NUCLEAR SAFETY OF LOW-FLUX AND HIGH-FLUX THORIUM MODE OF CANDU TYPE REACTOR

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The U-233 accumulation in Th-232 in CANDU reactor with low and high neutron flux is studied. The effect of U-233 increase in high flux is explained by the fact that U-233 is produced through intermediate short-lived Pa-233. The growth of multiplication factor after reactor shutdown connected with problem of nuclear safety is estimated.

1. INTRODUCTION

During long time the experts of different countries devoted many efforts to study thorium cycle of atomic power engineering. It is completely obvious that this study and the realization of thorium cycle along with uranium nuclear cycle or instead of one claims the weighty basis from the view point of strategic prospect and receipt of ponderable advantages. This research specify following basic features of thorium cycle. Firstly, this is essential expansion of fuel base of atomic power engineering, as the thorium resources much surpass uranium resources. Secondly, the production of long-lived actinides in thorium cycle is much less than in uranium cycle. This important circumstance facilitates the decision of a problem of ultimate storage of radioactive wastes of atomic power engineering. Thirdly, for initial stage of thorium cycle, preliminary accumulation of a great amount of U-233 is necessary.

These important features give the basis to continue researches of thorium cycle.

From basic view point, thorium cycle do not differ from ordinary uranium fuel cycle by physical characteristics. In both cycles there is the nuclear fuel and fertile material in which new fissile nuclides are formed. However there is some distinction, connected first of all to production of different radioactive nuclides. In uranium cycle, fissile (U-235) and fertile (U-238) materials belong to the same chemical element. In thorium cycle, especially at initial stage of U-233 accumulation, fertile material is Th-232 and fuel material can be U-235 or Pu-239. In this connection there is the number of physical problems having practical interest.

Main requirement to nuclear fuel used in thorium cycles is high enrichment of a fissile nuclide. In uranium cycle, enrichment of nuclear fuel by U-235 is rather small (several percents). For thorium cycles, the fuel nuclide should have high enrichment, as necessary reactivity should be created in this case not only for fuel lifetime but also for conversion of Th-232 into U-233. Highly enriched uranium or weapon-grade plutonium can be used as fuel material. Afterwards, in accordance with expansion of thorium cycle scale, U-233 can be used as fuel nuclide.

It is necessary to pay attention also to problem of control in thorium cycle, as the fuel nuclides Pu-239 and U-233 have smaller share of delayed neutrons than U-235.

However, one of main problems connected with thorium cycle is formation of Pa-233. This problem has two parties: accumulation of a maximum amount of U-233 and nuclear safety.

The amount of produced U-233 essentially depends on neutron flux density. At small flux density typical for power reactors, Pa-233 has time to decay in U-233, which burns up effectively. Therefore its accumulation is small, it makes about 1-1.5 % with respect to initial Th-232. If neutron flux density is high, then Pa-233 has no enough time to decay in U-233 at irradiation, and the amount of Pa-233 is several times more than U-233. At the same time the effect of burnup is much lower, and the accumulation of U-233 is more than at low neutron flux.

The problem of nuclear safety lies in the fact that after finish of irradiation, there is the decay of accumulated Pa-233 in U-233 and increase of multiplication factor.

The reactor CANDU has the high neutronic characteristics due to use of heavy water as moderator and coolant. The additional advantage is connected to opportunity to operate in mode of continuous refueling on-load. Because of this reason there is no need in reactivity compensation. Due to these advantages, the CANDU reactor can operate safely in thorium cycle for U-233 production both at usual and high neutron flux.

In the paper, the amount of nuclides formed from Th-232 is calculated and increase of a reactivity after reactor shutdown is estimated for CANDU reactor operation at usual and high neutron flux.

2. USUAL NEUTRON FLUX

The design of a reactor is described in [1]. Fuel assemblies contain 37 pin fuel elements with zirconium cladding surrounded by zirconium tube. Fuel assemblies are located in square lattice with pitch 23.5 cm. A coolant and moderator is heavy water.

It was considered that in thorium cycle, there are two types of assemblies in active core: fuel assemblies and fertile assemblies. Fuel assemblies contain main fuel, U-235 or plutonium. They determine main multiplicating property of a reactor. The continuous reloading of fuel assemblies creates mode of operations with constant neutron flux. Fertile assemblies contain Th-232. U-233 is accumulated there.

For calculations of nuclides production, it was accepted that the irradiation of fertile assemblies occurs at constant flux density $5 \cdot 10^{13}$ neutr/cm^{2}c. Neutron spectrum hardness (share of epithermal neutrons) typical for heavy-water reactor $\gamma = 0.1$.

For estimation of reactivity increase after reactor shutdown, calculation of two values of multiplication factor K in elementary cell with fertile assemblies with Th-232 and accumulated nuclides U-233, U-234 and other was performed. The first value K(T) corresponded to a moment of the finish of irradiation (T - time of irradiation). Neutron absorption in Th-232, U-233, U-234, U-235, U-236, FP was taken into consideration. FP - fission products of U-233 and other fissile nuclides formed from Th-232. Absorption in Xe-135 and Sm-149 was separately taken into account. The second value $K(T+\infty)$ corresponded to complete decay of Pa-233 in U-233 and complete decay of Xe-135. Increase of multiplication factor in elementary cell with fertile assemblies permits to evaluate indirectly influence of Pa-233 decay on nuclear safety of a reactor as a whole. The more detailed calculations should include specific core load by fuel and fertile assemblies, that can be a subject of further research.

In table 1, relative amount of nuclides are presented at Th-232 irradiation during time T = 1, 2, 3 years. These data are normalized by one initial nucleus of Th-232 in beginning of an irradiation.

Table 1. Nuclide production at neutron flux density $5 \cdot 10^{13}$ neutr/cm^{2}c

Nuclid	T, year		
	1	2	3
Th-232	0.987	0.973	0.960
Pa-233	1.40-3	1.38-3	1.36-3
U-233	7.61-3	1.07-2	1.17-2
(Pa-233) + (U-233)	9.02-3	1.21-2	1.31-2
(Pa-233)/(U-233)	0.184	0.129	0.116
U-234	5.69-4	1.47-3	2.32-3
U-235	4.31-5	1.87-4	3.72-4
U-236	2.19-6	2.05-5	6.67-5
FP	3.82-3	1.29-2	2.40-2

The data submitted show that the burnup of Th-232 is very low. The U-233 production during 2-3 years of an irradiation makes a little bit more than 1 %. The extension of an irradiation from 2 till 3 years insignificantly increases amount of U-233. Increase of U-233 at the expense of Pa-233 decay makes about 12 %. The amount of U-234 and more heavier nuclides is small. The amount of FP at T = 2 years is close to U-233, and at T=3 year it twice exceeds U-233. This fact indicates that the burnup of U-233 essentially reduces its accumulation. The irradiation longer than 2 years is not effective for U-233 production.

In table 2, values of multiplication factors K(T), K(T without Xe) and K(T+∞) and also share of absorption of neutrons in FP $q(FP) = \Sigma_{FP}/\Sigma_f$ are given. Σ_{FP} is macroscopic cross-section of absorption in fission products FP, Σ_f is macroscopic cross-section of fission in elementary cell. The value K(T without Xe) corresponds to absence of Xe-135 and shows increase of multiplication factor in short time after finish of an irradiation when Xe-135 is completely decayed but Pa-233 is not decayed in U-233. The calculations of reaction rates and multiplication factor in elementary cell were performed using code TRIFON [2]. 37-pin fertile assembly was presented for calculation as coaxial 4-ring assembly with preservation of volumes of all fertile and structural materials. Volume of Th-232 was accepted equal to volume of U-238 in usual fuel assembly with uranium fuel. The absorption of neutrons by fission products was taken into account by means of "effective fission fragment" [3].

Table 2. Multiplication factors in elementary cell with Th-232

Characteristics	T, year		
	1	2	3
K(T)	0.6574	0.7528	0.7505
K(T without Xe)	0.6709	0.7707	0.7679
K(T+∞)	0.7435	0.8237	0.8161
K(T+∞) - K(T)	0.0861	0.0709	0.0656
q(FP)	0.117	0.251	0.379

The data presented show that the elementary cell with Th-232 and accumulated U-233 is subcritical. The decrease of K(T) and K(T+∞) at transition from T=2 to T=3 years is explained by competition between weak growth of U-233 and essential growth of fission products. After reactor shutdown, multiplication factor of an elementary cell is increased at the expense of Xe-135 decay by 0.013-0.018 and additionally at the expense of Pa-233 decay by 0.05-0.07. Complete increase of multiplication factor makes 0.06-0.08

3. HIGH NEUTRON FLUX

The high neutron flux in CANDU reactor can be created at the expense of increase of heat power removed from 1 kg of fuel and at the expense of reduction of amount of fuel at the same number of fuel rods. The reserves of heat removal increase are limited. A reasonable estimation would be the fact that it is possible to increase heat power 2 times on account of more intensive heat removal. Then to increase neutron flux from $5\cdot10^{13}$ up to 10^{15} neutr/cm^{2}s it is necessary to reduce 10 times fuel amount in fuel rods and Th-232 amount in rods of fertile assemblies. At the same time the neutron spectrum in reactor will become essentially more thermalised than before reduction of fissile and fertile materials.

For calculations of nuclides production in high flux mode of CANDU reactor, a neutron flux density 10^{15} neutr/cm^{2}s and neutron spectrum hardness $\gamma = 0.01$ was accepted. Production of short-lived Pa-234 was taken into account. It decays into U-234 with half-life 6.7 hours and has a large fission cross-section. At low neutron flux, the role of this nuclide is negligible.

In table 3, relative amounts of nuclides are submitted at irradiation of Th-232 during time T = 10, 20, 30, 50 days. These data are normalized by one initial nucleus of Th-232 in beginning of an irradiation.

Table 3. Nuclide production at neutron flux density 10^{15} neutr/cm^{2}c

Nuclid	T, days			
	10	20	30	50
Th-232	0.993	0.987	0.980	0.967
Pa-233	5.75-3	9.98-3	1.31-2	1.70-2
U-233	6.58-4	2.04-3	3.60-3	6.34-3
(Pa-233) + (U-233)	6.41-3	1.20-2	1.67-2	2.33-2
(Pa-233)/(U-233)	8.74	4.89	3.64	2.68
Pa-234	3.93-6	6.93-6	9.14-6	1.19-5
U-234	1.20-4	4.55-4	9.55-4	2.26-3
U-235	3.16-6	2.17-5	6.18-5	2.04-4
U-236	6.82-8	9.74-7	4.33-6	2.56-5
FP	1.15-4	7.22-4	1.97-3	6.14-3

The data presented show that the burnup of Th-232 is very low. The production of U-233 in sum with Pa-233 for 20 days of an irradiation makes 1.2 %, for 50 days - 2.3 %. The amount U-233 grows after reactor shutdown at the expense of Pa-233 decay 4-10 times. The amount of U-234 and more heavier nuclides is very small. The amount of FP is essentially less than U-233 in sum with Pa-233. This indicates that the burnup of U-233 influences on its accumulation rather slow. It is possible to irradiate longer than 50 days however with smaller effect.

Thus in high flux mode, the production of U-233 grows about 2 times in comparison with usual mode. This effect is not rather great as we would like to expect. It is explained by an essential thermalisation of a neutron spectrum at transition to high flux mode and reduction of fuel and fertile materials.

In table 4, values of multiplication factors K(T) and K(T+∞) with additional account of short-lived Pa-234 and q(FP) are given. At calculation K(T), fission in Pa-234 is taken into account. At calculation K(T+∞), it is considered that Pa-234 decays into U-234.

Table 4. Multiplication factors at neutron flux density 10^{15} neutr/cm^{2}c

Characteristics	T, days			
	10	20	30	40
K(T)	0.0829	0.2278	0.3627	0.5372
K(T+∞)	0.5644	0.8530	1.0132	1.1541
K(T+∞) - K(T)	0.4815	0.6252	0.6505	0.6169
q(FP)	0.0474	0.0965	0.148	0.259

The data presented show that the elementary cell with Th-232 and accumulated U-233 is subcritical at irradiation but can become overcritical after finish of an irradiation. At increase of an irradiation time, K(T) and K(T+∞) are regularly increased. Thus, the absorption in fission products influences on multiplication factor rather slightly. The role of Xe-135 is also insignificant because of small amount of fission products. The contribution of Xe-135 decay and decay of Pa-234 in U-234 after finish of irradiation in K(T+∞) makes less than 0.01. Total increase of multiplication factor makes 0.48-0.65.

4. CONCLUSION

The research performed shows that the opportunity to increase an amount of accumulated U-233 is connected with increase of a neutron flux density. It is explained by the fact that at high neutron flux, Pa-233 has no time to decay in U-233, and the decay occurs after finish of an irradiation. However in CANDU reactor, increase of neutron flux density should result in essential thermalisation of neutron spectrum. Because of this reason, the effect of neutron flux increase is not rather high. At transition from neutron flux $5\cdot10^{13}$ to 10^{15} neutr/cm^{2}s, The accumulation of U-233 per unit of Th-232 in neutron flux 10^{15} neutr/cm^{2}s is about 2 times greater than in neutron flux $5\cdot10^{13}$ neutr/cm^{2}s.

At the same time, the decay of Pa-233 in U-233 after reactor shutdown influences on nuclear safety. However, the increase of a reactivity occurs slowly and the complete duration of this process makes several periods of Pa-233 half-life, that is several months. The increase of a reactivity after reactor shutdown appears essential at high neutron flux. After irradiation 50 days at neutron flux density 10^{15} neutr/cm^{2}s, multiplication factor in elementary cell with thorium load grows from 0.54 up to 1.15. Rather significant increase of multiplication factor after finish of an irradiation claims development of special measures. In regular mode of operation with refueling on-load, irradiated thorium assemblies are removed from reactor and do not influence on its safety. However after unloading they should be located in conditions excluding possibility to be overcritical at the further manipulation with them. The main danger is at emergency shutdown of a reactor, when the irradiation stops without thorium assemblies removal. For these cases, reliable system should be designed to introduce necessary amount of absorber into reactor for chain reaction suppression.

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