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Design and Analysis of Molten Salt Reactor Fueled by TRU from LWR

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This study assesses the feasibility of designing a finite once-through Molten Salt Reactor (MSR) fed with trans-uranium isotopes (TRU) from LWR spent fuel to be critical and to have a low peak-to-average radiation damage to graphite. The study also quantifies the transmutation effectiveness of this MSR considering the following measures: fractional transmutation of all actinides, of ^{239}Pu and of ^{237}Np and its precursors, radio-toxicity and decay-heat.

Three-dimensional design optimization with MCNP resulted in a graphite reflected critical core having a remarkably small peak-to-average graphite damage rate of 1.079. The optimal graphite-to-MS (C/MS) volume ratio is between 2 and 3. The transmutation characteristics of this reactor strongly depend on the MS feed-rate. They also depend on the C/MS ratio. There is no universally preferred neutron spectrum – to minimize radiotoxicity and decay heat, harder spectrum is preferred during the first few dozen years following discharge from the MSR as well as in the ten thousand and hundred thousand years time range. However, softer spectrum is preferred for periods in-between. Softer spectrum is also preferred for minimizing the inventory of fissile Pu and of ^{237}Np and its precursors and for maximizing the graphite lifetime.

KEYWORDS: Molten salt reactor, NaF-ZrF$_4$, transmutation, transuranium isotopes, LWR spent fuel, radiotoxicity, decay-heat

1. Introduction

Previous studies [1,2] reported that it is possible to design once-through Molten Salt Reactors (MSR) that are fed with the trans-uranium isotopes (TRU) from LWR spent fuel. These studies were based on the analysis of unit cells that were finite in the axial direction but infinite in the radial direction. They identified the spectrum that maximizes k_{eff} under the constraint of the solubility limit of actinides in the NaF-ZrF$_4$ molten salt (MS).

One objective of the present study is to assess the feasibility of designing a finite once-through MSR fed with transuranium (TRU) isotopes from LWR spent fuel to be critical.

Another objective is to optimize the reflectors so as to flatten, as much as possible, the fast neutron flux across the core. As the core lifetime is dictated by the radiation damage rate to the graphite moderator and structural material, and as this radiation damage is proportional to the fast neutron flux, it is desirable to minimize the peak-to-average fast flux across the core.

An additional objective is to quantify the transmutation effectiveness of this MSR. Six measures are used to quantify this effectiveness: fractional transmutation of all actinides, of fissile Pu and of ^{237}Np and its precursors, radio-toxicity and decay-heat.

An improved version of the methodology described in references [1,2] is used for the analysis. All the neutronic calculations are done with MCNP. Fission products are extracted as soon as they are generated. The effect of finite extraction time is to be addressed in a follow-on study. The small effect of delayed neutrons emission out of the core is ignored.

2. Core and reflector optimization

2.1 Reference Unit Cell

The unit cell concept is that proposed by Bowman [3]; it is made of an hexagonal prism of graphite with a central MS channel. The top and bottom 10 cm of the graphite is loaded with boron so as to minimize thermal neutrons induced fissions in the MS plenum. The molten salt is made of equal moles of NaF and ZrF_4. The MS is continuously fed at a rate of 1.067 liters per day per GW_{th} along with TRU from spent fuel from LWR from which the U isotopes were extracted. The TRU feed rate is adjusted so as not to exceed the actinide solubility limit [1]. The actinides composition fed into the MSR is defined in reference [1]. The average power density is 390 watts per cm^3 of molten salt.

The previous study [1] found that under the actinides solubility limit constraint, the multiplication factor, k, peaks for a graphite-to-molten salt volume ratio (C/MS) of approximately 3. A MS channel diameter of 7 cm was recommended [1].

2.2 Finite Core Considered

In order to minimize the number of MSR dedicated for transmutation and in order to minimize the neutron leakage probability the target thermal power for the finite core was set to be approximately 6000 MW_{th}. This is close to the thermal power output of the new French 1600 MW_e PWR. But whereas the PWR is highly pressurized, the MSR is not pressurized at all and is not subject to fuel meltdown accidents.

The core height-to-diameter ratio was assumed to be 0.924; according to one-group diffusion theory this ratio minimizes the neutron leakage probability from a given volume core. The core chosen have 955 unit cells that are 4 m long. The corresponding total reactor power is 5733 MW_{th}.

A 27 cm thick MS plenum is assumed located below and above the core. Graphite reflectors are located on the other side of the plenum. A graphite reflector also surrounds the core in the radial direction. The heat exchangers are assumed to be located beyond it.

The average MS and graphite temperature is assumed to be 650 ^oC. The graphite lifetime is assumed to correspond to a fluence limit of $3 \cdot 10^{22}$ neutrons of E > 0.01 MeV per cm^2.

2.3 Radial Reflector

Graphite of different thickness was tried for the radial reflector. MCNP was used to calculate three parameters: k_{eff}, power distribution and fast (> 0.01 MeV) neutrons flux distribution. Partial results of the parametric study are summarized in Table 1.

Table 1 Effects of graphite radial reflector thickness

Graphite thickness [cm]	k_{eff}	Radial leakage probability	Peak-to-average power density
0	1.00247± 0.00067	2.56449E-02	2.33431
40	1.01916	2.73610E-02	1.32359
60	1.02574	2.84630E-02	1.20721
80	1.02848	2.78310E-02	1.28900

It is found that, as expected, k_{eff} increases with increasing reflector thickness; the incremental increase beyond 80 cm does not justify considering thicker reflectors. Unexpectedly, the radial neutron leakage probability of the reflected cores is larger than that

of the bare core. This is due to the radial power flattening effect of the graphite reflector, displayed in Figure 1.

Figure 1 shows that the flattest radial power distribution is obtained with a reflector that is approximately 60 cm thick. In fact, the MSR core can accommodate a large power peaking factor because its fuel is in a liquid state and serves also as the heat transport fluid. What is of concern, though, is the radiation damage rate to the graphite. As the actinides solubility limit is very small, the neutron flux level needed for attaining desirable core power density is very high – in the range of 10^{15} n/cm^2-s. With such large fluxes, the graphite lifetime was estimated [1,2] to be on the order of 1 to 2 years if the fast neutron flux is perfectly flat across the core.

The flattest radial fast neutron flux distribution, shown if Figure 2, was obtained using a 80 cm thick reflector. The peak-to-average ratio is less than 10%!

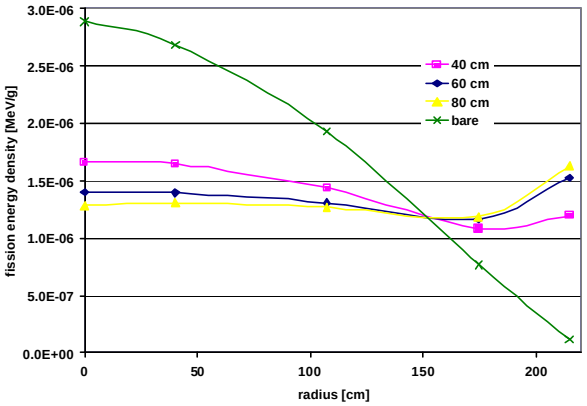

Fig. 1 Radial fission energy deposition comparison for different thicknesses of graphite

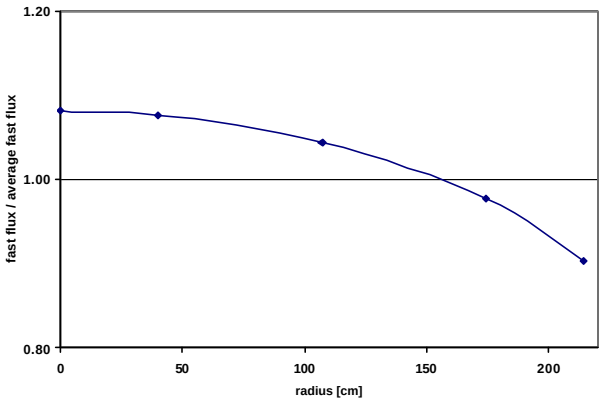

Fig. 2 Fast flux-to-average fast flux ratio in the radial direction

2.4 Axial Reflector

More complicated is the optimization of the axial reflector. This is due to the need to incorporate a MS plenum at the entrance to the core channels and at the exit from these channels. The plenum was modeled as a cylinder of a diameter of the reflected core and assumed to be 27 cm thick. This thickness makes the maximum MS flow velocity in the plenum equal to the MS velocity in the core channels. The boron was completely removed from the graphite near the axial core boundaries.

The MS in the plenums was found to be a poor reflector, with the actinides dissolved in it. Moreover, with a 27 cm MS buffer between the core and an axial reflector, graphite was found to be highly ineffective. Introduction of graphite in the plenum while increasing the plenum thickness so as to preserve the MS flow cross-section area was found ineffective as well.

The best solution found is to increase the C/MS ratio in a relatively short section near the core entrance and exit. This is obtained by reducing the radius of the MS channels. A parametric study was undertaken to find the optimal combination of MS channel radius reduction and the length of the section with the reduced diameter.

The optimal solution found is a reduction of the MS channel diameter from 7 cm to 5 cm over a 30 cm section from the core axial bases. Figure 3 shows the resulting axial power distribution relative to that of the bare core. 60 cm thick graphite is assumed on the other side of the MS plenums. The resulting total peak to average fast flux ratio is 1.079! The k_{eff} value

is 1.04047± 0.00043. That is, we have approximately 4% of excess reactivity to compensate for the effect of non-instantaneous removal of fission products and of the fraction of the delayed neutrons that are emitted out of the core. The increase in the pumping power as a result of the narrowing of the channels was estimated to be acceptable.

Figure 4 compares the average spectrum in the MS of the optimal MSR (finite model) with that of the unit cell. The spectra are quite similar; more than we expected. It turns out that the spectrum softening effect introduced by the radial and axial reflectors is compensated in the thermal energy range and more than compensated at high energies by the harder spectrum in the plenums.

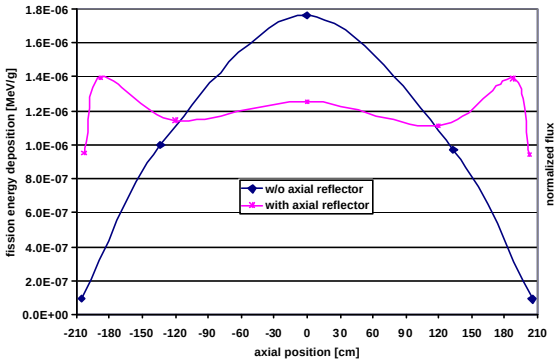

Fig. 3 Axial fission energy deposition without and with axial reflector

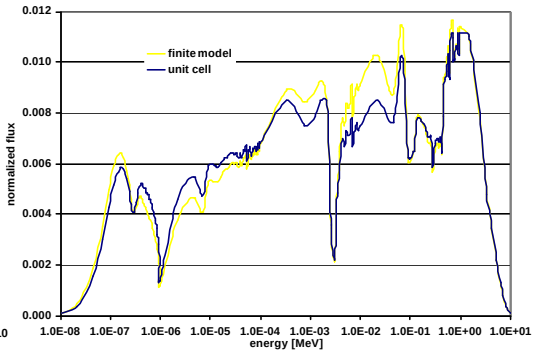

Fig. 4 Neutron spectrum in the MS of the infinite and the finite models

3. Transmutation capability

3.1 Methodology

The transmutation capability of the MSR considered in this work is measured in terms of the following characteristics: (1) Fractional transmutation – the fraction of the actinides that are fed into the MSR from LWR spent fuel that is transmuted in one pass through the core. (2) Radiotoxicity. (3) Decay heat.

All these parameters are calculated for MSR the actinides composition of which reached equilibrium; that is, the rate of introduction of a given actinide due to the continuous feed, to radioactive decay of parent nuclei and to transmutation equals the rate of elimination of that actinide due to continuous extraction, decay and transmutation.

The equilibrium composition is calculated from the following balance equation:

$$\sum_j \sigma_{j\to i}\,\phi\,\frac{N_j}{2} + \sum_j \lambda_{j\to i}\,N_j - \lambda_i\,N_i - \sigma_i\,\phi\,\frac{N_i}{2} + F_i - R\,N_i = 0 \qquad (1)$$

where N_i, σ_i, λ_i and F_i are, respectively, the atomic density, the effective one-group absorption cross section, the decay constant and the feed rate of the i^{th} actinide, $\sigma_{j\to i}$ is the effective one-group cross section for conversion of actinide j into actinide i, $\lambda_{j\to i}$ is the decay constant of actinide j into actinide i and R is the fractional removal rate of the MS from the reactor.

Equation 1 is solved iteratively. The flux and the cross-sections are first calculated with MCNP for an assumed initial actinides composition and are plugged into Equation 1. This equation is solved using a MATLAB program to obtain a new composition. This new composition is used for another MCNP run and so on. The process converges when the variation of k_{eff} and actinides concentration in two successive iterations is small enough.

The study considers hexagonal unit cell that is finite in the axial direction and infinite in the radial direction. The MS plenums are accounted for. 34 actinides are considered; from U to Cf. The actinides solubility limit is assumed to be 1.56 mole %. Three design variables are considered in this parametric study: C/MS, MS feed rate and power density.

3.2 Effect of MS Feed Rate

Table 2 shows the effect of the MS feed rate on the fractional transmutation of all actinides along with three design parameters. While the feed rate is changed from 1.07 to 0.13 liter/day-GW_{th}, the actinides mole ratio in the feed is increased so as to make the equilibrium concentration as close as possible to the solubility limit. It is seen that the fractional transmutation (FT) increases as the MS feed rate decreases. This is due to an increase in the actinides residence time in the core (Table 2) as the MS extraction rate drops. k_{eff} tends to slightly increase with the MS feed rate while the graphite life time is practically independent of the feed rate.

Table 2 Effects of MS feed rate. Power density = 390 W/cm^3. C/MS = 3.

MS feed rate [liter/(day-GW_{th})]	1.07	0.53	0.27	0.13
Fractional transmutation	89.56 %	94.49 %	97.20 %	98.60 %
Residence time [years]	13	26	53	105
k_{eff}	1.02351	1.02031	1.01760	1.01869
Graphite life time [years]	1.379	1.374	1.371	1.371

Feed rate variations also effect the actinides composition; the composition shifts towards heavier mass isotopes.

3.3 Effect of Neutron Spectrum

Table 3 shows the effect of neutron spectrum variation, brought about by changing the graphite-to-MS volume ratio, on the characteristics considered. As expected, the fractional transmutation (FT) of all actinides is practically independent of the C/MS ratio; the recoverable energy per fission only slightly depends on the fissioning isotope and one fission reaction destroys one actinide nucleus. However, the fractional transmutation of ^{237}Np and its precursors and of fissile Pu is enhanced with spectrum softening. k_{eff} peaks for C/MS around 2, and the graphite lifetime increases with spectrum softening.

Table 3 Effects of graphite-to-MS volume ratio. 390 W/cm^3. 1.07 liter/day-GW_{th}.

Graphite-to-MS volume ratio	1	2	3	4
FT of all actinides	89.56 %	89.56 %	89.56 %	89.57 %
FT of ^{237}Np and precursors	0.860	0.876	0.906	0.927
FT of ^{239}Pu	0.957	0.972	0.982	0.987
k_{eff}	1.01526	1.02559	1.02351	1.01666
Graphite life time [years]	0.632	0.998	1.379	1.734
(Pu^{239} + Pu^{241})/ Pu	0.352	0.259	0.176	0.128

3.4 Effect of Power Density

Table 4 summarizes selected results obtained with a10 fold reduction in the power density – from 390 down to 39 w/cm^3 of MS. Comparing these results to those of Table 2 it is observed that whereas the fractional transmutation is practically unaffected, there is, as expected, an

order of magnitude increase in the graphite lifetime. Unfortunately, k_{eff} value has dropped 6% to 7% and is below unity; it is due to a reduction in the probability to fission short-lived actinides, such as ^{238}Np, before they radioactively decay to other actinides having higher capture-to-fission ratio. These results are consistent with findings reported in reference [1].

Table 4 Effects of MS feed-rate. 39 W/cm^3. 1.07 liter/day-GW$_{th}$.

MS feed rate [liter/(day GW$_{th}$)]	1.07	0.53	0.27	0.13
Fractional transmutation	89.47 %	94.37 %	97.15 %	98.55 %
k_{eff}	0.96159	0.95514	0.95443	0.95481
Graphite life time [years]	13.91	13.89	13.88	13.88

3.5 Radiotoxicity

The radiotoxicity of the actinides leaving the MSR core with the extracted MS is measured by the ingestion-toxicity index – the volume of water with which the actinides must be diluted so that drinking the water will result in accumulation of radiation dose at a rate no greater than 0.5 rem/year.

Figures 5 and 6 show the time evolution of the radiotoxicity for two different feed rates and for graphite-to-MS volume ratio of between 1 and 4. Time zero corresponds to the moment of actinides extraction from the MSR. It is found that the radiotoxicity is not sensitive to the C/MS ratio in the C/MS range considered. There is no uniquely preferred spectrum – whereas shortly after extracting the actinides from the hardest spectrum (C/MS=1) core gives the highest radiotoxicity, in the 100 years time frame it gives the lowest radiotoxicity. These fluctuations continue several more times. They are due to differences in the relative concentration of different radionuclides. These differences are illustrated in Figures 7 and 8. Both correspond to a feed rate of 1.07 liter/day-GW$_{th}$.

Also shown in Figures 5 and 6 is the radiotoxicity of the actinides fed into the MSR. At a 1.07 liter/day-GW$_{th}$ feed rate, the radiotoxicity of the feed is lower than that of the waste for the first 30 years. This is due, primarily, to the buildup of ^{244}Cm (Figures 7 and 8). Thereafter the radiotoxicity of the actinides that come out from the MSR is smaller by up to an order of magnitude. At a feed rate of 0.13 liter/day-GW$_{th}$, the radiotoxicity of the actinides coming out from the MSR is always lower than that of the actinides from LWR spent fuel.

3.6 Decay-heat

Similar trends are observed for the decay heat, as illustrated in Figures 9 and 10. The dominant contributors to the decay heat, shown in Figure 11, are quite different, though.

4. Conclusions

Three-dimensional design optimization with MCNP resulted in a critical core having a remarkably small peak-to-average graphite damage rate of 1.079. This flatt flux was achieved by using an 80 cm thick radial graphite reflector and by narrowing the MS channels diameter at the outermost 30 cm section on both axial core ends from 7 cm to 5 cm. The C/MS value chosen is 3 although the peak k_{eff} is expected to be between C/MS of 2 and 3.

The transmutation characteristics of this reactor strongly depend on the MS feed-rate. They also depend on the neutron spectrum, i.e., on the C/MS ratio. There is no universally preferred spectrum – to minimize radiotoxicity and decay heat, harder spectrum is preferred during the first few dozen years following discharge from the MSR as well as in the ten thousand and hundred thousand years time range. However, softer spectrum is preferred for in-between

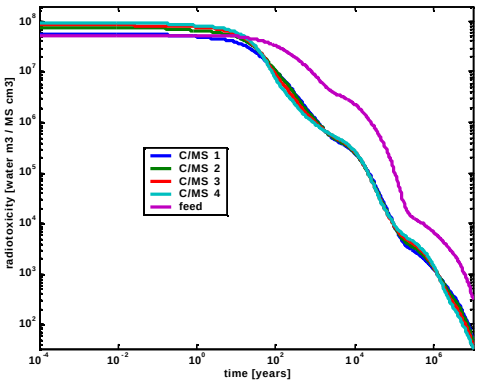

Fig. 5 Radiotoxicity time evolution for feed rate 1.07 liter/day-GW_{th}

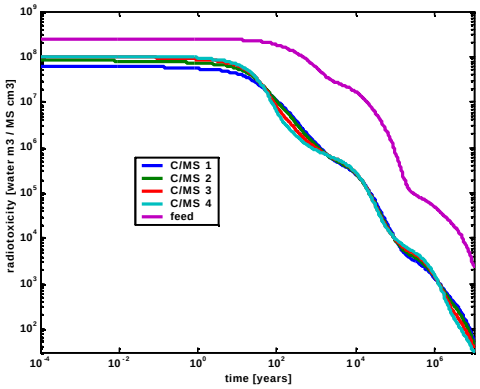

Fig. 6 Radiotoxicity time evolution for feed rate 0.13 liter/day-GW_{th}

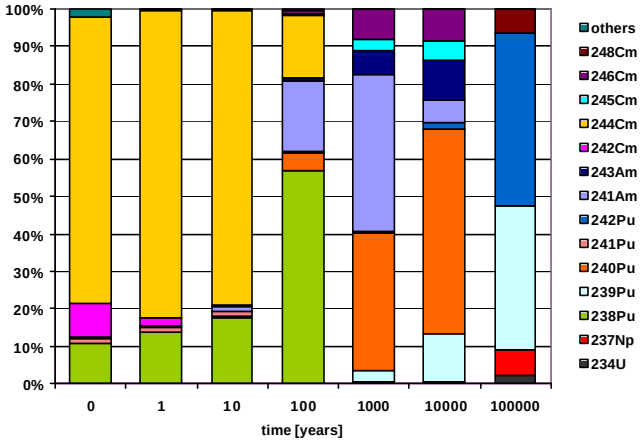

Fig. 7 Fractional radiotoxicity at selected times following extraction from MSR. C/MS=3.

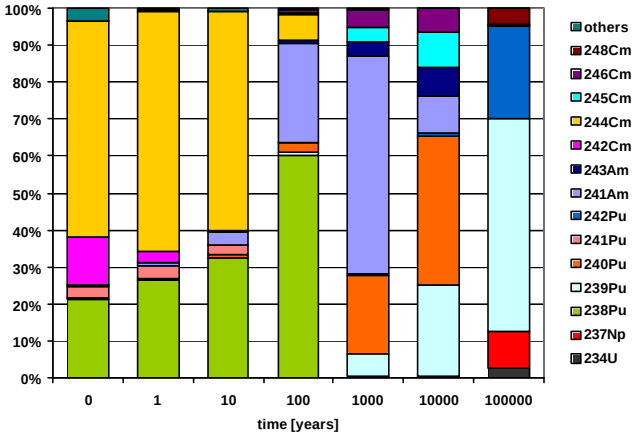

Fig. 8 Fractional radiotoxicity at selected times follo wing extraction from MSR. C/MS=1.

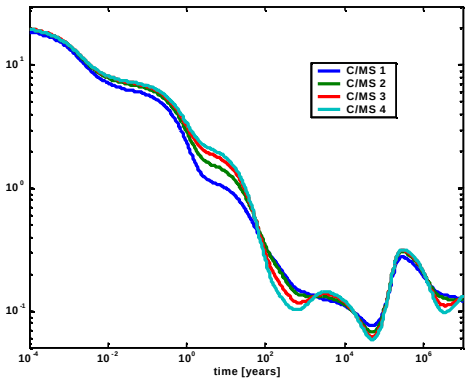

Fig. 9 Waste-to-feed decay heat ratio time evolution for feed rate 1.07 liter/day-GW_{th}

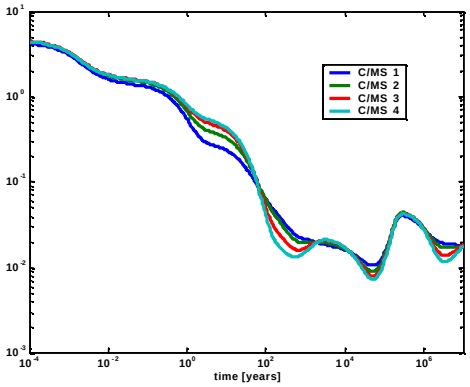

Fig. 10 Waste-to-feed decay heat ratio time evolution for feed rate 0.13 liter/day-GW_{th}

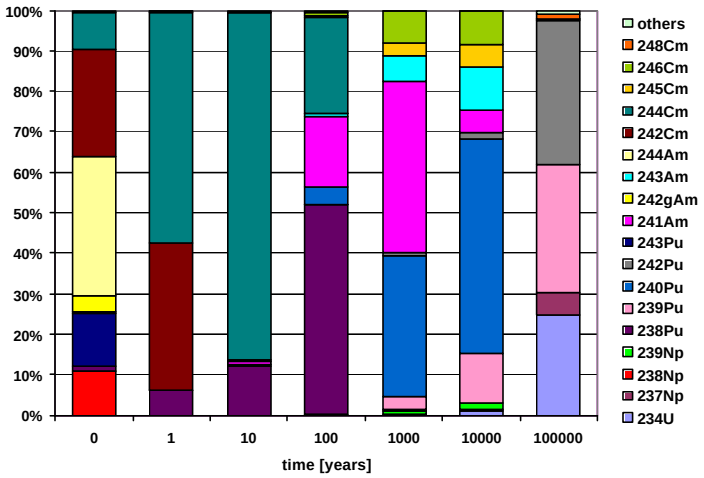

Fig. 11 Fractional decay heat at selected times following extraction from MSR. C/MS=3.

periods. Softer spectrum is also preferred for minimizing the inventory of ^{237}Np and its precursors and of fissile Pu and for maximizing the graphite lifetime. Firmer conclusions should await analysis of fission products effects.

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