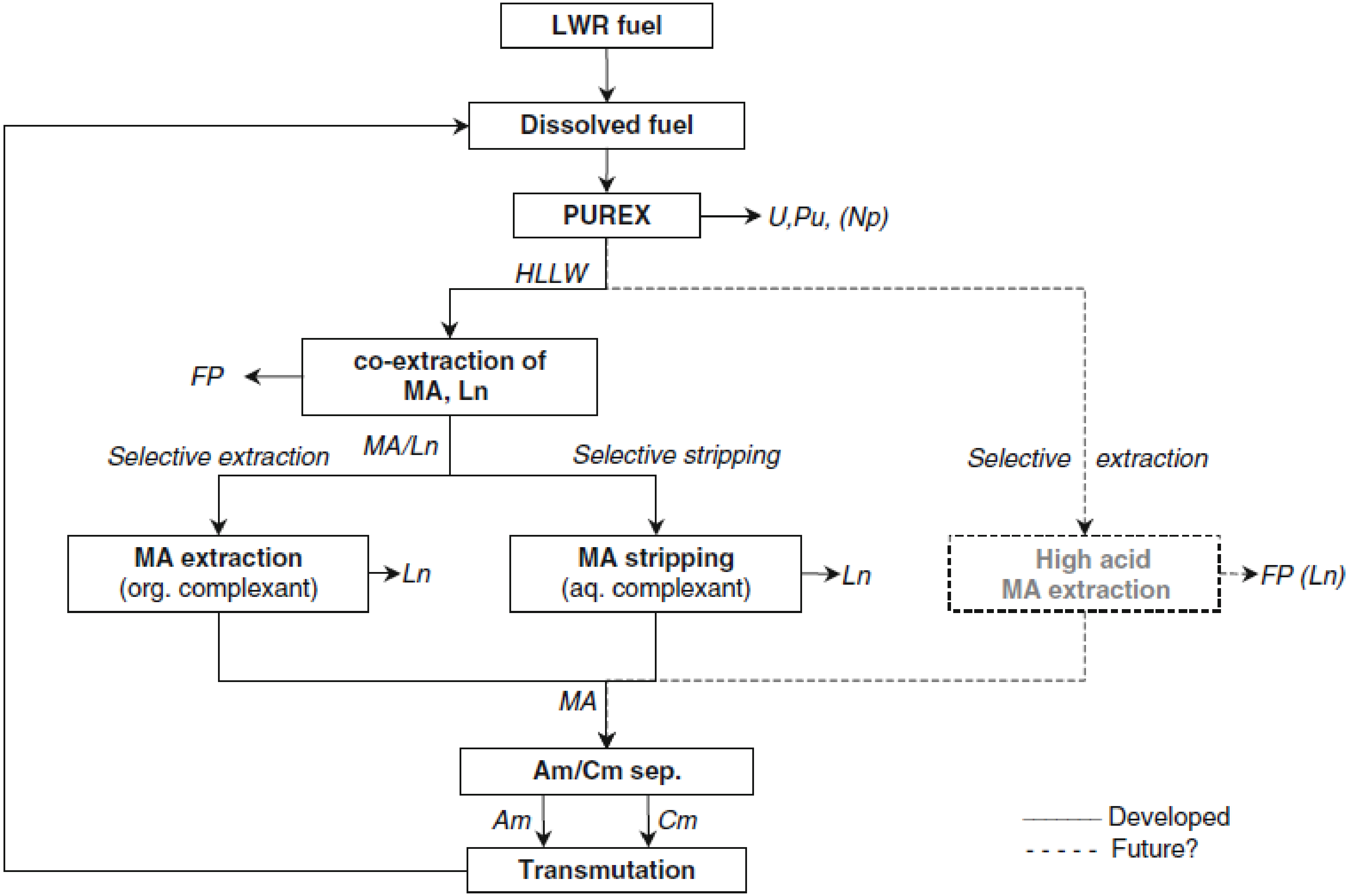


Figure 17

Strategies for the separation of the minor actinides from high level liquid waste (HLLW)

*D.G. Cacuci, “Handbook of Nuclear Engineering”, Springer, 2010.

5. Chemistry in reprocessing of spent nuclear fuels
-Advanced method: UREX+, for example-

- The UREX+ process consisted of five solvent extraction processes:

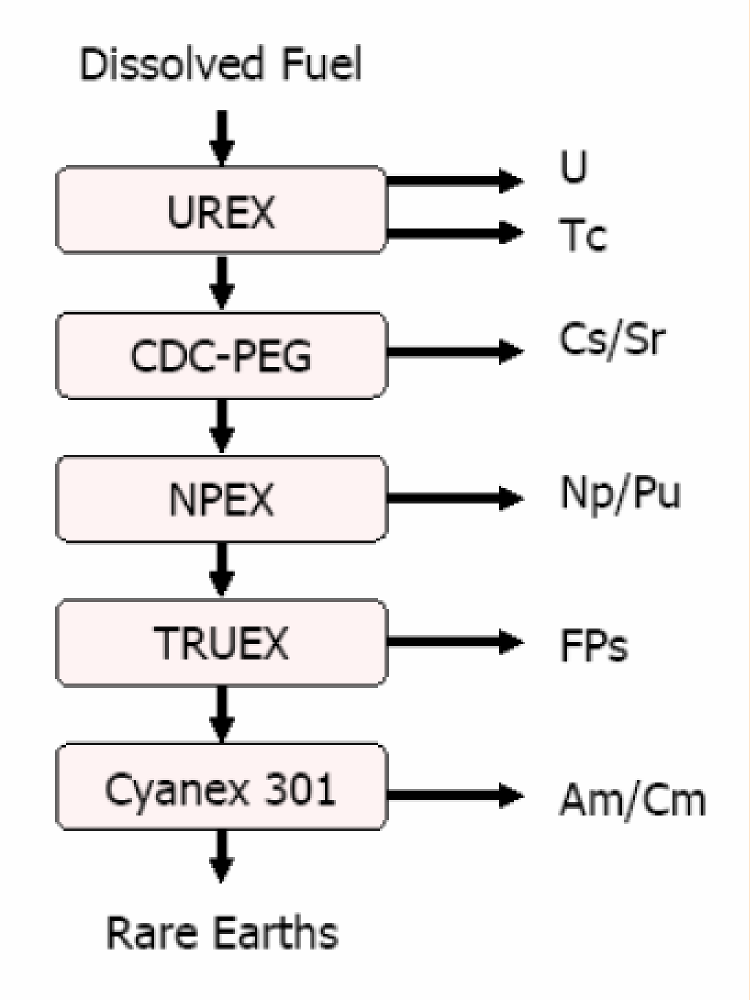
(1) UREX (U and Tc removal);

(2) CDC-PEG (Cs and Sr removal);

(3) NPEX (Pu and Np removal);

(4) TRUEX (Am, Cm, and rare-earth-fission-product removal), and;

(5) Cyanex-301 (separation of Am and Cm from the rare earths).



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