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Aspects on Thorium Utilization in Heavy Water Reactors

ASPECTS ON THORIUM UTILISATION IN
HEAVY WATER REACTORS

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Introduction

As natural uranium fuelled PHW reactors are well developed the thorium fuelled PHWRs seem to be the fastest attainable advanced fuel cycle. In support of this statement, the following arguments could be given :

- this fuel cycle avoids expenses associate with the development of a new reactor concept,
- it may utilise domestic thorium resources,
- it can provide some insurance against severe uranium shortage ; even for less severe shortage it seems to provide an economically acceptable option,
- reactor component manufacturing capabilities developed in connection with the first generation PHWRs could be used without significant changes.

1. SELF-SUFFICIENT EQUILIBRIUM THORIUM (SSET) CYCLES IN PHWR

Two types of thorium cycles in PHWR are considered:

- the SSET cycles, which can operate at a reasonable burnup without any external fissile topping, and
- the high or intermediate burnup thorium cycles, which can operate enriched uranium or plutonium topping.

There are three basic reasons for our interest in SSET cycles in PHWR :

- its low uranium requirments , the inventories permit the application of a direct or semi-remote mode of fabrication and, thus, the reduction of the low burnup penalty;
- its nuclear characteristics; the SSET cycles are valuable as reference, limiting case, for other thorium cycles in PHWR.

The main problem of SSET cycles in PHWR is the sensitivity of burnup estimates to uncertainties in estimates or allowance for :

- neutron leakage
- η values for fissile materials
- fissile material losses due to reprocessing and refabrication.

Fig. 1 shows the sensitivities of estimated burnup (see Appendix 1) to neutron leakage for different fissile material losses.

The effects of the well known uncertainties in the η values of U 233 for 2200 m/s and for the resonance range energies are presented in the same figure.

The neutron leakage is a critical parameter for high fissile material losses. This suggests that the SSET cycles apply only to large and both radially and axially reflected reactor cores. The beneficial effect of a more neutron absorbing fuel is largely reduced by the Pa-233 induced flattening of the spatial neutron flux distribution as demonstrated by Milgram /1/. This effect introduces an extra penalty for higher channel powers. For a 1 GW(e) core and 6.5 MW maximum channel thermal power 1.5 to 2% leakage losses are considered in this preliminary draft.

The burnup estimate sensitivities to η values for U-233 (both the maxwellian and resonance energy ranges) are very important. The corresponding entry data uncertainties (± 0.008 for η^0 and ± 0.06 for $\eta-1/E$) lead to $\pm 20\%$ burnup uncertainties.

Effects of fissile material losses at reprocessing and refabrication are also very important and they became critical at high values of neutron leakage. But addition in refabrication costs may be accepted, which should guarantee the limiting of these losses at about 0.5%.

The xenon over-ride is considered 3 times lower than for natural uranium fuel and the possible use of either thorium-enriched or recycled uranium booster rods or thorium absorbing rods needs no reactivity allowance for.

Under these conditions, the burnup lies between 10,000 and 13,000 MWd/t if no extreme values for η are taken into consideration. The maximum limit should correspond to initial plutonium inventory and the minimum limit to initial U-235 inventory,

the penalty due to U-236 being able to exceed 2000 MWd/t.

In this stage, penalties connected with limitations imposed by the fuelling strategy are not taken into consideration due to the fact that in case of Th-U the limitations concerning fuelling rate are more slight because of a higher burnup as in case of natural U, and eventually the economic optimum will be closer to an idealized fuelling mode. The existence of other improvement possibilities of neutron economy such those mentioned in [2] i.e. a lower H_2O content in D_2O , improved pressure tubes, multizonal cores^{*)}, etc., are sufficient margins for the acceptance of these burnup values even in the case of disadvantageous η_3 values.

An 10% extra fissile material for the inventory for example of U-235 utilization, is considered as sufficient margins for any case, having in view that a 10% reduction of burnup may be compensated adding 0,5% per cycle (at about 2,7 years). This quantity will be enough for 60 years. An extension of this period with other 30 years will be possible by reducing the out of core time with 1,3% per cycle, reaching 14% in 30 years i.e. 10,5 months instead of one years.

2. THORIUM AND URANIUM REQUIREMENTS FOR A 1 GW(e) REFERENCE THORIUM FUELLED PHWR

For the analysis concerning the 1995-2025 scenario, a reference PHWR was chosen to permit operation either with natural uranium or with thorium fuel; this was selected in order to permit an eventually optimal solution choice for a transition from one cycle to another. The selected reactor of which the basic data are very similar to those of CANDU current designs, has the following characteristics :

*) This is a characteristic of SSET-cycles, as will be seen in the subsequent report. In once-through nat.U cycles the multizonal operation leads to specific burnup penalties.

Core

Fission Power	3450 MW
Channel number	624
Axial flux ratio	0,77
Radial flux ratio	0,85
Coolant entry temp.	265°C
Coolant exit temp.	315°C
Lattice pitch	28,6 cm
Active core length	600 cm
Active diameter	800 cm
Axial reflector thickness	50 cm
Radial reflector thickness	70 cm

Channel

Maximum power	6,5 MW (thermal)
pressure tube inner diam.	10,5 cm
coolant cross section area	34 cm ²

Fuel element

Pin number per bundle	37
Fuel element length	50 cm
Nominal diameter of (Th,U)	
O ₂ pellets	1,2 cm
Sheath thickness	0,04 cm
Th + U weight per bundle	17 kg
Th + U weight per reactor	127 t
Medium specific power in fuel	27 MW/t

Two starting possibilities are considered for SSET :

- U-235 initial fissile inventory
- Pu " " "

In the first case a 2% U-235 (93% enrichment) total fissile inventory was obtained using the method presented in Appendix 1. This corresponds to about 4 t/GW(e), i.e., 800 t nat.U (for 0,2% tail assay) for a year that discharged material remains in fuel cycle before reload.

In the second case the corresponding fissile inventory is about 2,3% Pu 239 + Pu 241 in 72% fissile content plutonium. This corresponds to about 5 t/GW(e); equivalent to 1700 t nat uranium Candu fuel element discharged at 7500 MWd/t. About 4/5 of this fissile plutonium is to be considered, 1/5 being the fissile plutonium discharge during transient.

An estimation for the case of application of denatured cycle is under analysis. The burnup will be lower or it may require an external fissile material topping for each pass.

Two thorium inventories were considered: the lower one of 200 t/GW(e) corresponding to the remote fuel fabrication and a higher one, of 1500 t, which implies 20 years storage of recycled thorium for thorium 228 decay corresponding to direct or semi-remote fabrication. 2 t/GW-year should be added at each cycle, out of which 1 t/GW-year to compensate for the reprocessing losses.

3. SAFETY FEATURES OF THE REFERENCE TH-PHWR

For the reference Th-PHWR we have to consider its particularities due to.

1. kinetic parameters,
2. fuel thermal properties,
3. Pa-233 decay after reactor shutdown

Three different fuels are considered, namely Th-U-233 fuel, Th-Pu fuel and Th-recycled uranium fuel.

1. Kinetic parameters

1.1. In Table 1 the delayed neutron fractions β of the main fissile nuclide in each of the three kinds of fuel are given.

The lower β -values increase the prompt supercritical risk compared with natural uranium equilibrium fuel PHWR and this must be taken into account in the control and shutdown system design.

Table 1(5)

Nuclide	β - value
U ²³⁵	0.0065
U ²³³	0.0026
Pu ²³⁹	0.0021
Nat.U-CANDU	0.0046

1.2. The average lifetime of thermal neutrons is expected to be lower in Th-PHWR than in nat.U-PHWR (due to higher absorption cross-section of the thorium fuel) and this negative effect on safety should be considered too.

1.3. The reactivity coefficient is considered for two fuel cases, i.e.

Th-recycled uranium fuel, and
Th-Pu fuel

For the first case, the U-233 2eV resonance can lead to a positive reactivity effect as it results from the (T/1000) power series coefficients of the Westcott g_f , given in Table 2 a_1 for U-233 is positive but its absolute values is 10^2 - fold smaller than Pu 239- a_1 . The Doppler coefficient of Th-232 is high enough (about 50% of that of U-238 (5) to compensate for the (10w) positive value of a_1 of U-233.

Table 2
(T/1000) - power series coefficients

$$\text{for } g_f = \sum_i a_i \left(\frac{T}{100} \right)^i \quad (6)$$

	a_0	a_1	a_2	a_3	a_4	a_5
U 235	0.983	-0.238	0.112	1.53	-8.98	2.96
U 233	1.00	0.00768	0.0779	0.0211	-0.0288	0.0103
Pu 239	1.04	0.592	0.913	0.136	-3.97	5.97

For the Th-Pu fuel it is expected that the maximum positive effect lies at about 0.3% Pu 239 as in the case of U-Pu 239 [7], and that for higher contents selfshielding would reduce this effect and restore the negative Doppler coefficient dominant effect.

The initial fissile plutonium concentration in Th-Pu is high enough (about 2,3%) to permit acceptable burnup and Pu depletion.

2. Fuel thermal properties

The thermal conductivity of ThO₂ is a little higher (~ 15%, (8)) than that of UO₂, and therefore (the thermal capacities being equal) the thermal diffusivity is higher, leading to less severe temperature excursions.

3. Pu-233 decay after reactor shutdown.

The 27 days half-life decay of Pu 233 leads, in case of long-duration shutdowns, to positive reactivity effects due to U 233 generation, and therefore needs an increased shutdown capability.

4. FUEL ELEMENTS FOR INTERMEDIATE AND HIGH BURNUP

The Th-PHWR cycle capability to have also economically acceptable options for less severe uranium shortage depends upon the development of a corresponding fuel element; the current CANDU design fuel element may presents important burnup limitations.

A fuel element with a lower expected fission product release (lower fuel temperature), even at very high linear power, has been described by Lewis [9]. This fuel element may lead to high radial thermal flux nonuniformities in the sheath at the graphite disc level.

The local temperature increases could be of about 20°C at the sheath outer surface and more than of about 50°C at the

sheath outer surface and more than of about 50°C at the sheath inner surface. Improvements are expected by broadening the thermal flux transfer area between graphite and sheath by proper design of the graphite discs and by change of the fuel pellet shape. A graphite tube between pellets, graphite discs and sheath is also taken into consideration. Estimates will be necessary for these solutions from a technological point of view.

The use of thinner rods which can reduce the fuel temperature is also taken in view and the corresponding problems (rod vibrations, rod fabrication cost penalty) should be analysed.

5. PECULIARITIES OF THORIUM FUEL REPROCESSING

For ThO₂ fuel in metallic cladding, the wet technology used for UO₂ fuel reprocessing remains valid with some modifications imposed by the physico-chemical properties of ThO₂ matrix and nuclear properties of Pa-233, U-232, Th-228 and Th-224.

Thorium fuel requires strongest solvents (more concentrated acid and fluorine presence) even in these conditions the low dissolving rate imposes a higher contact time. The utilization of acid mixtures complicates the next steps of the process such as feed adjusting, solvent extraction and management of wastes due to strong corrosion.

After dissolution an intermediate step for removing the acid excess is compulsory and the adjusting of feed composition for the solvent extraction process. The possibility of the third phase formation imposes the lowering of thorium concentration against normal purex process and a more pretentious operation of the equipment.

The main steps of the technological process for ThO₂ - UO₂ or ThO₂ - PuO₂ fuel reprocessing in zircaloy cladding is similar to UO₂ with the modifications already shown.

1. The first operation is the mechanical or chemical fuel decladding in view of dissolution :

- Zirflex process using ammonium fluoride and nitrate mixture.

- HF + O₂ fragilization at 625° leading at ZrO₂
- H₂O + O₂ fragilization at 825° using N₂ as catalizer, process known as "Thermox process"
- Zircex process with HCl at 500°C leading to ZrCl₄.

Mechanical decladding represents 5% from the chemical process being more advantageous for the solid waste management (easier manipulation after a compating stage).

Another benefit is that used equipment for UO₂ fuel may be easily adjusted for ThO₂ fuel.

2. ThO₂ dissolution is optimal in 13 M nitric acid with 0,04 - 0,07 Mg fluoride ions at boiling. Thorium presence in the solution reduces the dissolving rate in spite of the big excess of nitric acid. The density of ThO₂ pellets affects the magnitude of dissolving rate (the increasing from 60% - 95% of the density leads to a rate 6 times smaller).

In ThO₂ - UO₂ system the rate is not affected by the presence of up to UO₂ 10%.

The dissolution of ThO₂ irradiated fuel is faster than the normal fuel especially for higher burnup.

For mechanical decladding, fine zirconium particles are dissolved in HNO₃.HF the rate being slightly diminished. The corrosion rate of the equipment may be reduced up to 20% adding 0.4 M Al (MO₃)₃.

3. Adjustment of crude feed is an obligatory step (stage) because of high free acidity (nearly 6.5 M) and the necessity of fission product conversion in unextractable forms. The acidity lowering is obtained by evaporation and followed by one hour heating at 155°C, when ruthenium passes into less extractable species and SiO₂ is dehydrated and is no more forming stable emulsions during solvent extraction.

The differences of uranium and thorium solvent extraction impose a characteristic flow-sheet against the normal Purex process. At pilot plant scale the following procedures were developed :

- U-233 process, hexone extraction from deficient acid solutions.
- Interim -23 process, TBP extraction from acid solutions.

- Thorex-1 process, Pa extraction with dibutyl carbitol, uranium extraction with 5% TBP and thorium extraction with 45% TBP.

- Thorex - 2 process, coextraction with 30-42% TBP and selective stripping of Thorium and Uranium of different concentrations.

- Thorex - 2 process is the most valuable because of the 99% recovering yield for fissile and fertile materials, the decontamination factors being satisfactory from the fuel physics point of view in addition smaller HLW are resulted. The inconveniences appear due to the third phase formation at an organic concentration higher than 95 g Th/l. for aliphatic diluents, as a consequence of chemical and radiolytical degradation of solvent making U-233 stripping more difficult.

The 34-40 stages for solvent extraction equipment are sufficient for a fuel cycle including the next sections :

- extraction
- aqueous scrubbing
- organic scrubbing
- thorium stripping
- uranium stripping

It is worth noticing that the contactor selection depends on the suspension presence in the system due to stable emulsion formation. In addition the equipment operation implies the rigorous control of acidity and flow rates in the stripping section due to U-Th partition operation sensibility. Final purification of U-233 is the standard tail - end for enriched uranium by ion exchangers, anorganic sorbents, or solvent extraction.

6. SPECIFIC PROBLEMS FOR RADIOACTIVE WASTES IN THORIUM CYCLE AND FOR IRRADIATED THORIUM STORAGE

The following radioactive waste sources will be analysed with respect to specific implications of Th-PHWR cycle in comparison with natural uranium cycle :

- fuel reprocessing
- fuel refabrication.

The fuel reprocessing produces the following types of radioactive wastes :

- gases (mainly fission products);
- high medium and low level liquid wastes;
- organic spent liquid resulting from extraction processing with solvents.

These wastes are resulted from

- fuel dissolution; this is the main source of solid wastes;
- uranium and thorium extraction, producing high level liquid wastes.

No special problems appear due to these wastes except the more pronounced corrosion effect due to free acidity of residual solution. This imposes the solidification of highly active wastes with a maximum one year delay and also a more safe storage form.

On the other hand, the cost of high active wastes treatment stored for decay (about 5 years) is lower in comparison with the cost of treatment in a short interval after the wastes were produced.

At fuel fabrication the highest quantities of wastes are the liquids & actives that result from the equipment decontamination process where γ radioactivity of uranium 232 daughters give not specific difficulties.

Recycled uranium decontamination is the main source of γ radiation.

The peculiarities that appear at fuel refabrication are the following :

- uranium solution purification (U^{233}) for U^{232} daughter removing, daughter that will be found in radioactive wastes;
- thorium storage for decay before conversion at ThO_2 .

6.1. Wastes from U^{233} purification solution

For daughter U^{232} purification solution for uranium conversion to UO_2 without γ protection two methods can be used ;

- an extraction method with organic solvents where from liquid wastes in aqueous solution will result, wastes that required

usual storage and treatment needing biological protection due to γ activity;

- a method by ion exchangers where from solid wastes in the form of spent ionic masses on which the daughters will be retained.

The problem appearing in the management of this wastes category are the same from the protection point of view, in addition the phenomenon of radiolytical degradation of this ionic masses, takes place. Radiolytical degradation imposes solidification in a short time interval after collection against the case of wastes from solvent extraction. This because a long period storage makes management from the point of view of ventilation and supplementary control measures, more expensive.

We must mention that in both cases it is cheaper to solidificate the wastes in a short time after collection rather than store them under the form resulting from the processes, even though after about 15 years the wastes activity is decaying at low levels.

6.2. Thorium solution storage for conversion at ThO_2

ThO_2 fabrication presents aspects connected with γ daughters existence (Th^{228}).

The natural thorium initial fabrication implies a low level that does not require protection or remote. Gamma activity associated with U^{233} recycle raises in time and becomes more intensive and more penetrating after chemical separation.

Thorium separated at reprocessing must be decontaminated by daughters for its recycle.

If the fuel is rapidly refabricated before daughters raising activity of U^{232} protections and remotes will not be necessary. When conversion at ThO_2 does not take place immediately thorium storage for an interval of 15-20 years for Th^{228} daughters decay is necessary.

Thorium fuel fabrication after a time period when daughters appear, protections are compulsory during storage, conversion and final assembly for operation staff protection.

The necessity of thorium solution storage up to decay followed by fabrication and recycle is revealed in order to avoid this direct implication on fuel fabrication cost.

More storage forms of recovered thorium from reprocessing installations may be used.

a) Storage under the form of primary solution ($\text{Th}(\text{NO}_3)_4$)

In this case the following aspects are revealed :

- accentuated corrosion due to free acidity (1-3 MHNO_3) that requires the use of special and expensive materials for tanks ;

- high storage volumes (1M Th/l of solution) that require higher prices because of greatest number of necessary tanks;

- radiolytical gas formation that implies off gas and control systems against explosion damage.

b) Storage under primary concentrated solution form by means of evaporation and neutralisation. In this case the following aspects are revealed :

- lower storage volume against the first case (about 50% reduction) so a smaller number of tanks ;

- a higher concentration (of about two times) of solution with free acidity removing and a low corrosion; this fact allows a less expensive storage ;

- radioanalytical gas formation that implies the same problems as in first case;

- the raising cost because of supplementary concentration necessity, partially contrabanced because of storage volume reducing.

c) ThO_2 form storage

In this case, the following aspects revealed :

- storage volume reduction (of about 50 times) with reduction of tanks number ;

- supplementary costs necessary for thorium nitrate to ThO conversion because refabrication ThO_2 with redissolution and²reconversion is imposed.

From storage solution analysis it may be appreciated that from the economical, necessary protection and operation

measures point of view, thorium storage under the form of primary concentrated and neutralised solution becomes more beneficial.

7. FABRICATION OF THE FUEL ELEMENTS

For the reference case, fuel elements with structure and dimension of CANDU fuel elements are taken into consideration.

The fuel elements for cycle starting are composed of fuel pellets of :

- ThO_2 (98%) - UO_2 (2%) solid solution, where uranium is highly enriched (93% U-235) or medium enriched at the level corresponding to a denatured cycle.
- ThO_2 (97%) - PuO_2 (3%) solid solution, where plutonium has a isotopic composition corresponding to the natural uranium fuel elements burned in CANDU and discharged at 7500 MWD/t.

The fuel elements for refuelling are composed of pellets of ThO_2 - UO_2 (2.5%) solid solution, where the uranium isotopic contents is that of the recycled uranium , i.e. 1.6% U-233; 0.6 U-234; 0.15 U 235 and 0.3 U-236 (in the case of U-235 initial inventory).

α and γ radioactivities specific to each pellet kind determines :

- Th - U-235 fuel element fabrication under similar conditions with those of natural uranium fuel element, paying special attention to fissile material recovery ,
- Th - Pu fuel element fabrication with glove box techniques ,
- Th - recycled uranium fabrication in glove boxes with shielding for uranium 232 - α and uranium 232 daughters α , γ radioactivities.

Thorium recycled uranium fabrication is the step of the cycle for which the lack of experience is illustrated in the very wide range of fabrication cost estimations.

Some recent estimations are presented in Table 1, for various types of fuel elements together with the ratios between their fabrication cost and natural uranium fuel element fabrication cost. Ratio between 2.5 and 9.3 for thorium-recycled uranium fuel elements may be explained by various refabrication modes that were taken into consideration at their estimations; the uranium 232 content in recycled uranium is the main parameter that determines the refabrication mode.

Table 1

PHWR - Fuel element fabrication costs estimates

Fuel element type	ORNL 1977/10/ dol/kg HE		ORNL 1977/11/ dol/kg HE		AECL5501/2/ dol/kg HE	
Nat. uranium	60	1	50	1	46	1
Th-U 235	140	2,3	100	2	50	1,1
Th-Pu	320	5,3	320	6,4	87	1,9
Th-recycled U	560	9,3	390	7,8	115	2,5

Estimations concerning the maximum uranium 232 content of cycle uranium for the reference case given before are plotted in figure 2 and an example of cycle by cycle raising of uranium 232 content is plotted in figure 3.

The main parameter that determines the uranium 232 of recycled uranium is the burnup and the first 1 - 2 cycles have sensible lower uranium 232 contents than maximum equilibrium value.

For the reference case a uranium 232 content of 22 ppm (heavy atoms) was taken into consideration.

The fuel specific radioactivity and the air volumes corresponding to the maximum permitted radioactivity concentration for 1 mg. fuel dust in comparison with natural uranium fuel and thorium - plutonium fuel is presented in Table 2.

Although thorium - recycled uranium fuel radioactivity is about 200 times less important than that of plutonium fuel, the glove boxes fabrication is generally accepted and taken into consideration by us.

Table 2

Fuel	Nat U	Th - Pu 3%	20 ppm U- 232 in Th-recycled U
The main radio- toxic isotope	U 238	Pu 241	U 232
Specific activity Ci/kg. fuel He	0.00034	150	0.4
Air concentration permits for profes- sional cond., Ci/m ³	7.10^{-11}	9.10^{-11}	3.10^{-11}
Air volume (m ³) for 1 mg. fuel dust.	5	3.10^6	$1.5.10^4$

Although thorium - recycled uranium fuel radioactivity is about 200 times less important than that of plutonium fuel, the glove boxes fabrication is generally accepted and taken into consideration by us.

The choice of refabrication mode (direct, semiremote or remote) requires a more careful analysis, taken into consideration the radioactivity of thorium recycled uranium fuel.

The radioactivity of the fuel is given by the daughters of uranium 232 presented in figure 4.

A method of the γ radioactivity temporary diminishing that was tested at ORNL and B et W and consists of extraction of these daughters by solvent extractions or by ion exchange separation. 1 - 2 days after this decontamination, the decay of the short - lived remaining radioisotopes reduces further the radioactivity before of the radiation grows. The total decontaminations ratio of about 400 for several years aged uranium have been obtained at the ORNL pilot plant /12/.

This reducing degree leads to an absolute level of radioactivity that is almost the same with that obtained at B et W as

it may be seen in figure 4.

This level is equal with the equilibrium level for a 200 up to 800 lower uranium 232 content.

The experiment at B et W with direct fabrication led to the following specific doses :

- 12 mrem/kg HE - general ;
- 25 mrem/kg HE - at hands

for a 1.2 ppm uranium 232 content in thorium + 48 - 72 days aged uranium, corresponding to a 22 ppm uranium content for the decontamination level given in figure 4.

Taking in consideration the maximum permitted rates of 7.5 mrem/hour - general - and 35 mrem/hour at hands, we have:

1,6 hours/kg HE - for general irradiation maximum dose rate,

0.8 hours/kg HE for hands irradiation maximum dose rate.

The experiments at ORNL with semiremote fabrication took in view the supplementary reducing of general irradiation at the ratio

$$\frac{\text{body irradiation}}{\text{hand irradiation}} = \frac{1}{15}$$

These experiments have demonstrated the possibility of a ratio value equal to 1/7 which should be compared with 1/2.3 obtained at B et W for direct fabrication.

The above means there is the possibility of reducing the general irradiation (even the hands irradiation) at an acceptable level with the semiremote fabrication mode..

A detailed evaluation for the PHWR - fuel element technology is under analysis.

8. CONCLUSIONS

In this preliminary report some of the main problems of the Th-PHWR cycle development, have been analysed .

The first problem consists of the burnup limitations introduced by SSET cycle conditions, i.e., no external fissile topping. Taking into consideration the actual equilibrium pro-

perties the neutron leakage seems to be the most important features ; this fact impose large reactor units. The most important uncertainty arises from those of η both thermal and resonance values of U-233.

Some safety problems are due to low delayed neutron fraction values of the same nuclide.

Conceptual level problems are presented by high and intermediate burnup fuel elements for low uranium price Th - cycle options.

One of the most important developments is required by the fuel reprocessing technology and the reprocessing equipment.

Few problems are posed by the specific radioactive wastes. The primary liquid form is accepted for radioactive thorium storage.

Fuel fabrication for SSET-cycles needs a careful analysis taking in view the preliminary evaluations of U-232 content which leads to a γ - radioactivity for fresh decontaminated recycled uranium low enough to permit semi-remote fabrication.

Appendix 1

SENSIBILITY ESTIMATES IN SELF-SUFFICIENT
EQUILIBRIUM THORIUM (SSET) CYCLES

The main characteristic of SSET cycles is the constant isotopic composition of uranium during irradiation.

The method applied at CRNL [2] for SSET cycles characteristic estimations is to repeat the isotopic content calculations, using a cell cod (LATREP) for an idealized fuelling scheme, until the isotopic content ceases to vary from iteration to iteration. With a such isotopic composition the evaluation of the characteristic sensitivity to various parameters is made.

A direct improvement of this method would result considering that to each value set of input parameters a specific isotopic composition corresponds and that for sensitivity evaluations we must compare the equilibrium cycles characteristics corresponding to the input applied values. This would require a considerable number of iterations if we take into account that for this comparison a special accuracy of these characteristics is required.

Another sensitivity evaluation method is that used by R.Naudet [13] which considers the pseudoequilibrium deduced from the burnup equations of nuclid row given in figure 5 in the case of indefinite burnup with maintaining the constant concentration of thorium atoms.

In our evaluations we have used the same method, a little developed in order to take into consideration the actual variation of the composition (that of thorium included) for limited burnup, the reprocessing losses and the eventual external topping estimations. The estimations of self-shielding effects for U-233 and Pa-233 obtained by both methods are given in fig. 2. The figure shows that the sensitivity estimates for real equilibrium may have different values and even signs when comparing it with the previous estimations.

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The nuclide balance for a limited burnup corresponding to a given reactor residence time t is (see fig. 5)

$$N_{Th}(t) - N_{Th}(0) = -n_{Th} = -n_{Th}^c - n_{Th}^n - 2n_{Th}^f - n_{Th}^{fis}$$

$$N_{Pa}(t) - N_{Pa}(0) = -\lambda_{Pa} \int dx \int_0^t N_{Pa}(x, t') dt' - n_{Pa} + n_{Th}^c$$

$$N_3(t) - N_3(0) = -n_3 + \lambda_{Pa} \int dx \int_0^t N_{Pa}(x, t') dt'$$

$$N_4(t) - N_4(0) = -n_4 + n_3^c + n_{Pa}$$

$$N_5(t) - N_5(0) = -n_5 + n_4$$

where $N_i(0)$ and $N_i(t)$ are the nuclide i fuel content before and after irradiation (3,4 and 5 represent U 233, U 234 and U 235), x is the volume element (dx) position inside the fuel volume, λ_{Pa} is the decay constant of Pa 233 and $N_i^{(j)}$ is the content change in nuclide i due to reaction j (c means (n, γ) ; f means fission,

$$n_i = \sum_j n_i^j \quad \text{i.e.}$$

$$n_i^j(t) = \int_{\text{fuel volume}} dx \int_0^t N_i(x, t') dt' \int_0^\infty \sigma_1^j(E) \phi(E, x, t') dE$$

The SSET cycles conditions require (b is the fraction of uranium lost per cycle) :

$$N_3(t) + N_{Pa}(t) - N_3(0) - N_{Pa}(0) = b | N_3(t) + N_{Pa}(t) |$$

$$N_4(t) - N_4(0) = b N_4(t)$$

$$N_5(t) - N_5(0) = b N_5(t)$$

and led to $| N_i = N_i(t) |$

$$n_3 + n_{Pa} = n_{Th}^c - b(N_3 + N_{Pa})$$

$$n_4 = n_3 + n_{Pa} - bN_4$$

$$n_5 = n_4 - bN_5$$

With v_i , a medium value for the neutrons generated per fission, we can define a value for η_i , the number of neutrons generated per absorption

$$\eta_i = v_i \frac{\int dx \int_0^t N(x, t') dt' \int_0^\infty \sigma_i^f(E) \phi(E, x, t') dE}{\int dx \int_0^t N(x, t') dt' \int_0^\infty \sigma_i(E) \phi(E, x, t') dE} = v_i \frac{n_i^f}{in_i}$$

With this n_4 and n_5 become :

$$n_4 = (1 - \frac{\eta_3}{v_3}) n_{Th}^c + \frac{\eta_3}{v_3} n_{Pa} - b | (1 - \frac{\eta_3}{v_3}) (N_{Pa} + N_3) + N_4 |$$

$$n_5 = (1 - \frac{v_3}{v_3}) n_{Th}^c + \frac{\eta_3}{v_3} n_{Pa} - b | (1 - \frac{\eta_3}{v_3}) (N_{Pa} + N_3) + N_4 + N_5 |$$

The number of absorbed neutrons in heavy elements (except (n,2n) reactions and thorium fissions) per neutron capture in thorium is given by

$$a_{HE} = \frac{n_{Th}^c + n_{Pa} + n_3 + n_4 + n_5}{n_{Th}^c} = 4 - 2 \frac{\eta_3}{v_3} + 2 \frac{\eta_3}{v_3} \gamma_{Pa} - \frac{b}{n_{Th}^c} | 2(1 - \frac{\eta_3}{v_3}) x$$

$$x (N_{Pa} + N_3) + N_{Pa} + N_3 + 2N_4 + N_5 |$$

where

$$\gamma_{Pa} = \frac{n_{Pa}}{n_{Th}^c}$$

The number of generated neutrons in heavy elements (except (n,2n) reactions and thorium fissions) per neutron captures in thorium is given by

$$y = \frac{\eta_3 n_3 + \eta_5 n_5}{n_{Th}^c} = \eta_3 + \eta_5 \left(1 - \frac{\eta_3}{\nu_3}\right) - \eta_3 \left(1 - \frac{\eta_5}{\nu_3}\right) \gamma_{Pa} - \frac{b}{n_{Th}^c} \left| \eta_3 + \eta_5 \left(1 - \frac{\eta_3}{\nu_3}\right) \right. \\ \left. \times (N_{Pa} + N_3) + \eta_5 (N_4 + N_5) \right|$$

The number of fissions, except thorium fissions, per neutron capture in thorium is given by

$$\varphi = \frac{\eta_3 n_3 / \nu_3 + \eta_5 n_5 / \nu_5}{n_{Th}^c} = \frac{\eta_3}{\nu_3} + \frac{\eta_5}{\nu_5} \left(1 - \frac{\eta_3}{\nu_3}\right) (1 - \gamma_{Pa}) - \frac{b}{n_{Th}^c} \left| \frac{\eta_3}{\nu_3} \left(1 - \frac{\eta_5}{\nu_5}\right) \right. \\ \left. \times (N_{Pa} + N_3) + \frac{\eta_5}{\nu_5} (N_4 + N_5) \right|$$

For the neutron cycle given in fig. 7, the fission product absorption per fission is for $K_{ef} = 1$

$$a_{FP} = \frac{\epsilon y (1-L) f - a_{HE}}{\varphi + \frac{\epsilon_{Th}^f - 1}{\nu_{Th} - 1} y}$$

where

$$\epsilon = 1 + (\epsilon_{Th}^f - 1) + (\epsilon_{Th}^{n2n} - 1) + (\epsilon_D^{yn} - 1)$$

L = leakage losses, and

f = fraction of neutron absorbed in heavy elements per neutron absorbed in heavy elements and reactor and fuel element structure materials, are input data evaluated with cell and core codes.

The capture ratio in Pa-233 and Th-232 may be obtained with the following relation, precisely enough if we take into consideration the low value of this ratio :

$$\gamma_{Pa} = \frac{\phi \sigma_{Pa}}{\lambda_{Pa} + \phi \sigma_{Pa}} \left(1 - \frac{1 - e^{-(\lambda_{Pa} + \phi \sigma_{Pa})t}}{(\lambda_{Pa} + \phi \sigma_{Pa})t} \right), \quad \phi t = \frac{n_{Th}^c}{\sigma_{Th}^c}$$

The initial fissil inventory may be evaluated considering that for k-irradiation of the transition we have (neglecting reprocessing losses) the following nuclid content increments :

$$N_3^k + N_{Pa}^k = N_3(t) + N_{Pa}(t) - N_3(0) = n_3 - n_{Pa} + n_{Th}^c$$

$$N_4^k = N_4(t) - N_4(0) = -n_4 + n_3^c + n_{Pa}$$

$$N_5^k = N_5(t) - N_5(0) = m_5^k - n_5 + n_4$$

where m_5^k is the k-cycle U-235 inventory make up.

Then we have :

$$n_{Pa} + n_3 = n_{Th}^c - N_3^k - N_{Pa}^k$$

$$n_4 = \left(1 - \frac{\eta_3}{\nu_3}\right) n_{Th}^c + \frac{\eta_3}{\nu_3} n_{Pa} - \left(1 - \frac{\eta_3}{\nu_3}\right) (N_3^k + N_{Pa}^k) - N_4^k$$

$$n_5 = m_5^k + \left(1 - \frac{\eta_3}{\nu_3}\right) n_{Th}^c + \frac{\eta_3}{\nu_3} n_{Pa} - \left(1 - \frac{\eta_3}{\nu_3}\right) (N_3^k + N_{Pa}^k) - N_4^k - N_5^k$$

and

$$a_{HE}^k = a_{HE} + \frac{1}{n_{Th}^c} \left\{ m_5^k - \left[1 + 2 \left(1 - \frac{\eta_3}{\nu_3}\right) \right] (N_3^k + N_{Pa}^k) - 2N_4^k - N_5^k \right\}$$

$$y^k = y + \frac{1}{n_{Th}c} \{ \eta_5 m_5^k - \eta_3 + \eta_5 (1 - \frac{\eta_3}{v_3}) | (N_3^k + N_{Pa}^k) - \eta_5 (N_4^k + N_5^k) \}$$

$$\begin{aligned} \phi^k = \phi + \frac{1}{n_{Th}c} \{ \frac{\eta_5}{v_5} m_5^k - \frac{\eta_3}{v_3} + \frac{\eta_5}{v_5} (1 - \frac{\eta_3}{v_3}) | (N_3^k + N_{Pa}^k) - \\ - \frac{5}{5} (N_4^k + N_5^k) \} \end{aligned}$$

where a_{HE} , y and ϕ are the corresponding equilibrium values.

For $a_{HE}^{(k)} = a_{HE}$, i.e., the same number of captures in fission products per pass during the transient, we have

$$a_{FP} = \frac{\epsilon(1-L)f [(y^k - y) - (a_{HE}^k - a_{HE})]}{[(\phi^k - \phi) + \frac{\epsilon_{Th}^{fis} - 1}{v_{Th} - 1} [(y^k - y) - (a_{HE}^k - a_{HE})]]}$$

if we neglect the variation of f , L and ϵ with k .

With (N_i = the equilibrium value of i nuclid content),

$$N_i = \sum_k N_i^k$$

we can obtain the total fissile initial inventory

$$m_5 = \sum_k m_5^k$$

as a function of the equilibrium SSET cycle characteristics and N_i , i.e.,

$$m_5 = N_5 - \frac{2 + a_{FP} \frac{\eta_5}{v_5} - \eta_5}{\epsilon(1-L)f\eta_5 - 1 - a_{FP} \frac{\eta_5}{v_5}} N_4 + \left[1 - \frac{\eta_3}{v_3} + \frac{\epsilon(1-L)f\eta_3 - (2 - \frac{\eta_3}{v_3}) - a_{FP} \frac{\eta_3}{v_3}}{\epsilon(1-L)f\eta_5 - 1 - a_{FP} \frac{\eta_5}{v_5}} \right] (N_3 + N_{Pa})$$

The main uncertainties in m_s evaluation are due especially to the uncertainties of N_i evaluations.

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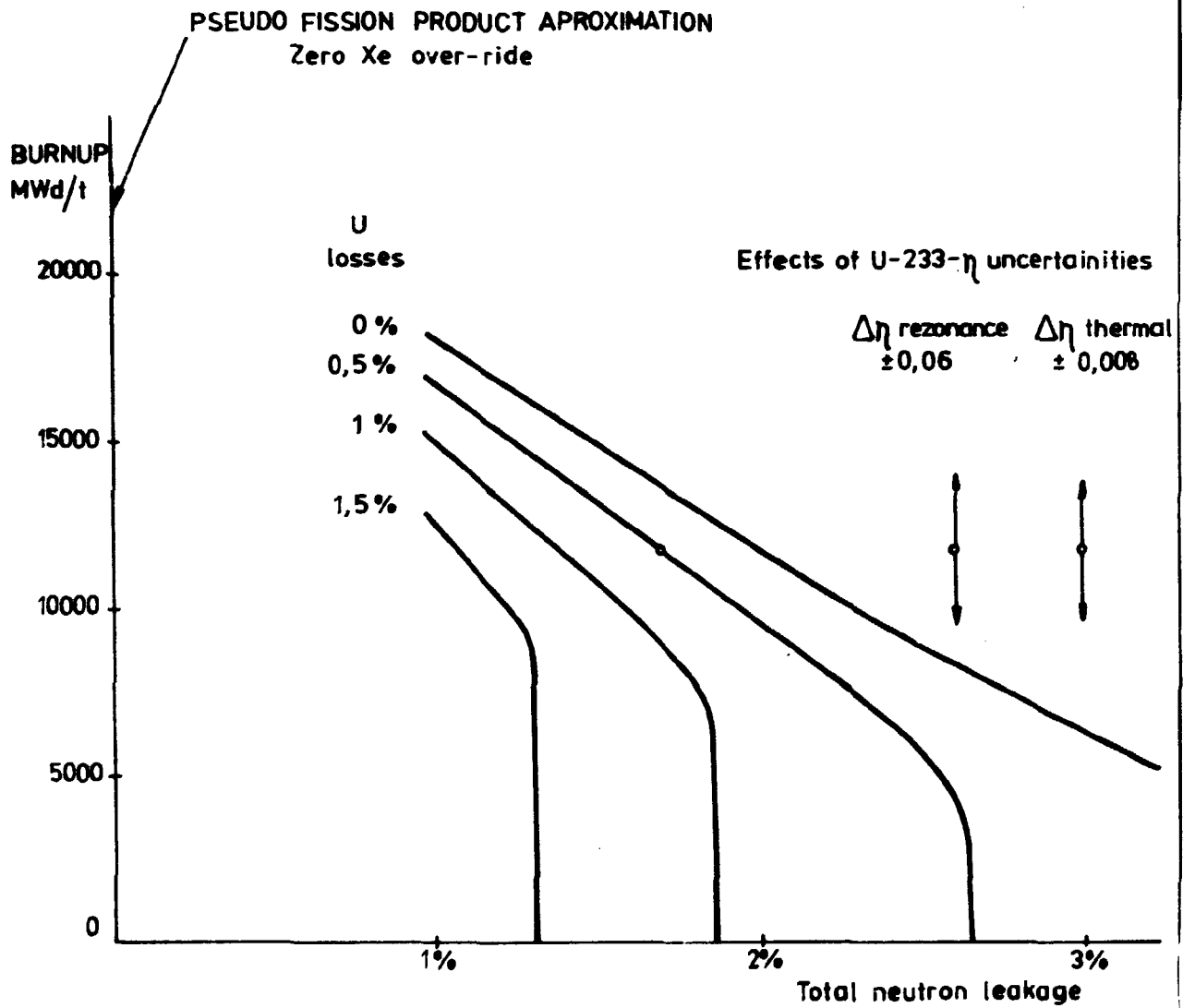


Fig. 1

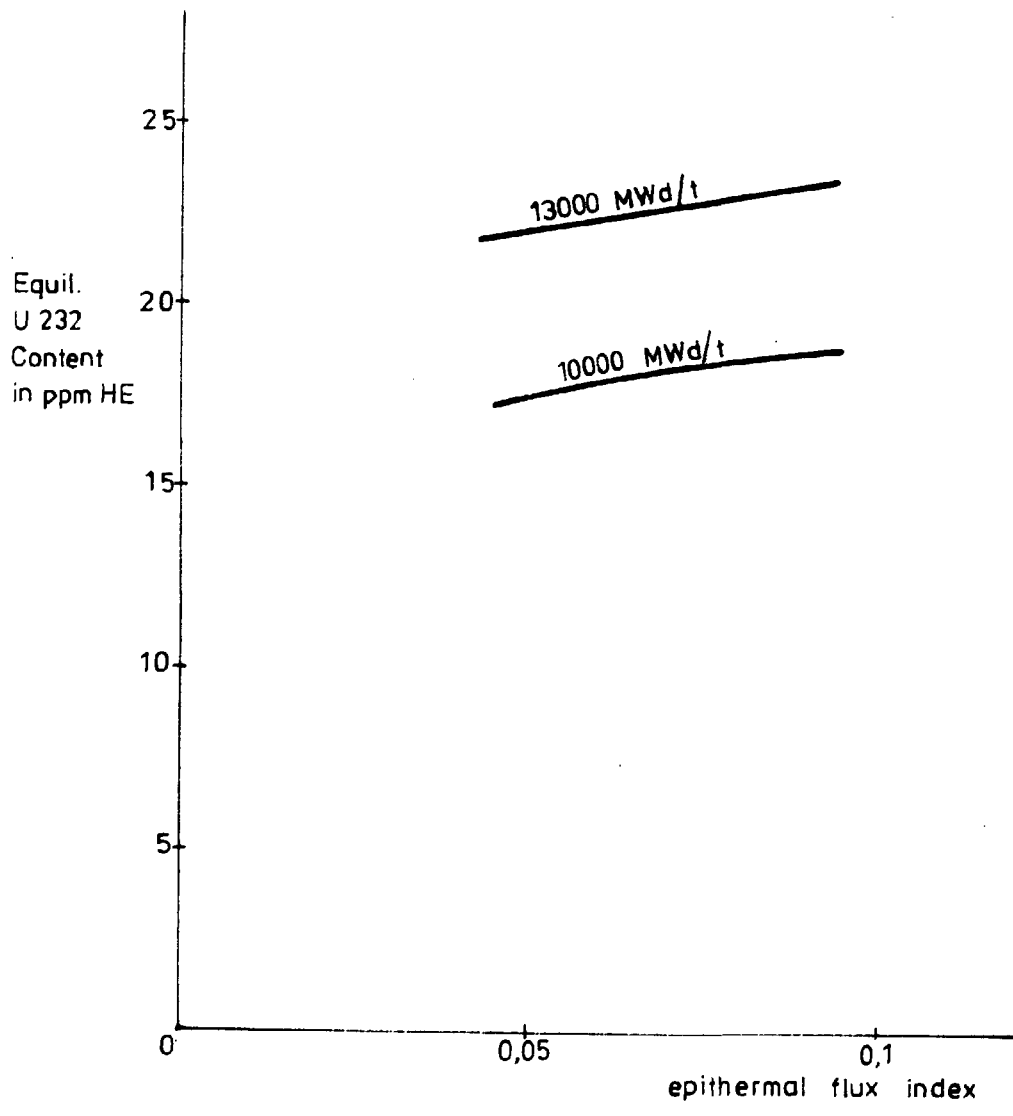


FIG. 2

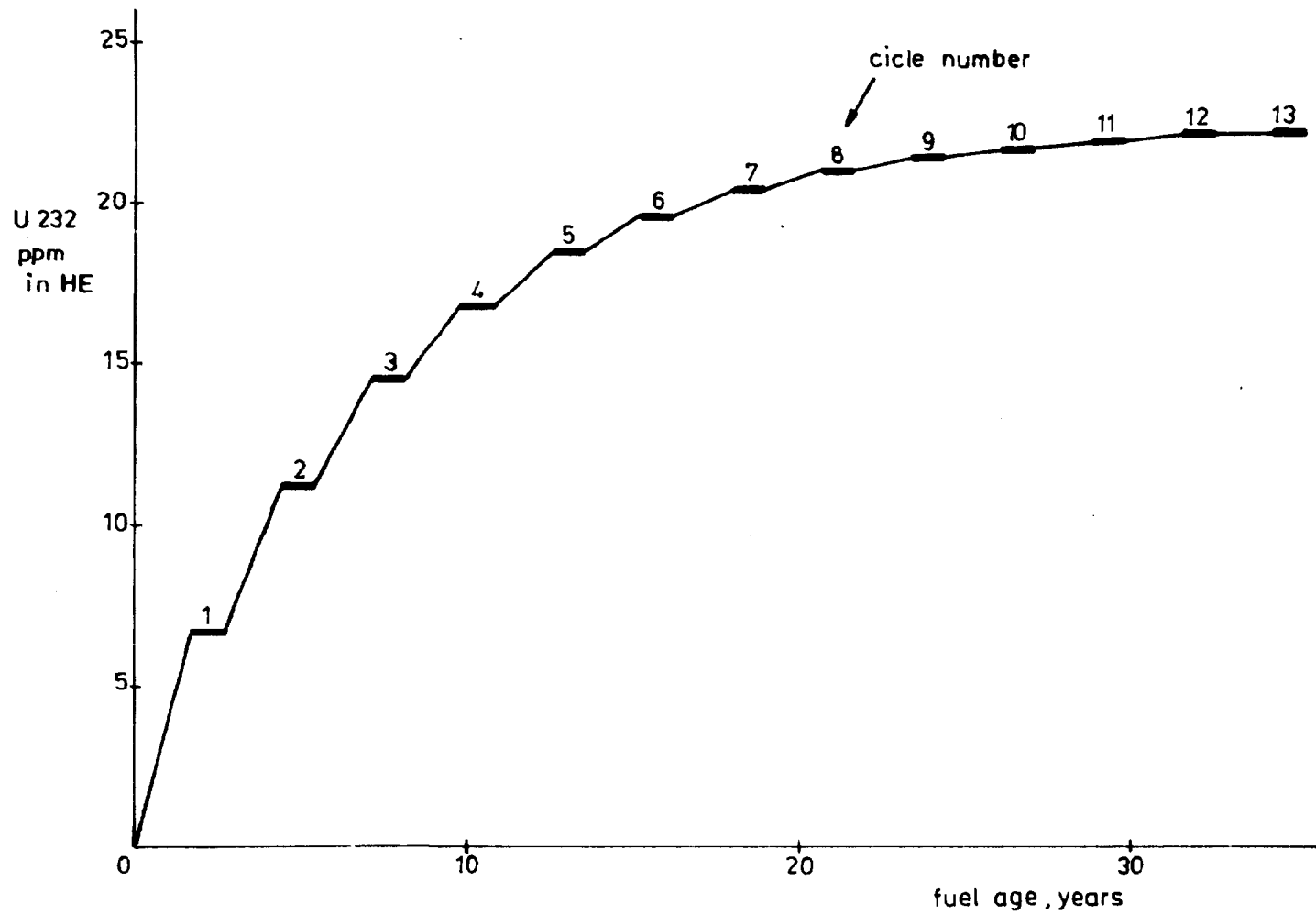


FIG. 3

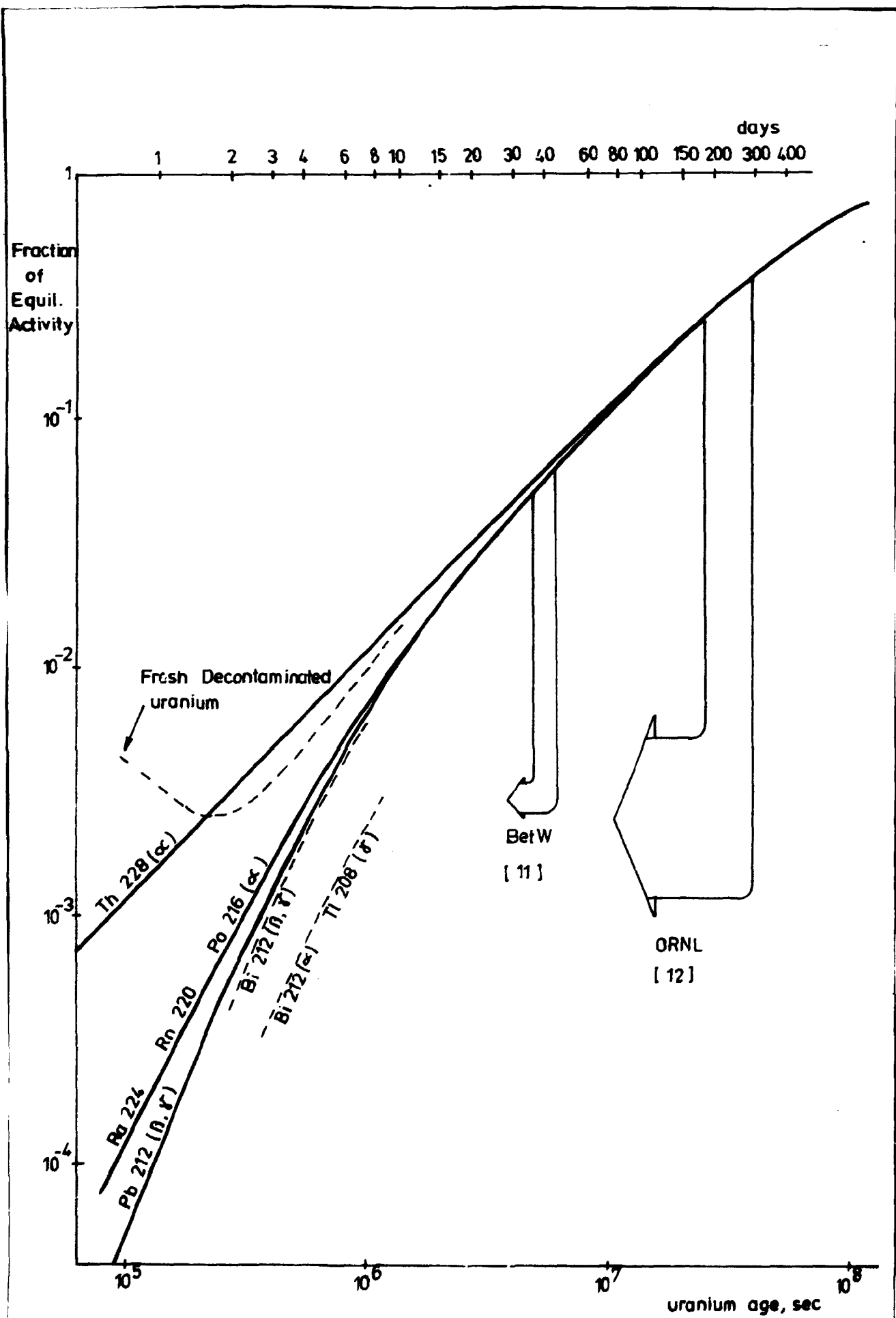


Fig. 4

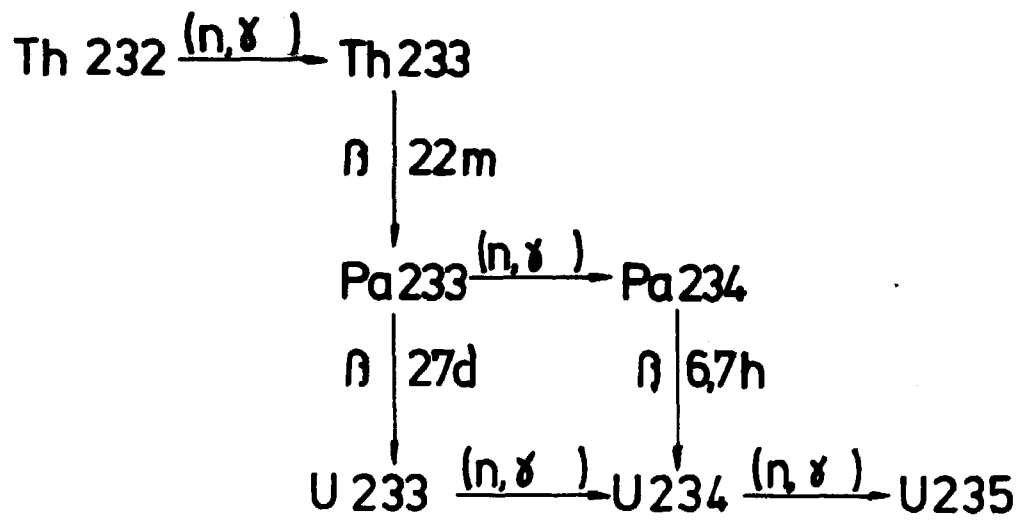


Fig. 5

----- Miligram and Walker AECL 5561, 1976

———— Our evaluations

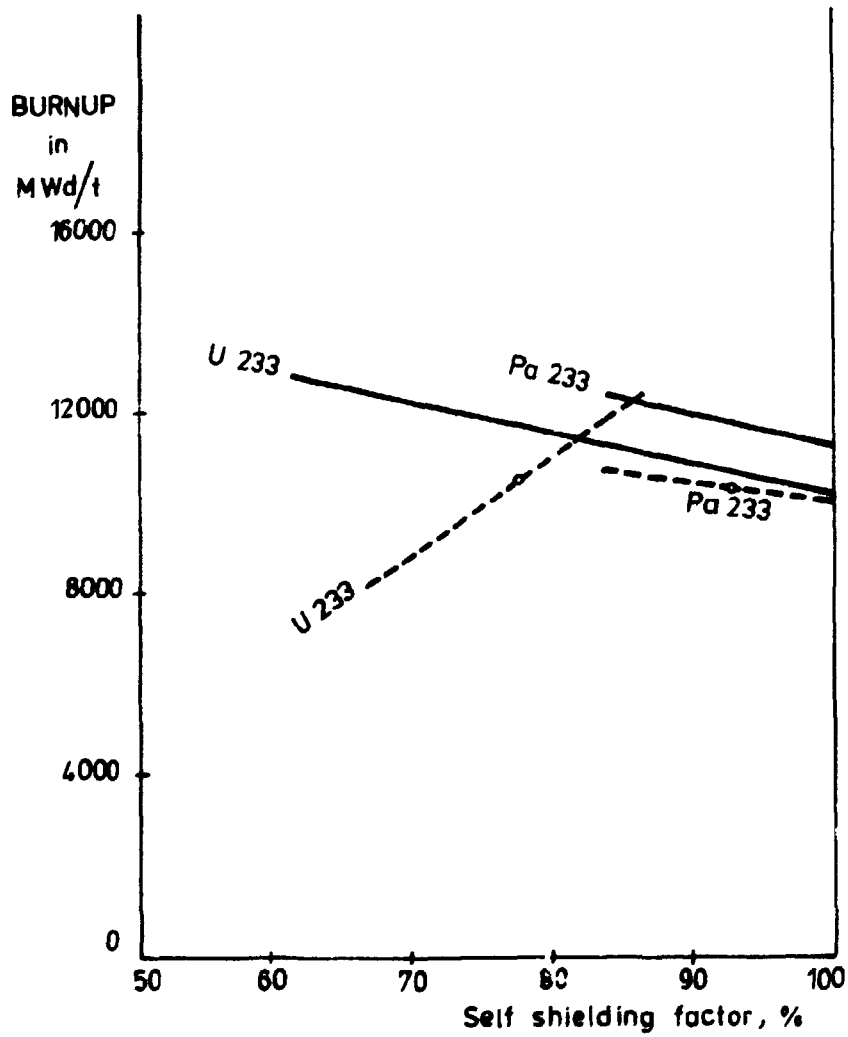


Fig. 6

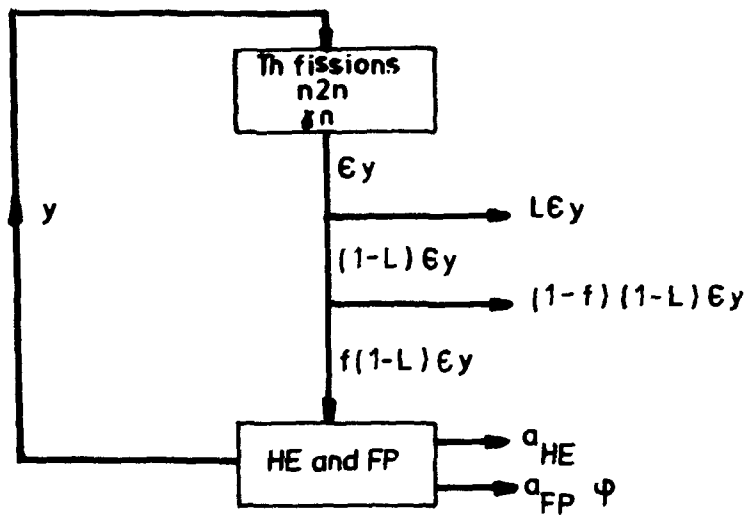


Fig. 7