IMPLICATIONS OF ADVANCED NUCLEAR FUEL CYCLES FOR NUCLEAR WASTE MANAGEMENT

PRESENTED AT THE NUCLEAR INTEGRATION PROJECT WORKSHOP

"THE BACK-END: HEALING THE ACHILLES HEEL OF THE NUCLEAR RENAISSANCE"

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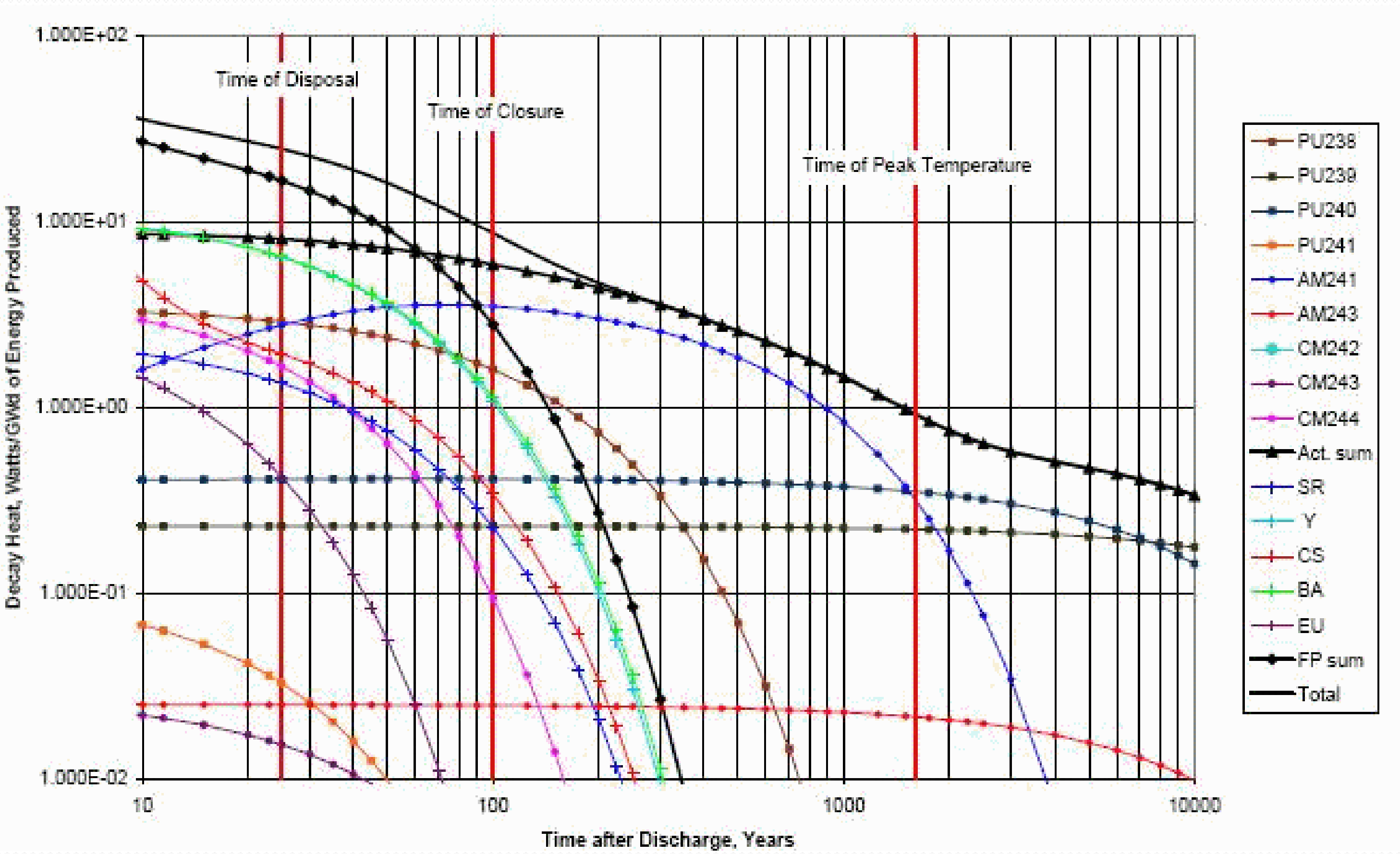
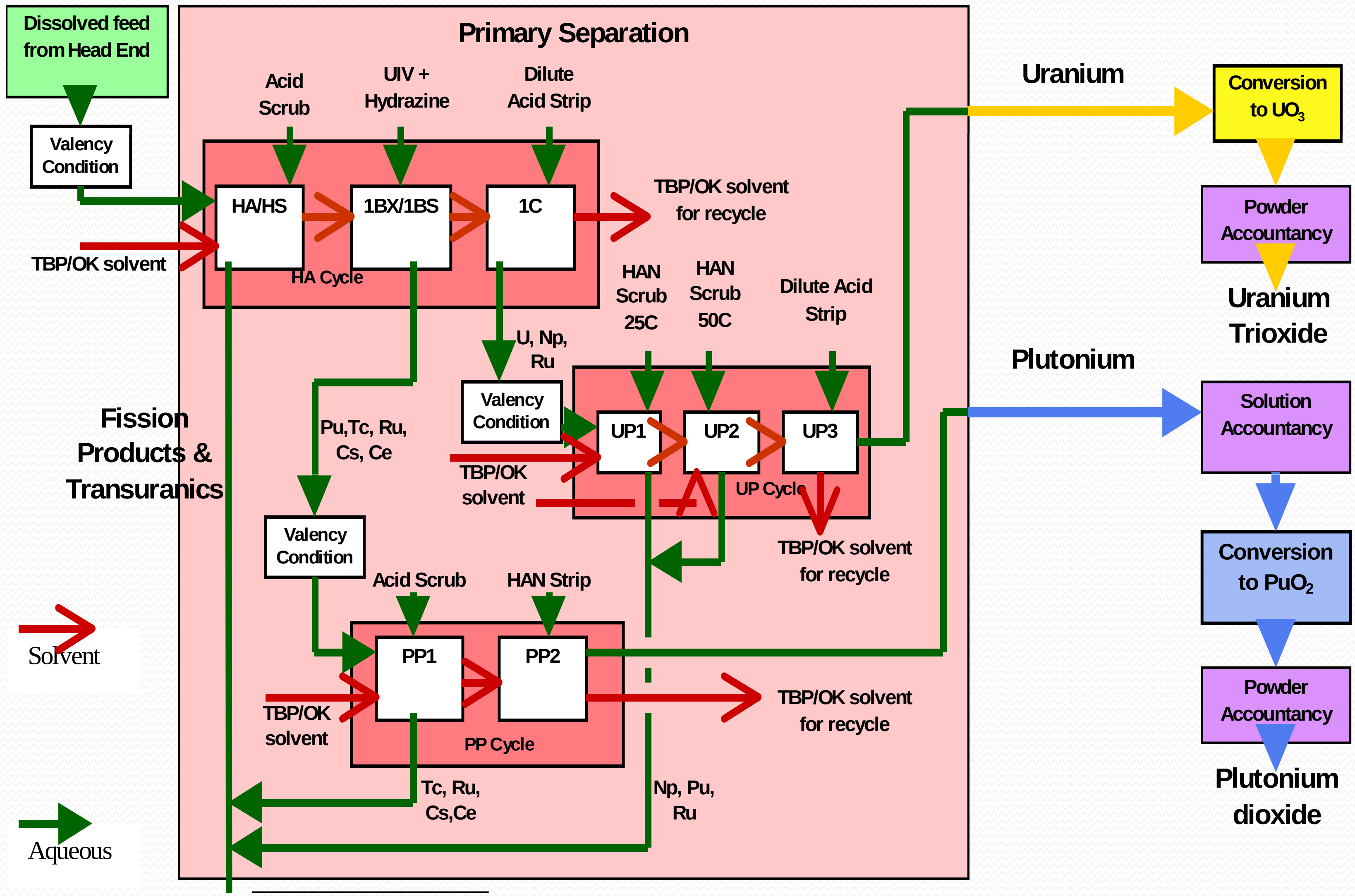
CONTRIBUTIONS OF SELECTED ACTINIDES AND FISSION PRODUCTS TO SPENT NUCLEAR FUEL DECAY HEAT GENERATION RATE



THORP REPROCESSING FLOWSHEET
A MODERN-DAY PROCESS



PROCESS WASTE STREAMS

- GASES AND VOLATILES: 3H, 85Kr, 14C, I, Ru, Xe
- 99Tc
- Cs/Sr
- OTHER FISSION PRODUCTS (INCLUDING LANTHANIDES)
- CLADDING HULLS AND FUEL ELEMENT STRUCTURAL MATERIAL
- SOLVENT CLEANUP WASTES

GASEOUS AND VOLATILE WASTES

- **3H**
  - **99.9% RELEASED FROM FUEL AND HULLS**
  - **100% OF RELEASED 3H MAY BE CAPTURED IN OFF GAS SYSTEM**
- **85Kr**
  - **100% RELEASED DURING FUEL DISSOLUTION**
  - **0.85% MAY BE CAPTURED IN OFF GAS SYSTEM**
- **14C**
  - **100% RELEASED DURING FUEL DISSOLUTION**
  - **0.99% MAY BE CAPTURED IN OFF GAS SYSTEM**

GASES AND VOLATILE WASTES (CONTINUED)

- I
  - 98.4% ENTERS OFF GAS AS MOLECULAR IODINE
  - 100% MAY BE CAPTURED IN OFF GAS SYSTEM
- **Ru** (2.19 KG OF STABLE ISOTOPES FROM ONE MTIHM WITH 33 Gwd BURNUP)
  - ESSENTIALLY COMPLETELY EVOLVED AS RuO_4 DURING SPENT FUEL DISSOLUTION
  - MAY BE CAPTURED ON SILICA GEL
- **Xe:** MAY BE CAPTURED WITH Kr (APPROX. 20 TIMES THE VOLUME OF ^{85}Kr)

FISSION PRODUCTS (INCLUDING LANTHANIDES)

- **FISSION PRODUCTS SEPARATED FROM ACTINIDES AND LANTHANIDES**
  - **ESSENTIALLY COMPLETE SEPARATION**
- **LANTHANIDES SEPARATED FROM ACTINIDES**
  - **99.9% SEPARATION FROM ACTINIDES**

CLADDING HULLS AND FUEL ELEMENT STRUCTURAL MATERIALS TREATMENT

- **FUEL ASSEMBLY HARDWARE MASS, kg**
  - **ZIRCALOY-4 (CLADDING, GUIDE TUBES) 108.4**
  - **STAINLESS STEEL 304 (END FITTINGS) 17.1**
  - **STAINLESS STEEL 302 (PLENUM SPRINGS) 21.9**
  - **INCONEL-718 (GRID SPACERS) 5.9**
  - **NICROBRAZE 50 (BRAZING ALLOY) 1.2**
    - **HARDWARE TOTAL: 154.5 KG**
- **COMPACTED FOR DISPOSAL (CONTAINS SOME UNDISSOLVED NOBLE METALS - ESTIMATED AMOUNTS)**
    - **Tc: ~ 15% OF TOTAL**
    - **Ru: ~ 50% OF TOTAL**
    - **Pd: ~ 20% OF TOTAL**
    - **Mo: ~ 40% OF TOTAL**
    - **Rh: ~ 10% OF TOTAL**

WASTE TREATMENTS

FISSION PRODUCT GASES AND VOLATILES

- **TRITIUM**
  - **VOLOXIDATION OF SHEARED FUEL SOLIDS RELEASES 3H; 3H CATALYTICALLY CONVERTED TO TRITIATED WATER**
- **85Kr**
  - **ISOLATED BY CRYOGENIC DISTILLATION**
- **Xe**
  - **CAPTURED WITH 85Kr (OPTIONAL)**
- **14C (EVOLVED FROM DISSOLVER AS CO2)**
  - **TRAPPED ON MOLECULAR SIEVES AND/OR AS CARBONATE IN ALKALINE SOLUTION**
- **129I**
  - **CAPTURED FROM DISSOLVER OFF GAS ON SILVER SORBENT OR IN ALKALINE SOLUTION**
- **Ru (SEVERAL STABLE ISOTOPES: 99Ru TO 104Ru)**
  - **SORPTION AS RuO4 ON SILICA GEL**

TECHNETIUM-99

- OCCURS IN SEVERAL PLACES AND AS SEVERAL SPECIES IN REPROCESSING FLOWSHEETS
  - DISSOLVER SOLIDS (PRESENT AS FINELY DIVIDED METAL WITH FUEL ELEMENT HULLS AND HARDWARE)
  - SEPARATED WITH URANIUM IN TBP SOLVENT EXTRACTION (SX) STEP
  - PRESENT IN DISSOLVER SOLUTION AS TcO_4^-
- POTENTIAL SEPARATION FROM DISSOLVER SOLUTION BY ION EXCHANGE OR SX
- CAN BE SEPARATED FROM URANIUM IF DESIRED DURING FIRST SX CYCLE

CESIUM AND STRONTIUM MAY BE TREATED BY SEVERAL OPTIONS

- **ION EXCHANGE**
- **SOLVENT EXTRACTION**
- **PRECIPITATION**

FISSION PRODUCTS

- REMAIN WITH RAFFINATE FROM FIRST SOLVENT EXTRACTION STEP
- SEPARATED FROM LANTHANIDES AND ACTINIDES IN ADVANCED FLOWSHEETS
- FISSION PRODUCTS CONVERTED FROM ACIDIC SOLUTION OF NITRATES TO OXIDES PRIOR TO VITRIFICATION

PROJECTED WASTE FORMS

- **I: SORBED, POSSIBLY ON SILVER MORDENITE (HYDRATED ALUMINOSILICATE SIEVE) AND GROUTED**
- **3H: CATALYTICALLY REACTED TO FORM WATER AND INCORPORATED IN POLYMER-IMPREGNATED CEMENT**
- **85Kr: CRYOGENICALY TRAPPED AND CONTAINED IN CYLINDERS**
- **14C: RECOVERED ON MOLECULAR SIEVES AND FIXED AS CaCO3 IN GROUT**
- **Xe: PROBABLY ACCOMPANIES 85Kr (OPTIONAL)**

PROJECTED WASTE FORMS (CONTINUED)

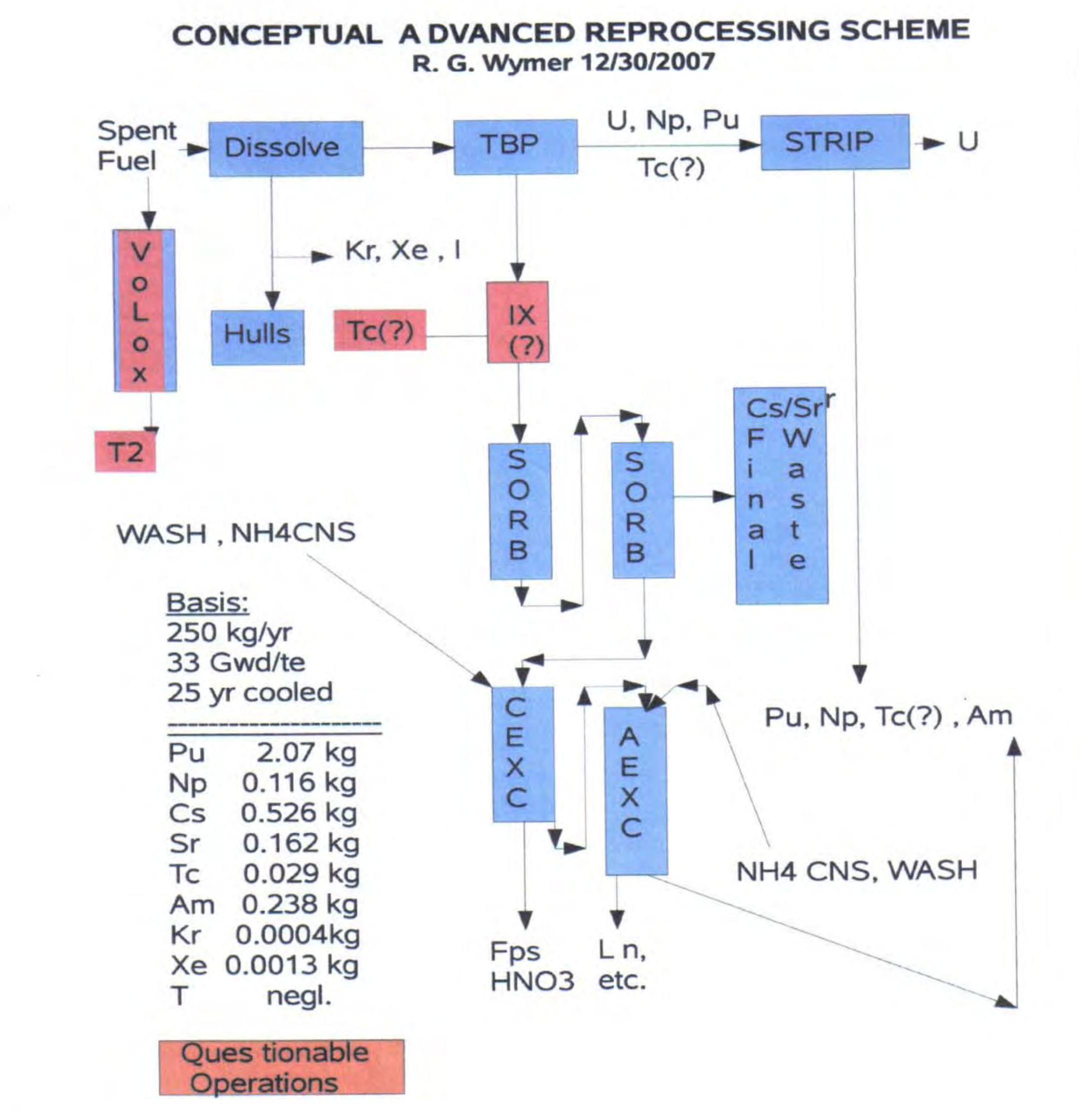
- **99Tc: FOLLOWS SEVERAL PATHS TO SEVERAL WASTE FORMS**
- **Cs/Sr: AN ALUMINOSILICATE**
- **FUEL CLADDING HULLS AND FUEL ELEMENT STRUCTURAL MATERIAL COMPACTED AS METAL**
- **OTHER FISSION PRODUCTS (INCLUDING LANTHANIDES) VITRIFIED AS BOROSILICATE GLASS**

ADVANCED REPROCESSING SCHEMES OTHER THAN THE UREXs ARE POSSIBLE AND MAY BE DESIRABLE

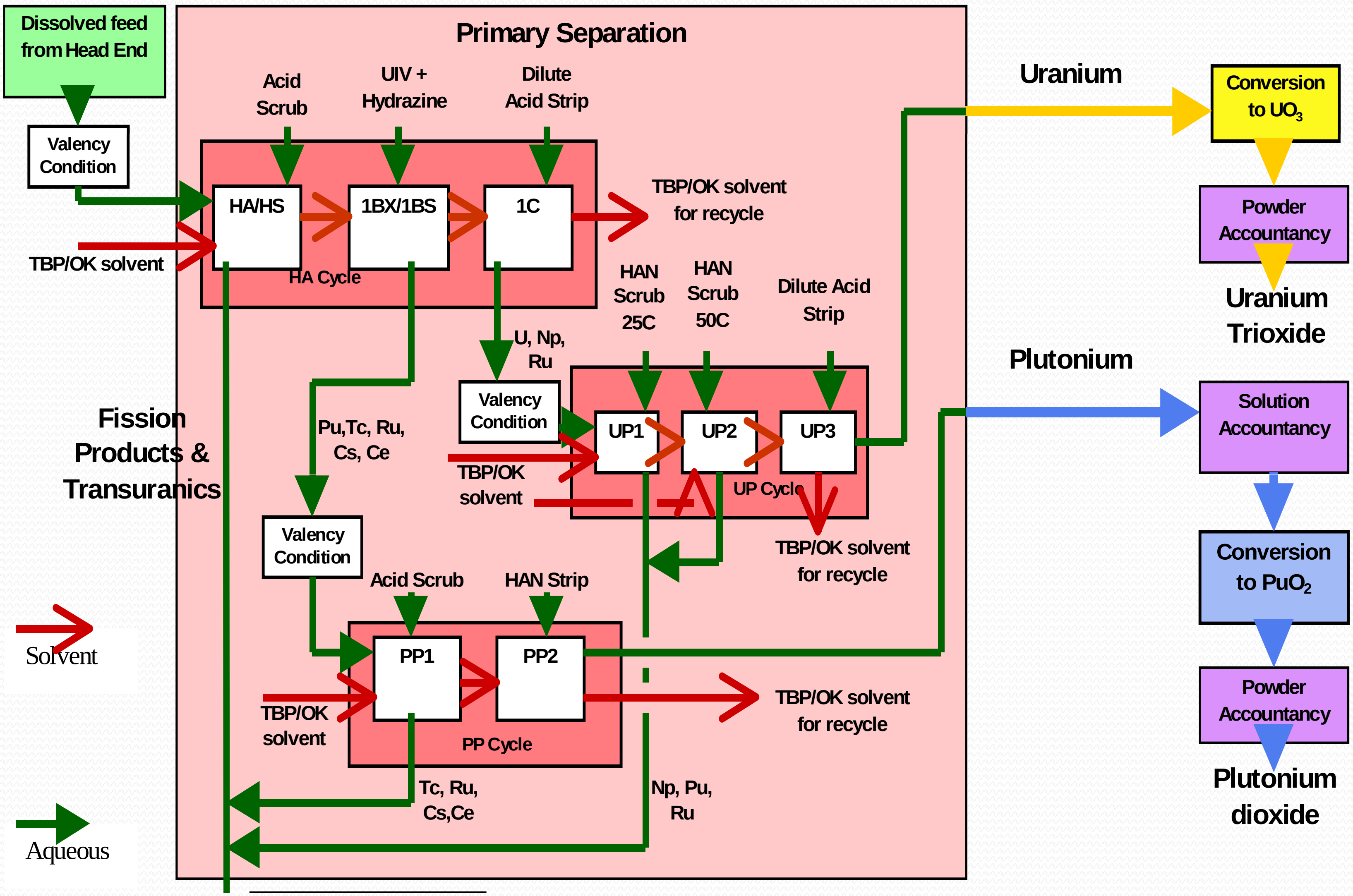
DESIRABLE ATTRIBUTES OF ADVANCED PROCESSES

- **MINIMUM NUMBER OF UNIT OPERATIONS (PROCESS STEPS)**
- **MINIMIZED WASTE PRODUCTION (RADIOACTIVE AND MIXED WASTES)**
- **ISOLATION OF 137Cs AND 90Sr (DESIRABLE FOR YUCCA MOUNTAIN REPOSITORY)**
- **ISOLATION OF ACTINIDES (AT A MINIMUM, SEPARATE Np & Pu ISOTOPES AND 241Am)**
- **CAPTURE 129I (A REQUIREMENT)**
- **ENSURES 99Tc REMOVAL MEETS REPOSITORY LICENSING REQUIREMENTS**

AN EXAMPLE OF A NON-UREX ADVANCED REPROCESSING SCHEME


CONCEPTUAL A DVANCED REPROCESSING SCHEME
R. G. Wymer 12/30/2007
Spent Fuel
Dissolve
TBP
U, Np, Pu
Tc(?)
STRIP
U
V o L o x
Kr, Xe , I
Hulls
Tc(?)
IX (?)
T2
S O R B
S O R B
Cs/Sr
Final Waste
WASH , NH4CNS
Basis:
250 kg/yr
33 Gwd/te
25 yr cooled
Pu 2.07 kg
Np 0.116 kg
Cs 0.526 kg
Sr 0.162 kg
Tc 0.029 kg
Am 0.238 kg
Kr 0.0004kg
Xe 0.0013 kg
T negl.
C E X C
A E X C
Pu, Np, Tc(?) , Am
NH4 CNS, WASH
Fps HNO3
L n, etc.
Ques tionable Operations

THORP REPROCESSING FLOWSHEET
A MODERN-DAY PROCESS



ADVANCED REPROCESSING SCHEME POTENTIAL ADVANTAGES

- REDUCES PROCESS COMPLEXITY
  - ELIMINATES MULTIPLE SX STEPS
- ELIMINATES SEVERAL WASTE STREAMS
- SEPARATES Cs/Sr DIRECTLY ONTO A POTENTIAL WASTE FORM
- CLEANLY AND EASILY SEPARATES LANTHANIDES FROM ACTINIDES
- USES ESTABLISHED UNIT OPERATIONS, e.g., ION EXCHANGE AND INORGANIC SORBENTS

ADVANCED REPROCESSING SCHEME POTENTIAL DISADVANTAGES

- **REQUIRES INCREASED UNDERSTANDING OF THIOCYANATE CHEMISTRY**
- **RADIATION DAMAGE TO THIOCYANATE ION IN THIS SYSTEM IS UNKNOWN**
- **EMPLOYS ORGANIC ION EXCHANGERS (IX) SUBJECT TO RADIATION DAMAGE AND REPLACEMENT AFTER MULTIPLE CYCLES AS WASTE**
- **REQUIRES PERIODIC REPLACEMENT OF Cs/Sr INORGANIC SORBENT COLUMN CONTENTS**

THE END

CURIES AND MASSES OF RADIONUCLIDES

- **BASIS OF VALUES LISTED:**
  - **33 Gwd/te BURNUP**
  - **25-YEARS COOLED**
  - **CURIES AND MASSES PRESENT IN ONE METRIC TONNE OF INITIAL HEAVY METAL (MTIHM)**
  - **3.2% 235U INITIAL URANIUM ENRICHMENT**

VALUES IN RED MERIT SPECIAL ATTENTION

CESIUM AND STRONTIUM

- **Cs**
  - **133Cs: STABLE; 1.132E+03 g**
  - **134Cs: 3.390E+01 Ci; 2.600E-02 g**
  - **135Cs: 3.500E-01 Ci; 3.010E+02 g**
  - **137Cs: 5.842E+04 Ci; 6.710E+02 g**
- **Sr**
  - **86Sr: STABLE; 4.000E-01 g**
  - **88Sr: STABLE; 2.500E+02 g**
  - **90Sr: 4.012E+04 Ci; 2.940E+02 g**

FISSION PRODUCTS INCLUDING LANTHANIDES

- **1.981E+05 Ci**
- **3.421E+04 g**

ACTINIDE ELEMENTS

- **U**
  - **232U: 2.770E-02 Ci; 1.300E-03 g**
  - **233U: 4.850E-05 Ci; 5.000E-03 g**
  - **234U: 1.290E+00 Ci; 2.060E+02 g**
  - **235U: 1.730E-02 Ci; 7.975E+03 g**
  - **236U: 2.570E-01 Ci; 3.965E+03 g**
  - **237U: 9.300E-01 Ci; 1.130E-05 g**
  - **238U: 3.180E-01 Ci; 9.441E+05 g**
- **Np**
  - **236Np: 3.300E-01 Ci; 4.630E+02 g**
  - **237Np: 3.200E-02 Ci; 4.120E-04 g**
  - **239Np: 1.700E-07 Ci; 1.700E+01 g**

ACTINIDE ELEMENTS (CONTINUED)

- Pu
  - 236Pu: 1.400E-03 Ci; 2.690E-06 g
  - 238Pu: 2.074E+03 Ci; 1.211E+04 g
  - 239Pu: 3.110E+02 Ci; 5.030E+03 g
  - 240Pu: 5.280E+02 Ci; 2.316E+03 g
  - 241Pu: 3.769E+04 Ci; 3.660E+02 g
  - 242Pu: 1.720E+00 Ci; 4.510E+02 g
  - 244Pu: 4.220E-07 Ci; 2.400E-02 g

ACTINIDE ELEMENTS (CONTINUED)

- **Am**
  - **241Am: 2.966E+03 Ci; 8.640E+02 g**
  - **242Am: 6.440E+00 Ci; 7.960E-06 g**
  - **242Am: 6.470E+00 Ci; 6.700E-01 g**
  - **243Am: 1.700E+01 Ci; 8.550E+01 g**
- **Cm**
  - **242Cm: 5.330E+00 Ci; 1.600E-03 g**
  - **243Cm: 1.150E+01 Ci; 2.200E-01 g**
  - **244Cm: 7.430E+02 Ci; 8.500E-01 g**
  - **245Cm: 1.500E+02 Ci; 1.500E-03 g**
  - **246Cm: 3.100E-03 Ci; 1.000E-01 g**

HALF LIVES OF RADIONUCLIDES IMPORTANT IN WASTE MANAGEMENT

RADIONUCLIDE	HALF LIFE, YRS	RADIONUCLIDE	HALF LIFE, YRS
3H	1.23E+01	236U	2.34E+07
14C	5.73E+03	238U	4.47E+09
85Kr	1.08E+01	237Np	2.44E+06
90Sr	2.88E+01	238Pu	8.77E+01
99Tc	2.11E+05	239Pu	2.41E+04
129I	1.07E+07	240Pu	6.56E+03
135Cs	2.30E+06	241Pu	1.44E+01
137Cs	3.01E+01	241Am	4.32E+02
235U	7.04E+08	244Cm	1.81E+01