SATURATION CONDITION AND EVOLUTION OF THE NUCLIDES FOR SUB-CRITICAL SYSTEM DRIVEN BY ACCELERATOR

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Abstract

At present work, under initial inventory with ^{232}Th and ^{nat}U, the evolution of nuclides in sub-critical devices under given thermal, fast, hardening fast and fission neutron field are studied without the detail structure of sub-critical device and the burn-up being considered. It is supposed that the sub-critical reactor consists of uniform in which the flux of neutron is homogeneous. The fissile nuclides breeding, equilibrium condition, minor activity (MA) accumulation and transmutation, are studied.

Introduction

In fission reactor, as a result of neutron reaction, certain amount of long-life high-level radioactive nuclei were generated in spent fuel. Besides U and Pu may recover in reprocessing, other is considered as waste. There are two kind wastes in spent fuel, minor actinide (MA) and fission product (FP). Their radio-toxicity is proportional to their activity and very long life time far beyond human society life span. To deal with the problem of disposal of these waste, R&D and some plans have been made to permanently store them in solid state in a deep stable geologic formation and as general consensus, this can be implemented safety, However, there is concern that geologic formation might change over millions of years, then this option still cause a potential risk to human's society.

The idea of direct transmutation of MA and some FP by medium proton bombardment had been considered. However this disadvantage of this direct transmutation option remains the need for a powerful accelerator with rather large energy consumption. To transmute waste by neutron irradiation is more efficient option.

Fission reactors provide ideal neutron field. To transmute MA, the nuclear absorb neutrons in converting to fissile then give some additional neutrons when it undergoes fission. Considering the balance between neutron consumption and production, one may calculate the neutron cost for transmutation of MA in neutron field with typical spectra at different flux. The restriction of transmutation of waste in well-developed normal reactors led to the idea of the hybrid system Accelerator driven sub-critical reactor system (ADS). [1-3] The ADS will greatly release these restrictions.

China begin to study the ADS at 1995, at November 1999, the project has got the national support. Based on the technical situation in China, Chinese scientists have proposed a two-stage plan. The first stage is to build a proton linac of 150 MeV beam energy and 3 mA intensity. And a conceptual validity device will be built based on the light water reactor located in China Institute of Atomic Energy. The second stage will be characterised by a proton beam of approximately 1 500 MeV energy and a few mA intensity. [4,5]

At present work, under the initial inventory with ^{232}Th and ^{nat}U, the evolution of nuclides in sub-critical devices under given thermal, fast, hardening fast and fission neutron field are studied without the detail structure of sub-critical device and the burn-up being considered. It is supposed that the sub-critical reactor consists of uniform in which the flux of neutron is homogeneous. The fissile nuclides breeding, equilibrium condition, minor activity (MA) accumulation and transmutation are studied.

Calculations

In fission reactor, as a result of neutron reaction, for ^{232}Th and ^{238}U, the nuclear evolution in the reactor via capture-decay chain in ref. [6,7] are adopted. A simplified burn-up equation and related data from CENDL [8,9] are adopted to calculate the ratio of converted ^{233}U and ^{239}Pu to ^{232}Th and ^{238}U as the function of neutron flux and operating time with out without the detail structure of sub-critical device and the sub-critical reactor consisting of uniform in which the flux of neutron is homogeneous.

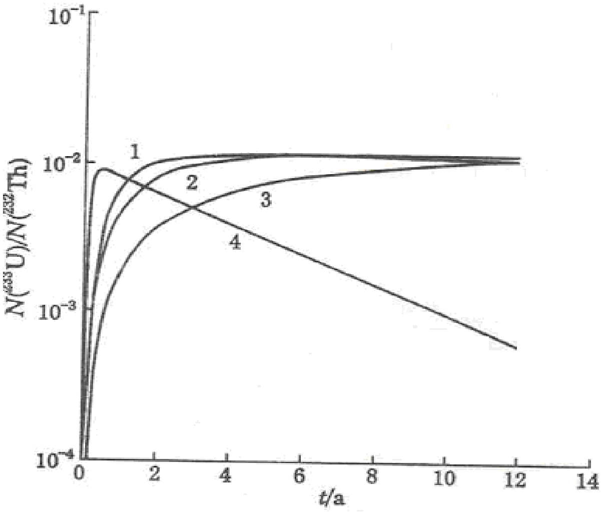

Fig. 1 The saturation condition of Th-U with thermal neutron

$E_n = 0.025\,3$ eV

$\varphi(\mathrm{cm^{-2} \cdot s^{-1}})$: 1——$5 \times 10^{13}$; 2——$3 \times 10^{13}$; 3——$1 \times 10^{15}$; 4——$1 \times 10^{15}$

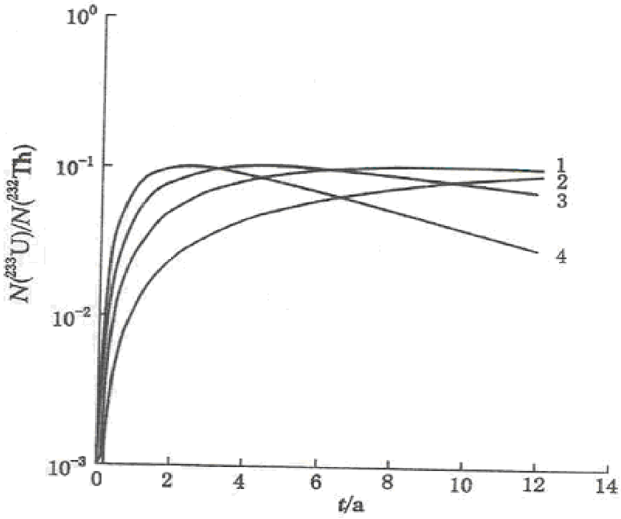

Fig. 2 The saturation condition of Th-U with fast neutron

$E_n = 0.455$ MeV

$\varphi(\mathrm{cm^{-2} \cdot s^{-1}})$: 1——$2.5 \times 10^{15}$; 2——$1 \times 10^{14}$; 3——$5 \times 10^{15}$; 4——$1 \times 10^{16}$

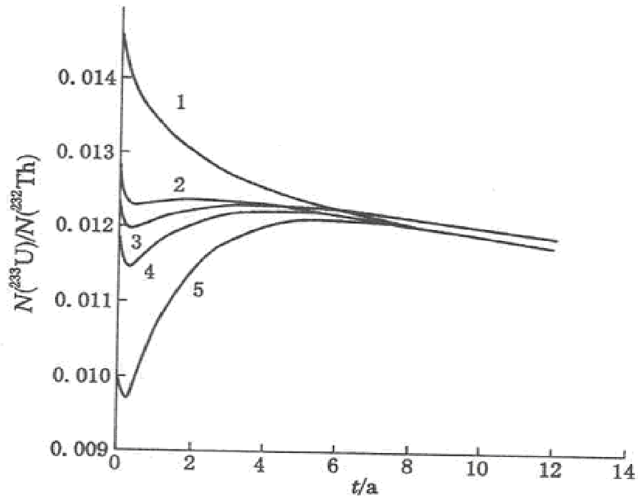

Figure 3. **Saturation curves of ADS loaded with ^{232}Th added by ^{233}U**

The ratio of ^{233}U to ^{232}Th at initial inventory: 1—1.5%; 2—1.3%; 3—1.25%; 4---1.2%; 5—1%.

Figure 1 gives the calculated ratio of coverted ^{233}U to ^{232}Th as the function of neutron flux and operating time at the thermal neutron flux assemble. We can find that the equilibrium ratio reaches only at neutron flux in range of $0.5 \sim 1.0 \times 10^{14}$ n/cm^2-sec. At higher flux, the burn-up of fissile is much faster and continuously feeding of fresh fissile is needed in order to keep long time operation of assembly with constant output of power. The conversion ratio reaches its equilibrium value after 3 years operation. The ratio of coverted ^{233}U to ^{232}Th is about 10^{-2}.

Figure 2 is shown the calculated ratio of converted ^{233}U to ^{232}Th as the function of neutron flux and operating time at the fast neutron flux assemble (average neutron energy is 0.455MeV). It is clear that the equilibrium ratio reaches only at neutron flux of 2.5×10^{15} n/cm^2-sec. At higher flux, the burn-up of fissile is much faster and continuously feeding of fresh fissile is needed in order to keep long time operation of assembly with constant output of power. The conversion ratio reaches its equilibrium value after 3 years operation. The ratio of converted ^{233}U to ^{232}Th is about 10^{-1}.

Equilibrium ratio of fissile to fertile inventory in the assembly.

$$\frac{^{233}U}{^{232}Th} = 0.0114, \quad \frac{^{239}Pu}{^{238}U} = 0.00256, \text{ for thermal assembly}$$

$$\frac{^{233}U}{^{232}Th} = 0.104, \quad \frac{^{239}Pu}{^{238}U} = 0.104, \quad \text{ for fast assembly}$$

In order to research the equilibrium ratio early, about 1.25% ^{233}U should be inputted into the thermal assembly at the beginning. Figure 3 shows the percent of the ration of the initial inventory ^{233}U to ^{232}Th.

For Th-U and U-Pu fuel cycle, the ratio of the accumulated U and Pu isotopes and MA to ^{232}Th and ^{238}U with the electronic power of 1 GeV for thermal, average neutron energies of 0.455, 0.7 and 1.8 MeV are shown in Table 1. For comparison listed is also the ratios of the yield from yearly unloading spend fuel from 1 000Mwe PWR [10] with 33 000MW-D/t burn-up. It is the same as the thermal assembly. That means the fast assembly is able to transmute MA's produced during its own operation. The physics basis for this difference is the due to much smaller α value in fast neutron energy region than that in thermal region.

The equilibrium inventory of fissile converted from fertile is roughly in factor of 3~10 higher for fast assembly than thermal one and operating neutron flux is much higher in fast assembly than thermal one. This makes the thermal output power much higher for fast assembly than thermal one. For Th-U fuel cycle, there are reasonable designs for both fast and thermal assembly. While for U-Pu fuel cycle only the fast system is the choice.

Summary and discussions

At present work, under initial inventory with ^{232}Th and ^{nat}U, the evolution of nuclides in sub-critical devices under given thermal, fast, hardening fast and fission neutron field are studied without the detail structure of sub-critical device and the burn-up being considered. It is supposed that the sub-critical reactor consists of uniform in which the flux of neutron is homogeneous. The fissile nuclides breeding, equilibrium condition, minor activity (MA) accumulation and transmutation, are studied.

Table 1. **The annual output per GWe of MA with Th-U and U-Pu, respectively**

Production of TRU in ADS with differential neutron spectrum						
Assembly	**Nuclei**	**PWR**	**thermal**	**0.455** (MeV)	**0.7** (MeV)	**1.8** (MeV)
Th-U	^{235}U		3.09	4.9	1.38	0.573
	^{238}U	2.13×10^{-4}	1.35×10^{-4}	5.42×10^{-6}	1.75×10^{-8}	
	^{238}Pu	4.52	3.06×10^{-3}	2.47×10^{-2}	3.41×10^{-4}	9.1×10^{-7}
	^{239}Pu	154	1.19×10^{-3}	2.6×10^{-3}	1.58×10^{-4}	2.26×10^{-9}
	^{240}Pu	61.5	4.17×10^{-4}	1.86×10^{-4}	1.56×10^{-7}	4.8×10^{-11}
	^{241}Pu	26.9	6.22×10^{-5}	8.53×10^{-6}	2.77×10^{-9}	6.1×10^{-13}
	^{242}Pu	16.04	3.24×10^{-5}	3.94×10^{-7}	5.48×10^{-11}	8.8×10^{-15}
	^{237}Np	13.2	1.54×10^{-2}	9×10^{-3}	6.58×10^{-3}	3.1×10^{-5}
	^{241}Am	18.1	3.16×10^{-6}	5.1×10^{-7}	1.77×10^{-10}	3.5×10^{-15}
	^{242}Am	0.026	5.0×10^{-9}	9.0×10^{-11}	1.80×10^{-14}	
	^{243}Am	3.3	6.32×10^{-9}	5.17×10^{-10}	9.7×10^{-15}	1.2×10^{-19}
	^{242}Cm	6×10^{-5}	1.1×10^{-6}	1.4×10^{-8}	3.03×10^{-12}	
	^{243}Cm	0.012	7.9×10^{-9}	1.02×10^{-9}	3.5×10^{-14}	8.5×10^{-19}
	^{244}Cm	0.69	5.42×10^{-9}	8.18×10^{-11}	2.32×10^{-16}	1.7×10^{-20}
	^{245}Cm	0.04	5.42×10^{-9}	8.18×10^{-11}	2.32×10^{-16}	1.7×10^{-20}
U-Pu	^{241}Pu	26.9	86	19.6	0.8	4.7×10^{-3}
	^{242}Pu	16.04	47	0.82	0.015	4.4×10^{-5}
	^{237}Np	13.2	4.1	3.42	1.1	2.1×10^{-2}
	^{241}Am	18.1	4.9	1.14	0.46	2.8×10^{-4}
	^{242}Am	0.026	0.006	4.6×10^{-4}	1.1×10^{-4}	1.3×10^{-8}
	^{243}Am	3.3	1.64	0.27	0.3	1.9×10^{-5}
	^{242}Cm	6×10^{-5}	8.6×10^{-4}	3×10^{-5}	7.4×10^{-6}	9.5×10^{-9}
	^{243}Cm	0.012	0.011	1.8×10^{-3}	4.1×10^{-6}	2.5×10^{-9}
	^{244}Cm	0.69	0.47	0.22	0.0044	1.6×10^{-5}
	^{245}Cm	0.04	0.183	9.3×10^{-3}	3.3×10^{-4}	5.3×10^{-8}

The best neutron flux is $\varphi \sim 10^{14}$ /cm^2/s for thermal sub-critical reactor and $\varphi \sim 10^{15}$ -10^{16} /cm^2/s for fast reactor. The fast reactor fissile nuclei ratio in equilibrium condition is larger than that of thermal reactor. The accumulation of MA in fast reactor is much lower than those of thermal reactor and initial inventory with ^{232}Th is much lower than those of inventory with ^{nat}U. Comparing with PWR, the accumulation of thermal reactor of initial inventory with ^{232}Th is much lower than PWR and approximately equals to those of PWR for the thermal sub-critical reactor with ^{nat}U. The MA is transmuted by absorbing neutron to convert into another MA in thermal neutron field and MA fission in fast neutron field. It is preferably to transmute MA in high flux field, especially when thermal neutron to be used, and hardening neutron spectra is preferably transmuting MA. By comparing the main characteristics of thermal sub-critical reactor and fast reactor, it shows that fast reactor has large power output, low production rate of long-lived actinides.

The equilibrium inventory of fissile converted from fertile is roughly in factor of 3~10 higher for fast assembly than thermal one and operating neutron flux is much higher in fast assembly than thermal one. This makes the thermal output power much higher for fast assembly than thermal one. For Th-U fuel cycle, there are reasonable designs for both fast and thermal assembly. While for U-Pu fuel cycle only the fast system is the choice.

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