THORIUM SELF-SUFFICIENT FUEL CYCLE OF CANDU POWER REACTOR

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ABSTRACT

This paper presents the results of calculations for CANDU reactor operation in thorium fuel cycle. Calculations were performed to estimate feasibility of operation of heavy-water thermal neutron power reactor in self-sufficient thorium cycle. Parameters of active core and scheme of fuel reloading were considered to be the same as for standard operation in uranium cycle.

Two modes of operation are discussed in the paper: mode of preliminary accumulation of $^{233}\text{U}$ and mode of operation in self-sufficient cycle.

For the mode of accumulation of $^{233}\text{U}$ it was assumed for calculations, that plutonium was used as additional fissile material to provide neutrons for $^{233}\text{U}$ production. Plutonium was placed in fuel channels, while $^{232}\text{Th}$ was located in target channels. Maximum content of $^{233}\text{U}$ in target channels was about 13 kg per 1 ton of ThO$_2$. This was achieved by irradiation for 6 years. The start of reactor operation in the self-sufficient mode requires content of $^{233}\text{U}$ not less than 12 kg/t.

For the mode of operation in self-sufficient cycle, it was assumed that all channels were loaded with the identical fuel assemblies containing ThO$_2$ and certain amount of $^{233}\text{U}$. It was shown that nonuniform distribution of $^{233}\text{U}$ in fuel assembly is preferable.

1 INTRODUCTION

Absence of isotope $^{233}\text{U}$ in nature does not exclude possibility of its use as a nuclear fuel. $^{233}\text{U}$ is a product of radioactive decay of $^{235}\text{Pa}$, which is formed by means of neutron capture by $^{232}\text{Th}$ with following $\beta$-decay. Thereby, thorium, existing in the nature in a single stable isotope $^{232}\text{Th}$, can be transformed using nuclear reactions into $^{233}\text{U}$, i.e. into the fuel of power reactors. Reserves of thorium in the world exceed that of uranium by several times. Cost of thorium mining is much less than that of uranium. This is due to approximately 100 times lower radiation danger in the process of mining of thorium in comparison with mining of uranium. Possibilities of commercial production of $^{233}\text{U}$ in nuclear reactors were many times considered in publications in connection with thorium-uranium fuel cycle. It should be
noted that even partial substitution of uranium-plutonium fuel cycle by thorium-uranium cycle is connected with significant difficulties. One of the difficulties is related to the necessity of changing of the process. However, there are also physical problems: half-life of the decay of $^{233}\text{Pa}$ in $^{233}\text{U}$, which is 27 days, greatly exceeds analogous half-life of the decay of $^{239}\text{Np}$ in $^{239}\text{Pu}$ in uranium-plutonium fuel cycle, and cross-section of capture of thermal neutrons in $^{232}\text{Th}$ is 2.8 times higher than analogous cross-section of $^{238}\text{U}$. These disadvantages of thorium fuel cycle were seemingly a reason that in publications of last years, thorium was considered only as a raw material for feed of nuclear reactors operating in uranium-plutonium fuel cycle [1]. Such mixed mode allows more or less to save natural uranium during electric power production. Possibility of operation of power reactor in thorium-uranium fuel cycle without feed by fissile materials was considered in preceding publications, for instance by [1, 2]. However, this possibility was only declared without demonstration of concrete ways of achievement.

It should be noted that self-sufficient mode is related with rather big effort in the extraction of isotopes of uranium from unloaded nuclear fuel. However, because of need of accumulation of required amount of $^{233}\text{U}$, this disadvantage is inherent not only to the self-sufficient mode, but also to thorium-uranium fuel cycle in any of its modifications. Accumulation of $^{232}\text{U}$ along with daughter nuclides affecting a radiation environment should also be mentioned as drawback of thorium fuel cycle.

Thorium-uranium fuel cycle nevertheless also has certain advantages. In particular, number of secondary neutrons in fission of $^{233}\text{U}$ by thermal neutrons is higher than for any other isotopes of uranium. This fact gives a hope for the possibility of operation of reactor in self-sufficient mode.

In this paper, results of calculations for self-sufficient mode for heavy-water power reactor are presented. This reactor is hereinafter referred to as "T" reactor. Parameters of active core and scheme of refuelling of current heavy-water power reactor HWPR (CANDU) with heat power of 2776 MW [3] were used for calculations.

Calculations were made using code complex MCCOOR, developed on the base of codes MCNP, COUPLE, ORIGEN-S.

## 2 MODE OF ACCUMULATION OF $^{233}\text{U}$

For reactor start-up, it is necessary to have certain amount of fissile material, in our case $^{233}\text{U}$. In this paper it is assumed, that $^{235}\text{U}$ for downstream operation is produced in the reactor "T" during the first period of operation. Although it is not improbable that accumulation of $^{233}\text{U}$ in the special reactor or in other power reactors, in particular, in blanket of fast breeder reactor may turn out to be more efficient.

In the mode of accumulation of $^{233}\text{U}$, tetragonal heavy-water lattice of reactor "T" is composed of channels of two types, evenly distributed over the active core. Half of channels are referred to as fuel channels, another half are referred to as target channels. Fuel channel contains fuel assembly composed of 37 fuel elements with power plutonium or enriched uranium. Target channel contains target assembly composed of 37 target elements. Each target element is made in the form of zirconium tube filled with pellets of ThO$_2$. Figure 1 demonstrates part of channel lattice with 16 channels and elementary cell accepted for calculations.

During the operation of reactor "T", power and neutrons are released in fuel channels due to fission of $^{239}\text{Pu}$, $^{241}\text{Pu}$ ($^{235}\text{U}$). In target channels in the beginning of operation, excess neutrons are captured with formation of $^{233}\text{Pa}$. As the $^{233}\text{Pa}$ decays in $^{233}\text{U}$, power releases in target channels too. It is assumed that power of fuel channels with plutonium or enriched uranium is maintained to keep constant power density per mass unit of heavy nuclei, equal to
that of current CANDU type reactor. This condition determines neutron flux density in targets.

Figure 1: Lattice of channels in active core in the mode of accumulation of $^{233}$U and elementary cell accepted for calculations

Results of calculation of elementary cell of active core for the variant using fuel containing 10% of power plutonium are presented in fig. 2 and 3.

Figure 2: Variation of concentration of $^{233}$U in ThO$_2$ in the mode of accumulation of $^{233}$U

Figure 3: Variation of multiplication factor for elementary cell in the mode of accumulation of $^{233}$U.
According to the data of fig. 2, maximum (equilibrium) content of $^{233}$U in targets is $\sim 13$ kg/t (these data are given in kilograms of $^{233}$U per one ton of ThO$_2$ with density of 10 g/cm$^3$). This result is close to that obtained in [4] for thorium-uranium lattice and neutron spectrum in the fuel of CANDU reactor. Time of irradiation till maximum content of $^{233}$U in targets is 5-6 years, time for achievement of maximum multiplication factor $K_\infty$ is about 2 years. During this time, content of $^{233}$U in targets reaches about 8 kg/t.

For start-up of reactor "T" in the self-sufficient mode, it is necessary to load ThO$_2$ with content of $^{233}$U not less than 12 kg/t in fuel elements of all assemblies. For the accumulation of necessary amount of $^{233}$U in the mode of accumulation, it will take, for instance, three cycles of irradiation of ThO$_2$ targets during 2 years. Therefore in the mode of accumulation of $^{233}$U, reactor “T” should work approximately for 6 years. After each cycle of irradiation, targets should be unloaded from the reactor and replaced by fresh targets without $^{233}$U. In order the reactor “T” does not become subcritical during targets reloading, replacement of irradiated targets by fresh targets without $^{233}$U should not be simultaneous. As $^{233}$U is accumulated in targets, total power of the reactor increases in accordance with given mode of operation, while power of fuel channels remains constant.

It is apparent that considered parameters of operation of reactor "T" in the mode of accumulation of $^{233}$U can be highly improved. It is possible to reduce time of accumulation of $^{233}$U and increase power of reactor by following measures:

- increase of heat load on fuel element, in particular, at the expense of use of fuel elements with the diameter less than in current CANDU reactor;
- compensations of reactivity increase (see fig.3) due to replacement of fuel channels by target channels;
- optimization of number and locations of target channels and fuel channels in the active core;
- optimization of scheme of reloading of target channels;
- increase of number of targets in the target assembly [1];
- reducing plutonium (uranium) load in fuel elements, that would result in increase of thermal neutron flux in targets at the same power of reactor "T".

3 SELF-SUFFICIENT MODE

In the self-sufficient mode, it is assumed that fuel elements containing ThO$_2$ with the given content of $^{233}$U are loaded in all channels of the reactor. Considered mode could be realized in the reactor "T", if two conditions are met (that is nontrivial task at the strict restriction of total amount of $^{233}$U).

Condition 1. Amount of $^{233}$U and its layout in channels must ensure overcriticality of the reactor "T" in the initial state. Overcriticality of the current CANDU reactor in the cold condition with the fresh fuel (natural uranium), calculated by means of above mentioned code complex, is $\Delta K \geq 0.1$. It is obvious that the reactor "T" should have overcriticality not less than this value.

Condition 2. In the reactor "T" under the acceptable burnup of $^{233}$U and other fissile isotopes, reproduction of $^{233}$U must ensure at least equality of content of fuel in the active core before the beginning and after the end of fuel lifetime. This condition together with the reactivity margin determines maximum burnup of fuel and fuel lifetime length. Breeding ratio and its variations during the fuel lifetime depend on the amount of fuel, formed in the mode of accumulation of $^{233}$U, and of its position in the active core of the reactor.

Fuel burnup and fuel lifetime in the reactor "T" was estimated by the method, which is suitable for CANDU reactors with continuous bidirectional refuelling. In this case, fuel
assemblies with different burnups in the range from zero to maximum burnup are present in active core during the whole lifetime. Such condition can be described by average multiplication factor determined by the leakage of neutrons from the active core, absorption of neutrons by structure materials of active core and by control rods. For the CANDU reactor with control rods in active core, this value is $1.045$, and for extracted control rods – $1.035$ [1]. Fuel burnup $W_0$ in reactor "T" was estimated in accordance with the parameter $\alpha$

$$\alpha = \int_0^{W_0} K_\infty dW / W_0$$  \hspace{1cm} (1)

where $W$ is fuel burnup. Dependence $K_\infty(W)$ was calculated in the interval between initial value $W = 0$ corresponding to the fresh fuel, and final value $W_0$ corresponding to the fuel unloaded from the reactor. In the process of calculation of $K_\infty(W)$, neutronic parameters were calculated over again at different values of $W$ with the step of $1$ MW·d/kg in order to take into account change of neutron spectrum in fuel.

Calculations were made for different burnups and for three variants of initial layout of $^{233}$U in the active core of the reactor "T".

In the first variant, accumulated $^{233}$U was distributed in equal parts over all fuel assemblies of the reactor with the content $C = 12$ kg/t in each fuel element.

In the second variant, accumulated $^{233}$U was distributed in equal parts over all fuel assemblies of the reactor. However, in each fuel assembly, $^{233}$U was placed only in 18 external fuel elements with the content $C = 24$ kg/t while 19 internal fuel elements were filled only with ThO$_2$ without $^{233}$U.

In the third variant, accumulated $^{233}$U was distributed in equal parts over all fuel assemblies of the reactor. However, in each fuel assembly, $^{233}$U was placed in 18 external fuel elements with the content $C = 16$ kg/t, and in 19 internal fuel elements with the content $C = 10$ kg/t. Balance on the uranium was ensured due to changing of density of $^{233}$U+ThO$_2$.

Results of calculations of $K_\infty(W)$ for considered variants of $^{233}$U layout in fuel assemblies are presented in fig. 4. Curves 1 – 4 were used for calculation of fuel burnup in the reactor “T” for different values of the parameter $\alpha$ (1). The curve 4 corresponds to the common-type CANDU reactor with natural uranium fuel.

![Figure 4: Multiplication factor for elementary cell in the self-sufficient mode: 1, 2, 3 – 1st, 2nd, and 3rd variant of $^{233}$U layout in fuel assemblies of the “T” reactor, 4 – CANDU reactor with natural uranium.](image-url)
Results of calculations of fuel pin respective power $P_{\text{int}}/P_{\text{ext}}$ in fuel assembly are presented in fig. 5 for 2nd and 3rd variants of $^{233}$U layout. $P_{\text{int}}$ is total power of 19 internal fuel pins with lower content of $^{233}$U, and $P_{\text{ext}}$ is total power of 18 external fuel pins with higher content of $^{233}$U in fuel assembly. These curves were used for estimation of intervals of heat power variation of the reactor “T”. Results of calculation of fuel burnup for three variants of $^{233}$U layout in fuel assemblies are given in the table 1.

![Figure 5: Fuel pin respective power $P_{\text{int}}/P_{\text{ext}}$ in fuel assembly. $P_{\text{int}}$ – total power of 19 internal fuel pins, $P_{\text{ext}}$ – total power of 18 external fuel pins in fuel assembly 1, 2 – 2nd and 3rd variant of $^{233}$U layout in fuel assemblies of the “T” reactor.](image)

<table>
<thead>
<tr>
<th>Reactor</th>
<th>Fuel burnup, MW·d/kg</th>
<th>Heat power for $\alpha=1.035$, MW</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\alpha=1.025$</td>
<td>$\alpha=1.035$</td>
</tr>
<tr>
<td>CANDU</td>
<td>9.4</td>
<td>8.1</td>
</tr>
<tr>
<td>“T”, variant 1</td>
<td>3.4</td>
<td>1.5</td>
</tr>
<tr>
<td>“T”, variant 2</td>
<td>17.5</td>
<td>14</td>
</tr>
<tr>
<td>“T”, variant 3</td>
<td>10.1</td>
<td>6.7</td>
</tr>
</tbody>
</table>

Calculated fuel burnup of the CANDU reactor with natural uranium is in good accordance with operating value of 8.3 MW·d/kg [3]. So, there is good reason to think that fuel burnups calculated for three variants of fuel layout in fuel assemblies of the reactor "T" using the same method with same neutronic constants, are close to real values. Data of table 1 demonstrate that under even distribution of $^{233}$U over all fuel elements of the reactor "T" (first variant), fuel burnup is unacceptably low, while initial overcriticality of active core is less than 10% (curve 1 in fig. 4). Fuel burnup can be increased to 15 MW·d/kg due to location of the same amount of $^{233}$U in the external fuel elements of the fuel assembly according to the second variant. In this case shielding of internal fuel elements allows to increase burnup.
approximately by 10 times. At nonequal load of $^{233}$U into external and internal fuel elements in each assembly according to the third variant, in which shielding of internal fuel elements is less than in the second variant, burnup decreases by 1.5 times with respect to the second variant. As one would expect, heat power of the reactor "T" for the third variant is much higher than for the second variant.

It is obvious that possibility of practical realization of self-sufficient mode in the reactor "T" depends mainly on the amounts of fissile isotopes in the fuel unloaded from the active core in the end of the next fuel lifetime. The curves in fig.6 describe changes of total amount of $^{235}$U, $^{233}$U, and $^{233}$Pa during the fuel lifetime for the second and the third variants of fuel layout (initial value is accepted as 100%).

![Figure 6: Variation of $Q$ - total mass of $^{233}$U, $^{235}$U, $^{233}$Pa during the fuel lifetime, 1 – 2nd variant of $^{233}$U arrangement in fuel assemblies of the “T” reactor, 2 – 3rd variant of $^{233}$U arrangement in fuel assemblies of the “T” reactor.](image)

These curves demonstrate that changes in contents of $^{233}$U, $^{235}$U, $^{233}$Pa in fuel assemblies during the fuel lifetime do not exceed 1-2% of initial amount, i.e. they are within the error interval of our calculations. So, these results can be considered as preliminary data, demonstrating possibility of operation of the reactor "T" in self-sufficient mode.

For obtaining additional data, calculations of multiplication factors $K_\infty(0)$ and $K_\infty(W_0)$ were made for the third variant of fuel layout in cold reactor "T" (table.2). Fuel burnup of 8 MW·d/kg was accepted in these calculations, that corresponded to the fuel lifetime length of 430 days. Nuclides $^{233}$U, $^{235}$U, and $^{233}$Pa extracted from unloaded fuel of each preceding fuel lifetime were used as fuel for the following fuel lifetime.

<table>
<thead>
<tr>
<th>Number of fuel lifetime</th>
<th>Beginning</th>
<th>End</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1.120</td>
<td>1.005</td>
</tr>
<tr>
<td>2</td>
<td>1.103</td>
<td>1.006</td>
</tr>
<tr>
<td>5</td>
<td>1.103</td>
<td>1.010</td>
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<tr>
<td>10</td>
<td>1.102</td>
<td>1.009</td>
</tr>
</tbody>
</table>

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In the table 3, composition of uranium isotopes extracted from unloaded fuel after the 1st, 5th, and 10th fuel lifetimes is presented. Total mass of all isotopes of uranium at the beginning of the first fuel lifetime is accepted as 100%.

Table 3: Isotopic composition of the uranium

<table>
<thead>
<tr>
<th>Number of fuel lifetime</th>
<th>$^{232}$U</th>
<th>$^{233}$U</th>
<th>$^{234}$U</th>
<th>$^{235}$U</th>
<th>$^{236}$U</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.002</td>
<td>99.7</td>
<td>6.5</td>
<td>0.4</td>
<td>0.02</td>
</tr>
<tr>
<td>5</td>
<td>0.060</td>
<td>100.3</td>
<td>23.1</td>
<td>3.6</td>
<td>0.9</td>
</tr>
<tr>
<td>10</td>
<td>0.085</td>
<td>100.5</td>
<td>34.7</td>
<td>6.0</td>
<td>3.1</td>
</tr>
</tbody>
</table>

The data of tables 2 and 3 are in fact calculating substantiation of feasibility of self-sufficient thorium mode for CANDU reactor.

4 CONCLUSIONS

The data submitted above demonstrate that self-sufficient thorium cycle can be realized in CANDU reactor. Starting amount of $^{233}$U can be produced in the CANDU reactor itself for about 6 years of operation in the mode with use of additional fissile materials – plutonium or enriched uranium. After that, accumulated $^{233}$U can be used in self-sufficient mode of operation. Non-uniform distribution of $^{233}$U over fuel pins within fuel assembly is favorable for increase of fuel burnup and heat power (table 1). Calculations of 10 consequent fuel lifetimes (tables 2 and 3) demonstrate that multiplication factor of the elementary cell and the amount of the $^{233}$U produced during fuel lifetimes are sufficient for further operation in the self-sufficient cycle. These results substantiate feasibility of self-sufficient mode for heavy-water power reactor of CANDU type, operating in thorium-uranium fuel cycle.

REFERENCES


