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ADS Physics: Physics and Dynamics.
Part IV

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Set III:
RADIO- WASTES AND TRANSMUTATION POTENTIAL OF HYBRIDS (ADS)

III.1. INTRODUCTION.

General characteristics of nuclear wastes and their origins: risks of the long-life wastes, activity and toxicity waste levels and their natural decays. Quantitative Measure of Radiotoxicity (R)

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SET III

III.1. INTRODUCTION

A major issue to secure the development of nuclear energy in future, is the radioactive waste minimization, both inside the fuel cycle and in the deep geological storages.

Most of the research activities have been devoted to assess the potential benefits of the so-called partitioning/transmutation technologies.

One needs to establish the physical principles which provide an "inherent" minimization of the radioactive wastes.

Several groups of the toxicity sources are considering as an object of studies: they are:

- **Actinides as a part of nuclear fuels which is remaining unusable after irradiation in Nuclear Power plants,**

- **Long-Lived fission products (LLFP) as the inevitable result of nuclear energy production,**

Quantitative Measure of Radiotoxicity (R)

For each radio-nuclide, the contribution dose is proportional to the integral activity **A** (in Bq), where the coefficient is called as the Dose Factor (F_d) "via ingestion" or "via inhalation" (in Sv/Bq):

$$R(Sv) = F_d(Sv/Bq) \times A(Bq)$$

Table. Periods and Factors of dose "via ingestion"

Nuclides	Life period (years)	F_d	Emission
Tc-99	2×10^5	0.64×10^{-9}	β-
I-129	1.6×10^7	0.11×10^{-6}	β-
Pu-238	88	0.23×10^{-6}	α
Pu-239	2.4×10^4	0.25×10^{-6}	α
Am-241	432	0.20×10^{-6}	α
Am-243	7.4×10^3	0.20×10^{-6}	α
Cm-244	18	0.16×10^{-6}	α

Nuclide chain evolution (a long-term case in Prof. P. Ravetto designation) can be described by the known Bateman equation to represent the time dependent number of each nuclide N_i:

$$\frac{dN_i}{dt} = -\lambda_i N_i + \sum_{j \neq i} \lambda_j R_{j \to i} N_j$$

Hence, the radio-toxicity of each nuclide varies with time as the following:

$$R_i(t) = F_d(i) \times \lambda_i N_i(t)$$

where N_i is defined by the Bateman equation.

Usually, this toxicity evolution value is normalized on either **electricity production** or on the initial mass unit.

III-1. Transmutation requirements: overall potential of the supplementary neutron production: thermal reactor cores versus fast reactors, general impact of reactor/fuel cycle parameters on the transmutation potential

Physics of the Toxicity Reduction via Transmutation/Incineration

Neutronics plays a most important role in the assessment of the transmutation potential of nuclear power (NP):

transmutation requires neutrons!

For example, long lived fission products (LLFP) incineration is directly defined by the neutron surplus available in the given nuclear system. Every fission produces a fraction of long lived nuclei, however,

the same fission can produce some neutron surplus for elimination of these long lived nuclei.

This "*incineration*" potential can be increased by an "*external neutron source*" using a part of fission energy released.

The assessment of the neutron needs for transmutation of all long lived fission products (produced by one fission in NP) gives a value of at least 0.3÷0.5 neutrons/fission (see later).

Taking into account the large inventory of the overall fission products release, one should provide a neutron surplus as high as possible. As for fuel waste (Minor Actinides - MAs) transmutation, this potential is very sensitive to neutron spectrum : in some spectra and for some MAs nuclei, transmutation needs a neutron surplus, in other cases it does not.

Moreover, the toxicity reduction factor after irradiation of an initial nucleus ("*father*") in a neutron flux depends strongly on nuclear properties of all their "*family*" in a given flux.

Hence, **it is important to know how many neutrons can be produced from the transmutation of every nucleus during its life under neutron flux and what is the level of "family"'s toxicity achieved at the end.**

Methods of assessments of the overall transmutation potential of NP

The management of transuranics (TRU) is a major challenge for the future of nuclear power development.

The physics approach is also useful here, and in order to intercompare different reactors at the "equilibrium" (after some transitions) one needs to solve the standard Bateman equations, taking into account different strategies:

fuel cycle type, intention for selective TRU reprocessing, fuel feeds, burn-up, etc.

The Neutronic Approach – D-Method

Why equilibrium?.....

One shall consider equilibrium states of all "*families*" whose "*fathers*" are used as a fuel feed of nuclear systems: nuclear critical reactors, subcritical accelerator driven hybrids, etc.

It allows understanding the role of each feed component in fuel waste toxicity production!

The following definitions are valid when considering an equilibrium state :

Definition 1:

J-family is a set of nuclei (including the initial nucleus J) which have been produced both by transmutation of the J-nucleus in a neutron flux Φ of as well as of its subsequent products, and by decays of these nuclei (no flux importance case).

Definition 2:

The equilibrium state of the J-family in a given nuclear system defines the concentration distribution N among J-family members for the asymptotic condition ($t \rightarrow \infty$) when a given system is continuously fed by the J-nucleus only.

The processes of transmutation of the incoming nuclides (the "fathers") under neutron flux according to their nuclear chains gives rise to "families" which are independent at the equilibrium.

Hence, one can consider the "transmutation behaviour" of each "family" separately. During its life in a core, the father and his family are producing neutrons up to their complete disappearance.

There are the following reasons for **family disappearance (sink)**:

- **via fission, at least one of its members**
- **natural decay,**
- **fuel cycle operational procedures, either discharge to storage or fuel losses during reprocessing.**

Multiple nuclear reactions lead to different branches of family members: some of these reactions consume neutrons through neutron capture (e.g., (n,γ) reactions), other paths produce more neutrons (e.g., fission, (n,2n) reactions) than consumed, and some (e.g., (n,n) reactions, decay) do not influence the total neutron balance.

The total number of neutrons D_j consumed by the given J-family can serve as an indicator of the capability of a core to achieve destruction of a given J-feed if there is neither "parasitic" neutron consumption nor neutron leakage.

In the case of a <u>negative D</u> value (i.e., when the J-family produces more neutrons than it consumes), the core fed by J-nuclides produces enough neutrons to destroy the "source material" at equilibrium if the neutron excess compensates for parasitic captures (e.g. by structures, fission products etc) and for neutron leakage.

In the case of a <u>positive D</u>, the neutron consumption in the fuel dominates over neutron production and the core requires a supplementary neutron source to support transmutation.

The procedure of the D_j calculation can be illustrated by the following scheme:

$$D_J = \sum_{J1_i} P_{J\to J1_i} \left\{ R_{J\to J1_i} + \sum_{J2_k} P_{J1\to J2_k} \times \left[R_{J1\to J2_k} + \sum_{J3_n} P_{J2_k\to J3_n} \left(\dots \right) \right] \right\}$$

where $P_{JNr \to J(N+1)s}$ is the probability of transmutation of the nuclide J_{JNr} (belongs to the number N of the chain J) into nuclide J(N +1)s (belongs to the number N+1 of the chain J). All these nuclides are the members of the J-family.

$R_{A\to B}$ is the neutron consumption factor showing the number of neutrons consumed during transition A→B and depending on the nuclear reaction type

(1)

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When calculating, the neutron consumption for each reaction type are defined as follows:

- **neutron capture (n,γ) with 1 neutron being captured,**
- **neutron capture and subsequent multiplication (n,mn) with (1-m) neutrons being produced,**
- **fission with (1-ν) neutrons being produced,**
- **natural decay with 0 neutron being captured,**
- **discharge and nuclide loss with 0 neutron being captured, however nuclide disappearance nas to be taken into account**

Because of all feed components (families) are irradiated independently, one gets the total neutron consumption of the core at equilibrium (D_{fuel}) as

$$D_{fuel} = \sum_J \varepsilon_J D_J \qquad (2)$$

where ε_J is the fraction of J-family in the feed stream.

Global neutron balance of a core must consider the total neutron production (consumption) of the fuel families, the parasitic captures of other core components (C_{par}) and of the accumulated fission products (C_{FP}) and neutron leakage (L_{core}).

The general equation for the Neutron Surplus (NS_{core}) then becomes:

$$- D_{fuel} - C_{par} - C_{FP} - L_{core} +$$

$$\text{EXT.SOURCE (ADS !)} =$$

$$NS_{core} \quad ; \qquad (3)$$

$$- D_{fuel} - C_{par} - C_{FP} - L_{core} = G_{core}$$

For a critical system, the neutron surplus must equal zero; and the neutron production of the fuel feed (as calculated in Eq. 2) must be sufficient to overcome the other loss terms.

Thus, D_{fuel} ,G_{core} are useful parameters for quantifying the transmutation potential
because one knows the overall neutron production by the fuel and by cores

Similar calculation can be realized respecting neutron consumptions of fission products

The "Equilibrium" E-Method

The standard Bateman equations can be **generalized** to account for the characteristics of specific fuel cycles and solved for the equilibrium concentrations of the actinide nuclides ("long-term" dynamics according to Prof. P. Ravetto definitions)

If a core is fed by a source S_j of J-nuclides, under neutron flux Φ, the fuel concentrations N_J the J-family are time-dependent and described by the following matrix-type – **Generalized Fuel Cycle equation**:

$$\frac{\overline{\partial N_J}}{\partial t} = \left(\hat{M} \times \Phi - \hat{\Lambda}_n - \hat{\Lambda}_{dl} \right) \overline{N}_J - \overline{S}_J \tag{4}$$

where $\overline{S}_J$ is the nuclei feed vector with a single non-zero component J, and $\overline{N}_J$ is the vector of the atomic concentrations of the J-family members including its "father".

$\hat{M}$ is the matrix related to **all nuclear interaction processes** under the neutron flux. It is composed of the one-group absorption cross section on the diagonal and off-diagonal terms representing the transmutation to different isotopes.

The $\hat{\Lambda}_n$ and $\hat{\Lambda}_{dl}$ are matrices of the family natural decays and of fuel discharge-losses **(!!!!).**

The λ_{dl} elements of the matrix $\hat{\Lambda}_{dl}$ are defined by the ratio of nuclides loss (the transfer to a repository or reprocessing technology wastes, nuclear decay during fuel "cooling" time interval τ_{cool}) and it depends also upon the discharge "frequency" τ_{core}.

A general expression λ_{dl} for each nuclide and for all fuel cycles is given by:

$$\lambda_{dl} = \frac{1}{\tau_{core}}\left\{loss + \left(1 - loss\right)\left[1 - \exp\left(-\lambda_n \ \tau_{cool}\right)\right]\right\} \quad (5)$$

where "loss" could be assessed as the fraction of the nuclide inventory which is lost during reprocessing or send to a repository ;

$\left(1 - loss\right)\left[1 - \exp\left(-\lambda_n \ \tau_{cool}\right)\right]$ describes the isotopic decay during storage and processing.

An open (once-through) fuel cycle for a nuclide means that this nuclide, being discharged, is sent to a repository and its loss term is equal to 1.

A closed fuel cycle for a given nuclide, means that this nuclide, after cooling and inevitable losses due to natural decay and after reprocessing with the relatively small losses, will return back to the core **(for example, loss = 0.001 if 0.10 % of this nuclide is lost during fuel reprocessing and 99.9 % is returning back to the core).**

Transition (due to the natural decay of a family during cooling time) to other nuclides can be taken into account by a matrix-type representation of the "operator" λ_n in Eq. 5.

The general solution of these equations is time-dependent and of an exponential type. **The asymptotic ($t \to \infty$)** solution (if all operators are time independent) corresponds to the "equilibrium" case i.e. when $\frac{\partial N}{\partial t} = 0$ in Eq. 4.

This equilibrium solution can be presented in the following matrix form,

$$\overline{N} = \hat{A}^{-1}\,\overline{S}$$

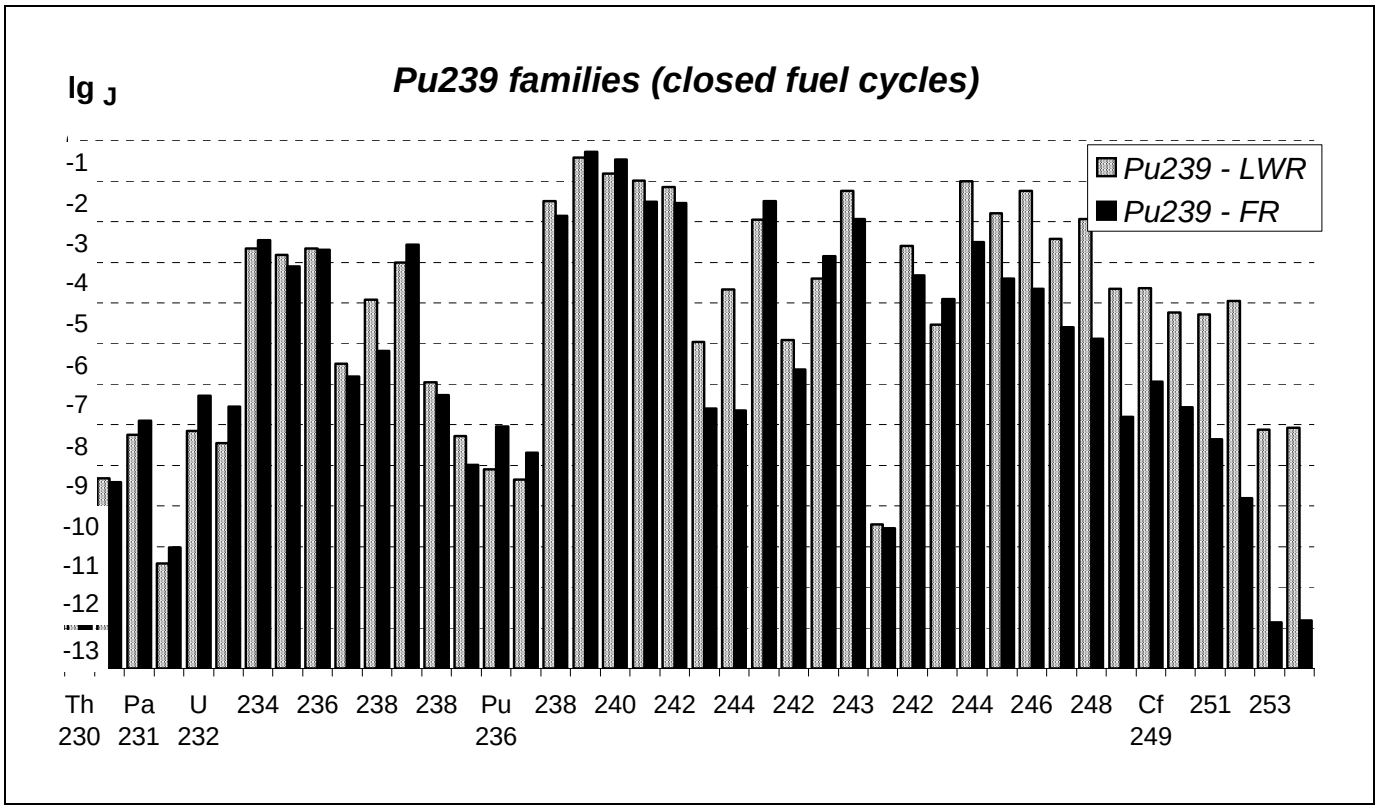
where $\overline{S}$ includes all "fathers" nuclei contained in the core feed and

$$\hat{A} \equiv \hat{M} + \hat{\Lambda}_n + \hat{\Lambda}_{dl}\ .$$

The linear character of the equation allows evaluation of the nuclear concentrations of all families independently, with a "unit" source for the corresponding feed:

$$S_J^{unit} = 1.$$

E-method presents the equilibrium composition which can be used to derive transmutation performance parameters and derived core parameters.



Neutron Balance Intercomparison

The physics approach has been applied to a large variety of reactors, both with thermal and fast spectra:

Table: D_{fuel} (net neutron consumption/fission) value for different isotopes in different systems

Isotope	MOX-LWR	He-cooled carbide fuel FR[3]	SUPER-PHENIX[3]	Lead-cooled nitride fuel FR[3]	Na-cooled oxide fuel FR[3]	Na-cooled metal fuel FR[3]
U-235	-0.38	-0.84	-0.86	-0.92	-0.95	-1.04
U-238	0.068	-0.63	-0.62	-0.71	-0.79	-0.90
Pu-239	-0.64	-1.44	-1.46	-1.58	-1.61	-1.71
Am-241	0.93 **requires neutrons**	-0.56	-0.54	-0.65	-0.77	-0.91
Am-242m	-1.56	-2.03	-1.87	-2.08	-2.10	-2.16
Am-243	0.36 **requires neutrons**	-0.84	-0.65	-0.85	-1.01	-1.15

The results allow comparison of the feasibility of transmutation of the different isotopes in each reactor concept. Significant variations in the D-factor are observed between the fast reactor concepts.

In general, a harder neutron spectrum leads to a more favorable neutron balance; thus, the metal-fueled FR provides the most excess neutrons for every actinide isotope.

Results confirm the trends that fast neutron spectrum systems have a marked advantage in terms of neutron balance over thermal systems.

Fuel Family Overall Neutron Consumption in a fast spectrumADS-blankets

Decay, irradiation and regular discharge (or losses) of every nucleus J (family "*father*") in a neutron flux produce at equilibrium (see equation (1)) the J-nucleus family.

Then one can calculate the total number of neutrons D_J which have been consumed by this family to be transmuted to a stable or short lived nucleus and, later on, extracted from this system).

If a given system is fed by a set of J-nuclei then the potential neutron surplus G will be defined by a sum of all D_J (neutron consumption), the neutron losses of this system (neutron leakage and neutron capture in construction materials,

(usually equal ~ 0.3 neutrons/fission for practical systems)

and supplementary neutrons (μ) (if this system is driven by some nuclear reaction as e.g. by spallation in ADS) :

$$G\,(neutrons/\ fission) = -\sum_J X_J \cdot D_J - 0.3 + \mu =$$

$$\sum_J X_J \cdot G_J + \mu = G_0 + \mu \quad ,$$

where X_J is a fraction of J-nucleus in fuel feeding.

REMARKS concerned the ADS further on motivations:

- ADS are able to compensate neutron balance production in the case of TRPu appearance in the cores of critical reactors;

On Irradiated Fuel Toxicity

The unit mass toxicity calculations at equilibrium (closed fuel cycle) show that **majority** of heavy nuclei, after irradiation in both fast and thermal fluxes, **increase their toxicity** per mass unit.

Open fuel cycle (with a feeding by relatively low toxic "initial" nuclei such as ^{235}U, ^{238}U and even toxic ^{239}Pu) cannot avoid this undesirable growth in fuel discharge toxicity unless an extremely high burnup case.

Ways to fuel toxicity reduction are:

- **closing the fuel cycle**
- **using fast spectra and**
- **providing sufficiently low levels of fuel losses during reprocessing.**

Table

Normalized toxicities of J-families T_J at equilibrium (Sv/gram) on Short ($10^2 \div 10^4$ years) and Long ($10^2 \div 10^6$ years) time intervals

(negligible losses rates)

	Thermal Reactors ($\Phi = 10^{14}$ n/cm^{2}s)		Fast Reactors ($\Phi = 10^{15}$ n/cm^{2}s)		Natural Toxicity ($\Phi = 0$ n/cm^{2}s)	
"*fathers*" of families	T_J Short	T_J Long	T_J Short	T_J Long	T_J Short	T_J Long
^{232}Th	21	4.6	38	11	0.0052	0.0052
^{233}U	221	53	117	62	111	46
^{235}U	372	19	92	19	0.037	0.48
^{238}U	97	1	841	5.7	0.0008	0.0068
^{237}Np	1436	71	318	65	28	39
^{239}Pu	4851	46	4290	28	2300	13
^{240}Pu	5952	50	6025	22	5730	0.41
^{241}Pu	5834	62	4506	63	1430	38
^{242}Pu	5139	58	3106	56	160	63
^{241}Am	3869	72	3826	67	1410	38
^{242m}Am	6222	60	5128	74	1180	85
^{243}Am	6277	55	6557	32	6500	17
^{242}Cm	2051	78	798	86	161	88
^{245}Cm	5942	101	7939	71	9640	52

On LLFP Toxicity Reduction

Rate of transmutation of a J-nucleus can be characterised by the time T_J^{transm} needed to incinerate the half of initial mass which is a function of the cross-section $\sigma_{n,\gamma}^J$ (barns) and of the neutron flux Φ (n/cm^{2}s) :

$$T_J^{transm} = \frac{\ln 2}{\sigma_{n,\gamma}^J \phi \times 3.15 \times 10^7} years$$

Transmutation of the toxic fission products in nuclear reactors and subcritical systems has a sense if rates of nuclear interactions with neutrons are **much higher than rates of natural decays**, which are defined by decay life-time $T_{1/2}$.

Hence, transmutation under neutron flux is reasonable if $T_{1/2} >> T_J^{transm}$.

There are at least 5 long lived isotopes :

$$^{99}Tc,\ ^{129}I,\ ^{93}Zr,\ ^{107}Pd,\ ^{135}Cs$$

which can be transmuted in a fast spectrum much faster than their natural decay.

TABLE

Parameters of long lived nuclei to be eventually transmuted in a fast and "thermal" spectra

with standard flux levels

Isotopes, *J*	$\sigma_{n,\gamma}^{J}$ (barns)		$T1/2$ (years)	T_J^{transm} (years)		Radio-toxicity in (Sv/g) at $t = T_J^{transm}$	Recommendation to transmutation
	fast spectrum	thermal spectrum		fast spectrum	thermal spectrum		
^{79}***Se***	0.03	0.1	6.5×10^{4}	7.3×10^{2}	2.2×10^{3}	6.0	*questionable*
^{90}***Sr***	0.01	0.14	29	2.2×10^{3}	1.6×10^{3}	-	*non-transmutable*
^{93}***Zr***	0.03	0.28	1.5×10^{6}	730	790	0.04	***transmutable***
^{94}***Nb***	0.04	2.2	2.0×10^{4}	5.5×10^{2}	1×10^{2}	9.0	*questionable or transmutable*
^{99}***Tc***	0.2	4.3	2.1×10^{5}	110	51	0.2	***transmutable***
^{107}***Pd***	0.5	0.3	6.5×10^{6}	44	730	0.0007	***transmutable***
^{126}***Sn***	0.005	0.05	1×10^{5}	4.4×10^{3}	4.4×10^{3}	4.0	*questionable*
^{129}***I***	0.14	4.3	1.6×10^{7}	160	51	0.5	***transmutable***
^{135}***Cs***	0.07	1.3	2.3×10^{6}	310	170	0.08	***transmutable***
^{137}***Cs***	0.01	0.02	30	2.2×10^{3}	1.1×10^{4}	-	*non-transmutable*
^{151}***Sm***	0.7	700	89	31	0.3	-	*non-transmutable or questionable*

As for

^{90}Sr, ^{137}Cs and ^{151}Sm,

natural decay is similar or much faster than transmutation and it is more reasonable to put them into interim storage for decaying.

On neutron consumption by LLFP

The overall neutron consumption D_J^* of the LLFP (*J*-family) can be defined as

the total D_J^* = number of neutrons which have to be spent to incinerate the whole family. Removal of stable and short lived fission products can be considered as a single sink of long-term radioactive fission products.

D_J^* value is given in terms of neutrons required per transmutation.

An algorithm of D_J^* calculation is very handy in this case

Two helpful parameters of neutron consumption definitions, depending on units choice - D_J^* (neutron/transmutation) and ***DJ*** (neutron/fission).

DJ can be obtained as the product of D_J^* and of the yield of *J* nuclide per fission, Y_J.

Taking into account LLFP yield, one can calculate the total neutron consumption needed to incinerate all "*transmutable*" and "*questionable*" long lived isotopes of fission products :

$$D_{Zr-93,Tc-99,Pd-107,I-129,Cs-135,Sn-126,Nb-94,Se-79} = \sum_J D_J^* Y_J = 0.22 neutron / fission$$

TABLE

Comparison of overall neutron consumption *D* (neutron/fission in NP) and LLFP transmutation effect (reduction of radio-toxicity) for groups of nuclei

Transmutable nuclides	***D* (neutron/fission in NP)**	**Radio-toxicity reduction at = 1000 years (Sv/GW$_e$×year) after discharge**
^{99}Tc	**0.055**	5500
^{129}I + ^{99}Tc	**0.064**	8160
^{129}I + ^{99}Tc + ^{135}Cs	**0.081**	9060
all isotopes of *Tc, I, Cs*	**0.15**	9060
all isotopes of *Tc, I, Cs, Zr*	**0.68**	9930
all isotopes of *Tc, I, Cs, Zr, Pd*	**0.98**	9935

Hence, to incinerate all *Tc, I, Cs* without isotopic separation, one needs about 0.15 (neutron/fission in NP) as minimum.

LLFP incineration requires an essential number of neutrons. Hence, fast neutron spectrum and external neutron source (e.g. Accelerator Driven subcritical Systems) are favourable for this application:

CONCLUSION:

Up to now, motivations in ADS application can be approved by:

- **necessity in supplementary neutron production for TRPu transmutation because parasitic neutron consumption of TRU;**
- **necessity in supplementary neutron production for LLFP transmutation (all spectra) because parasitic neutron consumption of LLFP;**
- **intrinsic safety enhancement in TRU-burners**
- **……**