Fast Reactor Physics

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Abstract

A very simplified model is used to simulate the equilibrium fuel cycle in a sodium-cooled fast reactor, considering (for the sake of comparison) two feed fuels: natural uranium and natural thorium, assuming recycling of all actinides under conditions of constant power density and constant fuel mass. The balance of reaction rates, equilibrium fuel composition, neutron balance, main safety parameters, as well as radiotoxicity and decay heat level of the equilibrium fuel are presented. The paper is a shortened version of the lecture given at FJOH'2013 summer school (Mikityuk, Equilibrium closed fuel cycle, 2013, [1]).

Introduction

A fast neutron critical reactor is a category of nuclear reactor in which the fission chain reaction is sustained by fast and resonance neutrons. Such a reactor needs no neutron moderator and typically uses stainless steel as a cladding and sodium as a coolant. Typical neutron spectra as well as the unit cell schematics are compared in Fig. 1 for a sodium-cooled fast reactor (SFR) and a pressurized water reactor (PWR).

The materials loaded in the reactor core exhibit nuclear transmutations, that is, conversions of one element or isotope into another due to reactions with neutrons (including fission, (n, γ), (n, 2n), (n, α) reactions, etc.) and due to radioactive decays (including spontaneous fission, α- and β-decays, electron capture). A transmutation of actinides with low fission probability (e.g., fertile U-238 or Th-232) into actinides with high fission probability (e.g., fissile Pu-239 or U-233) is called breeding or conversion. Owing to the nuclear properties of actinides in a high neutron energy region, a fast-spectrum nuclear reactor can be designed to enable efficient indirect burning of actinides available in nature (U-238 and Th-232) and recycling of all actinides, producing as final waste fission products only.

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In general, the nuclear fuel cycle is the progression of nuclear fuel from mining to final disposal through enrichment, fabrication, irradiation, cooling, and reprocessing (which is, in fact, separation of elements) [2]. The actinides periodically added to the fuel cycle are called feed fuel. The feed fuel can include natural uranium and natural thorium, enriched and depleted uranium, and weapons-grade actinides from nuclear weapons. The fuel cycle based on U-238 as a feed fuel is called the uranium fuel cycle, whereas the fuel cycle based on Th-232 as a feed fuel is called the thorium fuel cycle.

An open (or once-through) uranium fuel cycle does not include fuel reprocessing and is currently used at nearly all nuclear power plants. To reduce the radiotoxicity and decay heat load on the final repository, all actinides, which mostly contribute to these parameters, could be separated and recycled in the fast reactor. Diagrams of the corresponding fuel cycles are shown in Fig. 2a and b for U and Th fuel cycles, respectively. In this option, only fission products and reprocessing losses go to the final repository.

Owing to the unavailability of the thorium cycle fissile element (U-233), fissile material (plutonium) should be borrowed from the uranium cycle to start the thorium fuel cycle until enough U-233 is generated.

In this paper, the equilibrium fuel cycle, which is closed on all actinides for both the U and Th fuel cycles (EQL-U and EQL-Th), is considered. The equilibrium cycle is a cycle with a sufficiently large number of repetitive cycles, after

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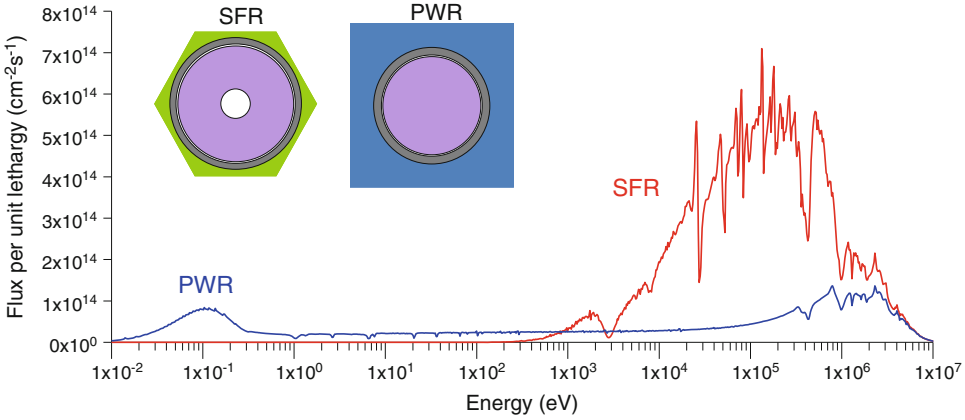

Fig. 1 Typical neutron spectrum of fast (SFR) and thermal (PWR) reactors as well as the schematics of the SFR and PWR unit cells [1]

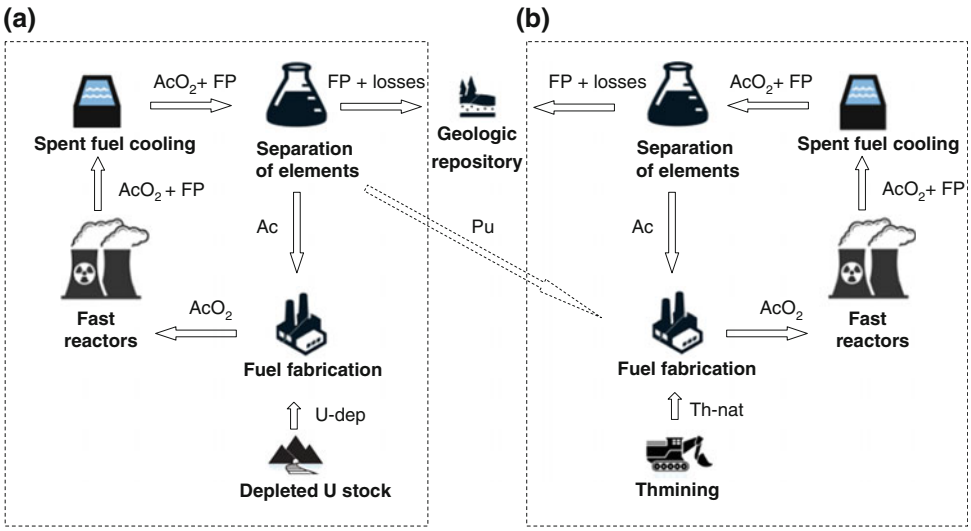

Fig. 2 Uranium (**a**) and thorium (**b**) fuel cycles closed on all actinides. *U-dep* depleted uranium; *Th-nat* natural thorium; *Ac* all actinides of the corresponding cycle; *FP* fission products

which the composition of actinides, averaged over the whole cycle duration from cycle to cycle, remains unchanged and the resulting reactor parameters can be considered as asymptotic characteristics of the reactor with imposed power, feed fuel, and reprocessing parameters. In this context, any reactor operation in repetitive fuel cycles can be considered as in transition from beginning-of-life to the asymptotic equilibrium state.

Calculation Tools, Assumptions, and Models

To study the mass flows, balance of reaction rates, neutron balance, radiotoxicity, and decay heat level of the equilibrium fuels, a simplified model of the equilibrium fuel cycles was developed, using the following assumptions and simplifications:

- The neutron spectrum is calculated for a unit cell, simulating the infinite fuel lattice of typical SFRs. No neutron leakage is considered;
- The power density and the total mass of actinides remain constant;
- The cooling and reprocessing times are assumed to be zero, which is equivalent to the assumptions of on-line feeding and reprocessing;
- The losses are assumed to be negligible and, therefore, only fission products (FPs) are removed from the system and replaced by the feed fuel to keep the mass of the actinides constant;
- The fission products are not considered in the neutron balance, mass evolution, evaluation of safety parameters, radiotoxicity, and decay heat.

We used the Serpent 1.1.16 Monte-Carlo code [3] for solving the eigenvalue problem and finding the infinite multiplication factor, the one-group cross sections, and integral flux for fuel region, by using the unit-cell model and an initial guess for fuel composition. After this step, we solved for all actinides a set of the steady-state Bateman equations, describing the evolution (transmutation) of actinides under the given irradiation conditions, characterized

by the set of one-group cross sections and integral flux as well as by decay constants for radioactive isotopes. The resulting fuel composition was then given to the unit-cell model and the Serpent calculation was repeated. When the iterations on k_∞ converged, the safety parameters as well as radiotoxicity and decay heat were calculated. The details of the procedure can be found in reference [1].

More detailed studies, in which neutron leakage as well as reprocessing and cooling times were explicitly simulated, can be found in references [4, 5].

Results for EQL-U and EQL-Th Closed Fuel Cycles in SFR

Reaction Rates

The balance of reaction rates in the EQL-U and EQL-Th fuel cycles closed on all actinides are shown in Fig. 3a and b, respectively. All reaction rates are normalized to the feeding rate, equal to 1000 U-238 and Th-232 atoms added to the system per unit time.

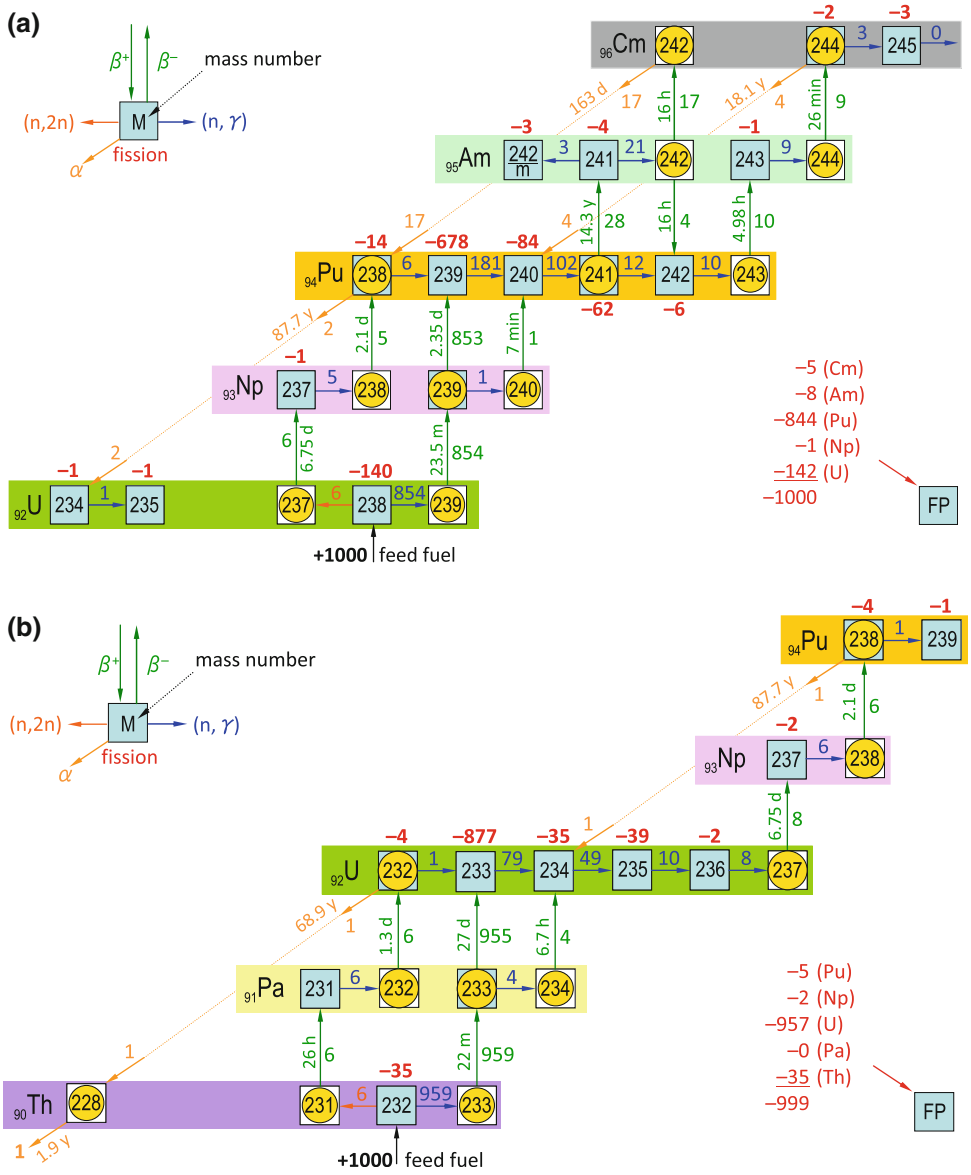

Fig. 3 Balance of reaction rates in the EQL-U (**a**) and EQL-Th (**b**) closed fuel cycles [1]

Table 1 Microscopic cross sections, fission-to-destruction ratios, and mass fractions for the main actinides of the EQL-U and EQL-Th fuel cycles in SFR [1]

	σ_{fc}(b)	σ_c (b)	λ/Φ (b)	F/(A + D)	EQL-U (wt%)	EQL-Th (wt%)
Th^{228}	0.02	0.30	8.69	0.00		0.04
Th^{230}	0.03	0.14		0.18		0.04
Th^{232}	0.01	0.31		0.04		85.6
Pa^{231}	0.25	2.72		0.08		0.06
Pa^{233}	0.07	0.86	224.8	0.00		0.12
U^{232}	2.04	0.59	0.24	0.71		0.05
U^{233}	2.51	0.23		0.92		9.56
U^{234}	0.31	0.50		0.38	0.07	2.98
U^{235}	1.85	0.53		0.78	0.01	0.60
U^{236}	0.10	0.39		0.21	0.04	0.63
U^{238}	0.04	0.27		0.14	81.59	
Np^{237}	0.32	1.49		0.17	0.10	0.13
Np^{239}	0.44	1.86	2474.7	0.00	0.00	
Pu^{238}	1.21	0.50	0.18	0.64	0.31	0.10
Pu^{239}	1.73	0.46		0.79	10.17	0.02
Pu^{240}	0.37	0.46		0.45	5.78	0.01
Pu^{241}	2.46	0.46	1.12	0.61	0.66	
Pu^{242}	0.26	0.46		0.36	0.55	
Am^{241}	0.27	1.74	0.04	0.13	0.36	
Am^{242m}	3.01	0.47	0.11	0.84	0.02	
Am^{243}	0.20	1.55		0.11	0.15	
Cm^{242}	0.62	0.45	35.8	0.02	0.01	
Cm^{244}	0.41	0.82	0.88	0.20	0.11	
Cm^{245}	2.65	0.51		0.84	0.03	
Cm^{246}	0.28	0.50		0.36	0.02	

In equilibrium, every feed fuel nucleus added to the system is burned: either as feed fuel or as another nucleus after one or several transmutations. The mass flow rate between actinides and the distribution of the fission rates over the actinides are determined by the ratio between fission, capture, and decay probabilities, characterized by a one-group fission cross section (σ_f), one-group capture cross section (σ_c) and the decay constant divided by the integral flux (λ/Φ). The fission-to-destruction ratio, $F/(A + D) = \sigma_f/(\sigma_f + \sigma_c + \lambda/\Phi)$, of an actinide characterizes its fissile quality. The higher this value is, the higher the fraction of the fission in the total reaction rate for a given isotope, and the lower the transmutation of this isotope to the higher actinides. As a rule, a lower equilibrium amount of higher actinides is beneficial for the neutron balance, safety parameters, decay heat, and radiotoxicity. In particular, the fuel composition for neutron balance, which is "closer" to the feed fuel composition, means that fewer neutrons are lost for captures. Microscopic cross sections and fission-to-destruction ratios for the main actinides in the EQL-U and EQL-Th fuel cycles are given in Table 1.

From the viewpoint of the source of penetrating radiation from actinides, a property that requires the protection of workers during fuel reprocessing, in the EQL-U cycle this is mainly a neutron source from spontaneous fissions of minor actinides, whereas in the EQL-Th cycle this is mainly a hard gamma source from the products of U-232 decay. It is worth remembering that the fission products are not considered in this study; thus, in the short-term, the gamma source from the fission products dominates [6].

Fuel Composition

The resulting fuel compositions in wt% are shown in Fig. 4 and Table 1 for the equilibrium uranium and thorium cycles. The first observation is that, in the case of the EQL-Th cycle, the fuel composition is "closer" to the feed fuel (Th-232).

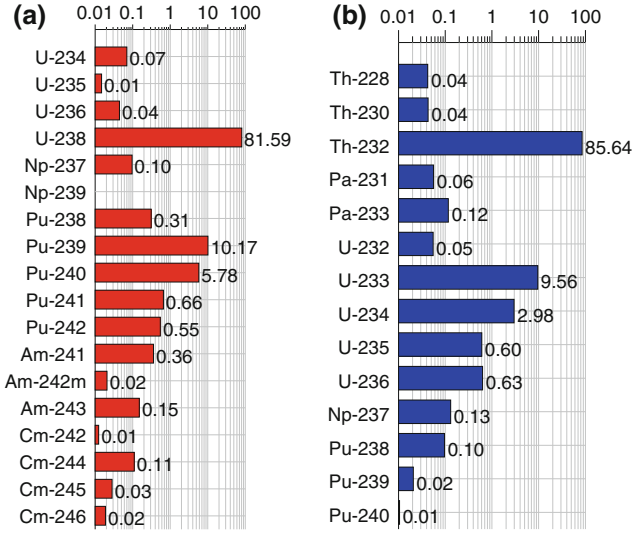

Fig. 4 Fuel composition in EQL-U (**a**) and EQL-Th (**b**) closed fuel cycles in SFR (wt%)

Because of the low mass number of Th-232, a very low amount of minor actinides (Np, Am, Cm, etc.) is generated in the thorium cycle. The second observation is a difference of about 5 % in feed fuel concentration (U-238 and Th-232), which in the equilibrium is determined by the ratio of the macroscopic fission cross section, Σ_f, divided by the microscopic total cross section of feed fuel, σ_t [1]. Although σ_t is about the same for U-238 and Th-232, Σ_f is about 5 % higher in the EQL-Th cycle.

The third observation relates to the ratio between the primary fissile (Pu-239 and U-233) and primary fertile (U-238 and Th-232) concentrations, which is about 12 % and 11 % in the U and Th cycles, respectively. Neglecting small bypass reactions (see Fig. 3a and b), this ratio can be estimated by dividing the capture cross section of the primary fertile by the total cross section of the primary fissile [1, 5].

Neutron Balance

To evaluate the individual contributions of actinides to the multiplication factor, k_∞, we use the following simple neutron balance equation:

$$k_\infty = P/A = \sum_i P_i / \left(\sum_i A_i + A_{CC} \right) \qquad (1)$$

where P is the total neutron production rate and A is the total neutron absorption rate. A can be presented as a sum of the absorption rates of individual actinides plus A_{CC}—the rate of neutron absorption by cladding and coolant (for the sake of simplicity we neglect the absorption by oxygen in fuel). Similarly, P can be presented as the sum of the neutron production rates of individual actinides. If A is normalized to 1, then P equals k_∞. Using this isotope-wise decomposition, we can consider the value P_i/A as a contribution of the isotope, i, to the total multiplication factor.

The isotope-wise decomposition of the numerator (red bars) and denominator (blue bars) of (1) is shown in Fig. 5. The main observation is a significantly lower k_∞ in the EQL-Th cycle compared with the EQL-U cycle, mainly explained by a lower fission rate of the primary fertile isotope Th-232 compared with U-238.

Safety Parameters

The inherent safety of fast-spectrum systems is based on the negative (leakage) component of the void reactivity effect,

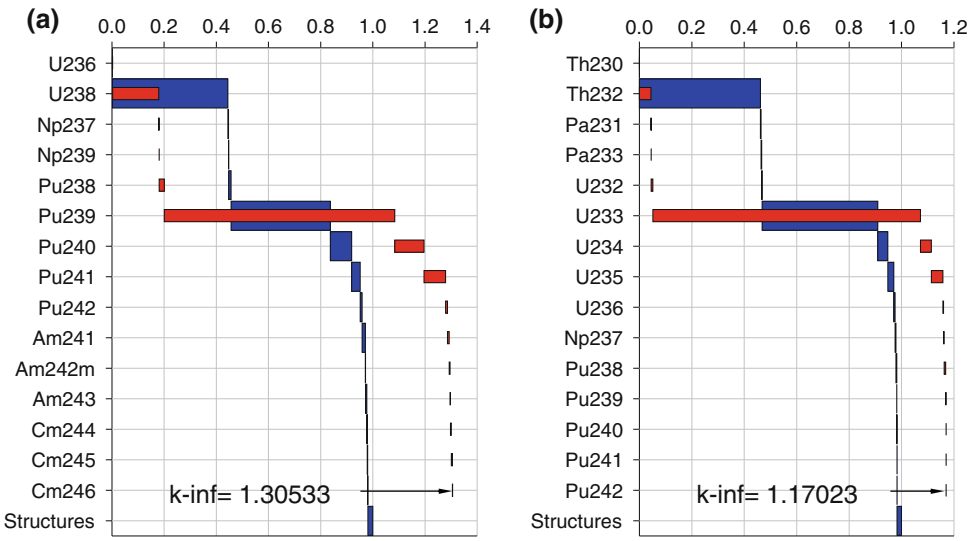

Fig. 5 Isotope-wise decomposition of neutron absorption and production rates normalized to a total absorption rate equal to 1.0 for the EQL-U and EQL-Th fuel cycles. *Blue bars* absorption; *red bars* production

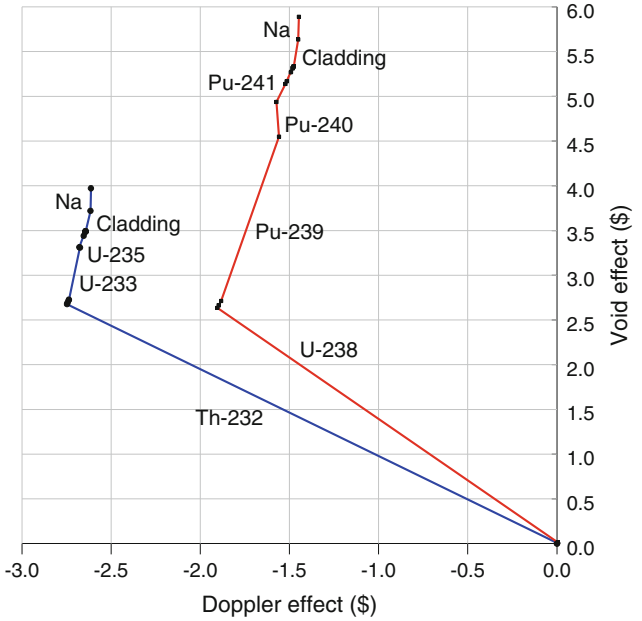

Fig. 6 Decomposition of Doppler and void effects for the EQL-U and EQL-Th fuel cycles

the negative fuel Doppler reactivity effect, and negative reactivity effects from thermal expansions of the in-reactor structures.

We consider the two most important safety parameters: Doppler and void effects.[1] To understand the contribution of individual actinides to the total effect, we made a simple actinide-wise decomposition assumption: the actinide contribution is evaluated as the change of the rate of absorption by the given actinide divided by the total neutron production rate, assumed constant. The results of this evaluation are shown in Fig. 6.

Radiotoxicity and Decay Heat

Radiotoxicity and decay heat for 1 g of the EQL-U and EQL-Th fuel were calculated, by using coefficients derived under framework of another study [7] (Fig. 7). Mainly, all Pu isotopes, Cm and higher actinides as well as their progenies determine the radiotoxicity and decay heat of the fuel in the EQL-U cycle, whereas U-232, Pu-238, and their progenies are responsible for radiotoxicity and decay heat of the EQL-Th fuel for the first millennium. After 1000 years, the progenies of U-233 and U-234 become the dominating isotopes in terms of radiotoxicity and decay heat.

[1]Doppler and void effects are evaluated as changes of the eigenvalue caused by the changes of fuel temperature from nominal to the melting point and of the coolant density from nominal to zero, respectively.

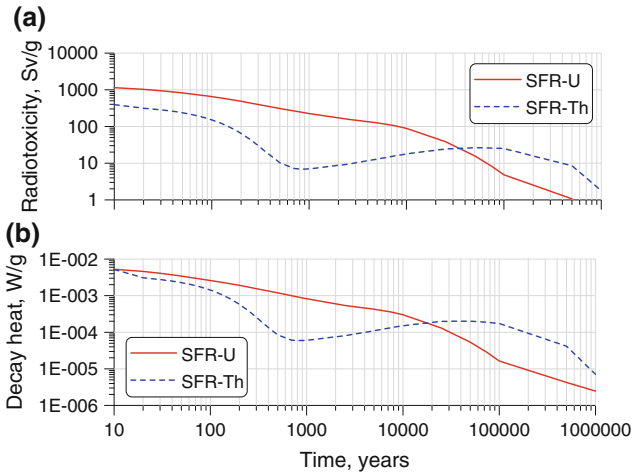

Fig. 7 Radiotoxicity (**a**) and decay heat (**b**) of the EQL-U and EQL-Th fuels

Summary

The advantages and disadvantages of implementation of the thorium cycle in fast reactors in comparison with the uranium cycle can be summarized as follows:

1. Past and current fast reactors were/are based on the uranium cycle. Operational experience with thorium fuel is very limited. Experience in thorium fuel manufacturing and reprocessing is significantly lower compared with that with uranium fuel. This is one of the main obstacles in introducing the Th cycle in fast reactors.
2. An important disadvantage of the Th cycle is that the fissile fuel for this cycle (U-233) is available neither from nature nor from previous reactor operation. Therefore, in order to start the Th fuel cycle, fissile fuel should be borrowed from the U cycle (Pu-239 or U-235).
3. Nevertheless, calculational analysis with state-of-the-art tools shows that fast reactors can operate in an equilibrium thorium cycle closed on all actinides. However, mainly owing to the lower fast fission probabilities of Th-232 compared with U-238, k_∞ of the EQL-Th fuel is significantly lower than k_∞ of the EQL-U one. This means that blankets of fertile material can be required in the thorium fast reactor, which is unfavorable from a non-proliferation viewpoint.
4. Owing to the low mass number of the feed fuel in the Th cycle (Th-232), a very low amount of minor actinides (Np, Am, Cm, etc.) is generated in this cycle and, therefore, the long-lived neutron source, decay heat, and radiotoxicity associated with minor actinides are very low. On the other hand, U-232—precursor of hard gamma emitters—is produced in the Th cycle via the (n, 2n) reaction of Th-232.

5. From a safety viewpoint, the Th cycle has a number of advantages. In particular, because of the nuclear cross sections of the primary fissile and fertile isotopes, the Doppler effect is stronger and the void effect is weaker in the EQL-Th fuel cycle compared with the EQL-U cycle. This trend can be weakened or even reversed for the start-up conditions, when Pu-239 has to be used as the fissile fuel in the Th cycle.
6. From the viewpoint of proliferation risks, the misuse of U-233 is, on one hand, prevented by presence of U-232 (predecessor of hard gamma emitters), but, on the other hand, the precursor of the primary fissile in the Th cycle—Pa-233—has a significantly higher half-life (27 days) compared with the precursor of the primary fissile in the U cycle—Np-239 (2.35 days). This potentially gives time for separation of protactinium, which decays to weapons-grade U-233 practically without the presence of U-232 (due to the low amount of Pa-232 in the equilibrium fuel).
7. Radiotoxicity and decay heat of the equilibrium Th fuel are lower for about the first ten thousand years of cooling compared with the equilibrium U fuel. However, they become higher after ten millennia due to the build-up of decay products.

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