Further Assessments of the Attractiveness of Materials in Advanced Nuclear Fuel Cycles from a Safeguards Perspective

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Outline

- Statement of Problem
- Figure of Merit (FOM)
- Correspondence of FOM with Attractiveness Levels
- Results:
  - UREX,
  - COEX,
  - ^{238}Pu Spiking,
  - PYROX,
  - THOREX.
- Conclusions

Statement of Problem

- Initial Charge: Compare SNM Attractiveness Levels for COEX and UREX+1A reprocessing schemes
- Refinement: Analyze SNM attractiveness for other reprocessing schemes, including:
  - DUPIC,
  - DIAMEX-SANEX,
  - GANEX,
  - FLOREX,
  - others.

Comparison will use the following Figure of Merit (FOM).

- The FOM is applicable to an adversary intending to build a stockpile of nuclear weapons without purifying the materials:

$$FOM = 1 - \log\left(M\left[\frac{1}{800} + \frac{h}{4500}\right] + \left[\frac{D}{500}\right]^{\frac{1}{\log 2}} \right)$$

  - M – bare critical mass in unpurified metal form (kg)
  - h – heat content in unpurified metal form (W/kg)
  - D – dose rate of 0.2•m @ 1 m (rad/h)
- The FOM only addresses the attractiveness of the material to make a stable threshold nuclear device capable of being stockpiled.
- There are several factors that determine proliferation risk. This study addresses merely one factor.
- The FOM should be useful for informed safeguards discussions and decisions for GNEP.

FOM and Attractiveness Levels† of DOE Nuclear Materials‡ are similar but not equivalent

FOM for metals	Utility	Designation On Plots
> 2	High	H
1-2	Moderate	M
0-1	Low	L
< 0	Off Scale	O

Attractiveness Level	
B	PURE PRODUCTS
C	HIGH-GRADE MATERIALS
D	LOW-GRADE MATERIALS
E	ALL OTHER MATERIALS

- Desirable FOM designations are L and O.
- Undesirable FOM designations are H and M.

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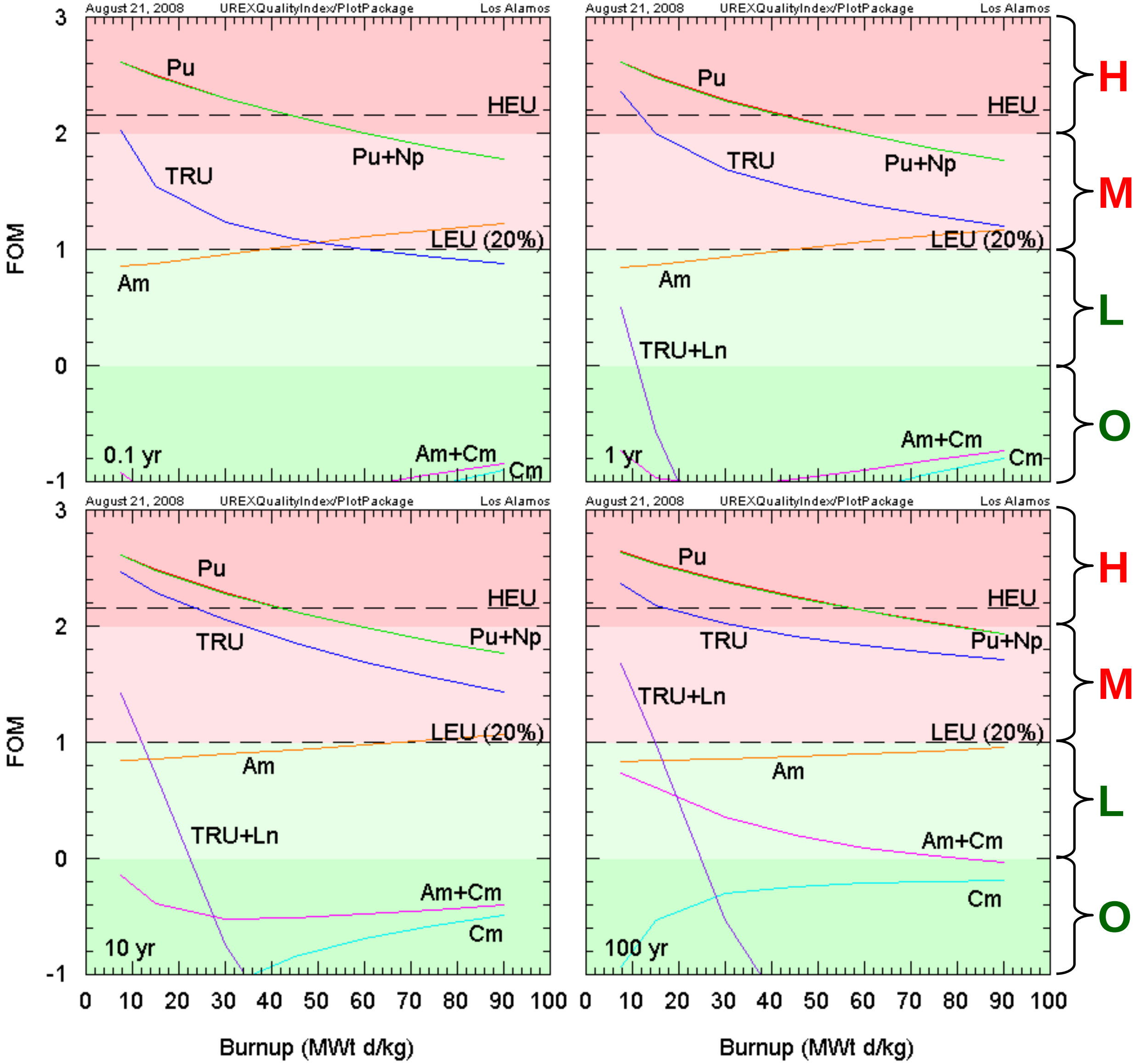
† "Nuclear Material Control and Accountability," U. S. Department of Energy manual DOE M 470.4-6 Chg 1 (August 14, 2006).

‡ Depleted, Enriched, and Normal Uranium; ^{233}U; ^{238}Pu; ^{239}Pu; ^{240}Pu; ^{241}Pu; ^{242}Pu; ^{241}Am; ^{243}Am; Bk; ^{252}Cf; Cm; ^{2}H; Enriched Lithium; ^{237}Np; Th; ^{3}H; and Uranium in Cascades.

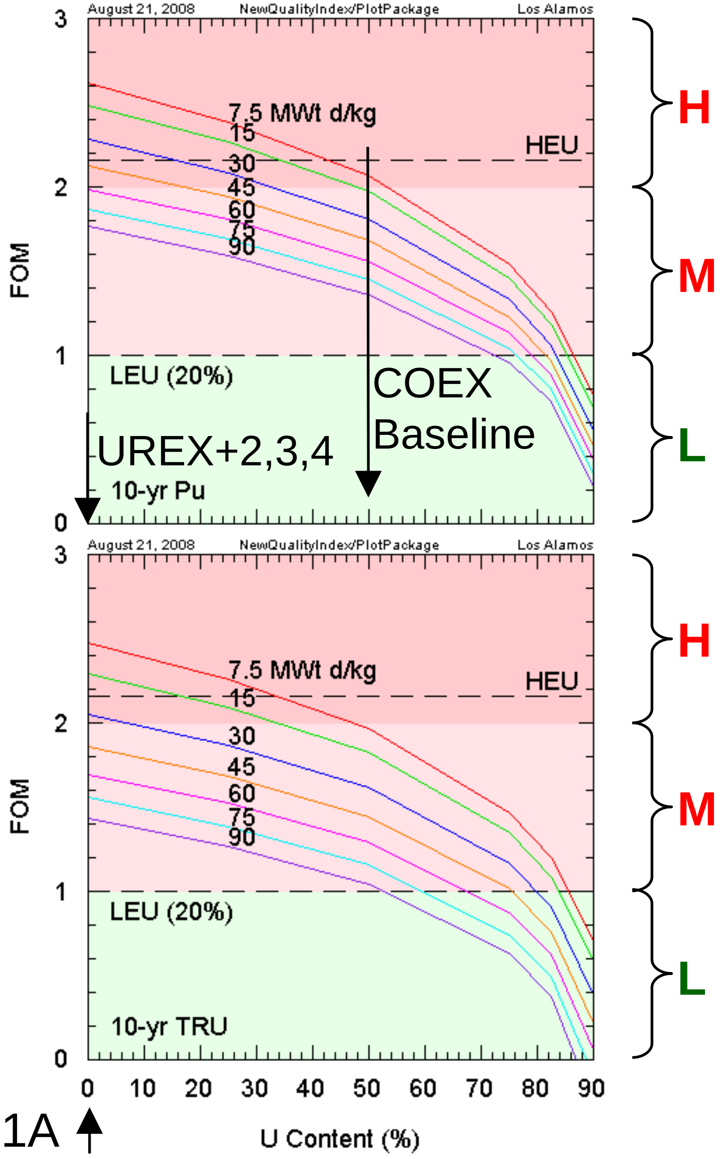
Pu and Pu+Np have the highest FOM of the UREX products.

- Co-extracting lanthanides with TRU can reduce the FOM by a whole letter grade relative to TRU.
- Co-extracting Np with Pu does not reduce the FOM relative to Pu.
- The FOM of TRU, Am+Cm, and Cm increases with time.
- It would be better to co-extract Am and Cm than to extract Am and Cm separately.



Diluting reprocessed Pu or TRU metal with U can reduce the FOM to L.

- For 10-yr, 45-MWt·d/kg Pu, an FOM of L requires ≥82% U.
- For 10-yr, 45-MWt·d/kg TRU, an FOM of L requires ≥75% U.
- DU or NU are as effective as the irradiated U used herein.



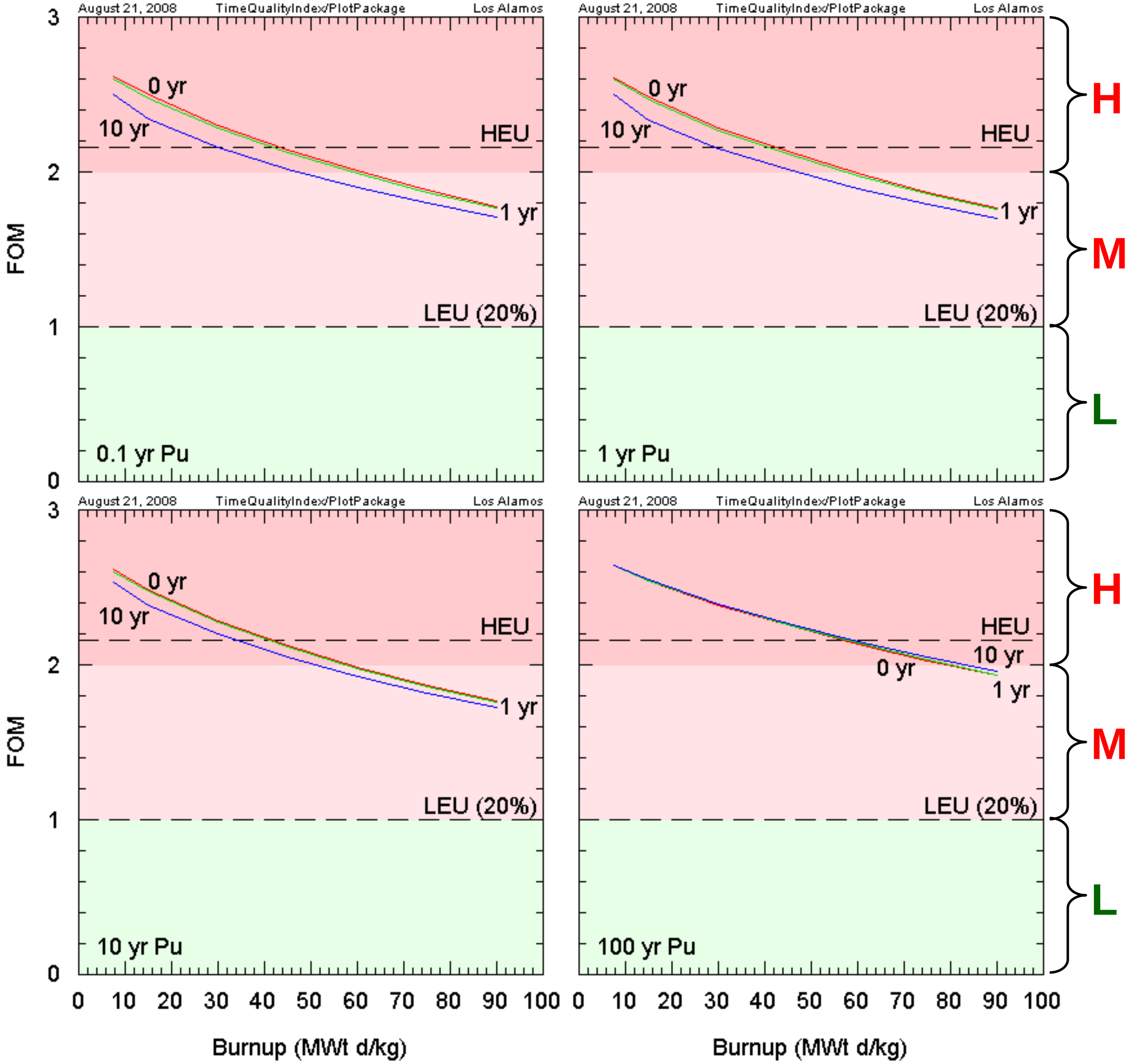
COEX Baseline

UREX+2,3,4

UREX+1A

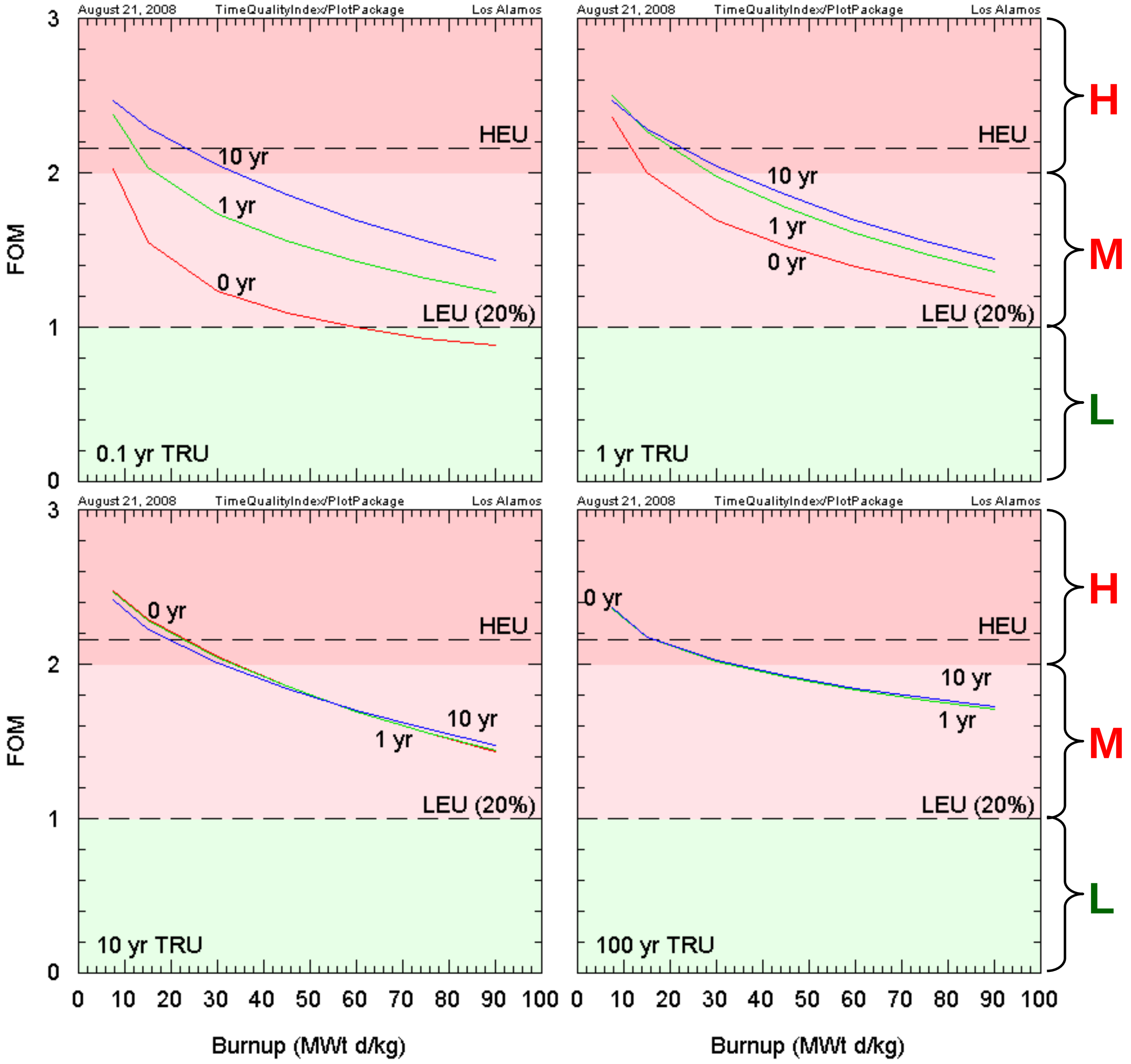
Generally, the FOM of separated Pu decreases slightly with time.

- The older the Pu when reprocessed, the smaller the decrease in the FOM with increasing post-separation age.
- For ≤10-yr Pu, the $^{241}\text{Pu}\rightarrow{}^{241}\text{Am}$ decay increases heating rate, whereas for old (≥ 100 yr) Pu, the $^{238}\text{Pu}\rightarrow{}^{234}\text{U}$ decay decreases heating rate, thereby decreasing or slightly increasing the FOM, respectively.



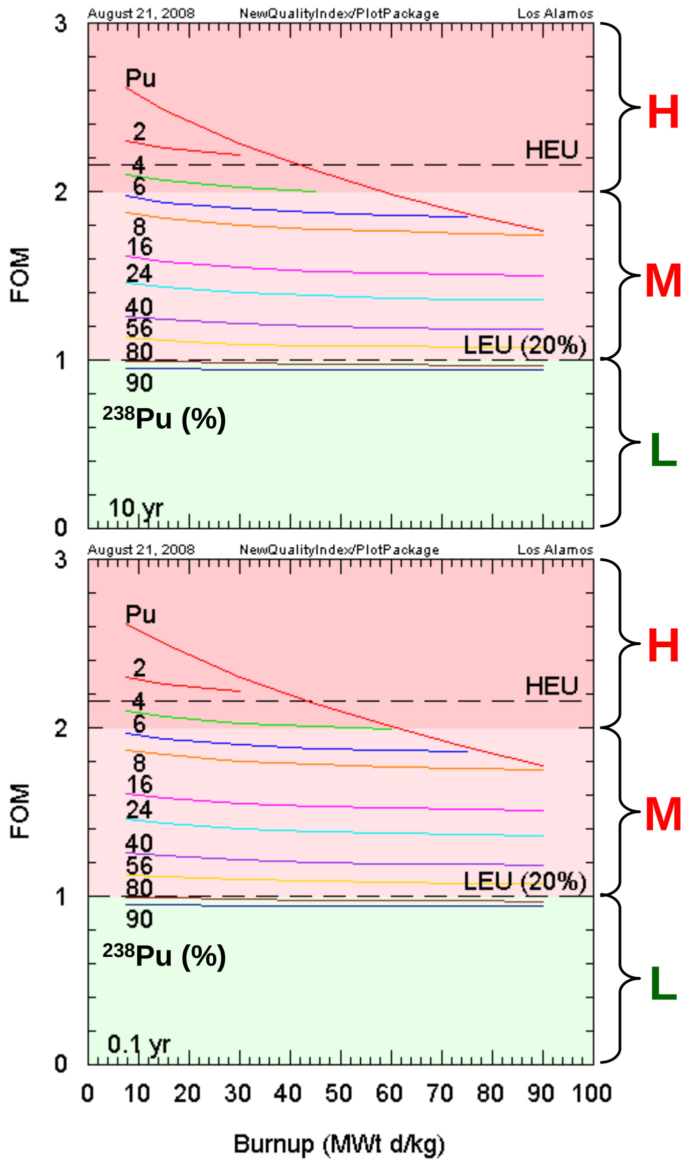
The FOM of separated TRU increases with time.

- The older the TRU at the time of separation, the smaller the decrease in the FOM with increasing post-separation age.
- For old (≥ 100 yr) Pu, the FOM increases slightly with increasing post-separation age.
- Increasing FOM for early separation times is the result of the loss of ^{242}Cm and ^{244}Cm.



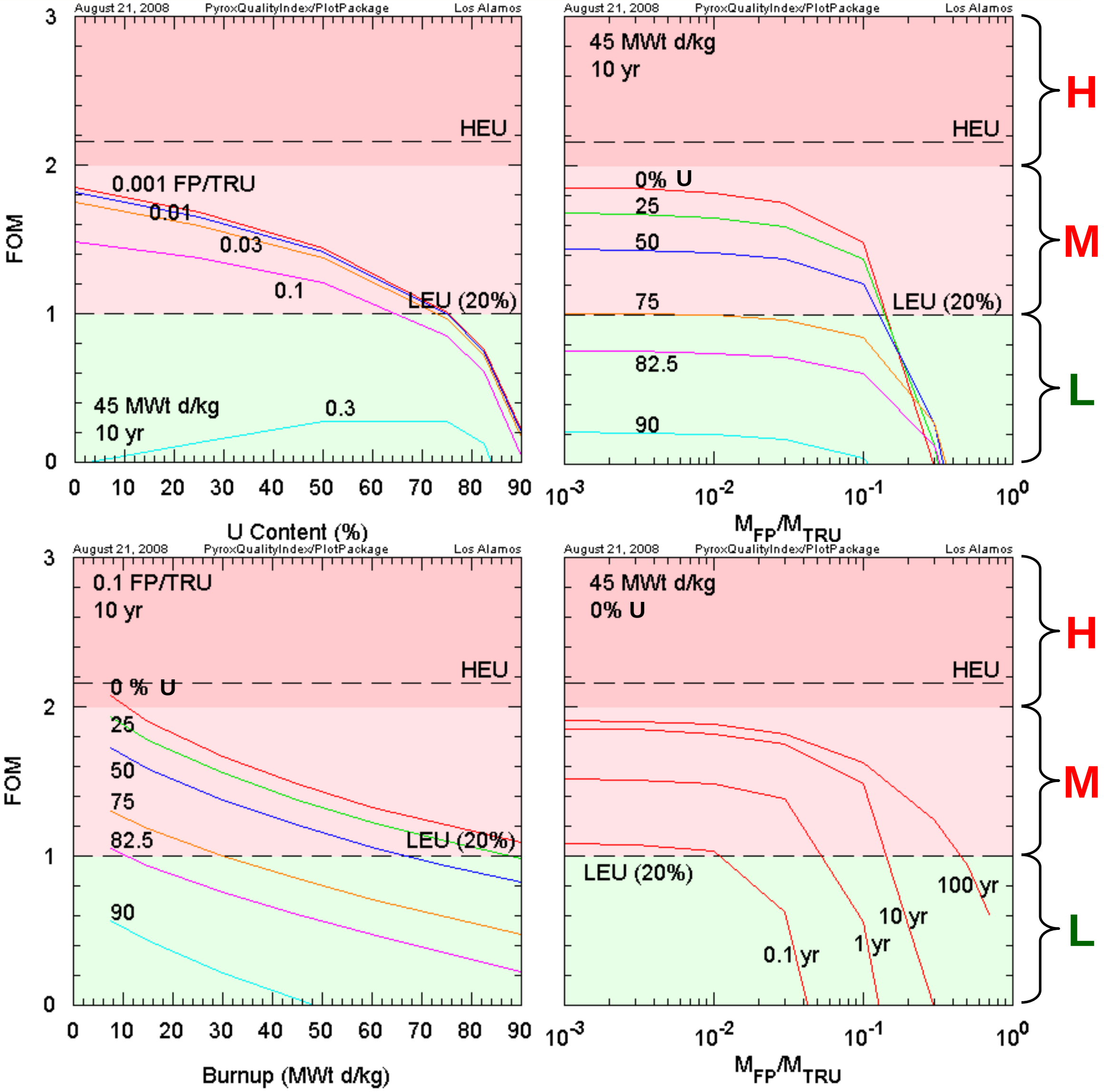
Spiking requires large amounts of ^{238}Pu to reduce the FOM to L.

- Reprocessed Pu is an end product of:
  - PUREX
  - COEX
  - UREX+2,3, and 4.
- The FOM of Pu can be reduced with:
  - higher burn-up,
  - dilution with ^{238}Pu.



Like TRU, the FOM of PYROX product increases with age and decreases with burn-up.

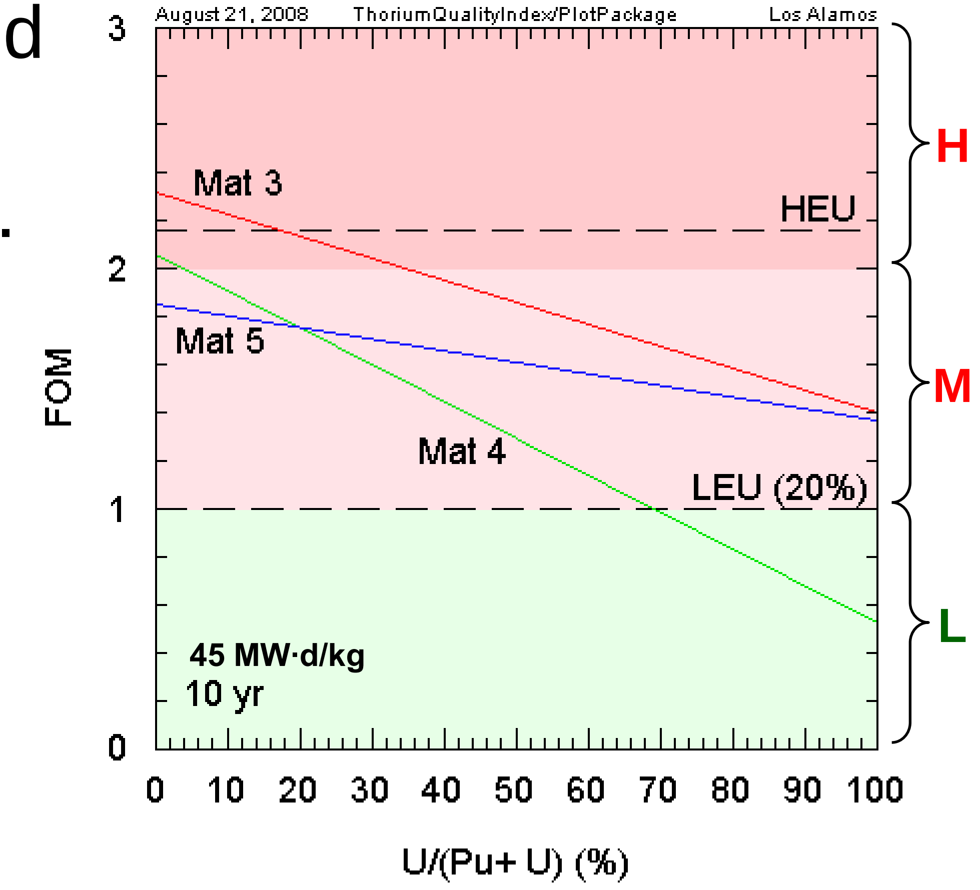
- The PYROX product can also be diluted with uranium to reduce the FOM.
- Increasing the relative fission product mass also reduces the FOM of the PYROX product.



For Th fuel, the THOREX products with the highest FOM are Pu and U.

- The FOM for Th fuels burned to 45 MW·d/kg is independent of cooling time.

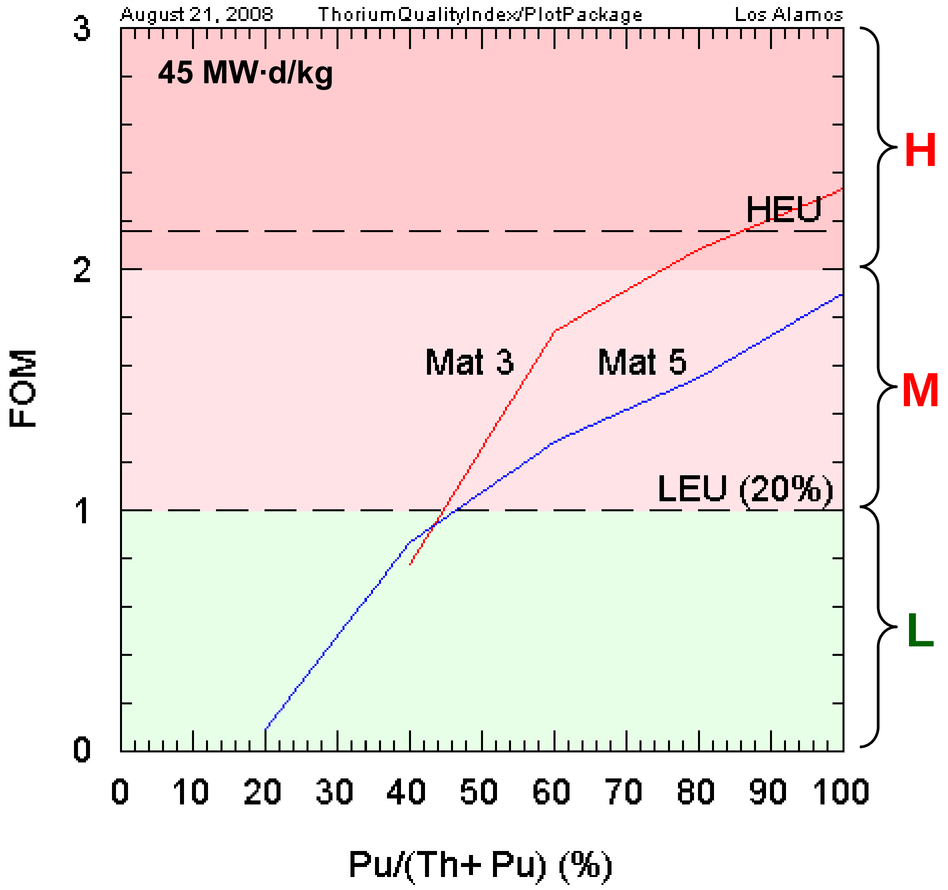
- Initial fuel mixes are:
  - Mat 3: 6.25-5-88.75 % Pu(94% ^{239}Pu)-U-Th.
  - Mat 4: 30.5-69.5 % U(19.9% ^{235}U)-Th
  - Mat 5: 10-5-85 % Pu(53% ^{239}Pu)-U-Th.



- The amount of Pu in spent Th fuel is lower than in spent U fuel.

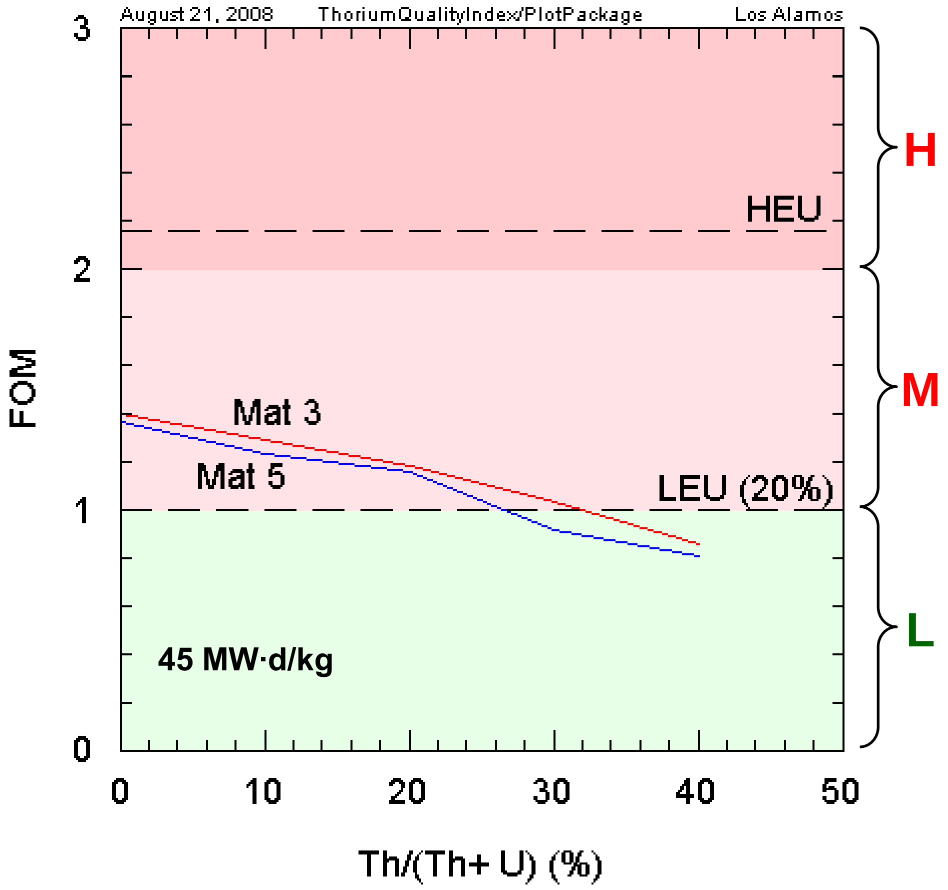
A Thorium fraction of >2/3 is required to reduce the FOM to L for THOREX Pu.

- The reprocessed Pu+Th mixture requires > 55% Th to reduce the FOM to a low utility (L).



A Thorium fraction of > ⅓ is required to reduce the FOM to L for THOREX U.

- The FOMs for U + Th mixtures for Mat 3 and 5 are nearly equivalent.



UREX Conclusions

- The FOM of UREX+1A material (TRU) is dependent upon age and burn-up.
- 10-yr, 45-MWt·d/kg UREX+1A material (TRU) requires a U content > 75% to reduce the FOM to L.
- The FOM of UREX+1A material (TRU) is sensitive to the post-irradiation decay time, and should be reprocessed as soon as practical.

COEX (and UREX+2,3,4) Conclusions

- Pu+Np has the same FOM as Pu:
  - Adding Np to Pu does not reduce the FOM of COEX material. This conclusion applies equally to UREX+2, UREX+3, and UREX+4.
  - Extracting just Pu puts Np into waste stream.
- A U content of ≥82% is required to reduce the COEX FOM to L.
- The FOM of Pu is not significantly affected by the post-irradiation decay time.

^{238}Pu-Spiking Conclusions

- The FOM of Pu with ^{238}Pu content < 80% is still at least M.
- Based on the FOM formula used in this study, there is not enough ^{238}Pu (nor Np for breeding ^{238}Pu) to reduce the FOM to L.

THOREX Conclusions

- The Thorium fuel cycle produces two potentially attractive materials: ^{239}Pu and ^{233}U.
- The Pu is of greater concern from a safeguards perspective.
- The Pu product can be rendered unattractive by retaining >2/3 Th fraction with it during/after reprocessing.
- The U product can be rendered unattractive by adding natural or depleted U to the fuel before irradiation, but may exacerbate the ^{239}Pu problem in the product.
- The U product can be rendered unattractive by retaining >1/3 fraction Th with it during/after reprocessing.

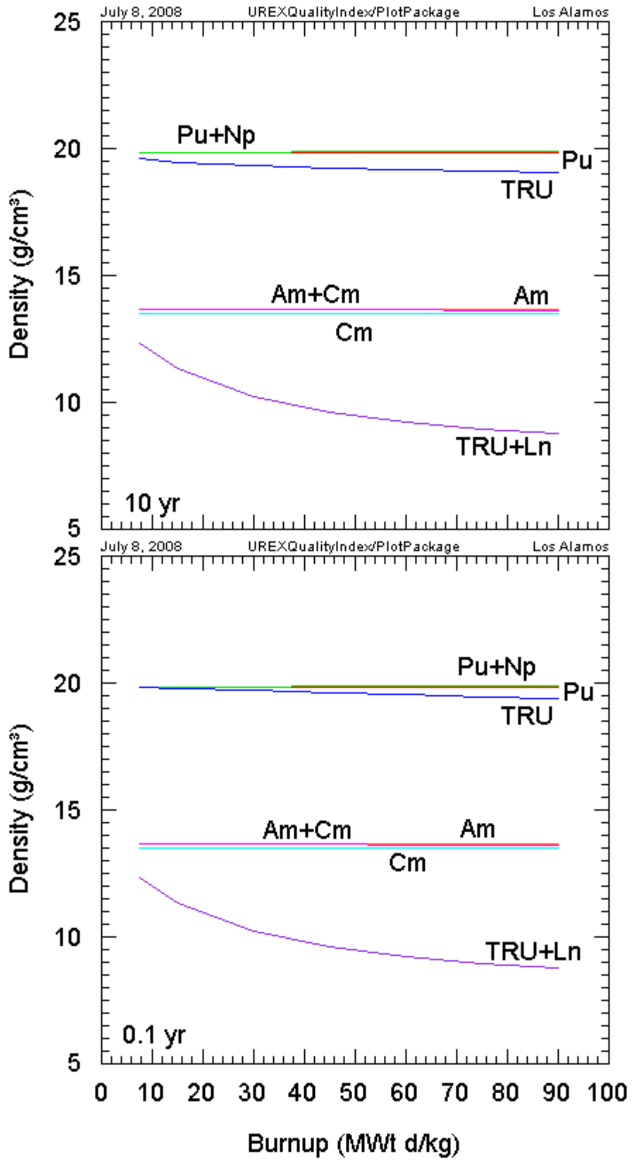
Generic Conclusions

- There is a safeguards and security benefit with respect to safeguards to diluting the reprocessing end products with:
    - Ln
    - U – reprocessed, natural, or depleted
- However – There is no silver bullet to solve the safeguards and security issue. None of the proposed flow-sheets examined to date justify reducing international safeguards or physical security protection levels. All reprocessing products evaluated need to be rigorously safeguarded and provided the highest levels of physical protection.

Background Slides

Many factors contribute to the behavior of the Quality Index of UREX forms.

- Generally, density is insensitive to time for most UREX products, except TRU.
- As spent fuel ages or as the burn-up increases, the composition of TRU shifts to higher-Z elements, which are less dense.

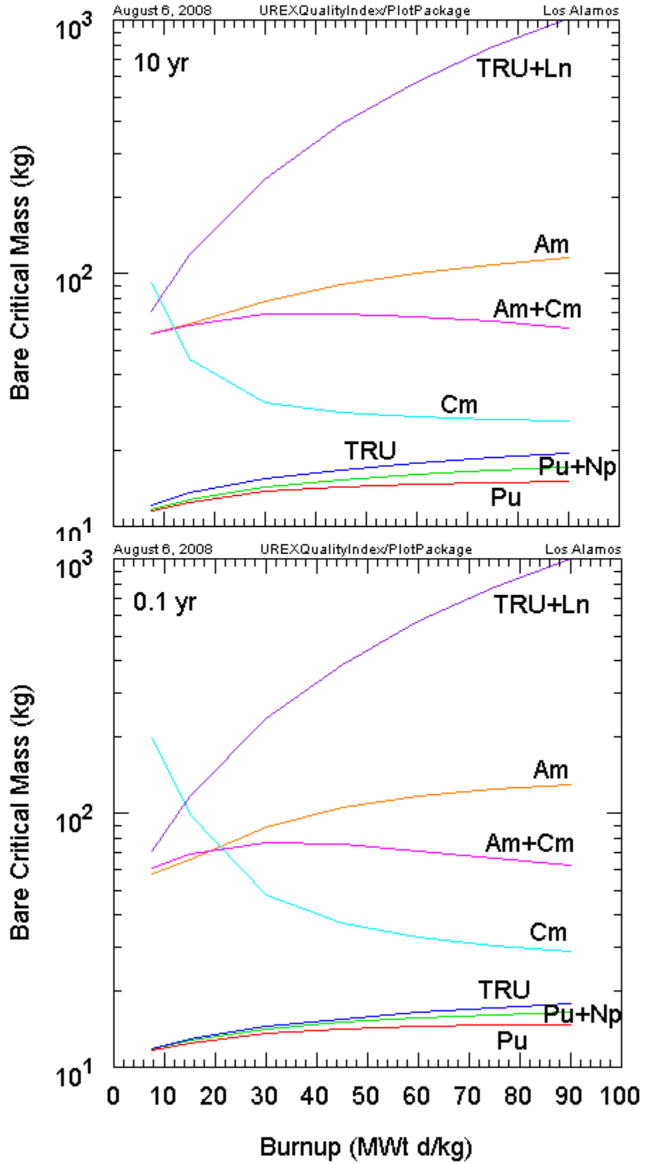


Many factors contribute to the behavior of the Quality Index of UREX forms (cont’d-1).

- Bare critical mass of Pu containing products is determined by ^{239}Pu + ^{241}Pu weight fraction:

Age (yr)	0.1		10	
Burnup (MWt·d/kg)	7.5	90	7.5	90
^{239}Pu	78	42	79	45
^{239}Pu + ^{241}Pu	84	57	84	55

- For TRU+Ln, the TRU weight fraction decreases with increasing burn-up from 68% at 7.5 MWt·d/kg to 34% at 90 MWt·d/kg at 10 yr.



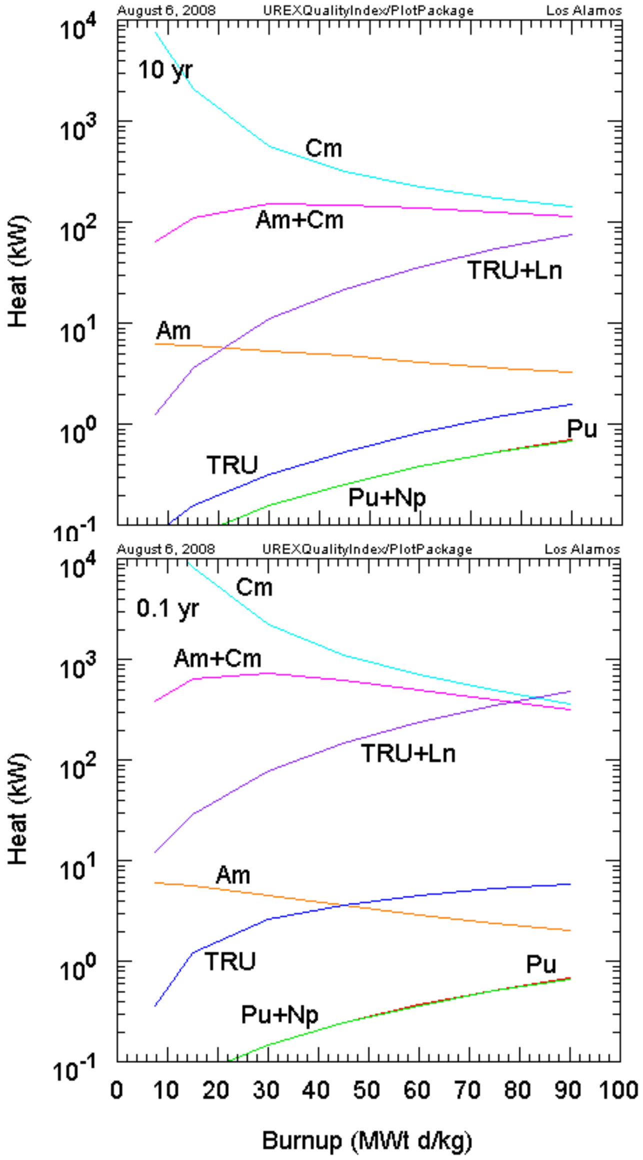
Many factors contribute to the behavior of the Quality Index of UREX forms (cont’d-2).

- Heating is not significant for Pu nor Pu+Np.
- Am:

Age (yr)	10	
Burnup (MWt·d/kg)	7.5	90
^{241}Am	94	20
^{243}Am	5	80

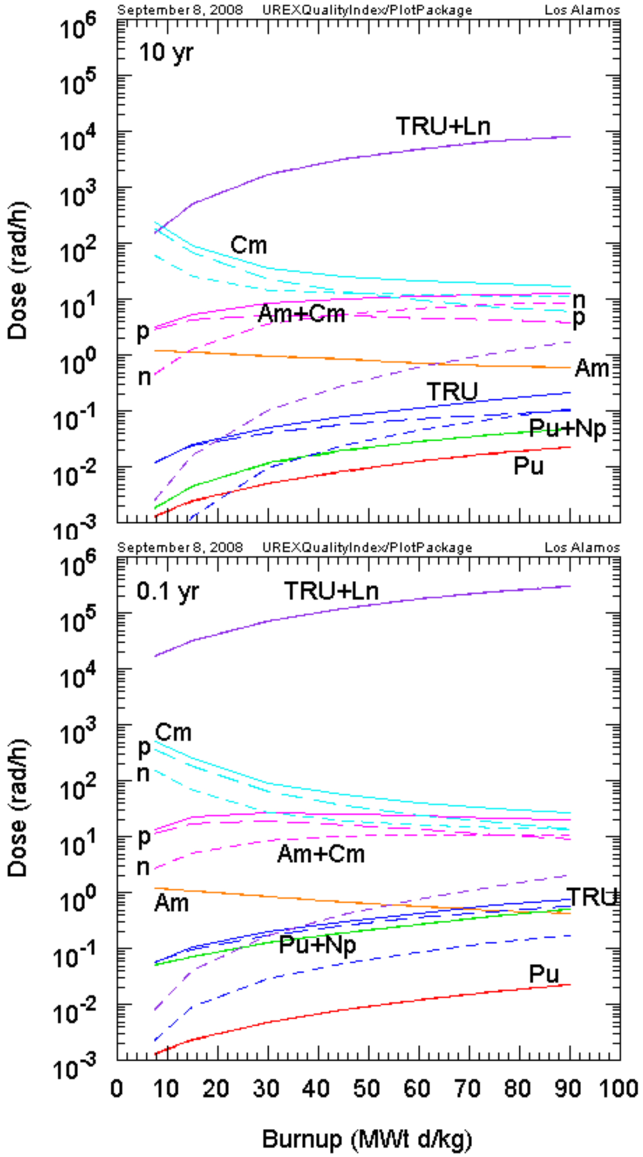
- Cm:

Age (yr)	10	
Burnup (MWt·d/kg)	7.5	90
^{242}Cm	68	2
^{244}Cm	29	91
^{245}Cm	0.4	5



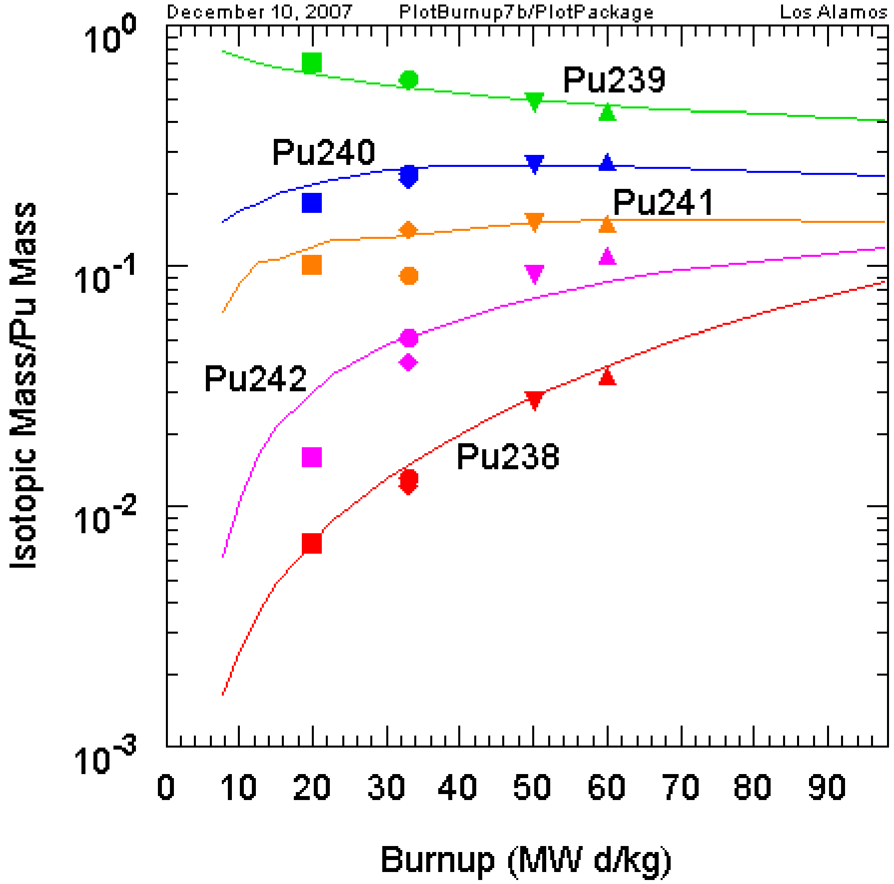
Many factors contribute to the behavior of the Quality Index of UREX forms (cont'd-3).

- The "self-protection" afforded by the Ln dose in TRU+Ln quickly dissipates.
- ^{242}Cm and ^{244}Cm are significant sources of dose.
- Dose type:
  - Total dose – solid
  - Photon dose – long dash
  - Neutron dose – short dash

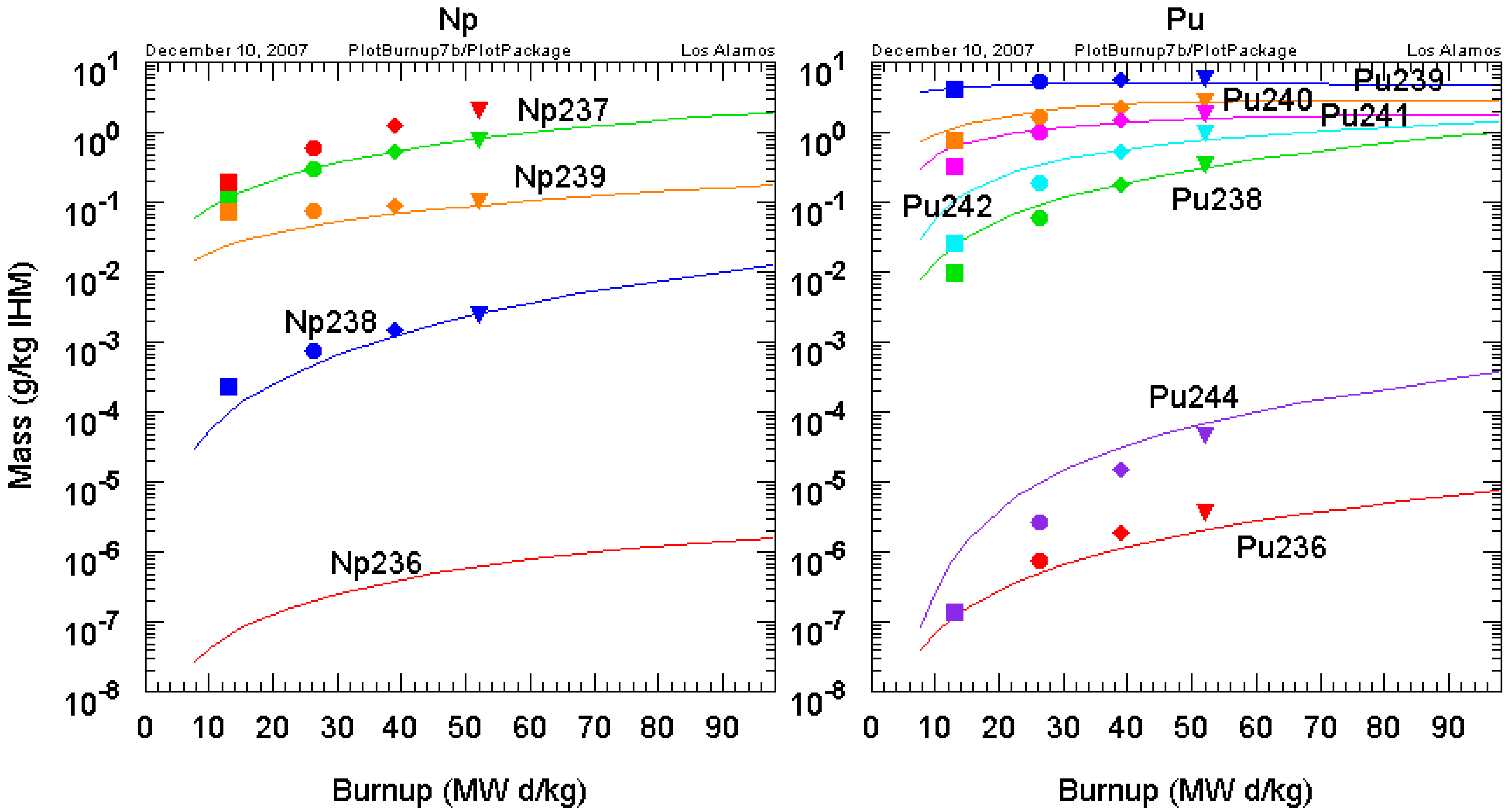


Calculated Pu isotopic composition at EOB compared to data from Pellaud.

- Data Points:
  - B. Pellaud, “Proliferation Aspects of Plutonium Recycling,” J. Nuc. Mat. Management XXXI, 30 (2002).
  - Initially, 4% ^{235}U/U.
- Calculations:
  - 4% enrichment is used only for 45 MW d/kg



Np and Pu isotopic composition at EOB compared to Neeb's data (4% enrich.).



Isotope Data

Isotope	$T_{1/2}$	Decay Mode	n/s/g	p/s/g	$\sigma_f^\dagger$	$\sigma_\gamma^\dagger$	W/g
^{232}U	68.9 y	α	1.300E+00	9.958E+10	77 b	74.9 b	7.08E-01
^{233}U	1.592E+5 y	α	8.600E-04	2.265E+07	531.2 b	45.3 b	2.81E-04
^{234}U	2.455E+5 y	α	5.020E-03	2.292E+07	6.2 mb	99.8 b	1.79E-04
^{235}U	7.04E+8 y	α	2.990E-04	1.013E+05	584.4 b	98.8 b	5.99E-08
^{236}U	2.342E7 y	α	5.490E-03	2.393E+05	61.3 mb	5.30 b	1.75E-06
^{238}U	4.468E9 y	α	1.360E-02	1.004E+03	11.8 μb	2.72 b	8.51E-09
^{236}Np	1.53E+4 y	ε		1.191E+09	2770 b	701.0 b	2.69E-05
^{237}Np	2.144E+6 y	α	1.140E-04	2.463E+07	22.49 mb	165 b	2.01E-05

† at 0.0253 eV

References:

http://atom.kaeri.re.kr/

http://www.nndc.bnl.gov/chart

http://www.nndc.bnl.gov/wallet/wccurrent.html

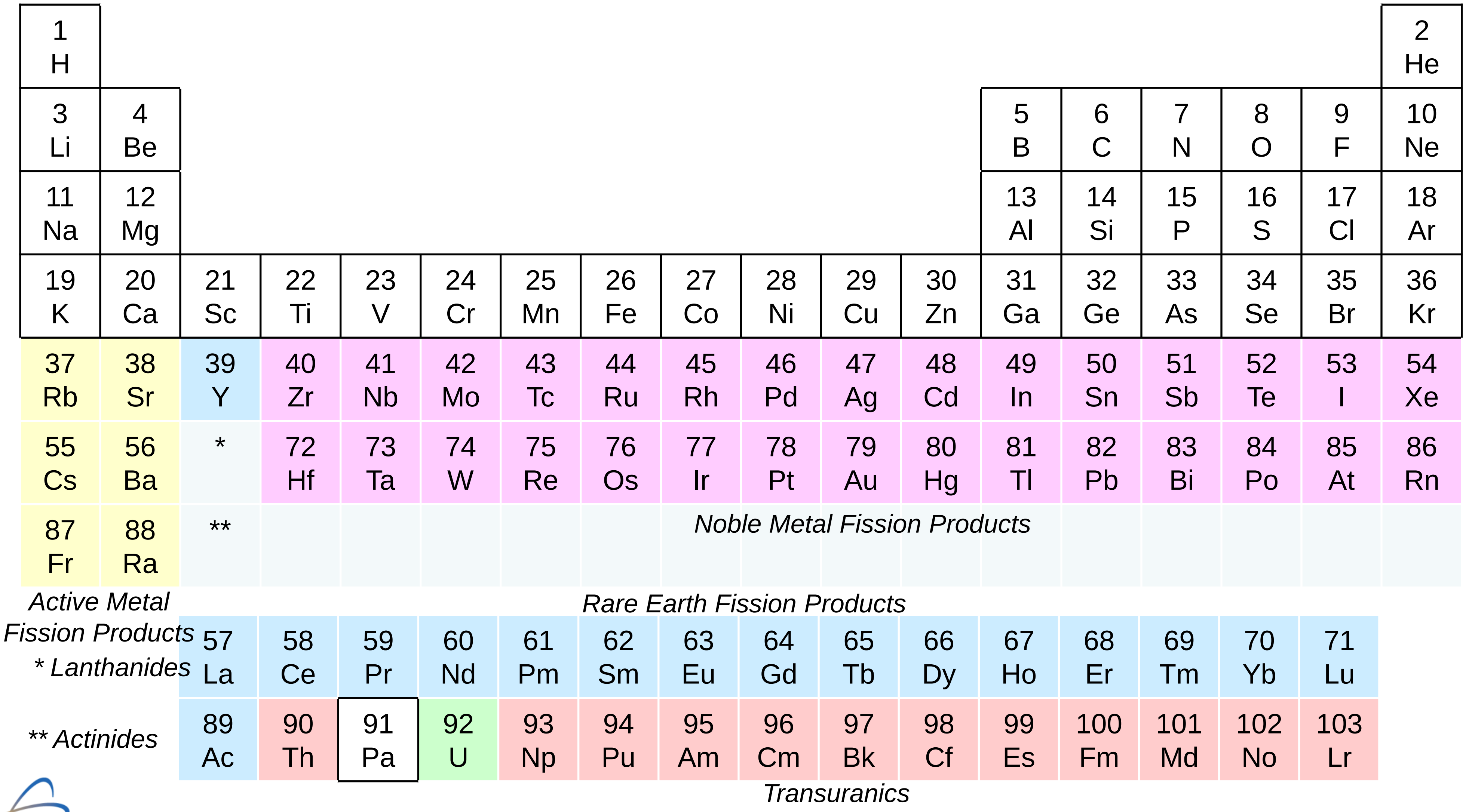
Isotope Data (cont'd-1)

Isotope	$T_{½}$	Decay Mode	n/s/g	p/s/g	σ_f	σ_γ	W/g
^{236}Pu	2.858 y	α	3.492E+04	2.586E+12	169.4 b	145.4 b	1.82E+01
^{238}Pu	87.7 y	α	2.590E+03	7.349E+10	17.9 b	540.3 b	5.68E-01
^{239}Pu	24110 y	α	2.180E-02	1.076E+08	747.4 b	270.3 b	1.93E-03
^{240}Pu	6561 y	α	1.020E+03	8.490E+08	58.8 mb	289.4 b	7.07E-03
^{241}Pu	14.290 y	β^-	5.000E-02	1.076E+08	1012 b	361.5 b	3.28E-03
^{242}Pu	3.75E+5 y	α	1.720E+03	1.255E+07	2.6 mb	18.8 b	1.17E-04

Isotope Data (cont'd-2)

Isotope	$T_{1/2}$	Decay Mode	n/s/g	p/s/g	σ_f	σ_γ	W/g
^{241}Am	432.6 y	α	1.180E+00	9.723E+10	3.02 b	600.4 b	1.14E-01
^{242M}Am	141 y	ITβ^-	1.424E+02	1.190E+11	7000 b	2000 b	4.23E-03
^{243}Am	7370 y	α	3.345E+00	7.196E+09	116.1 mb	78.50 b	6.43E-03
^{242}Cm	162.8 d	α	2.100E+07	1.401E+13	5.064 b	15.90 b	1.21E+02
^{243}Cm	29.1 y	α	2.665E+02†	2.402E+12	617.4 b	130.2 b	1.89E+00
^{244}Cm	18.1 y	α	1.080E+07	3.003E+11	1.037 b	15.10 b	2.83E+00
^{245}Cm	8500 y	α	1.113E+02†	8.364E+09	2001 b	346.4 b	5.71E-03
^{246}Cm	4760 y	α	8.897E+06	1.049E+09	140.1 mb	1.291 b	9.97E-03
^{247}Cm	1.56E+7 y	α		2.831E+06	81.79 b	57.20 b	2.87E-06
^{248}Cm	3.48E+5 y	α	4.349E+07	1.149E+07	370.0 mb	2.570 b	1.19E-04

In PYROX the concentrations of Active Metal and Rare Earth Fission Products, Nobel Metal Fission Products, Uranium, and the Transuranics are manipulated separately.



1 H																	2 He
3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
55 Cs	56 Ba	*	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
87 Fr	88 Ra	**															
* Lanthanides		57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu	
** Actinides		89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr	

Noble Metal Fission Products

Active Metal Fission Products

Rare Earth Fission Products

Transuranics