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Fuel cycle aspects of MSR

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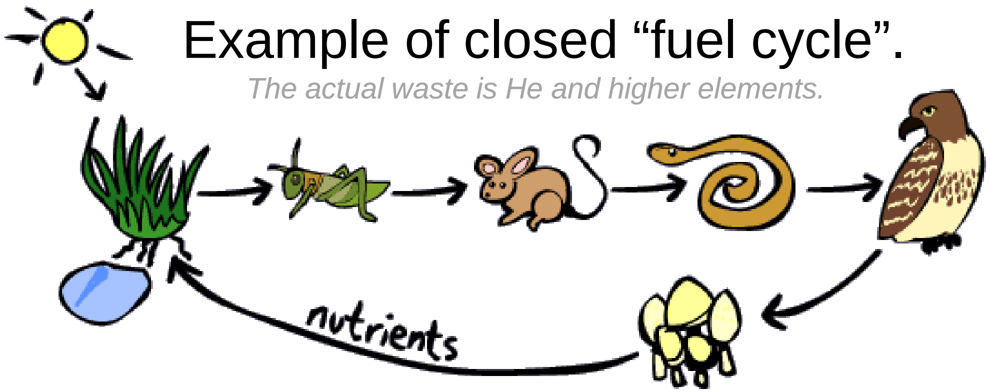
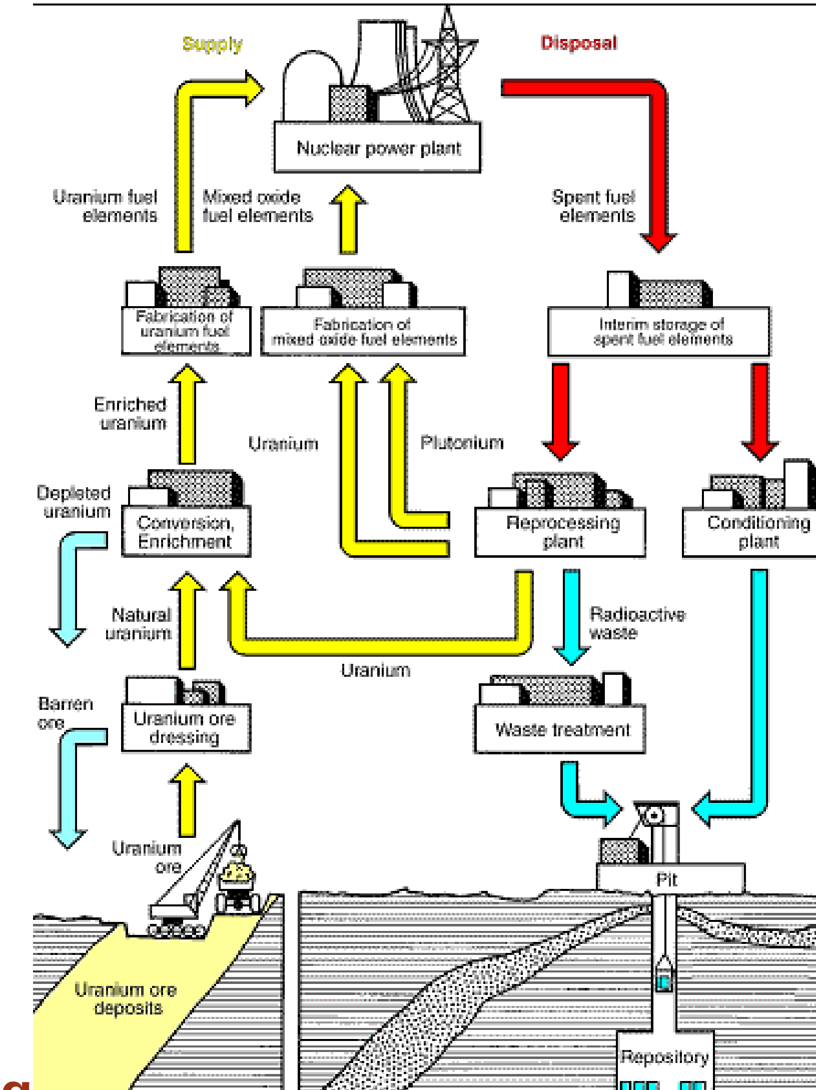
What is fuel cycle? – process chain to obtain energy

Example of closed “fuel cycle”.

The actual waste is He and higher elements.





https://www.euronuclear.org/info/encyclopedia/n/nuclear-fuel-cycle.htm

Nuclear Fuel cycle

Front end:	Back end:
o Exploration	o Interim storage
o Mining	o Transportation
o Milling	o Reprocessing
o Conversion	o Partitioning
o Enrichment	o Transmutation
o Fabrication	o Waste disposal

This presentation covers the reactor physics aspects of irradiation and recycling.

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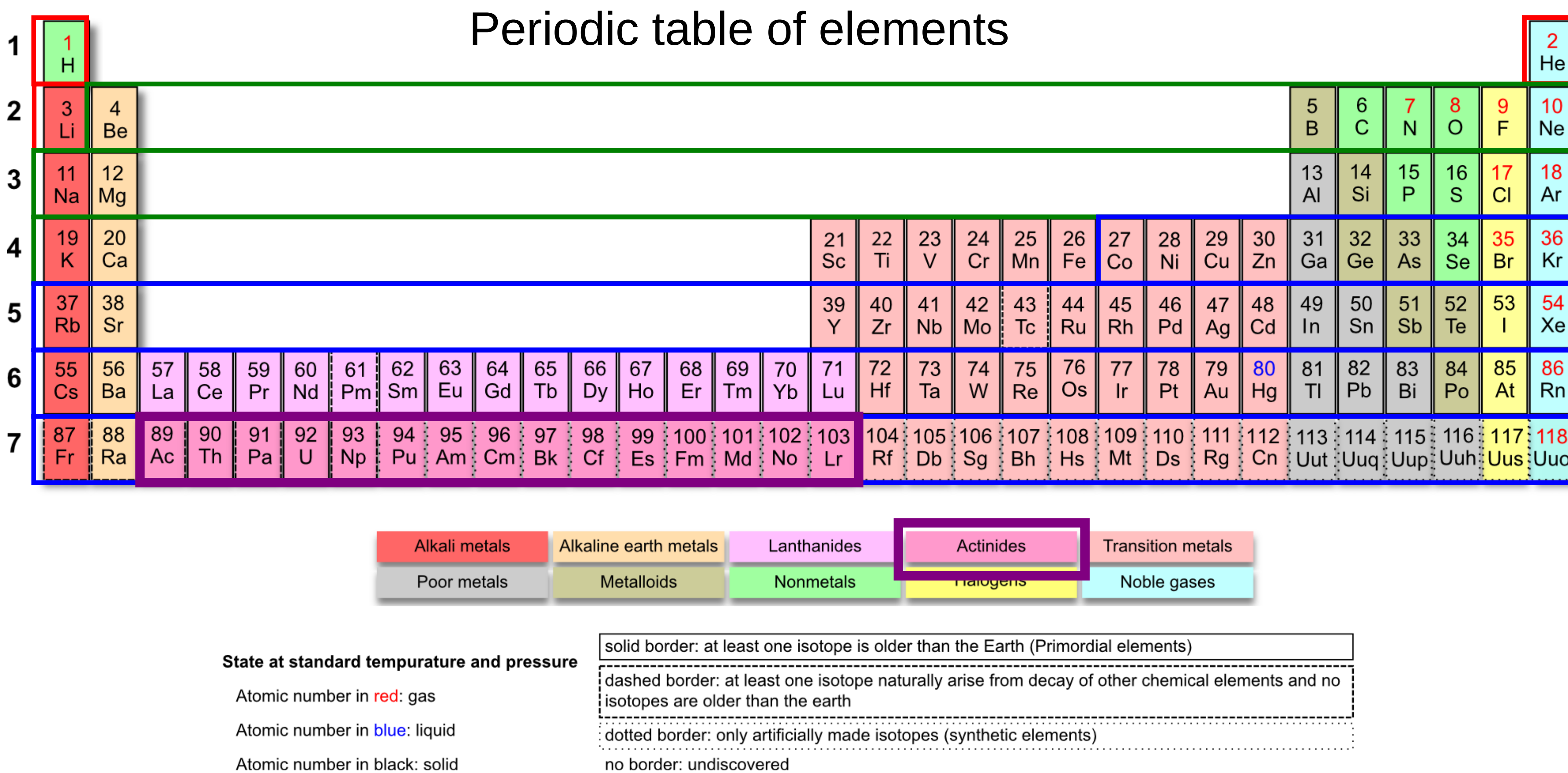
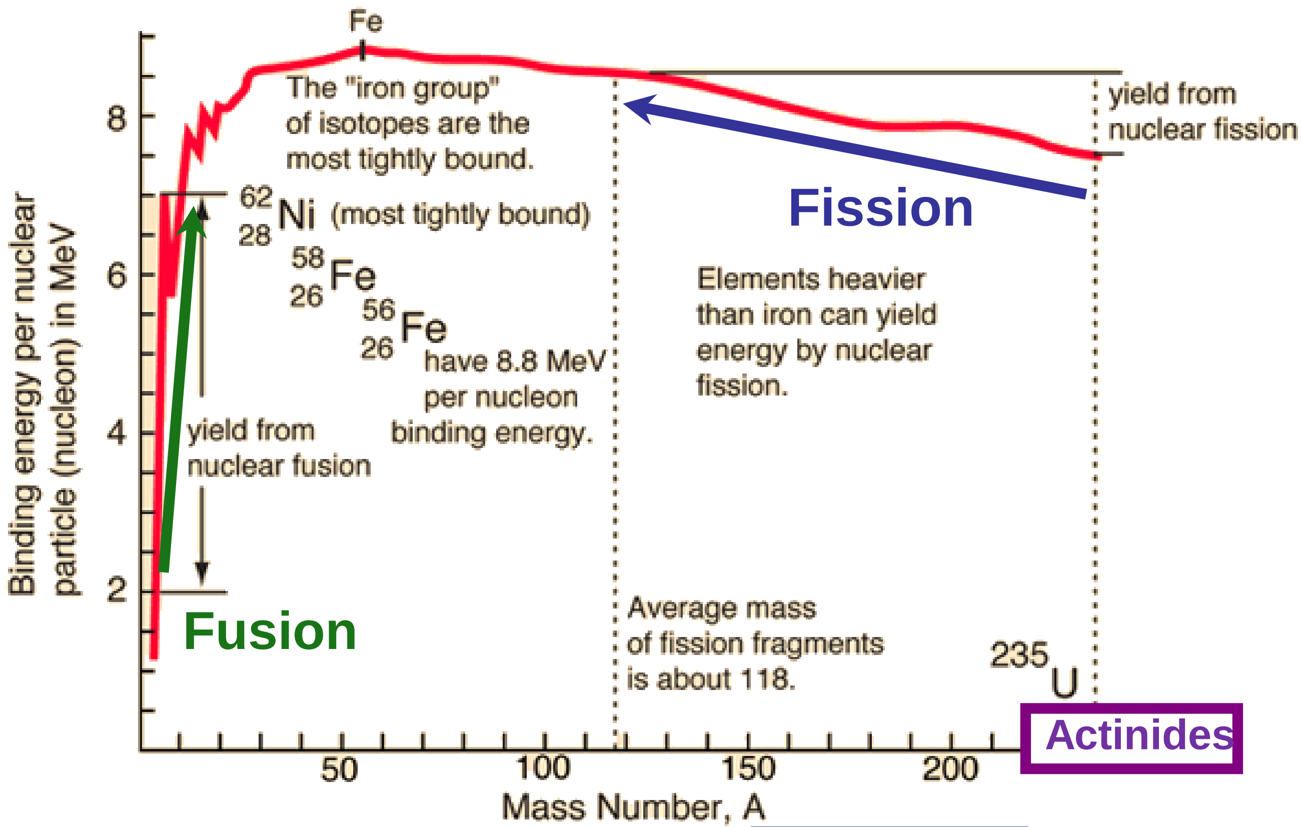
Nuclear fuel (resources) => Elements and their origin



Originated by: Big Bang, Stellar, and Supernova nucleosynthesis.

http://www.sciencegeek.net/tables/tables.shtml

“Nuclear” Energy and Nuclear forces



The Liquid Drop Model:

$$E_b(MeV) = 15.76A - 17.81A^{2/3} - 0.711\frac{Z^2}{A^{1/3}} - 23.7\frac{(N-Z)^2}{A} \pm 34A^{-3/4}$$

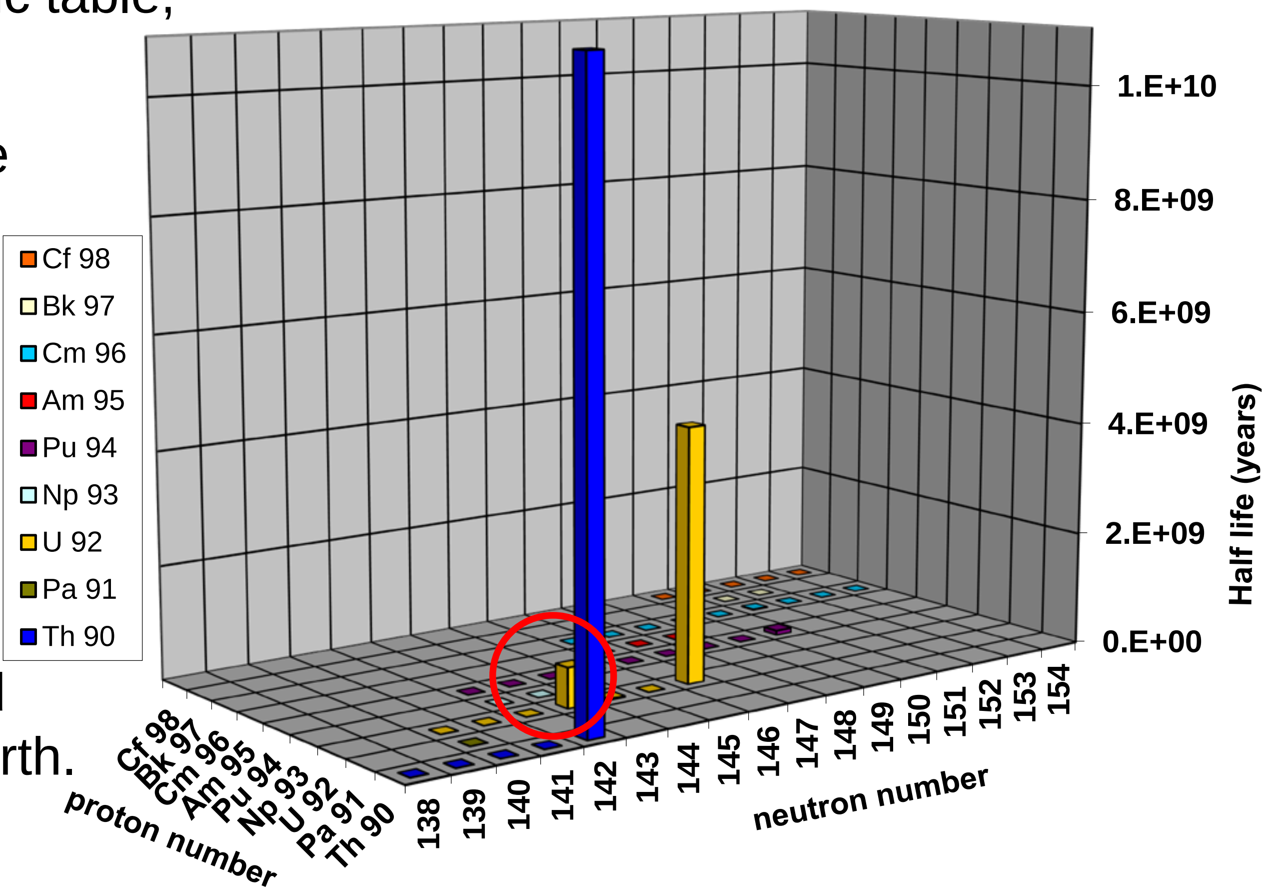
The Coulombic electrostatic repulsion is a barrier for fusion.

The reduced Coulombic electrostatic repulsion “drives” the fission.

http://www.nuclear-power.net/nuclear-power/fission/liquid-drop-model/

Actinides as nuclear fuel => are all unstable

- Actinides, the heaviest primordial elements in the periodic table, are all unstable.
- But three of them have relatively long half-life:
  - ^{235}U: 0.7 x10^{9} years
  - ^{238}U: 4.5 x10^{9} years
  - ^{232}Th: 14 x10^{9} years
- Accordingly: they are still present in nature.
- For 1 kg of ^{238}U there are 3-4 kg of ^{232}Th and 7.2 g of ^{235}U on the Earth.
- ^{235}U is the only fissile nuclide and its reserves are the smallest.



Actinides half-life in linear scale.

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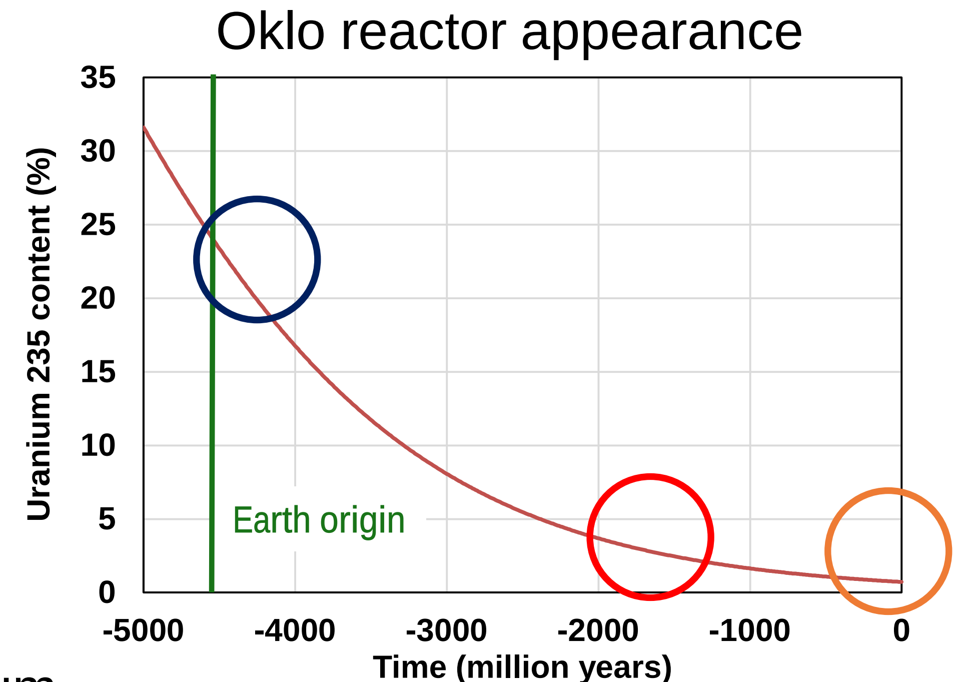
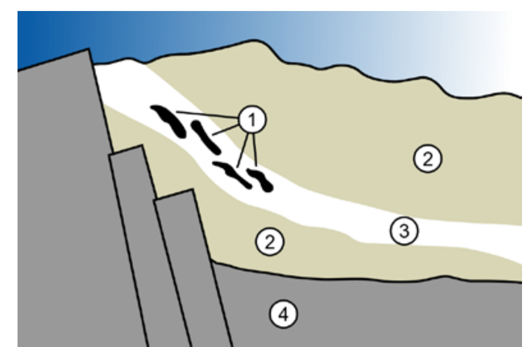
Natural uranium evolution – Oklo reactor

- ^{235}U and ^{238}U half-lifes differ. Accordingly the ^{235}U content in natural uranium is evolving.
- 1.7 billions years ago it enabled water moderated natural nuclear fission reactor in Oklo (Africa).
- Why not earlier? Several Dissolution–Precipitation cycles were necessary for the geological concentration of uranium.
- What about fast reactor? What if the geological concentration in the earth outer core was faster? If so, it may be still running on U-Pu cycle.
- Nowadays there is **only 0.72%** of ^{235}U in natural uranium. ☹





① Nuclear reactor zones, ② Sandstone, ③ Uranium ore layer, ④ Granite

https://en.wikipedia.org/wiki/Oklo

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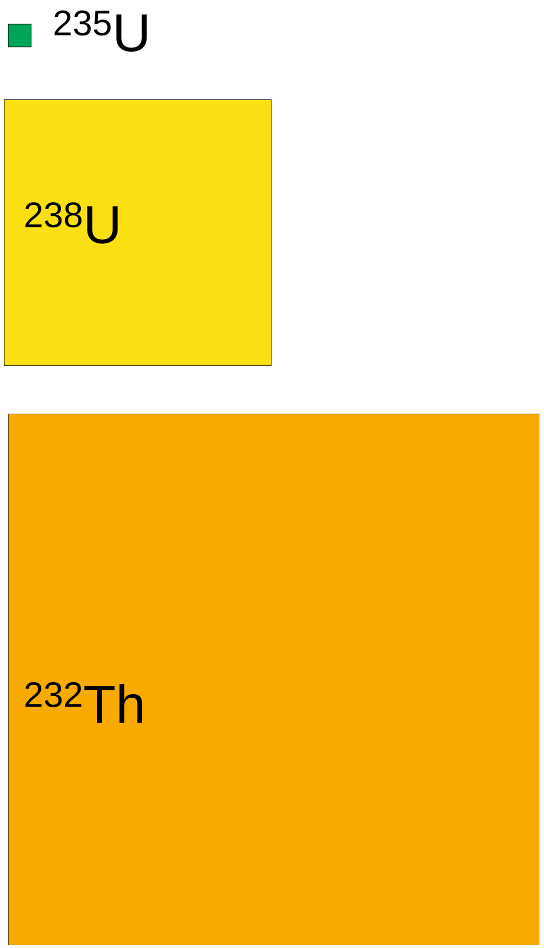
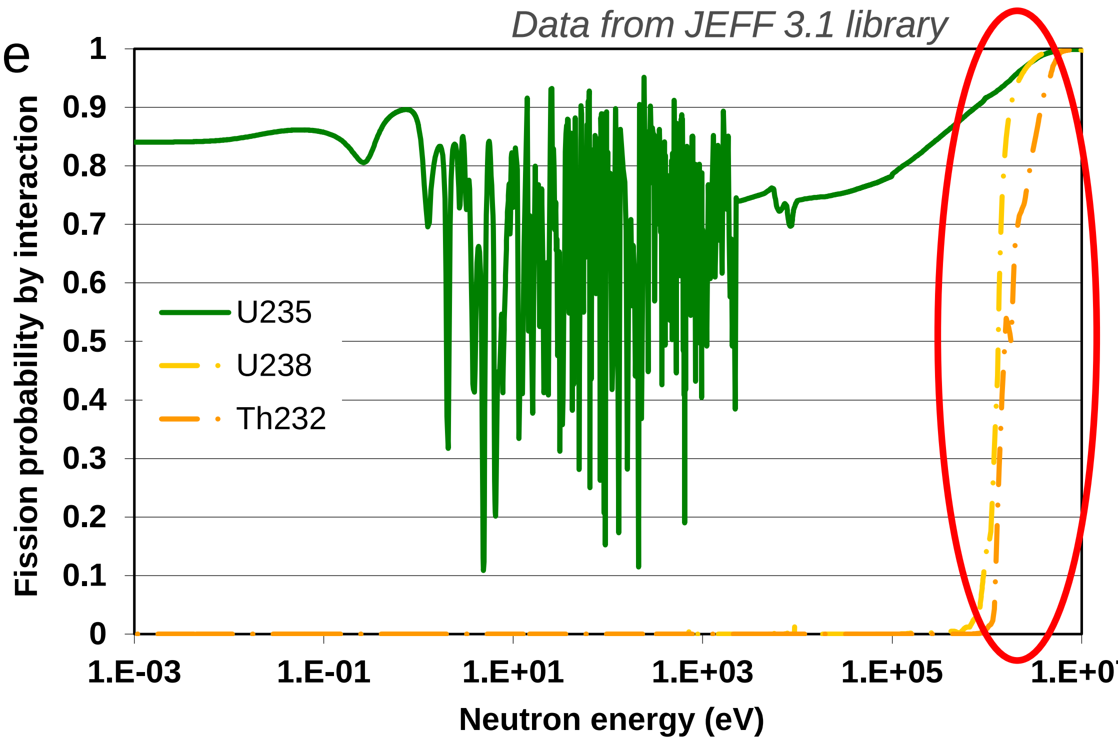
Sustainability = maximal resources utilization

- Reserves of actinides on the earth are not renewable. *Aim: their max. utilization!*



Relative size (surface) of actinides reserves.



- ^{235}U is the only primordial fissile nuclide and it is now the main working horse.
- ^{232}Th and ^{238}U are **fissionable** by fast neutrons (^{238}U up to 5x more than ^{232}Th).
- Both of them are mainly capturing neutrons, what leads to their transmutation.

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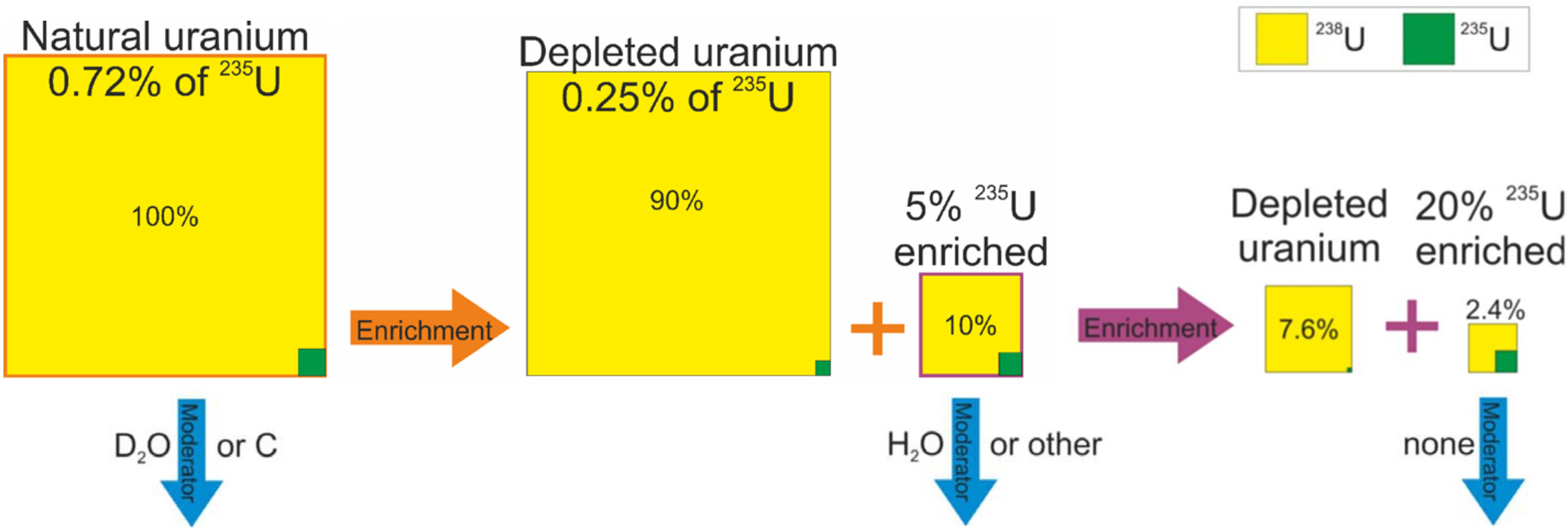
Sustainability of initial ^{235}U fueled reactors is low



CANDU	PWR, BWR	SFR, LFR, GFR

Burn-up of the fuel in **FIMA* %** (GWd/t_{hm}):

0.65-0.75 (6.5-7.5)	**3.3-5.0** (33-50)	**10-15** (100-150)

Burn-up in % of the original mass of natural uranium:

0.65-0.75	**0.33-0.5**	**0.24-0.36**

Sustainability?

Any ^{235}U fueled reactor has low sustainability

(not even 1% of natural uranium is utilized)

* *FIMA = **FI**ssion **MA**terial = actinides = heavy metals*

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Sustainability = ^{238}U and ^{232}Th catalytic burning

- *One neutron is needed for* ***next fission****.*
- *One of the new neutrons may be also captured by fertile ^{238}U or ^{232}Th.*
- *Then they will be transmuted to fissile ^{239}Pu or ^{233}U.*
- *This transmutation is also called conversion or breeding.*
- ***^{239}Pu or ^{233}U*** *may actually act as an intermediary (catalyzer)*
- *and* ***^{238}U or ^{232}Th*** *indirectly as a fuel.*
- ***Very tight neutron economy****.*

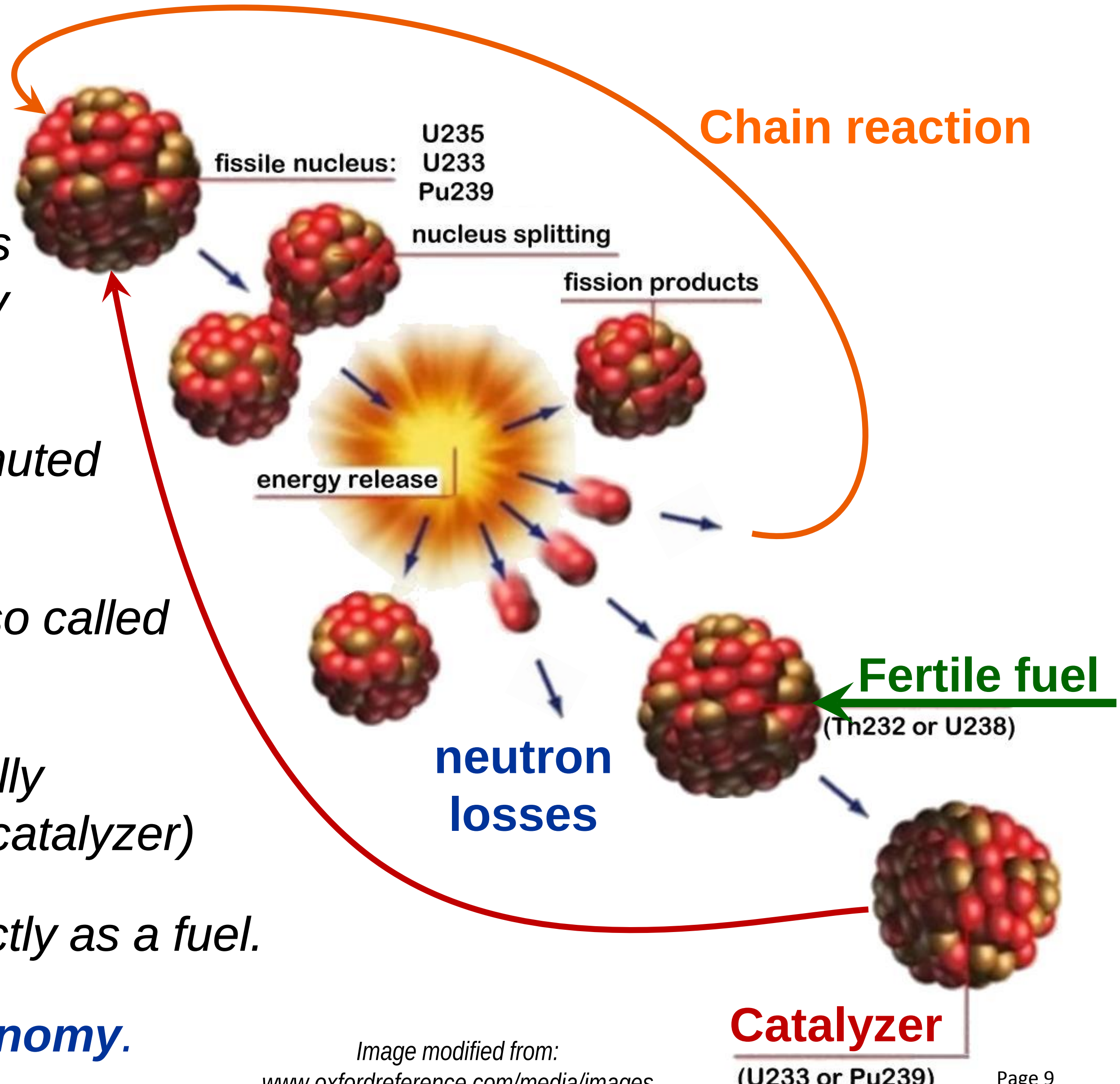

Image modified from: www.oxfordreference.com/media/images

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^{233}U and ^{239}Pu: synthetic (secondary) fissile elements

- Transmutation of fertile ^{232}Th and ^{238}U create fissile ^{233}U and ^{239}Pu:

$$^{232}_{90}Th + ^{1}_{0}n \longrightarrow ^{233}_{90}Th \xrightarrow{\beta^- 22\,\text{min}} ^{233}_{91}Pa \xrightarrow{\beta^- 27\,day} ^{233}_{92}U$$

$$^{238}_{92}U + ^{1}_{0}n \longrightarrow ^{239}_{92}U \xrightarrow{\beta^- 24\,\text{min}} ^{239}_{93}Np \xrightarrow{\beta^- 2.4\,day} ^{239}_{94}Pu$$

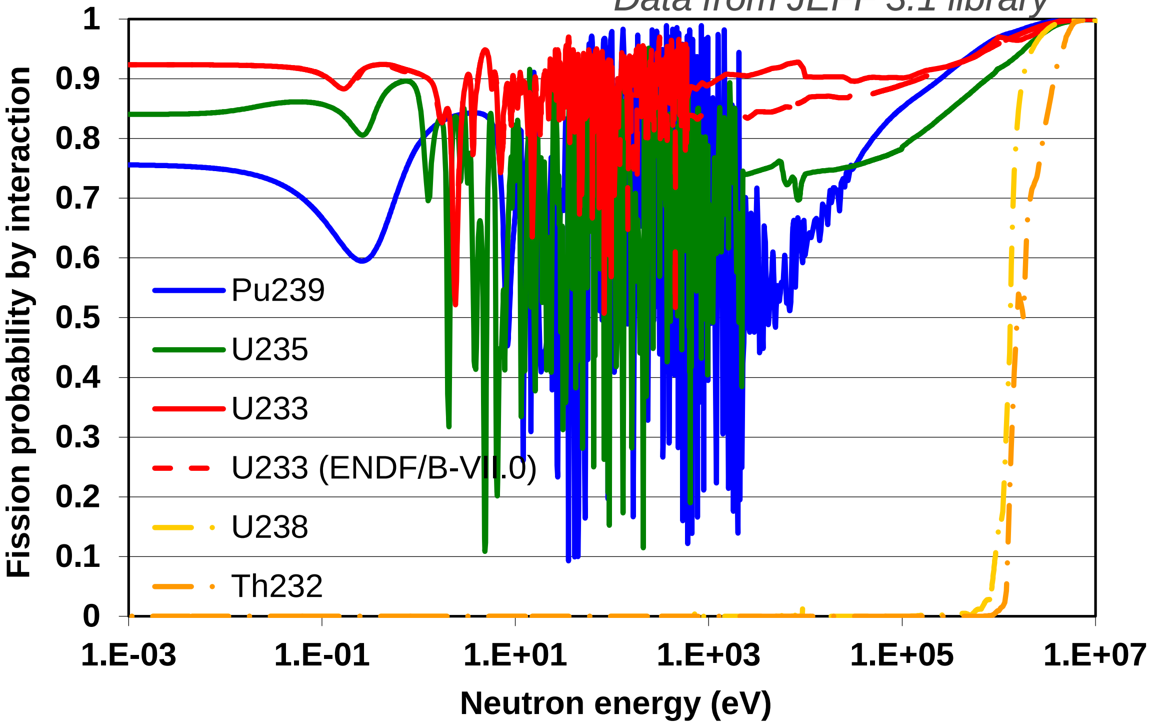
- In general, transmutation which increases fission probability is called **Breeding.**

(BTW: burning = fission)

- High fission probability up to **90%** is the biggest advantage of **^{233}U**. (for **^{239}Pu** it is **60-75%**)

(JEFF 3.1 X ENDF/B VII.0)

Data from JEFF 3.1 library



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^{234}U, ^{236}U, and ^{234}Pu: secondary fertile elements

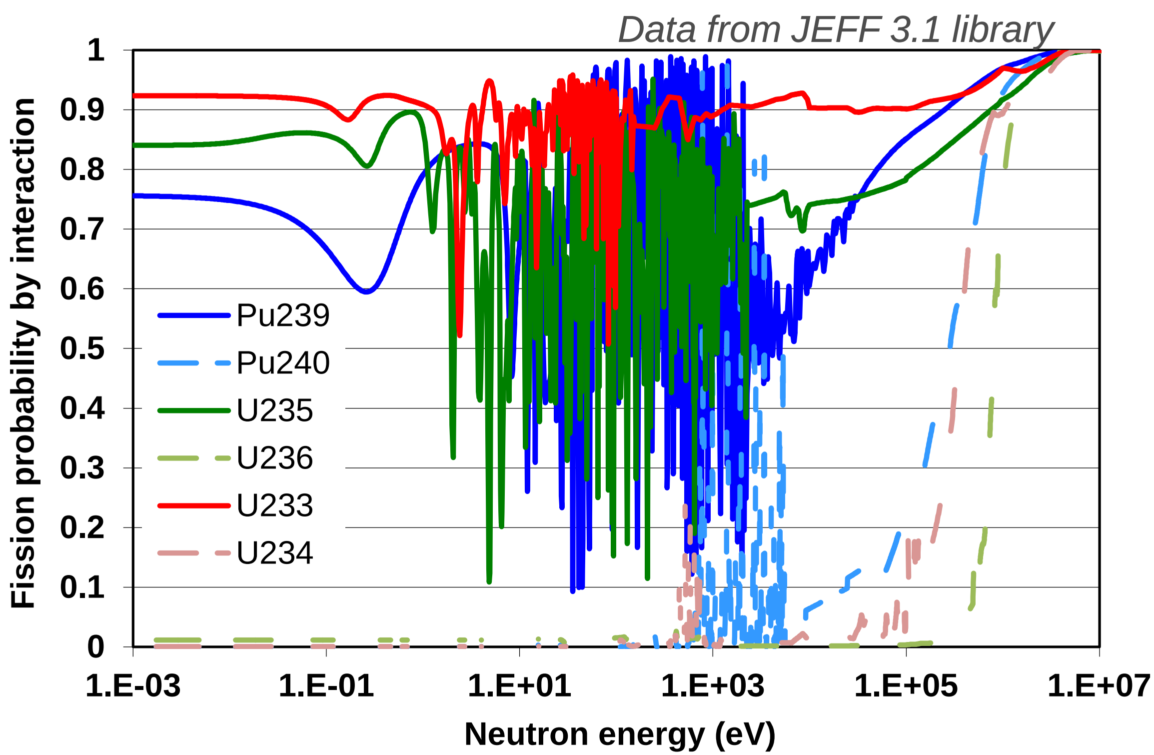
- Transmutation of fissile ^{233}U, ^{235}U, and ^{239}Pu create fertile ^{234}U, ^{234}U, and ^{240}Pu:

$$^{233}_{92}U + ^{1}_{0}n \longrightarrow ^{234}_{92}U$$

$$^{235}_{92}U + ^{1}_{0}n \longrightarrow ^{236}_{92}U$$

$$^{239}_{94}Pu + ^{1}_{0}n \longrightarrow ^{240}_{94}Pu$$

- When fissile nuclide captures neutron the products is typically fertile, thus it is called: **Parasitic capture.**
- The secondary fertile element needs to absorb one additional neutron to became fissile!



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Fissile or fertile? (fission barrier X binding energy)

- There exist pairing effect described even by the Liquid Drop Model:

$$E_b(MeV) = a_V A - a_S A^{\frac{2}{3}} - a_C \frac{Z^2}{A^{\frac{1}{3}}} - a_A \frac{(A-2Z)^2}{A} \pm \delta(A,Z)$$

where: $\delta(A,Z) = \begin{cases} +\delta_0 \; for \; Z,N \; even \\ 0 \\ -\delta_0 \; for \; Z,N \; odd \end{cases}$ (or actually $\pm 34A^{-3/4}$)

- Hence the interacting neutron brings different binding energy to each nuclide.

Nuclide: $^{232}_{90}Th + ^{1}_{0}n \longrightarrow ^{233}_{90}Th \xrightarrow{\beta^- \, 22\,\mathrm{min}} ^{233}_{91}Pa \xrightarrow{\beta^- \, 27\,day} ^{233}_{92}U$

	$^{232}_{90}Th$	$^{233}_{90}Th$	$^{233}_{91}Pa$	$^{233}_{92}U$
Neutron nr.:	142 (even)	143 (odd)	142 (even)	143 (odd)
Fissile:	no	yes (poorly)	no	yes

$^{233}_{92}U + ^{1}_{0}n \longrightarrow ^{234}_{92}U + ^{1}_{0}n \longrightarrow ^{235}_{92}U + ^{1}_{0}n \longrightarrow ^{236}_{92}U$

$^{234}_{92}U$	$^{235}_{92}U$	$^{236}_{92}U$
142 (even)	143 (odd)	144 (even)
no	yes	no

- Fission: binding energy > **fission barrier**. However, with growing nucleon number the barrier is **decreasing** => yes or no is not black and white.

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Uranium and Thorium fuel cycles

Cycle label:	U-Pu	Half-life	Th-U	Half-life
Main fertile:	^{238}U	4.5e9	^{232}Th	14e9
Main fissile:	^{239}Pu	2.4e4	^{233}U	1.6e5
Secondary fertile:	^{240}Pu	6500	^{234}U	2.5e5
Secondary fissile:	^{241}Pu (β^-)	14	^{235}U	7.0e8
Tertiary fertile:	^{242}Pu or ^{241}Am	3.7e5 or 432	^{236}U	2.3e7
Tertiary fissile:	^{243}Pu (β^-) or ^{242}Am		^{237}U (β^-)	
4th fertile:	^{244}Pu or ^{243}Am		^{237}Np or ^{238}Pu	
4th fissile:	^{245}Cm or ^{244}Am (β^-)		^{239}Pu	

- Th-U cycle produces less Minor Actinide - MA (Am, Cm, Np (from ^{235}U) etc).
  It is based on **^{239}Pu** position. It has implication on the waste radiotoxicity.

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^{233}Pa and ^{239}Np: intermediate products

- Transmutation of fertile ^{232}Th and ^{238}U goes through fertile ^{233}Pa and ^{239}Np:

$$^{232}_{90}Th + ^{1}_{0}n \longrightarrow ^{233}_{90}Th \xrightarrow{\beta^- 22\,\mathrm{min}} ^{233}_{91}Pa \xrightarrow{\beta^- 27\,day} ^{233}_{92}U$$

$$^{238}_{92}U + ^{1}_{0}n \longrightarrow ^{239}_{92}U \xrightarrow{\beta^- 24\,\mathrm{min}} ^{239}_{93}Np \xrightarrow{\beta^- 2.4\,day} ^{239}_{94}Pu$$

- It may happen that ^{233}Pa and ^{239}Np capture neutron:

$$^{233}_{91}Pa + ^{1}_{0}n \longrightarrow ^{234}_{90}Pa \xrightarrow{\beta^- 6.7\,h} ^{234}_{92}U$$

$$^{239}_{93}Np + ^{1}_{0}n \longrightarrow ^{240}_{93}Np \xrightarrow{\beta^- 65\,\mathrm{min}} ^{240}_{94}Pu$$

- The capture probability depends on cross-section and number of atoms *N*.
- After some time equilibrium will establish where the ^{232}Th and ^{238}U capture rates (*CR*) and the ^{233}Pa and ^{239}Np decay rates (λN) are equal: ***CR= λN.***
- Based on the different decay constants λ , there will be **11x more ^{233}Pa** than ^{239}Np in the core with the same transmutation rate.

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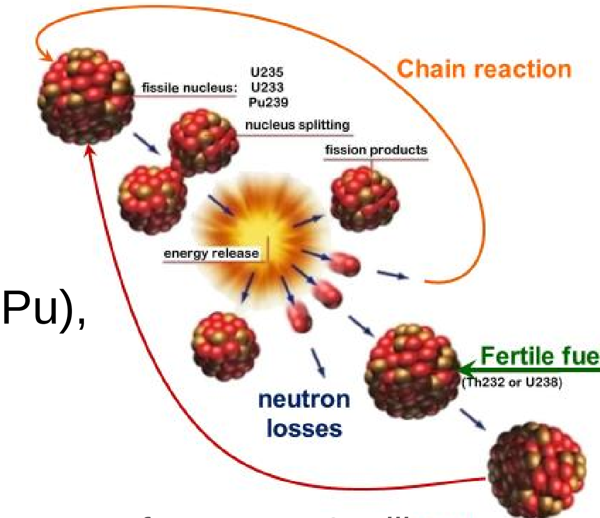
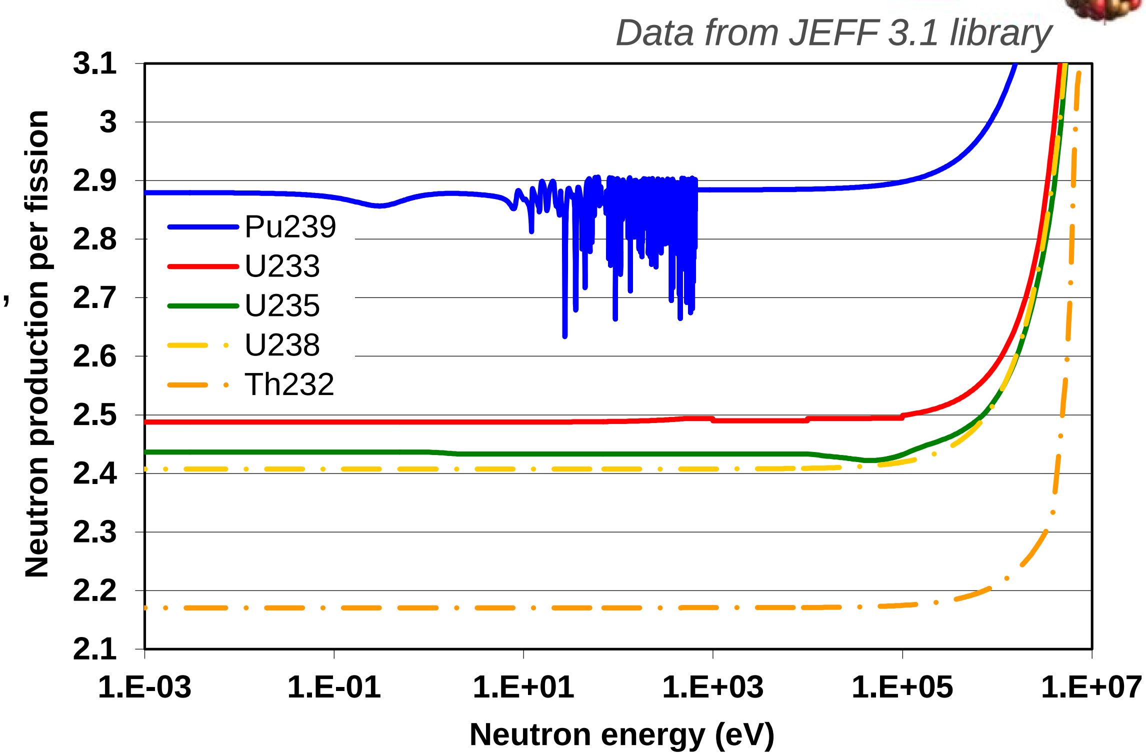
How many neutrons are available from fission?

- Number of average neutrons per fission (v-bar or $\overline{\nu}$) is function of interacting neutron energy and differs between actinides.
- From 5 basic isotopes (^{232}Th, ^{238}U, ^{233}U, ^{235}U, and ^{239}Pu), it is highest for ^{239}**Pu**: around **2.9 neutrons**.
- Second best is the ^{233}**U** with only **2.5 neutrons**.
- ^{235}**U** with 2.43 neutrons is the worst from the major fissile isotopes.
- ^{232}Th and ^{238}U, if fissioned, also produce neutrons.





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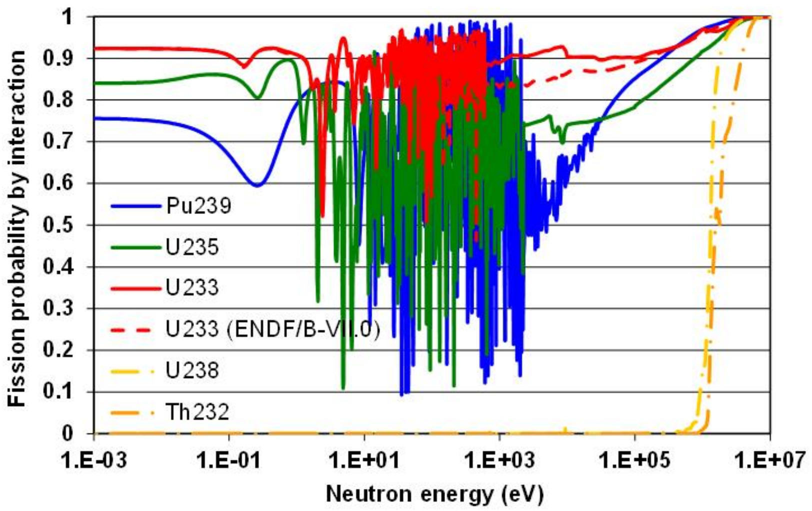
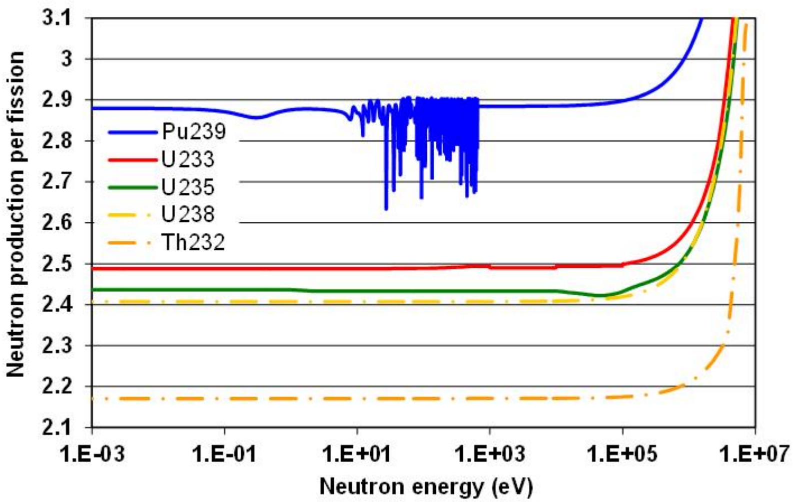
η – fission probability X neutrons originated per fission

- Eta (**η**) as the neutron generation factor describes generally the number of neutrons emitted by isotope/fuel per neutron absorption.
- It was introduced by Enrico Fermi around spring 1941* as a part of the 4-factors formula for the fuel as a whole and solely for thermal neutrons.
- It is often used for discussion of single isotope breeding capability.
- In this case, it is a product of fission probability and v-bar:



X



Enrico Fermi, Physicist, book by Emilio Serge

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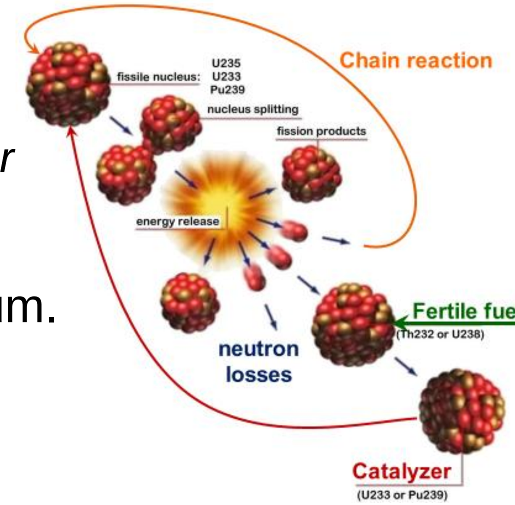
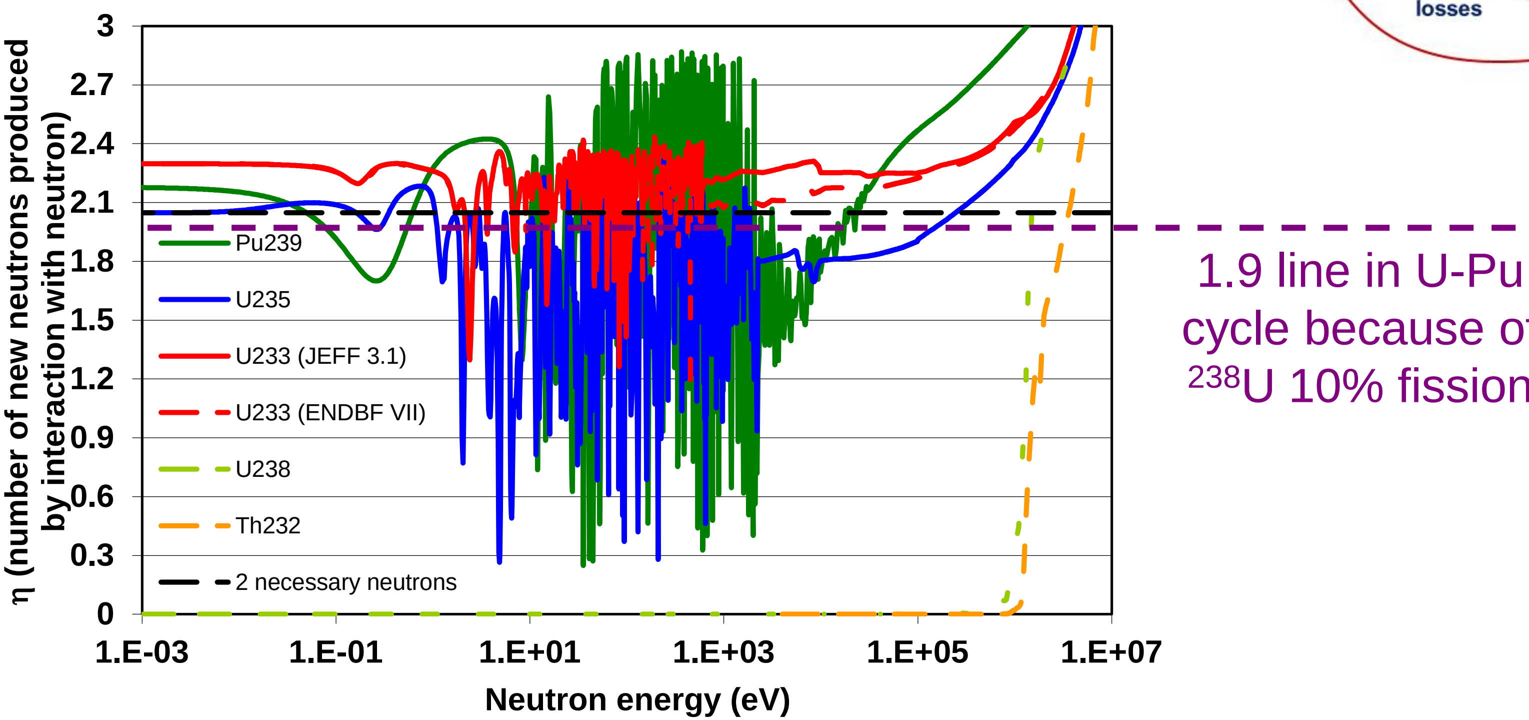
η – fission probability X neutrons originated per fission



- Recalling the trivial neutron economy, we need:
  1 neutron *to maintain the* ***fission chain reaction*** *and another*
  1 neutron *to* ***breed*** *new fissile isotope from the fertile one.*

- Hence **η** should be higher than **2** in the respective spectrum.



- It does not accounts for ^{238}**U** and ^{232}Th fission

- and for different properties of secondary fertile and fissile isotopes.

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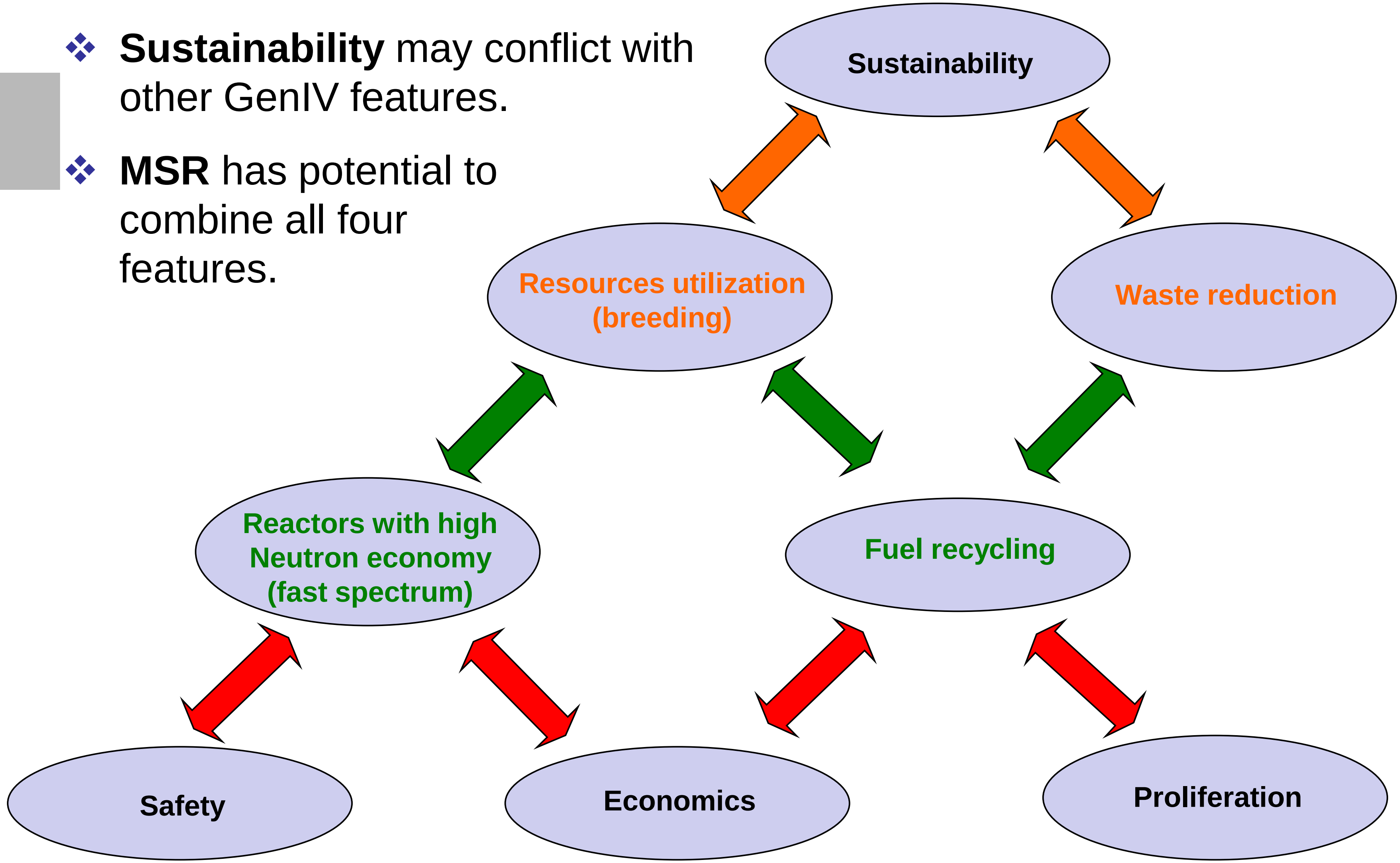
GenIV reactors = Sustainable reactors

- **Sustainability** may conflict with other GenIV features.
- **MSR** has potential to combine all four features.



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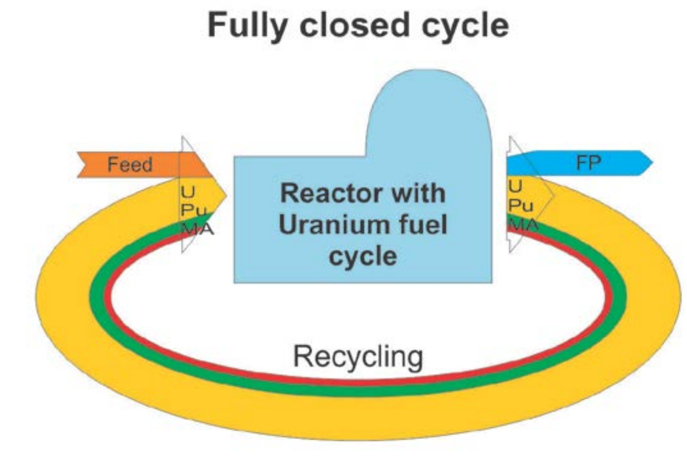
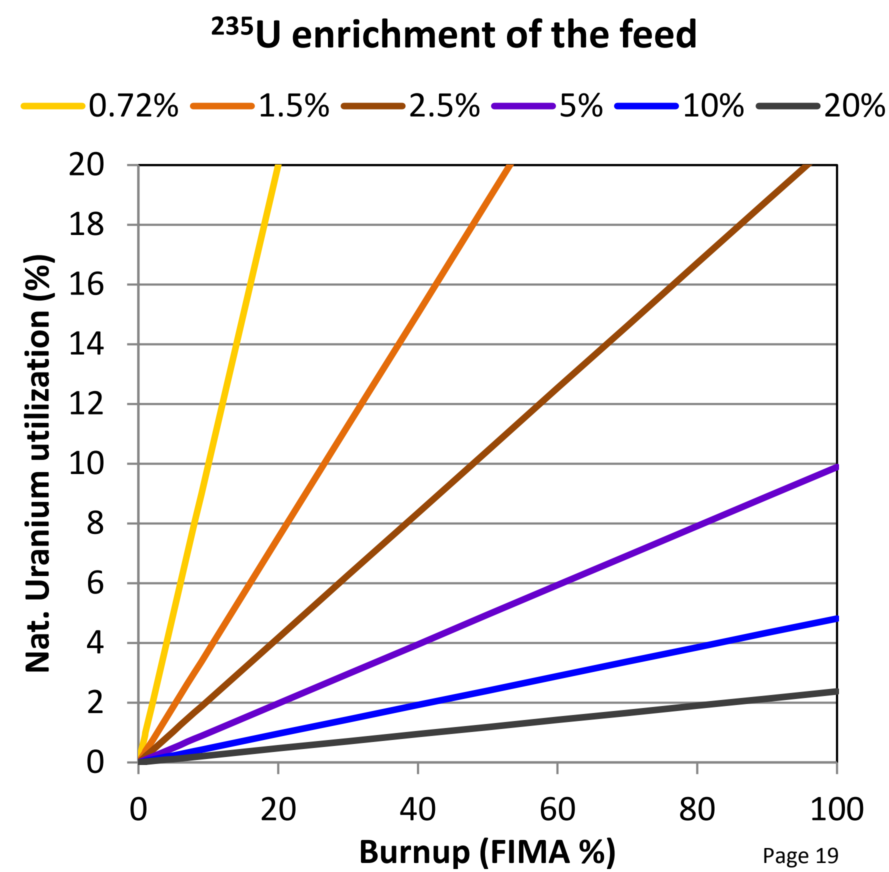
Recycling ≤ Sustainability

- Even if **all actinides** from spent fuel will be **recycled**, the utilization of natural resources can be still relatively low.
- The "**make-up fuel**" (US English) or actually the **feed** (EU English) should **not** be **enriched uranium**.
- Whenever **enriched uranium** is used as the feed, **sustainability** is strongly **decreased**.
- Even its 100% utilization by recycling will not help.
- Lets recall here than in ^{235}U fueled reactor without recycling it is at maximum **0.75%**.





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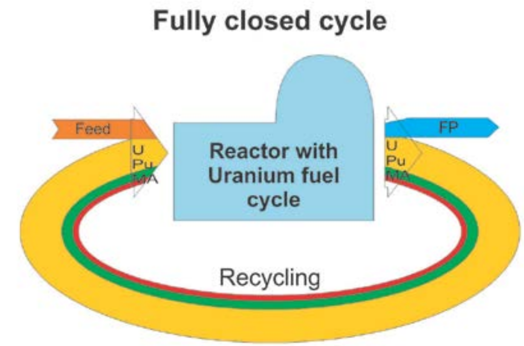
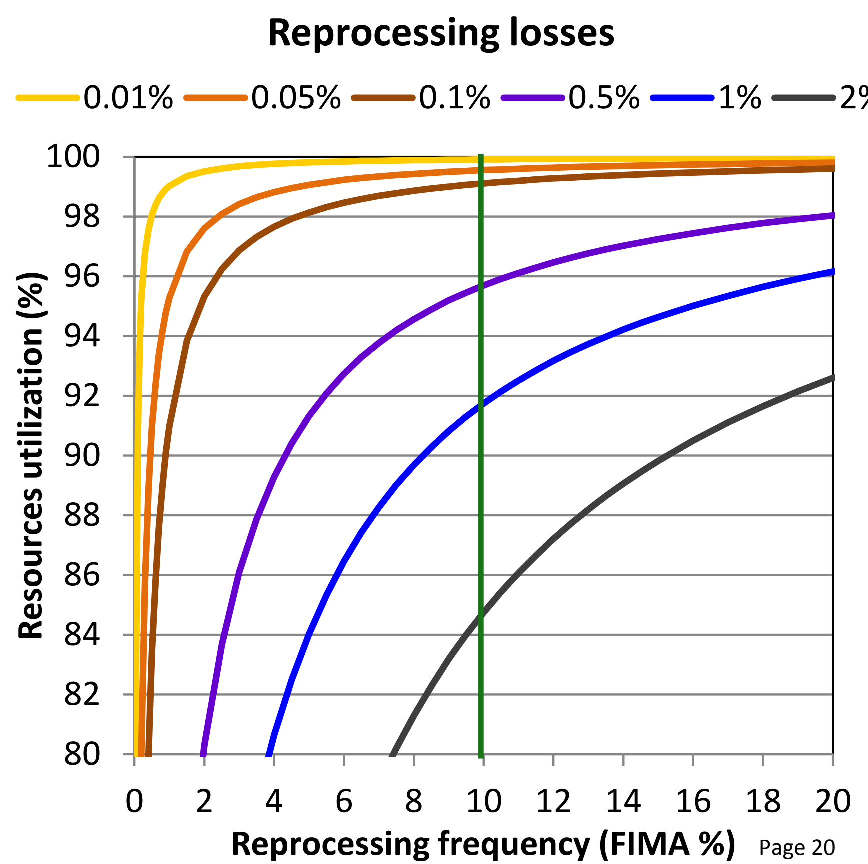
Sustainability = ^{238}U and ^{232}Th catalytic burning



- With natural uranium or thorium feed high **sustainability** can be achieved by **recycling**.
- Nonetheless, due to the **reprocessing losses,** it will be always **below 100%.**
- It depends on reprocessing method **losses (L)** and on the **reprocessing frequency (F)** (both expressed in fuel %).
- Typical fuel burnup in solid fuel fast reactor is **10% FIMA**. In MSR the discharge burnup may be lower.
- Homework: please cross-check if it was derived correctly:

$$Utilization = 1 - losses = 1 - \frac{L(1-F)}{1-(1-L)(1-F)}$$



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GenIV: Sustainability versus Safety

- **Sustainability** often requires **fast neutron** spectrum.
- In fast neutron spectrum **coolant does not moderate** the neutrons.
- **Coolant removal** or **fuel compaction** leads to **reactivity increase**.
- **GFR** has quite low positive void. It is, however, hard to cool in case of coolant depressurization.
- **SFR** has strong positive void; nonetheless, it can be minimized by neutron leakage maximization in voided core. Still, there is an issue with sodium fire.
- **LFR** has very strong void coefficient, but lead is not so easy to void.
- In general **SFR** and **LFR** are **low pressure** system and the metallic coolant has **retention potential** for some problematic fission products.
- **MSR** combines coolant and fuel in one **liquid**. It can be designed with **negative void** coefficient and **fuel compaction / collection** may be **prevented**. (moderated MSR breeder may have positive graphite temperature feedback coefficient)

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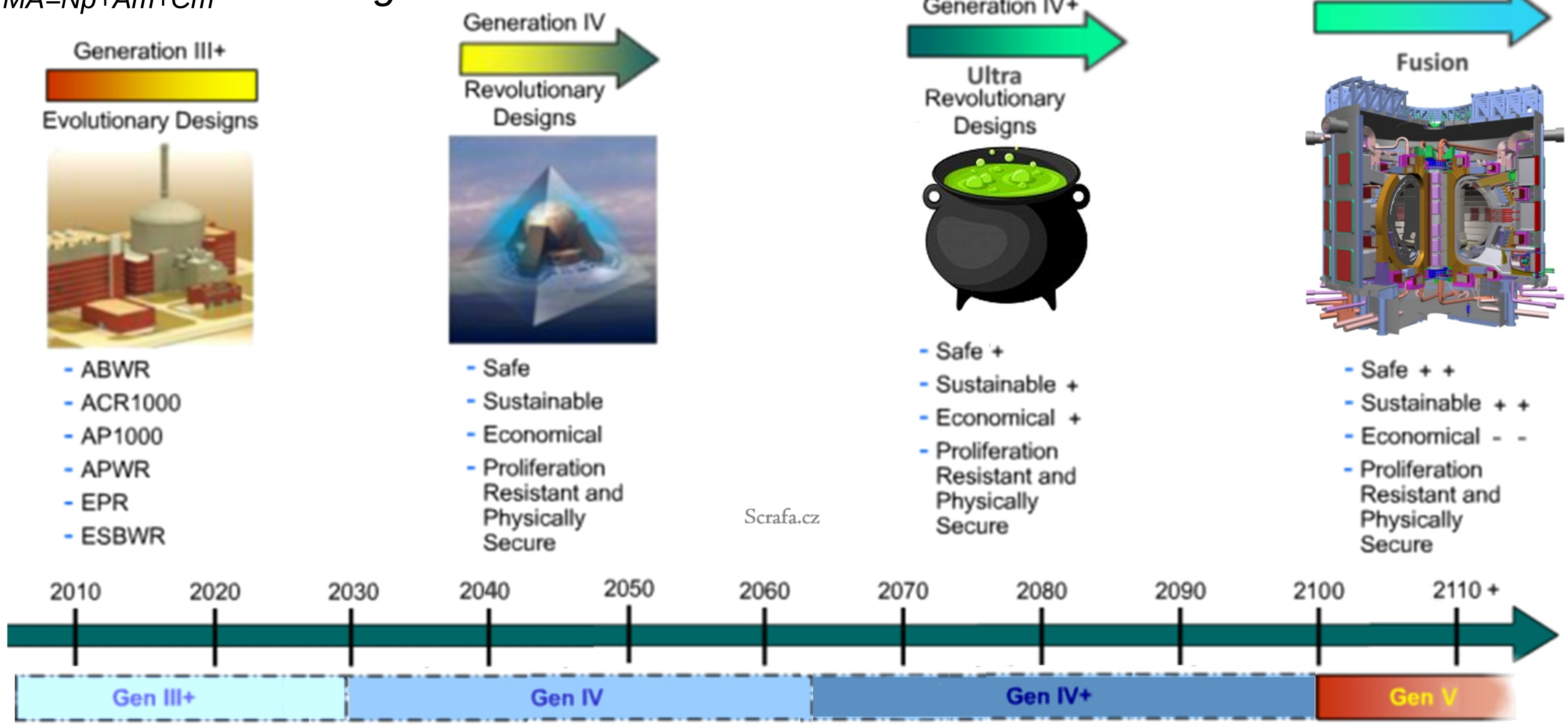
Is Gen IV the last one? No, let's add some more ☺

Fuel / Cycle:	^{235}U (U-Pu)	U-Pu / closed	Th-U / closed	D-T / Li
Sustainability*:	100-200 years	5 000 years	20 000 years	200 000+ years
Radioactivity:	**Gen III+**	**Gen IV**	**Gen IV+**	**Gen V**
Activated mat.:	yes	yes	yes	yes
1t burned fuel:	1000kg FP	1000kg FP	1000kg FP	800kg He
+ byproducts:	200kg Pu 20kg MA	50kg MA	15kg ^{237}Np+^{238}Pu	

MA=Np+Am+Cm



*Just kidding; it is hard to estimate energy consumption even in 100 years from now. ☺

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Two basic fuel cycle issues related to sustainability

How to start it

1. Any reactor capable of burning for ^{238}U and ^{232}Th should be started by ^{235}**U** or by products from ^{235}**U fueled reactor**.

How to maintain it

2. Any sustainable reactor for ^{238}U and ^{232}Th catalitic burning should be capable to operate with **equilibrium fuel composition**. (secondary fertile and fissile, tertiary fissile and fertile, etc.…)

3. There should exist technology to regularly **separate** the **fission products** (FPs) from the fuel.

Separation of FPs is usually not possible without complete fuel decomposition. Hence, what other industries call ***recycling*** *is called “****reprocessing****”.*

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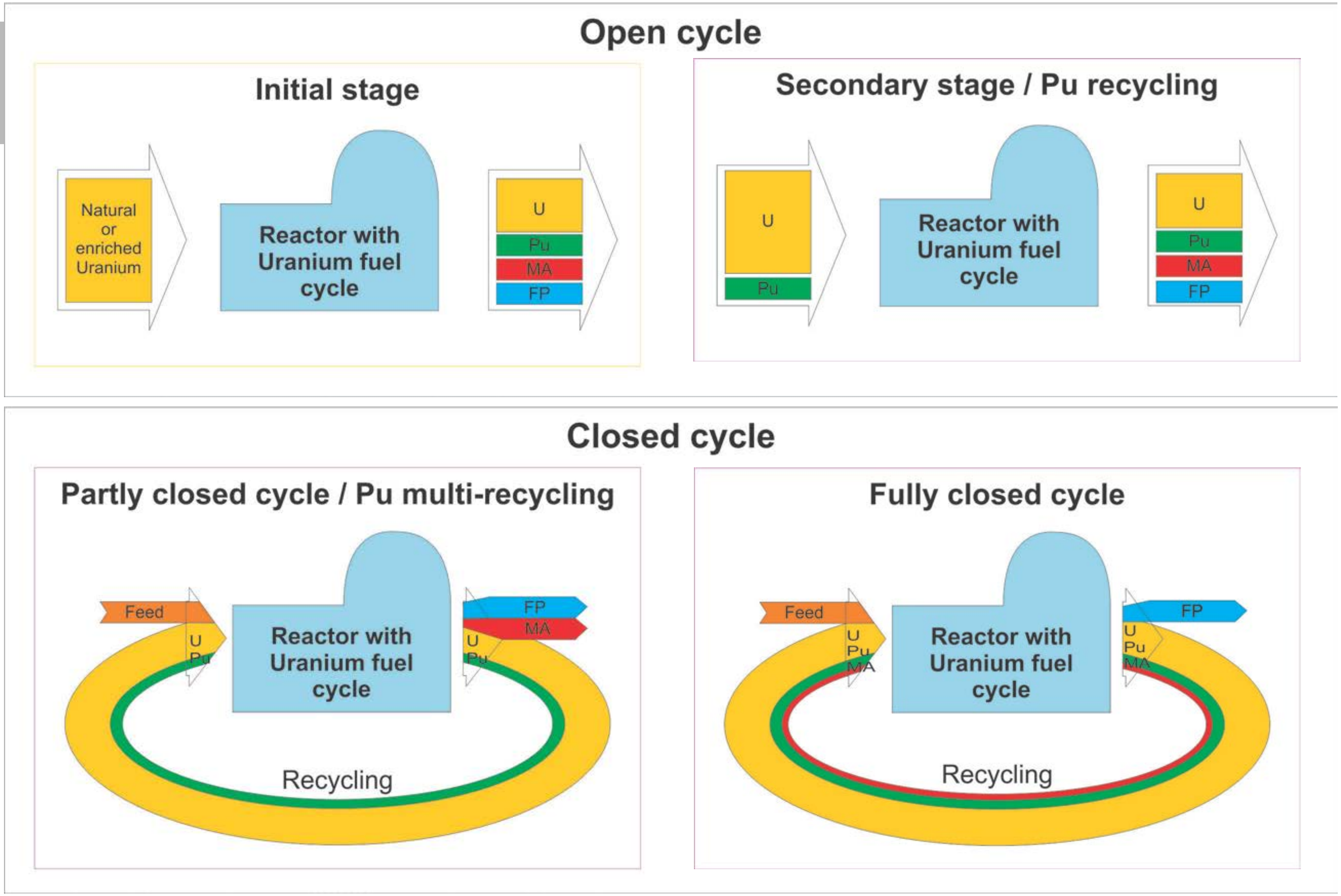
Initial and secondary stages & open versus closed cycle

Uranium-Plutonium cycle



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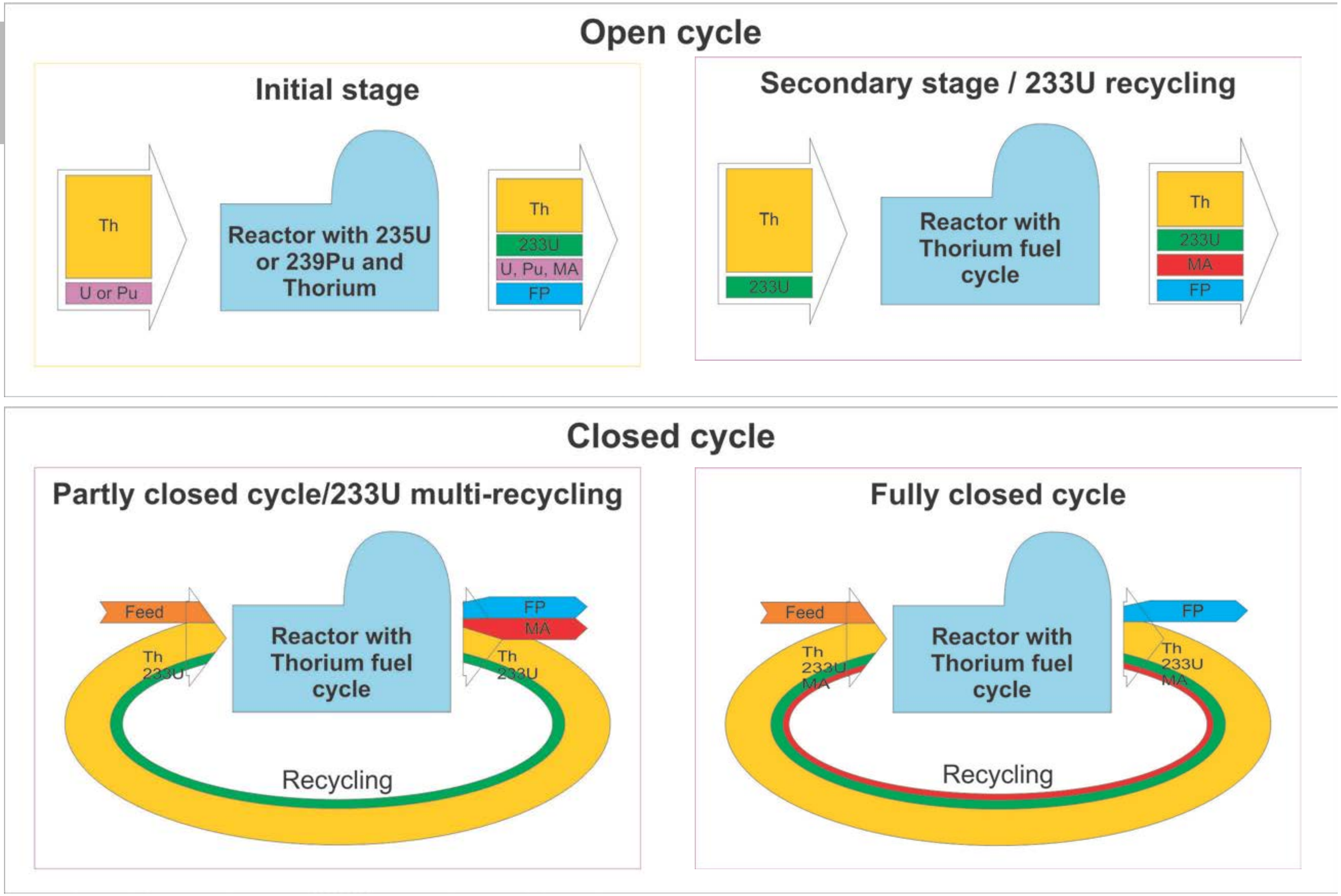
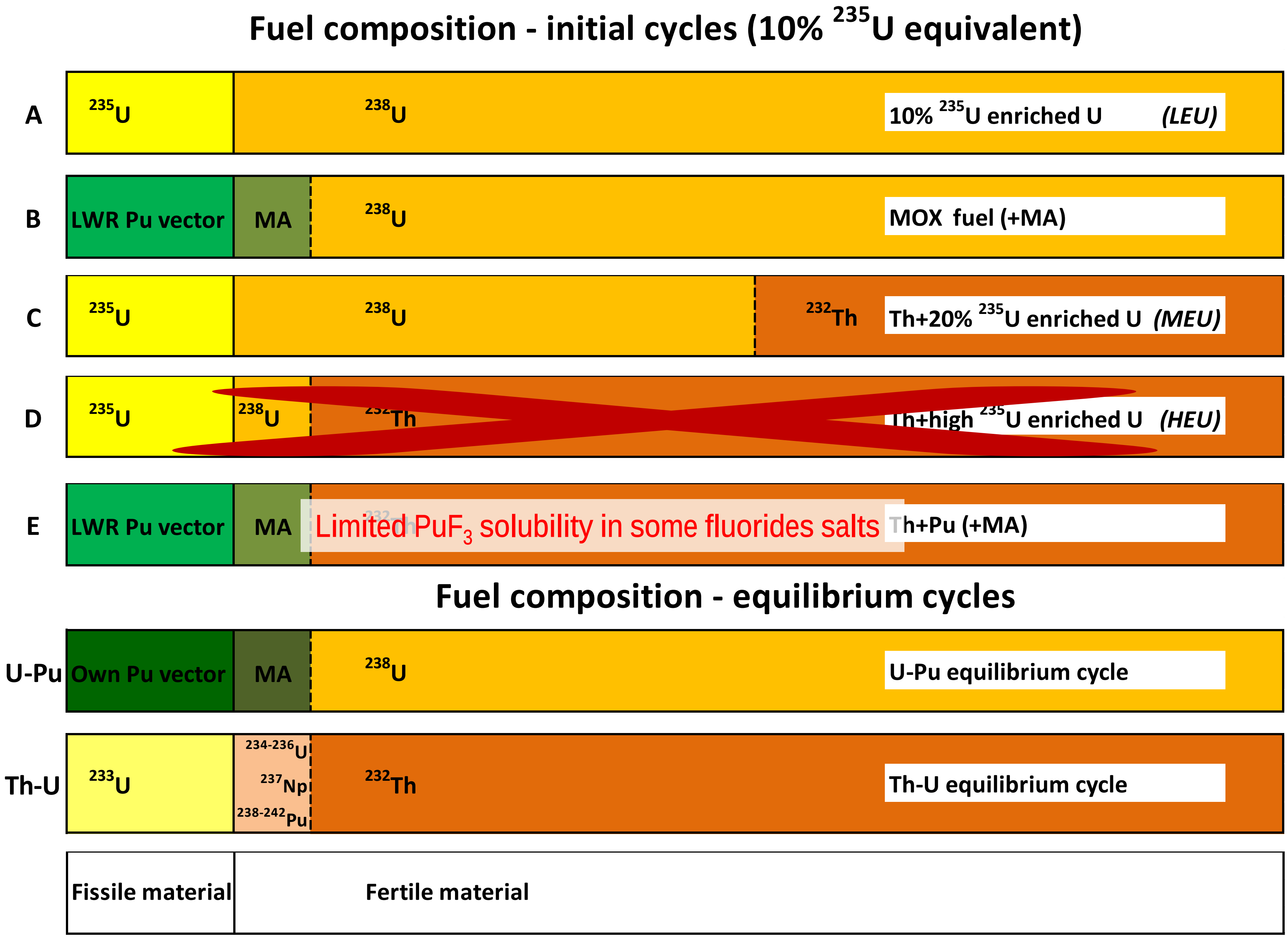
Initial and secondary stages & open versus closed cycle

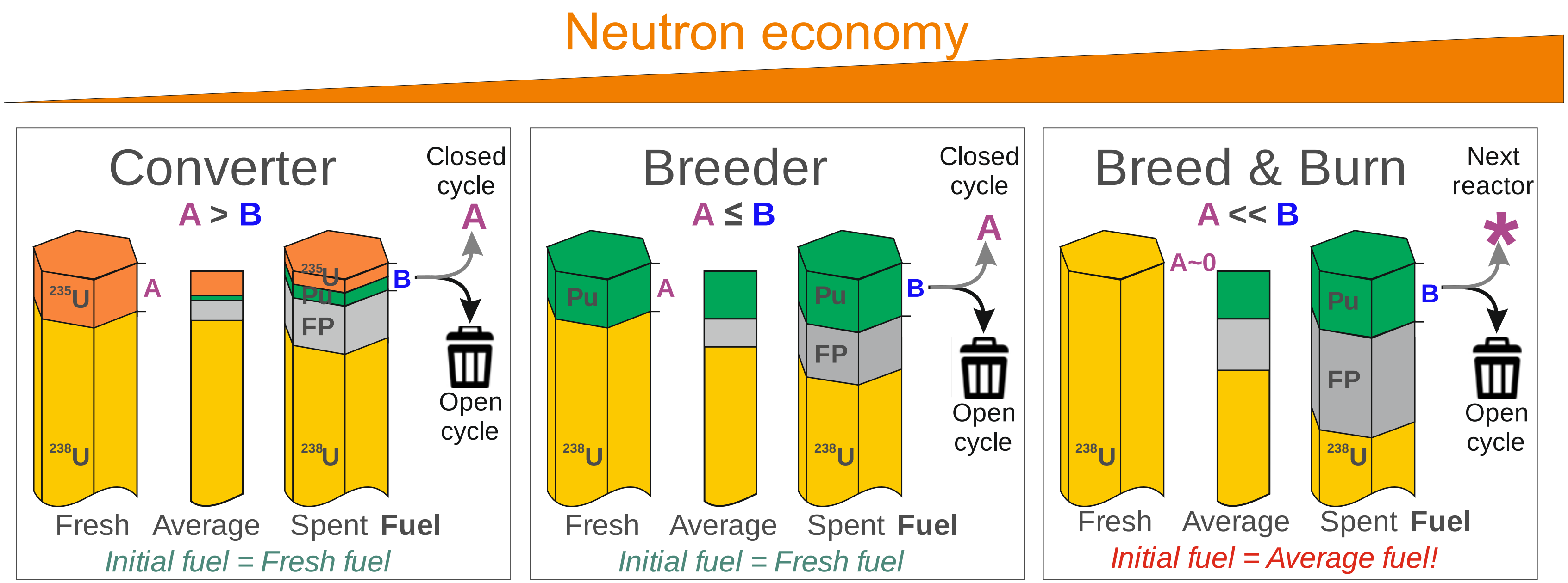
Thorium-Uranium cycle



Example of initial fuel composition equivalent



Equilibrium cycle operation – neutron economy



- **Convertor**, e. g. PWR or IMSR, is operated **usually** in **open fuel cycle**.
- **Breeder** profit from neutronics advantages only in the **closed cycle**.
  For Iso-breeding (EU) or Break-even (US) reactor => A=B.
- Extreme breeder can be operated in **Breed-and-Burn** mode.
  It can have **high fuel utilization** even **without reprocessing**.

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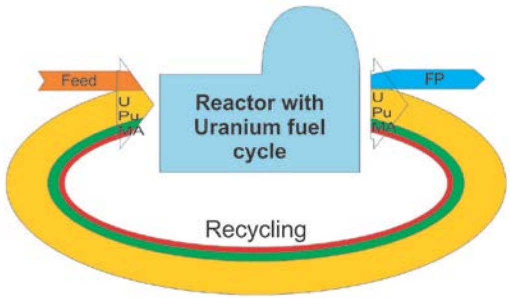
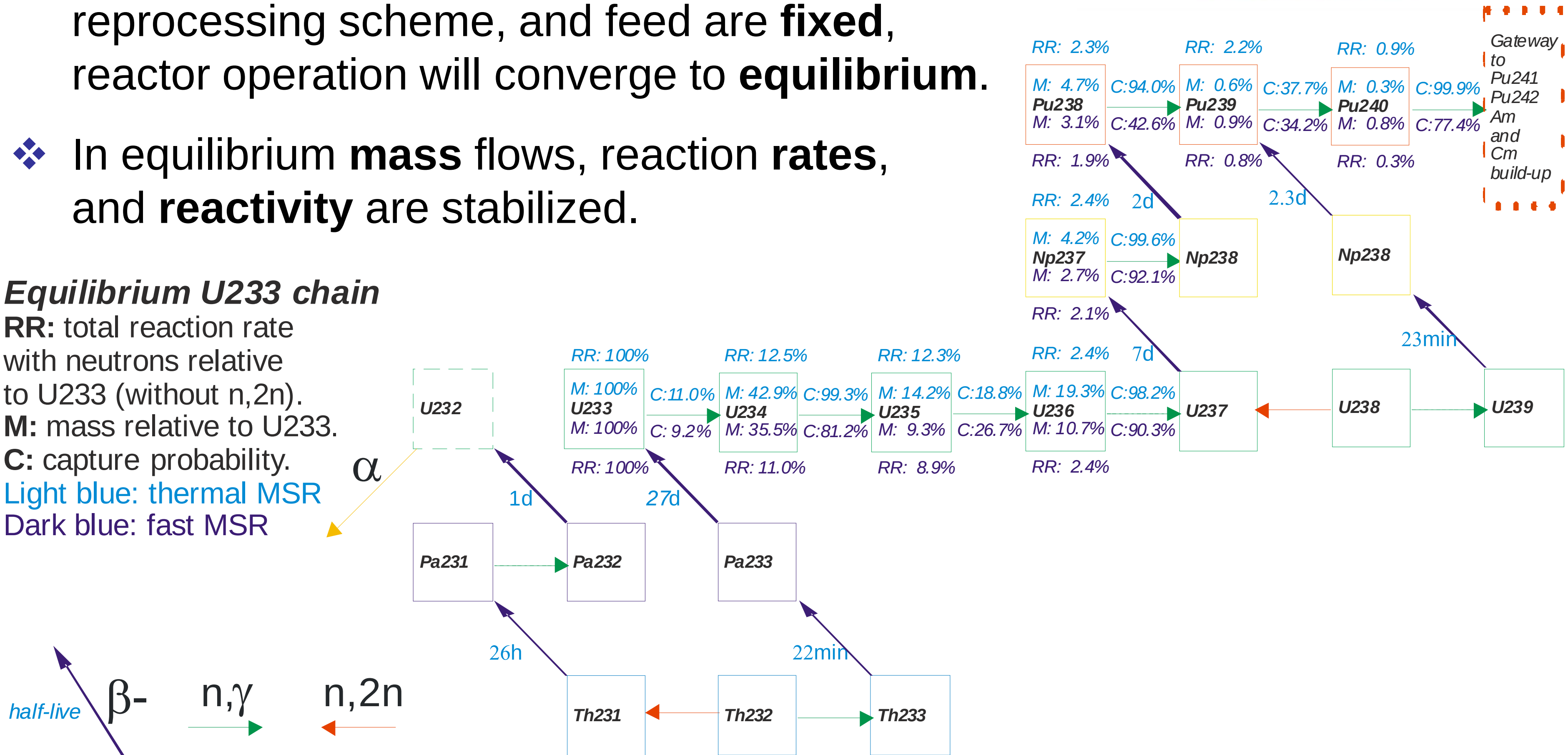
Breeding capability – excess reactivity in equilibrium

- **Breeding capability** can be estimated from **excess reactivity in equilibrium** fuel cycle.
- If **fuel cycle properties** like: power (or flux), reprocessing scheme, and feed are **fixed**, reactor operation will converge to **equilibrium**.
- In equilibrium **mass** flows, reaction **rates**, and **reactivity** are stabilized.

Fully closed cycle



Equilibrium U233 chain
RR: total reaction rate with neutrons relative to U233 (without n,2n).
M: mass relative to U233.
C: capture probability.
Light blue: thermal MSR
Dark blue: fast MSR



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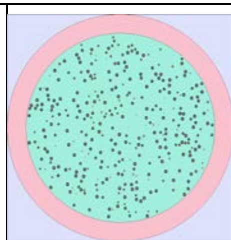
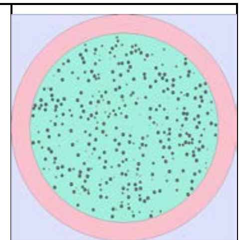
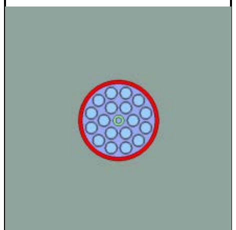
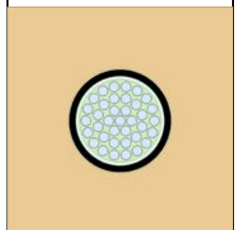
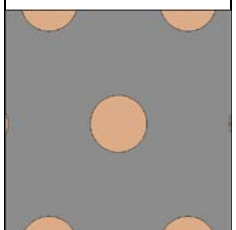
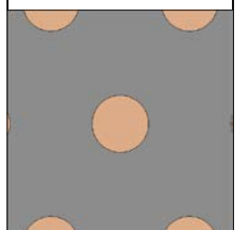
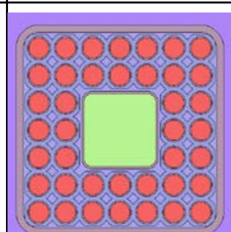
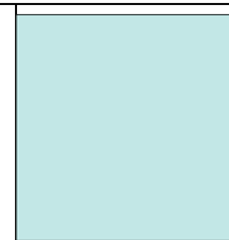
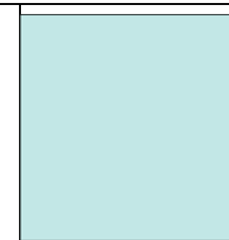
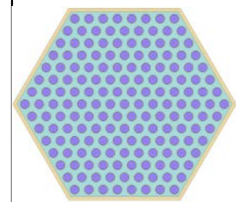
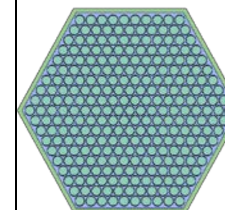
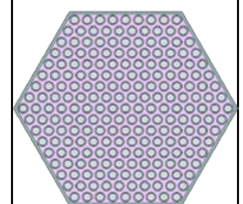
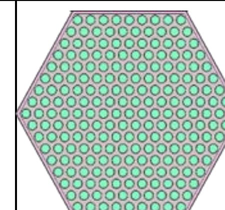
Comparison of 16 reactors: 8 thermal & 8 fast

FHR 192.0 W/g_{HM}		**HTR 96.8 W/g_{HM}**	
RBMK 13.7 W/g_{HM}		**PHWR 32.1 W/g_{HM}**	
MSR-FLIBE 41.1 W/g_{HM}		**MSR 41.1 W/g_{HM}**	
HPLWR 25.3 W/g_{HM}		**LWR 41.1 W/g_{HM}**	

MSFR 41.1 W/g_{HM}		**MSFR-FLIBE 41.1 W/g_{HM}**	
LFR 54.8 W/g_{HM}		**SFR 48.8 W/g_{HM}**	
GFR 40.1 W/g_{HM}		**MFBR 178.6 W/g_{HM}**	
NaCl-AcCl4 salt 54.8 W/g_{HM}		**AcCl4 salt 54.8 W/g_{HM}**	

- The simplified designs were adopted as is without optimization.
- If the core consists of assemblies with identical geometry but different fuel composition only one assembly was simulated.
- If the geometry differs, all cases have been simulated, but only one selected is presented.

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Assumptions for equilibrium cycle simulation

- **Infinite lattice** cell level simulation.
- Reactor specific power given by burnup in **FIMA %** (FIssile MAterial %) and **fuel residence time.**
- Neglecting fission products.
- Zero reprocessing losses (L=0).
- Continuous feed of fertile material (^{232}Th or ^{238}U).
- ENDF/B-VII.0 nuclear data library.

- With these assumptions we obtained equilibrium **fuel composition** and equilibrium **reactivity**.
- The excess **reactivity** should be **high enough** to compensate for: *neglected reprocessing losses, neutron leakage and fission products parasitic captures.*

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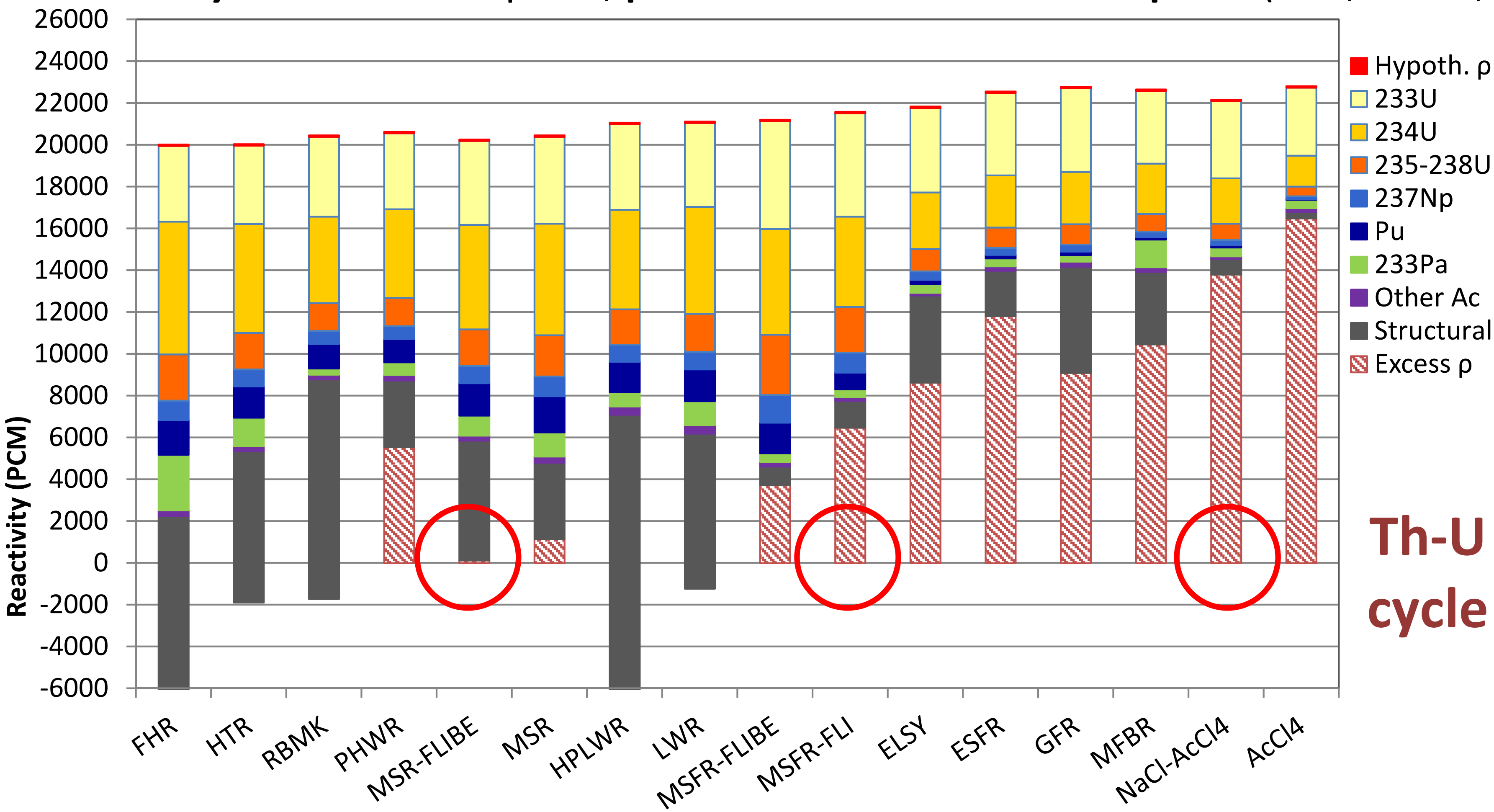
Excess reactivity in eql. cycle for Th-U cycle

- **Excess reactivity** for eql. fuel composition quantifies the **closed cycle capability**.
- Comparison of **16 reactors** is based on infinite lattice calculations with no FPs.
- **Th-U cycle**: **low ^{233}U** capture, **power effect** due to **^{233}Pa capture** (FHR, MFBR,…).



Krepel, J. at. al., Chapter for IAEA document, Near Term and Promising Long Term Options for Deployment of Thorium Energy, in preparation.

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Excess reactivity break-down method

Neutron balance eq.:

$$k_{\text{inf}} = \frac{R_P^{total} + 2R_{n,2n}^{total}}{R_F^{total} + R_C^{total} + R_{n,2n}^{total}}$$

Four assumptions:

1) $R_P^{total} = \bar{\nu} R_F^{total}$ ***2)*** $R_C^{total} = R_C^{^{232}Th} + R_C^{other}$

3) $R_{n,2n}^{total} = R_{n,2n}^{^{232}Th}$

main fertile represents at least 90% of all (n,2n) reactions

Valid only in equilibrium:

4) $R_C^{^{232}Th} + R_{n,2n}^{^{232}Th} = R_F^{total} - R_F^{^{232}Th}$

total other-than-fertile actinides destruction (total fission rate without the fertile isotope fission rate) should be in equilibrium equal to the total other-than-fertile actinides production (capture or (n,2n) reactions on the main fertile element)

Result:

$$\rho = \frac{(\bar{\nu} - 2)R_F^{total} + R_F^{^{232}Th} + 2R_{n,2n}^{^{232}Th} - R_C^{other}}{\bar{\nu} R_F^{total} + 2R_{n,2n}^{^{232}Th}} \cong \frac{\bar{\nu} - 2}{\bar{\nu}} + \frac{R_F^{^{232}Th} + 2R_{n,2n}^{^{232}Th}}{\bar{\nu} R_F^{total} + 2R_{n,2n}^{^{232}Th}} - \frac{R_C^{other}}{\bar{\nu} R_F^{total} + 2R_{n,2n}^{^{232}Th}}$$

Available neutrons *Bonus from fertile* *Parasitic captures*

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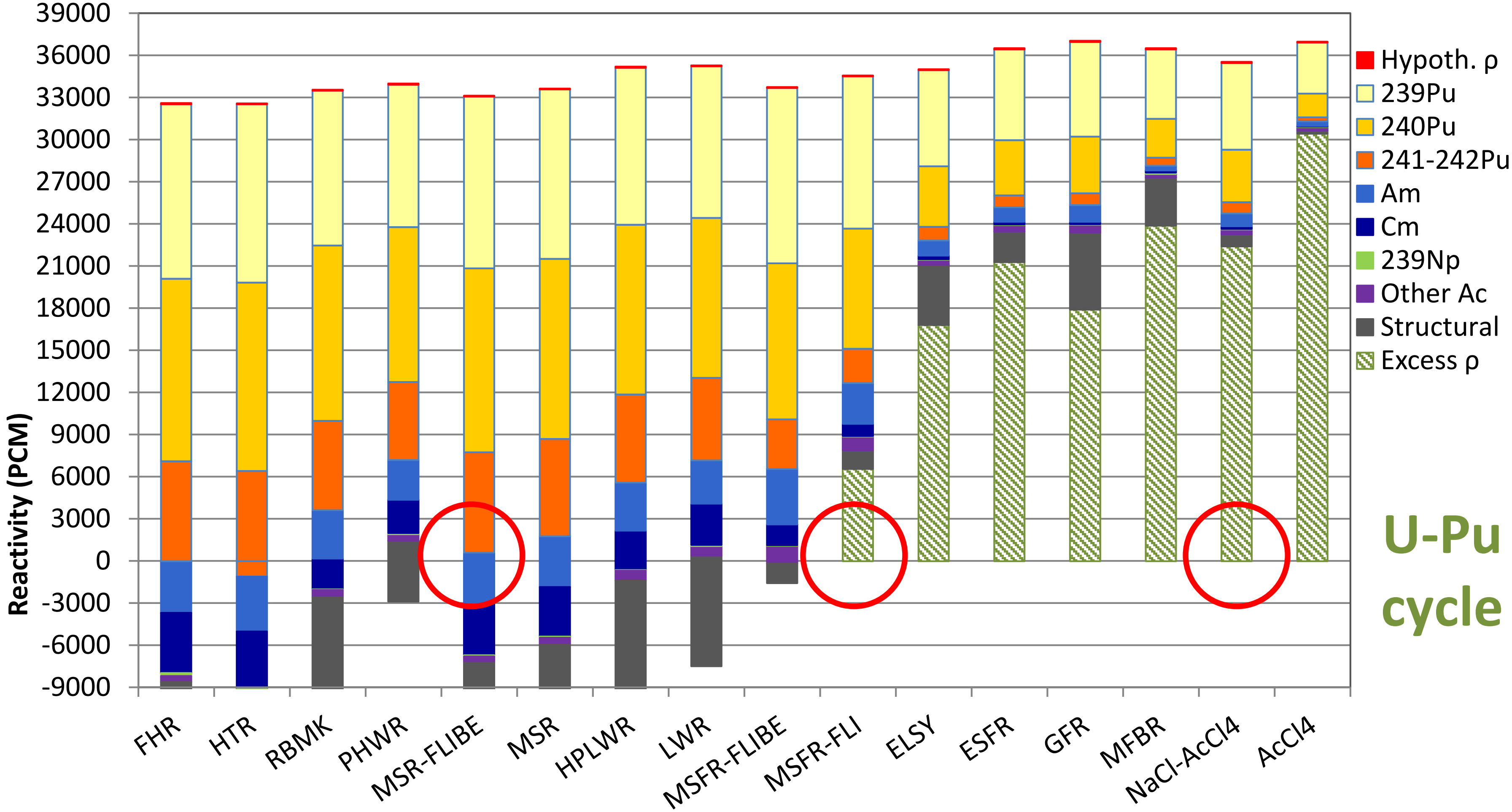
Excess reactivity in eql. cycle for U-Pu cycle

- Low ^{239}Pu fission probability: ^{239}Pu: 65-75% x ^{233}U: 90% => ~~thermal reactors~~.
- Excess reactivity is higher for fast reactors: ^{239}Pu: ν=2.9 x ^{233}U: ν=2.5
- U-Pu cycle has better neutron economy, Th-U cycle better neutron efficiency.



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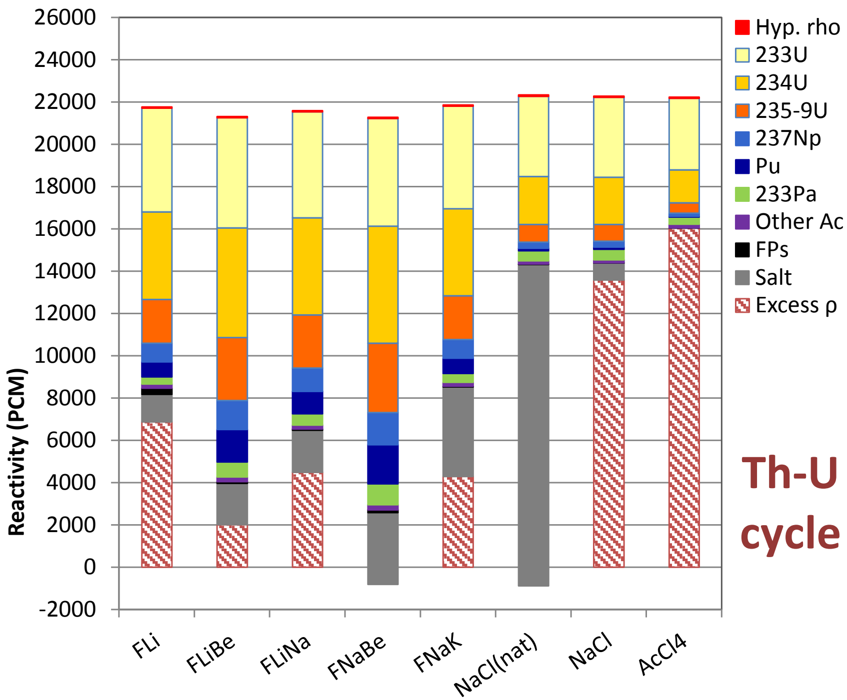
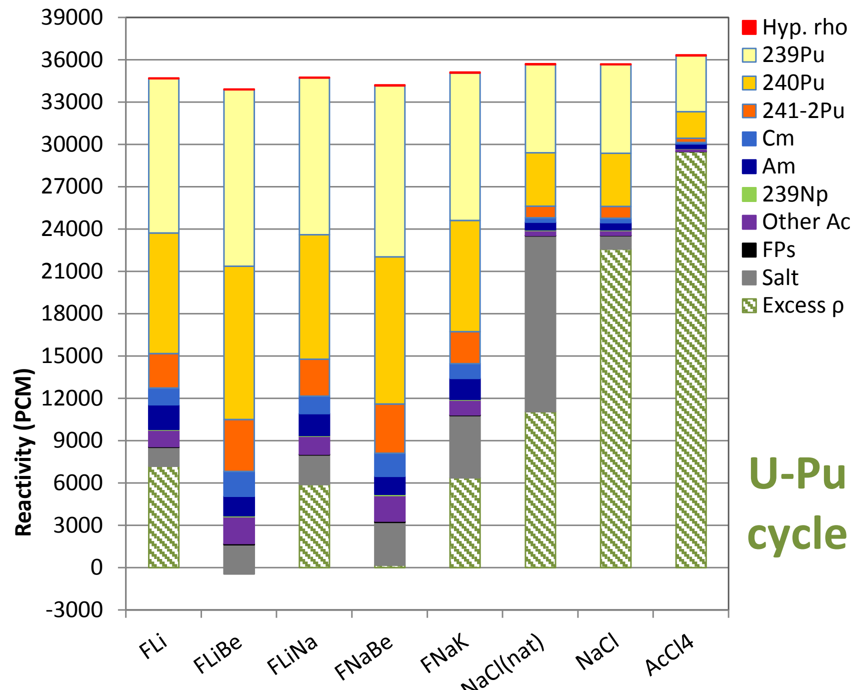
8 Fast MSR (salts) comparison - inclusive FPs

- **8 selected salts** were compared (**infinite medium** of fast reactor with FPs).
- **U-Pu** and **Th-U** equilibrium closed cycles were evaluated (by excess reactivity).
- It confirmed that for **U-Pu** cycle **chlorides** are preferable.
- The **reactivity excess in chlorides** may enable **breed and burn** mode.
- **Th-U** cycle has two favorites ^{7}**LiF** and **Na^{37}Cl** carrier salts.

^{233}**U**: *v*=2.5, fission probability 90% X ^{239}**Pu**: *v*=2.9, fission probability 65-75%





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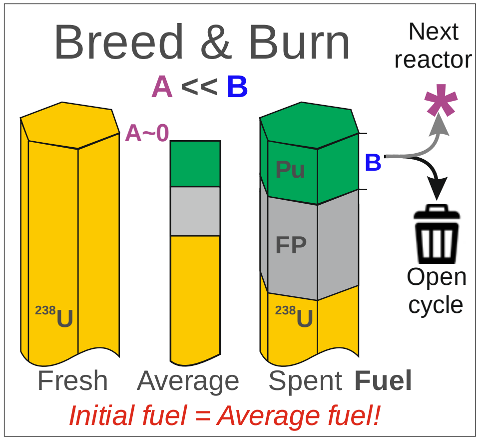
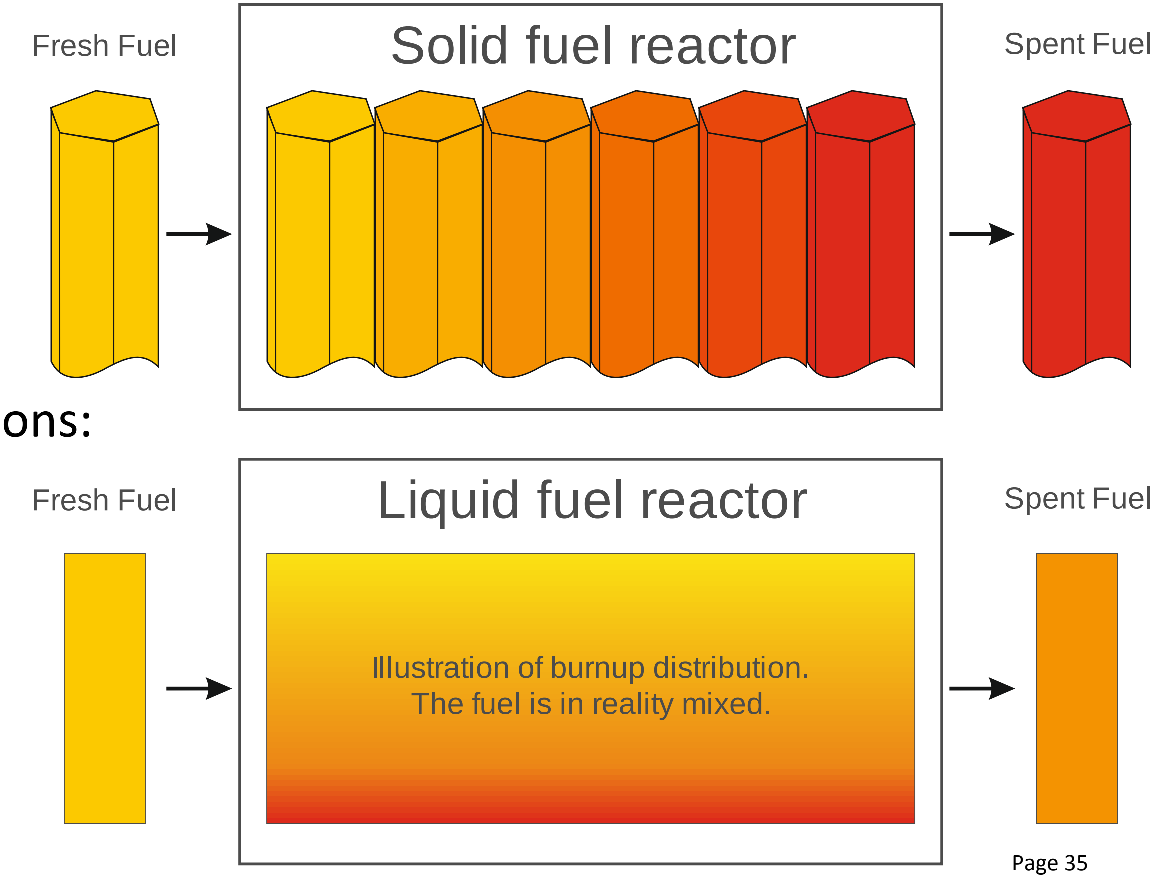
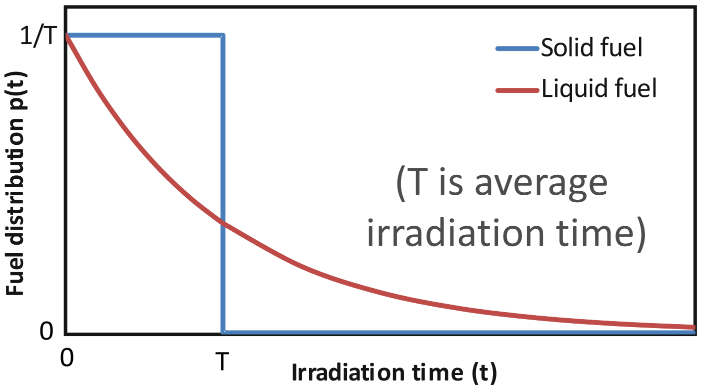
Breed and burn fuel cycle mode



- In case of a super breeder, fuel based only on fertile ^{238}U or ^{232}Th can be loaded to the reactor.
- The fissile fuel will be produced (**Breed**) in the reactor.
- Later during its **burn**ing it will supply enough neutrons to breed new fuel from new fresh fertile assemblies.
- In solid fuel case it looks like this =>
- The situation for liquid is different. Everything will be homogenized.
- Different burnup distributions:





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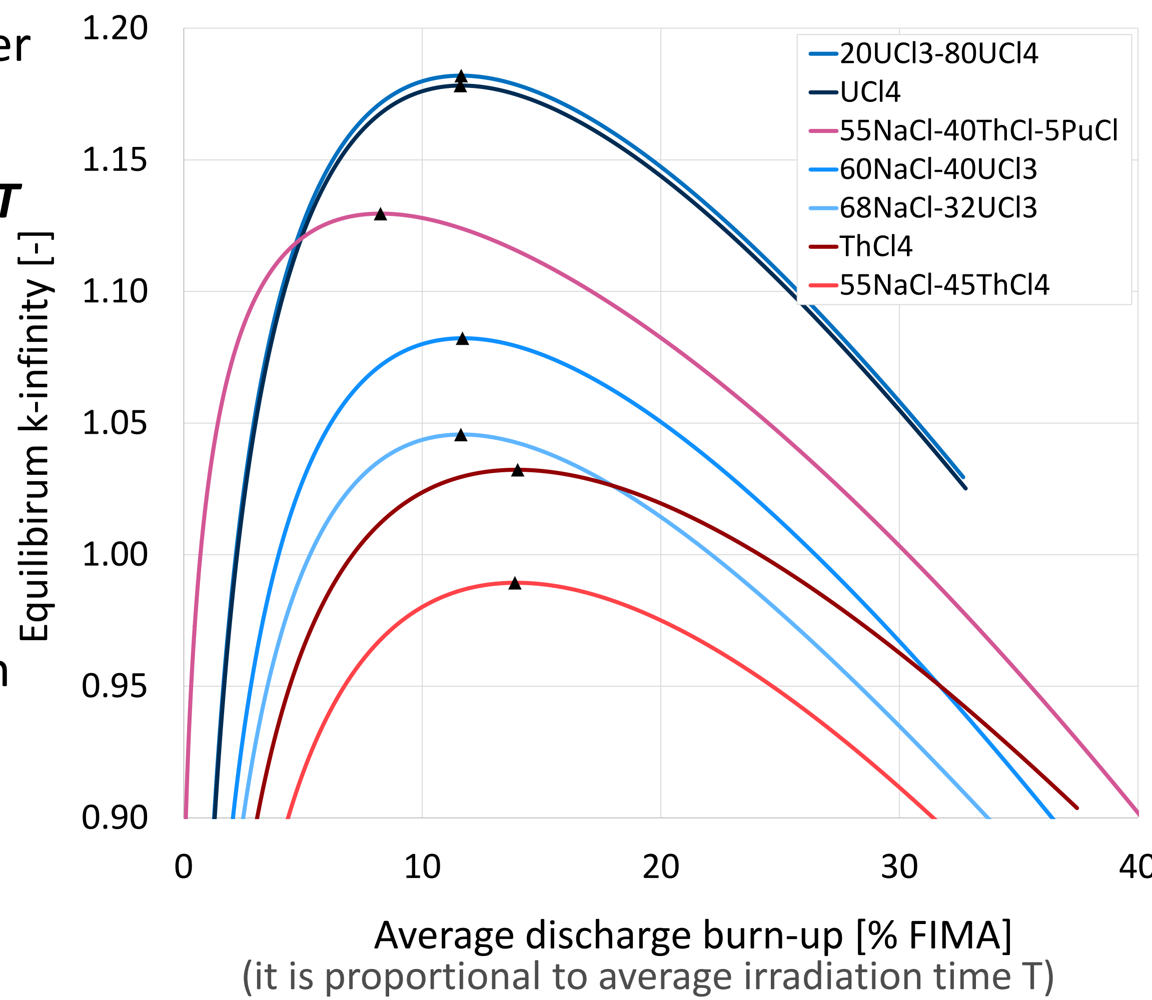
MSR in Breed-and-Burn (B&B) mode

- Method can be developed on the **burnup distributions** (which differ between solid and liquid fuels).
- The **average k-infinity** for given ***T*** can then be computed using a simple cell depletion calculation:

$$\bar{k}_{\infty}^{T} = \int_{0}^{\infty} p^{T}(t) k_{\infty}(t) dt$$

- **Main results on cell level:**
  - **B&B** mode is possible only with **enriched ^{37}Cl** based MSR.
  - U-Pu cycle is better that Th-U.
  - B&B in **Th-U** cycle may require **fissile support** (e.g. LWR Pu).

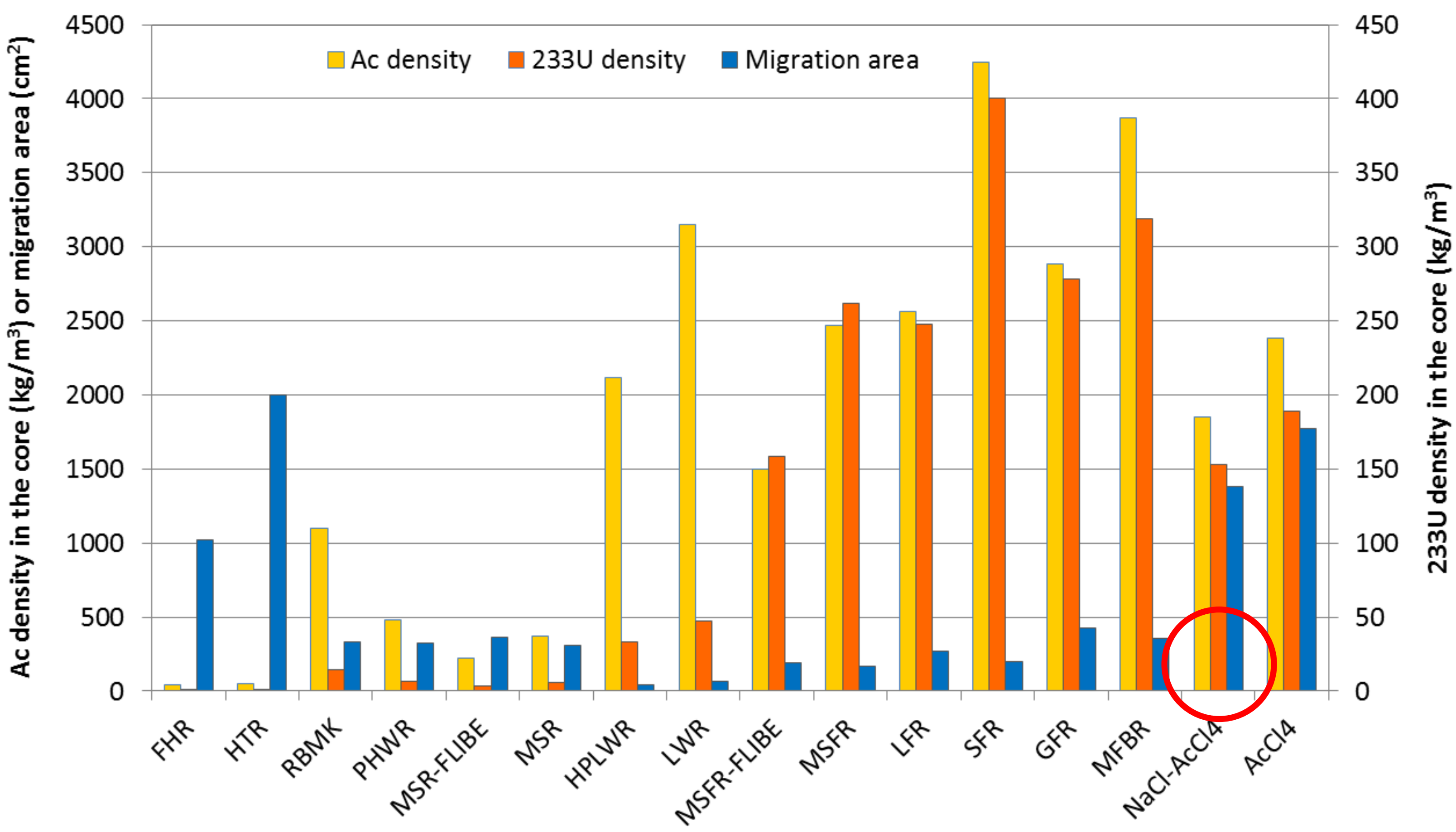


- **B&B mode** represent **open fuel cycle** with up to **20% resources** utilization.

Hombourger, B. et. al., Fuel cycle analysis of a molten salt reactor for breed-and-burn mode, ICAPP 2015, Nice

Chlorides disadvantage: density and migration area

- Chlorides salts have lower specific Ac density and higher migration area.
- Chlorides area transparent for neutrons (absence of scattering).
- High migration area => high leakage => blanket or reflector or bigger reactor.



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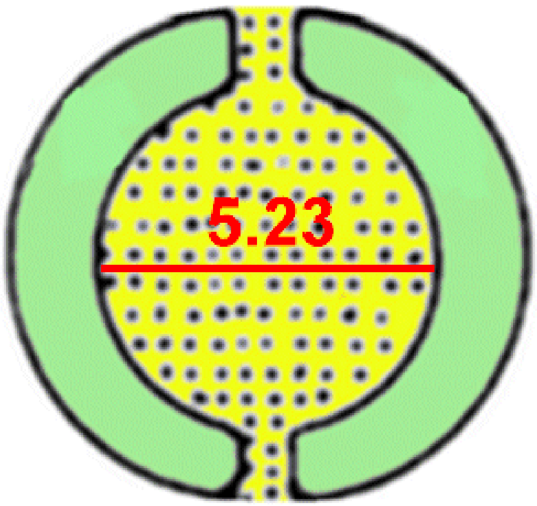
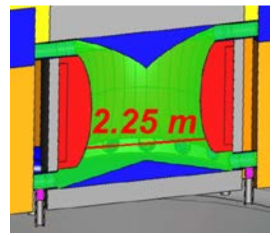
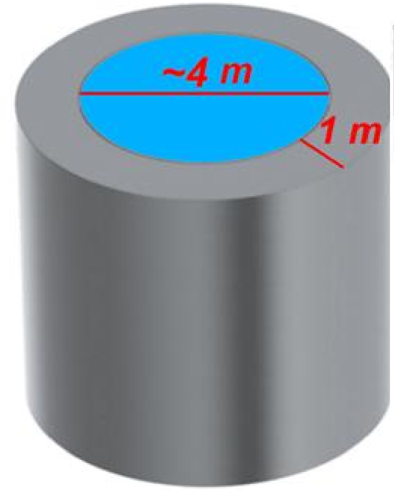
MSR Breed-and-Burn: core level







B&B – PSI test design

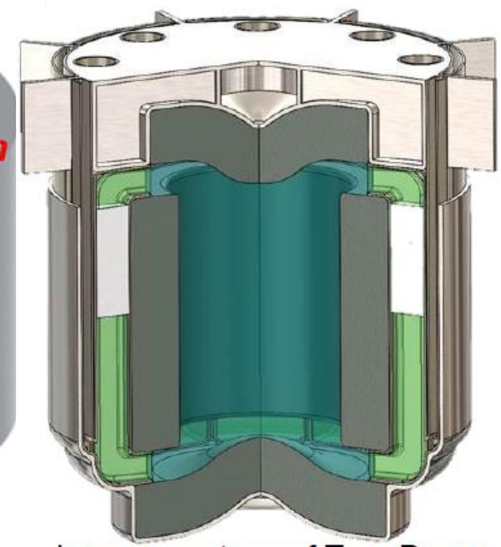


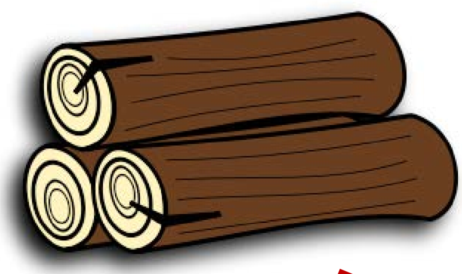
Image courtesy of TerraPower

Concept	SOFT-1980	MSFR	B&B - PSI	MCFR
B&B / salt	No / nat. chlorides	No / fluorides	Yes / enr. chlorides	Yes/ enr. chlorides
Core dimensions	5.23 m	2.25 m x 2.25 m	4 m x 4 m	?
Core volume	75 m^3	9 m^3	50 m^3	?
Blanket / cycle	None / U-Pu	7.3 m^3 / Th-U	None/ U-Pu, Th-U+Pu	None/ U-Pu
Reflector	$CaCl_2$-NaCl & steel	Axial only - Hastelloy	**Yes – lead / Enr. lead**	**Yes - ?**
Processing	Volatile & Soluble FP	Volatile & Soluble FP	Volatile FP only	Volatile FP + ?
Processing flow	0.25 L/s	3-8 L/day	2 L/day	?
Cycle time	?/continuous electrolysis	6-16 years	52 years	?

Breeding reactor - in ideal case = never-ending fire

Works only if emissions (FPs) are (continuously) removed

Fuel: ^{238}U or ^{232}Th



Energy

Reactor
("catalyzed" by ^{239}Pu or ^{233}U)

http://www.clipartpanda.com/categories/fire-clip-art-free-download

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Issue with closed cycle: reprocessing & fabrication

- Since **FPs absorb neutrons**, they sooner or later poison the reactor.
- Thus the **fuel**, which is highly radiotoxic, must be **reprocessed**, which is demanding and complicated.
- In **U-Pu** cycle recycling of Pu and U is **technologically mastered** and practically available in several countries.
- The by-products of the U-Pu, minor actinides **Am** and **Cm** emit α (heat source) and neutrons (mainly Cm). Their recycling may thus strongly **complicate the fabrication** of solid fuel.
- Similar technological experience as for U-Pu is missing for Th-U cycle.
- Furthermore, **irradiated Th fuel** emits more **hard gammas** from the ^{232}U decay chain, mainly from ^{208}Tl and ^{212}Bi.
- Recycling of **solid Th fuel** may be **demanding**.
- **Liquid fuel** (no fabrication) **can accommodate** both **MA** from **U-Pu** and **^{232}U** from **Th-U** cycles (recycling by volatilization) more easily.

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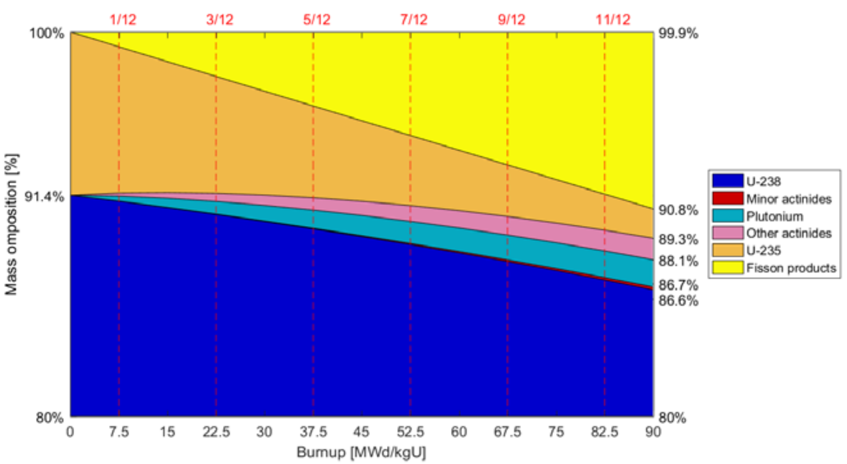
Fission products sensitivity

- **Fast spectrum systems** are **less sensitive** to fission products **FPs**…
- **Why?**
- The FPs **cross-section** are **higher** in thermal spectrum.
  (Isn't it valid for all cross-section, not only for FPs?)
- There is also **second reason**: FPs to fissile fuel ratio.
- In typical **thermal burner** (e.g. LWR or HTR *see the figure*) initial fissile load may be between **5-8%**. After discharge there are usually **2%** left.
- In fast breeder reactor (SFR) the fissile isotopes can represent **10%** of fuel and it is **the same** after **discharge**.
- The FPs to fissile ratio is thus:
  5/2 for LWR and **10/10** for SFR.
  (FPs replace fertile absorbers)
- Hence, thermal reactors, especially burners, are more sensitive to fission products.

HTR fuel with initial 8% enrichment



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Fuel irradiation time and need of recycling

In solid fuel reactors the irradiation time is limited by:

1. *Limited cladding lifetime caused by irradiation.*
2. *Fissile element load in burners (breeders can be self-sustaining).*
3. *Gaseous Fission Products (FPs) pressure.*
4. *Core poisoning by FPs neutron capture.*

In liquid fuel reactor:

1. *There is no cladding.*
2. *Breeders are self-sustaining and fuel or Th can be continuously added.*
3. *Gaseous and volatile FPs are continuously removed form the core.*
4. *Remaining FPs are still poisoning the core by neutron capture.*

- *In MSR case there is **not another reason** for fuel reprocessing **than FPs removal**.*

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Sal components and FPs removal

MSR fluoride salts components:

1. *Carrier salt (LiF, LiF-BeF_2, NaF-BeF_2, NaCl, etc.)*
2. *Fertile actinides (^{232}Th and ^{238}U).*
3. *Fissile fuel (mainly U or Pu vector).*
4. *By-products (MA).*
5. *FPs.*

FPs removal

- *There is not a simple method how to separate FPs from the fuel salt.*
- *Furthermore, even if several methods are combined, FPs are usually the last separated component.*
- *Practically in every MSR design study or simulation, the spent fuel salt is removed from the core for reprocessing being immediately replaced by the same cleaned salt.*
- *Since the whole fuel salt mix must be removed from the core, the question is what should be recycled, why, and for what price?*

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Motivation for salt recycling and recycling strategies

Why to recycle salt components:

1. *From a reactor physics point of view, it is* ***important to recycle*** ^{233}***U or*** ^{239}***Pu*** *as the main fissile elements; the other components are not substantive.*

2. *From a* ***sustainability*** *point of view, it may be important to recycle the main fertile elements* ***Th*** *or* ***U*** *and possibly some rare elements* ***(Li, Be****).*

3. *From* ***economy*** *point of view, it may be interesting to* ***recycle all*** *components. Nevertheless, it will depend on their price and on the* ***reprocessing costs****. In some cases their* ***direct disposal****, e.g. by vitrification,* ***can be cheaper****.*

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Operations with liquid fuel

Four possible basic operation with the liquid fuel:

1. *Salt removal from the core.*
   (no direct impact to the core neutronics)
2. *Salt cleaning inside of the core.*
   (direct impact to the core neutronics)
3. *Salt cleaning or reprocessing outside of the core.*
   (no direct impact to the core neutronics)
4. *Salt refilling into the core.*
   (direct impact to the core neutronics)

Recycling strategies:

Salt removal from the core	Removed salt share	Fissile fuel recycling (U-vector)	Fissile fuel return after reprocessing	Carrier salt cleaning	Carrier salt return after reprocessing	Reprocessing waste immobilization
Continuous or Batch-wise	From 0.1% to whole salt volume	In-situ or Ex-situ	ASAP or with months or years of delay	In-situ or Ex-situ	ASAP or with months or years of delay	In-situ or Ex-situ

- *Two extremes: on-line recycling – everything in-situ and ASAP, and off-line recycling – everything ex-situ and with years of delay.*

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Comparison of 7 similar salt treatment schemes (Th-U)

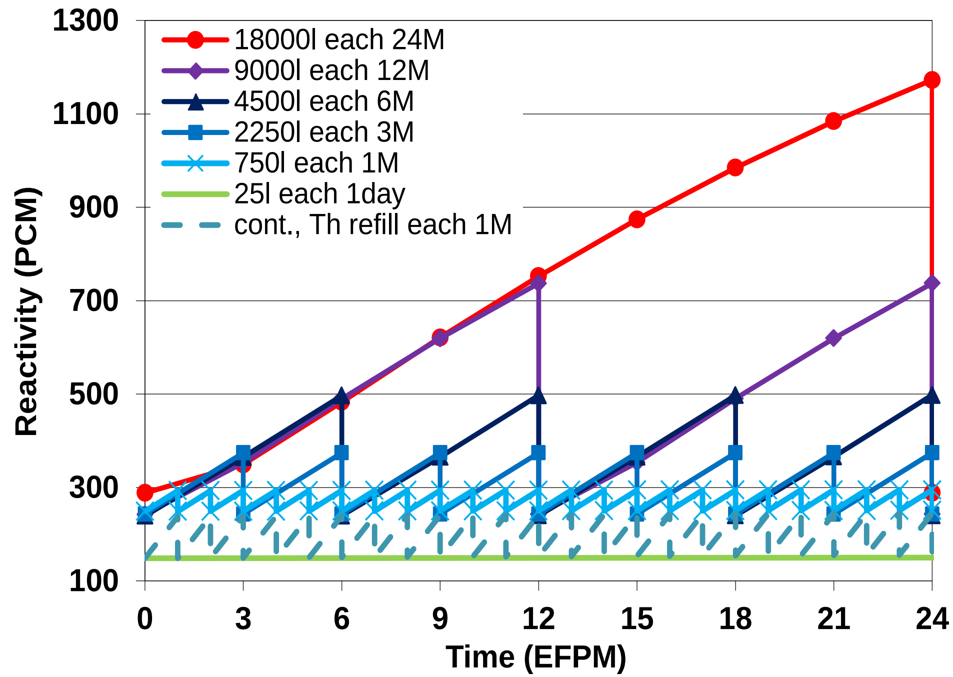
Assumptions:

- *Reprocessing unit capacity **25 l/day**.*
- *The volume for reprocessing is taken from **core** (cases 6 and 7) or from **temporary storage tank** (cases 1-5).*

Strategy Nr.	Salt clean-up from FPS	Th refilling	Min. salt volume for operation
1	18000l each 24M	each 24M	36 m^3
2	9000l each 12M	each 12M	27 m^3
3	4500l each 6M	each 6M	22.5 m^3
4	2250l each 3M	each 3M	20.25 m^3
5	750l each 1M	each 1M	18.75 m^3
6	25l each 1day	each 1day	18 m^3
7	continuous	each 1M	18 m^3

Main conclusions:

1. *Reactivity **swing is positive** and proportional to the reprocessing time.* *(decreasing Th mass = +2.2 PCM/kg; increasing FPs mass = -2.0 PCM/kg)*
2. *Continuous **Th refilling** can be used as **reactivity control**, independently off the selected salt clean up treatment.*
3. *The strategy with **longest reprocessing time** has **lowest average FPs content**.* *(it has also highest breeding gain)*
4. *Its **disadvantage** is the **biggest salt volume** (initial load) necessary for reactor operation.*



Reactivity swing for 7 recycling strategies

Krepel, J. at. al., Comparison of Several Recycling Strategies and Relevant Fuel Cycles for Molten Salt Reactor. ICAPP 2015 Nice

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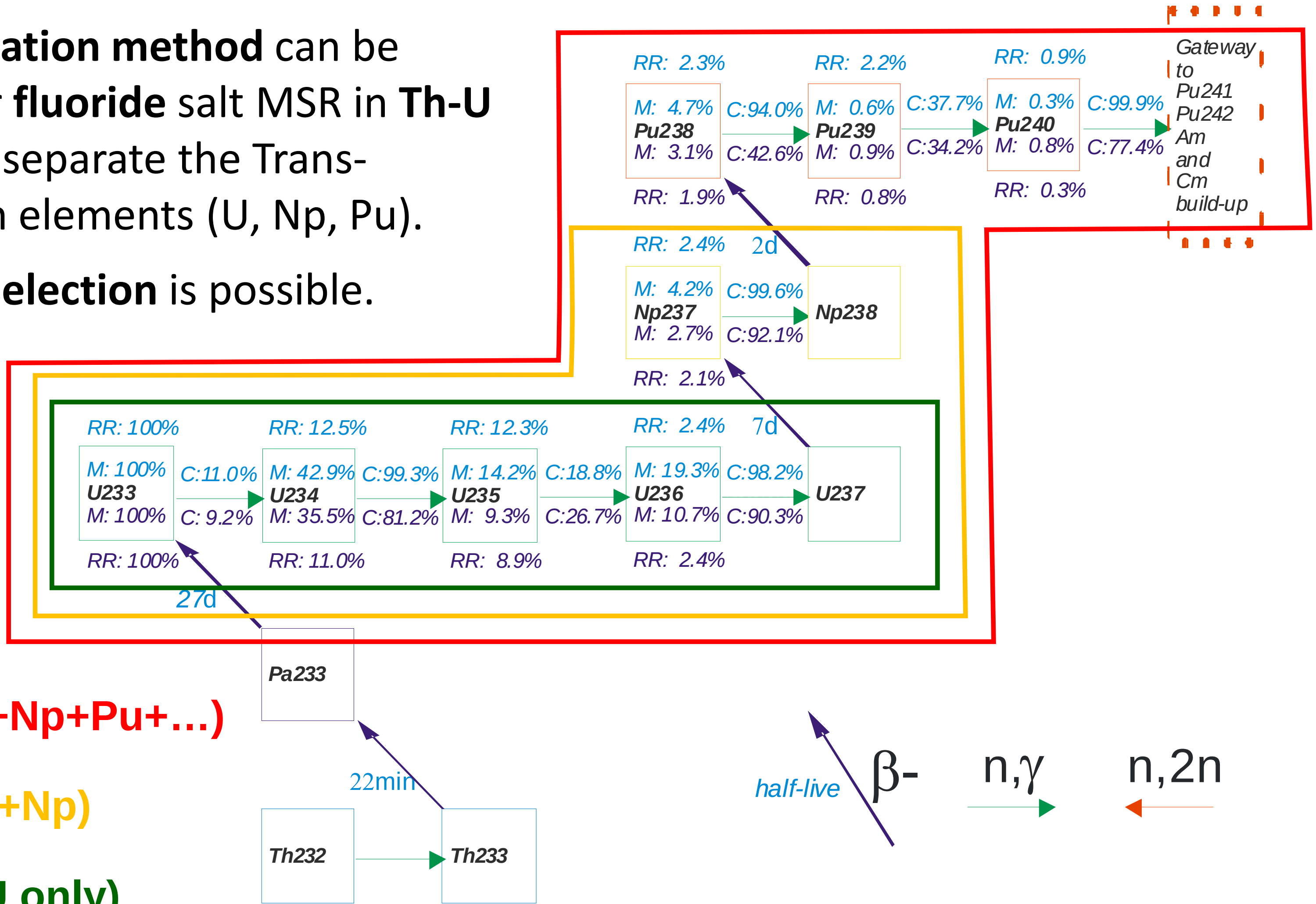
Combined recycling: fissile in-situ ASAP, the rest ex-situ

- **Volatilization method** can be used for **fluoride** salt MSR in **Th-U** cycle to separate the Trans-Thorium elements (U, Np, Pu).
- **Partial selection** is possible.



Scenario I. (U+Np+Pu+…)

Scenario II. (U+Np)

Scenario III. (U only)

Assumption for the simulation: repetitive application of reprocessing: U 0% other Ac 1% loss. FPs 99% removal efficiency are replaced every 12 months by Th.

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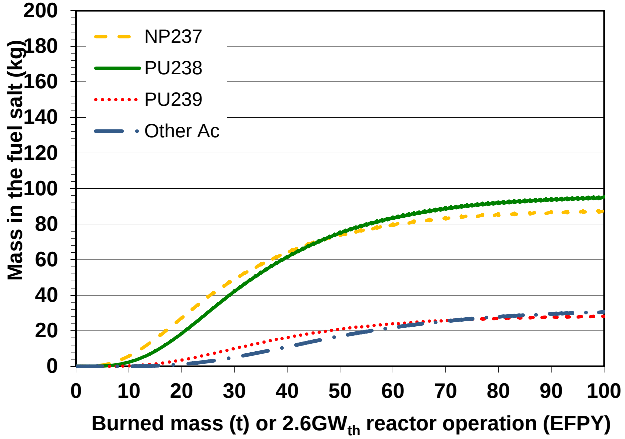
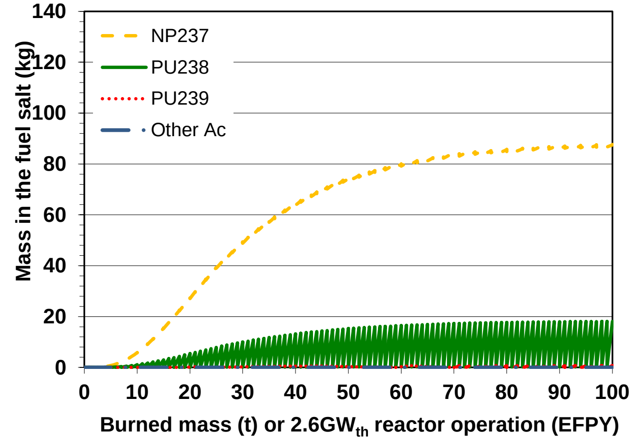
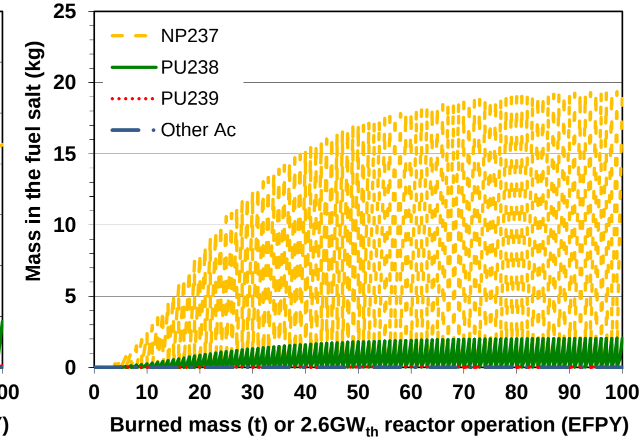
Combined recycling: Fissile in-situ ASAP, the rest ex-situ

Fast spectrum MSR core

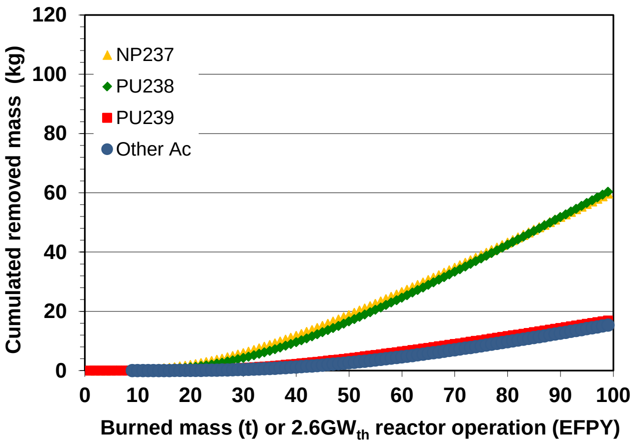
Scenario I. (U+Np+Pu+…) **Scenario II. (U+Np)** **Scenario III. (U only)**

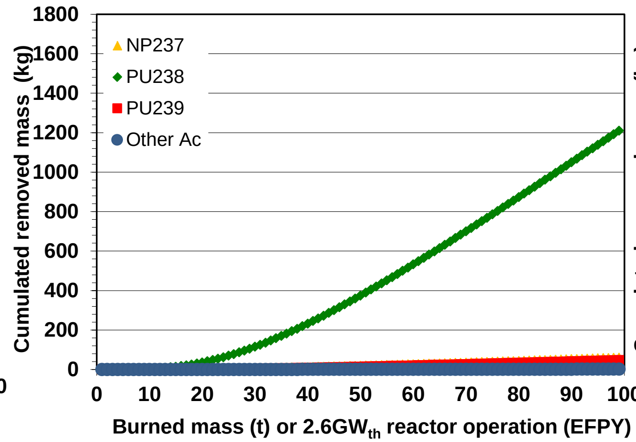


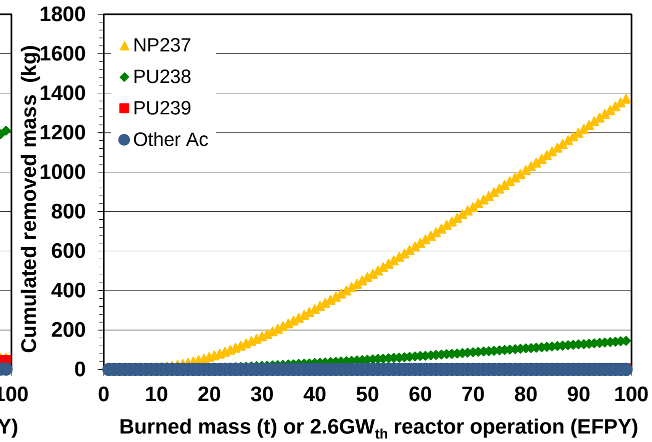




Ac mass in the core







Cumulative Ac mass in the waste 100EFPY

Krepel, J. at. al., Molten Salt Reactor with Simplified Fuel Recycling and Delayed Carrier Salt Cleaning. ICONE 2014 Prague

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Combined recycling: radiotoxicity of cumulated waste

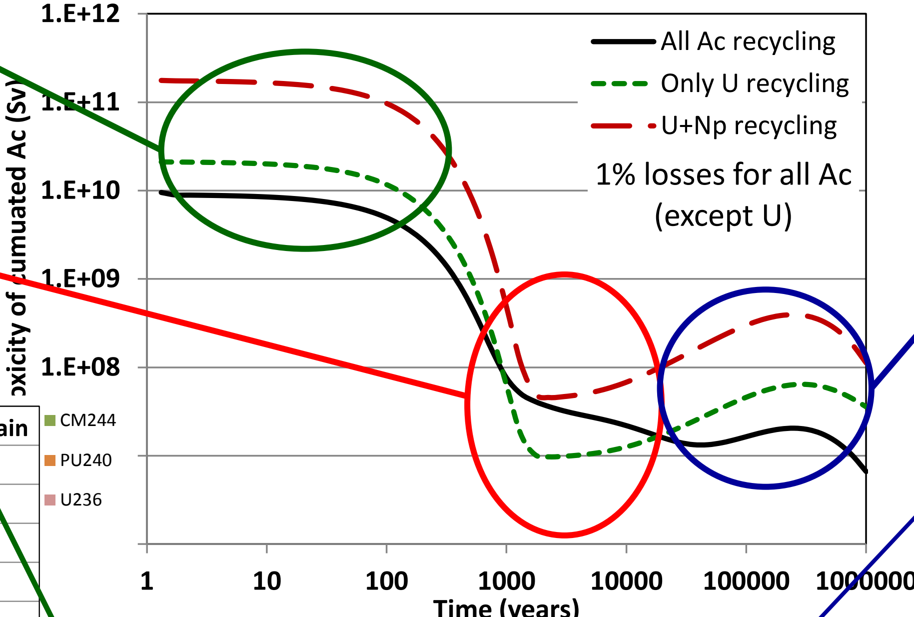
Difference due to ^{238}U decay chain

Mainly ^{238}Pu

Difference due to ^{232}Th and ^{235}U decay chains

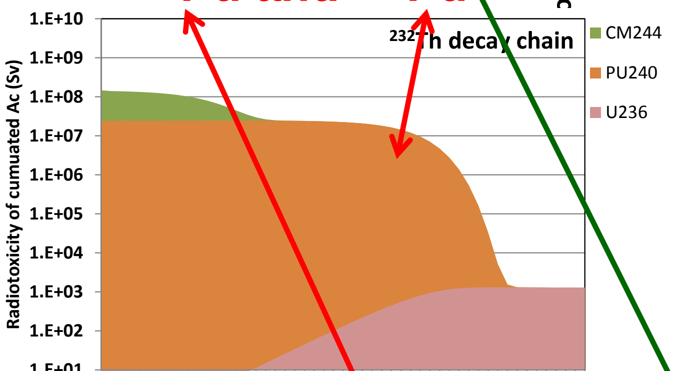
Mainly ^{239}Pu and ^{240}Pu

Fast spectrum MSR core

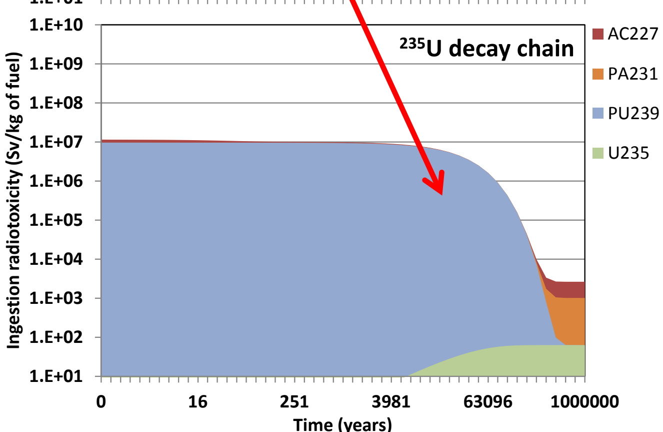


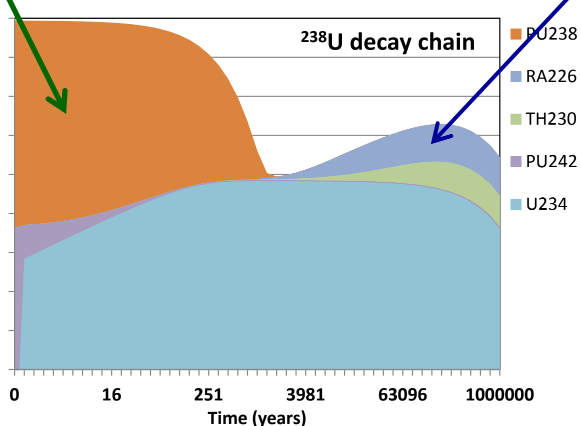
Difference due to ^{238}U and ^{237}Np decay chains

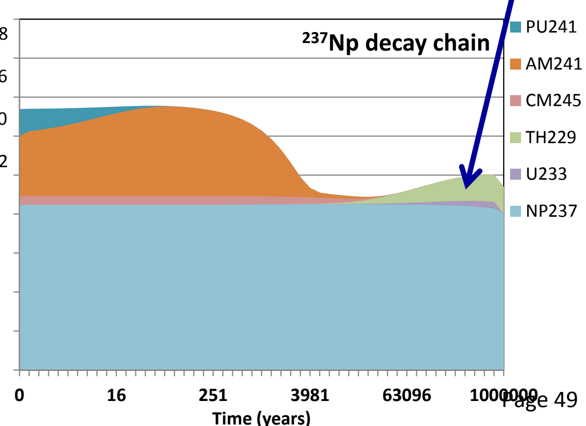
Mainly ^{226}Ra and ^{229}Th



All Ac recycling case







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Conclusion

- Sustainability of ^{235}U fueled reactors is low.
- ^{232}Th and ^{238}U can be burned in fast (^{232}Th possibly also in thermal) breeders; with fuel recycling high sustainability may be achieved.
- Fast spectrum breeder with solid fuel may have safety issues.
- These issues may be eliminated by liquid fuel.
- The liquid fuel state also provide fuel cycle flexibility.
- Several cleaning, reprocessing, and refilling / removing techniques may be applied to liquid fuel.
- Solubility and other thermochemical properties may be the limiting factor.
- MSR may combine sustainability with acceptable safety, economy and proliferation resistance.

MSR is a very promising energy source.

It can combine unparalleled safety features with high fuel utilization.

It can also provide us enough time for mastering of the nuclear fusion! ☺

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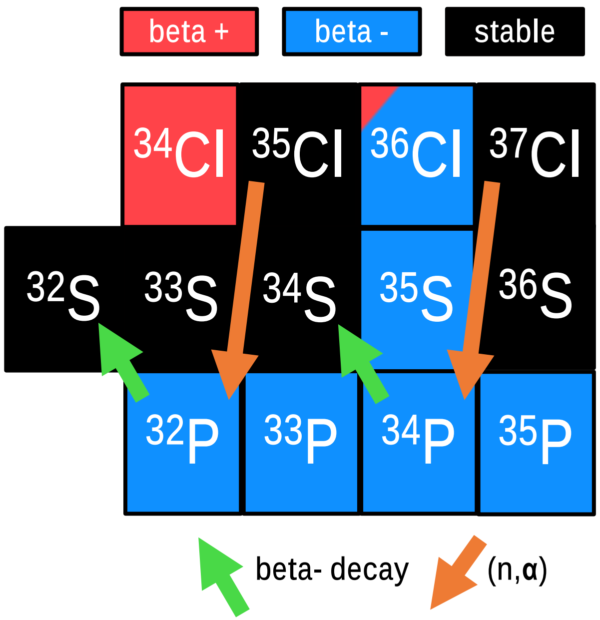
Sulfur production in Chloride MSRs

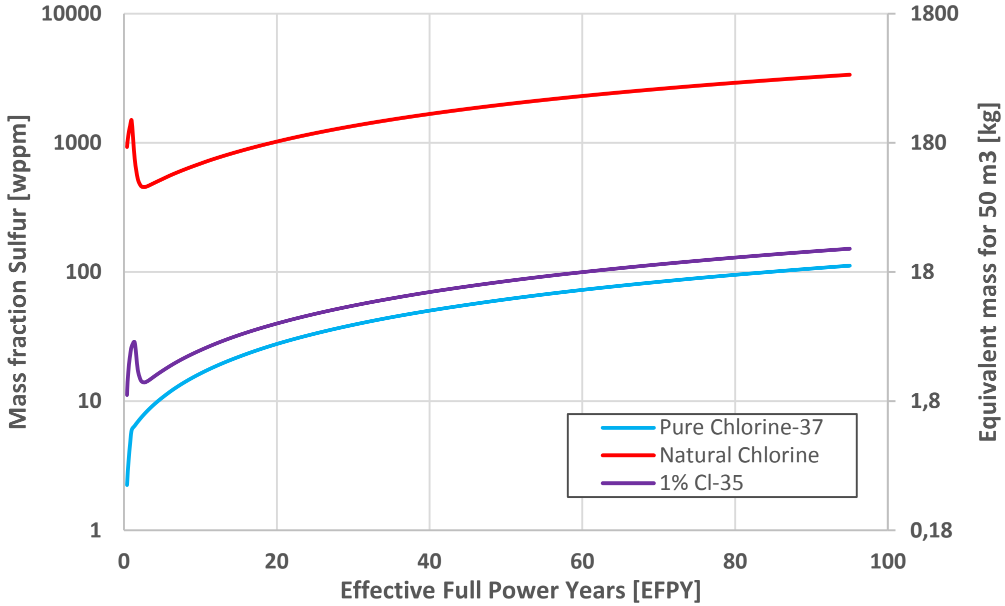
- Sulfur embrittles steels & nickel alloys
- Main production paths:

$$^{35}Cl + n \rightarrow {}^{32}P + \alpha \rightarrow {}^{32}S$$

$$^{37}Cl + n \rightarrow {}^{34}P + \alpha \rightarrow {}^{34}S$$

- Enrichment in Cl-37 substantially decreases Sulfur production.





- Investigation on the speciation of Sulfur were carried out at EIR in the seventies.

[1] IANOVICI, E., TAUBE, M., Chemical behaviour of radiosulphur obtained by 35Cl(n, p)35S during in-pile irradiation, J. Inorg. Nucl. Chem. **37** 12 (1975) 2561.

[2] IANOVICI, E., TAUBE, M., Chemical State of Sulphur Obtained by the 35Cl(n,p)35S Reaction during In Pile Irradiation, EIR-Bericht 267, Eidgenössisches Institut für Reaktorforschung, Würenlingen, Switzerland (1974).