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A Physics Study on the Recycling of Mixed Thorium/Uranium Fuel in a CANDU Reactor by Dry Process Technology

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Abstract

Dry process technology has high proliferation-resistance which can be utilized not only in the existing nuclear system but also in a future nuclear system. In this study, a homogeneous ThO_2-UO_2 fuel cycle option was analyzed for a Canada deuterium uranium (CANDU) reactor based on the dry process to recycle the spent fuel. In order to assess the feasibility of a closed fuel cycle in the CANDU reactor, basic physics parameters such as reactivity coefficients and isotopic content variation were obtained for various fuel conditions. The results of the physics calculations showed that it is feasible to recycle thorium/uranium fuel by the dry process, which in turn significantly reduces the natural uranium consumption. It is, however, required to further investigate a dry process option that is technically feasible for the thorium-abundant dioxide fuel.

I. Introduction

There are many research activities to minimize nuclear waste in next generation nuclear technology development. In the early 1990's, a closed fuel cycle concept was proposed for a liquid metal fast reactor system in conjunction with the pyrometallurgical process for the extraction of actinides. [1] In the closed fuel cycle, it is possible to keep most of the actinides in the reactor system throughout the plant lifetime, if an appropriate reprocess technology is applied. In this study, we are investigating the feasibility of a closed fuel cycle in thermal reactor systems, which are currently operating as commercial reactors.

In the closed fuel cycle system, the fuel should breed the minimum amount of fissile material in order to sustain the excess reactivity during the recycle. We have chosen thorium as the fertile material because there are several advantages of using ^{232}Th as a nuclear fuel. First of all, the thermal absorption cross-section of ^{232}Th is higher than that of ^{238}U (7.4 vs. 2.7 barns at 0.0253 eV), and it produces a very good fissile isotope ^{233}U. Secondly, the ^{233}U produced from ^{232}Th has a higher yield than other fissile isotopes. The number of neutrons released per one neutron absorption in ^{233}U (η) is greater than 2 over almost the entire

thermal and epithermal energy range, which in turn contributes to a reduced burnup reactivity swing. Thirdly, thorium produces less transuranic isotopes than ^{238}U does, which is a strong advantage from the viewpoint of long-term waste management. [2]

The reprocess technology considered in this study is a dry process, which can be an alternative to the expensive aqueous process. One of the examples is the oxidation and reduction of oxide fuel (OREOX) process, which is based only on the thermo-mechanical process. [3] The OREOX process repeats two or three oxidations at 450°C in air and a reduction at 650°C in H_2. The OREOX process recovers all the actinides and some of the fission products from the spent fuel. Because the process does not include any aqueous material and a melting step, this process is inherently very proliferation-resistant.

II. Fuel Burnup Model

The reactor system considered in this study is a 713 MWe Canada deuterium uranium (CANDU-6) reactor, which has a very high thermal flux level (1.9×10^{14} n/cm^{2}sec) due to a heavy water moderation. The CANDU-6 reactor was originally designed to use natural uranium as the fuel and pressurized water as the coolant. This saves on the high initial capital expense of uranium enrichment and fuel reprocessing plants, although D_2O production plants are required. However an important feature of the CANDU concept is that it can evolve to use different coolants and fuels, resulting in an improvement of the fuel cycle. For example, the use of thorium can substantially reduce the uranium requirements. [4]

In the CANDU reactor analysis, it is possible to estimate the fuel discharge burnup using only the lattice code once the reactor characteristics (reactor size, effective fuel length, reactivity device, etc.) are appropriately described in the lattice calculation. The CANDU reactor is refueled daily and, therefore, the fuel burnup is distributed from the fresh to the discharge state over the entire core. The way of estimating the discharge burnup is to solve an integral equation of the excess reactivity for w_{dis},

$$\int_0^{w_{dis}} k_{eff}(w)\, dw = 0 \tag{1}$$

where w_{dis} is the discharge burnup.

If the fuel type is different from that of natural uranium, the discharge burnup can be estimated in the same manner. However the effect of the neutron spectrum change on neutron leakage and absorption should be considered to a certain extent. Therefore a simple formulation was derived for the calculation of an effective multiplication factor that can be estimated from the lattice calculation as follows:

$$k_{eff} = \frac{\text{Production rate}}{\text{Loss rate}}. \tag{2}$$

Here the loss term can be divided into three components: neutron absorption by the reactivity devices and structural materials, neutron absorption by the fuel lattice, and neutron leakage. For convenience, the k_{eff} can be written as follows:

$$\frac{1}{k_{eff}} = \frac{A_d}{k_\infty A_c} + \frac{L}{k_\infty A_c} + \frac{1}{k_\infty} \tag{3}$$

where A_d, A_c, and L are parasitic absorptions in reactivity devices and structural material,

absorptions in the fuel lattice, and leakage terms, respectively. The k_∞ and A_c can be calculated from the lattice calculation. The leakage term can also be estimated using the buckling approximation. Because the parasitic absorptions occur mostly in the moderator region where the thermal flux is dominant, the fast absorptions can be neglected in A_d. Therefore the above equation can be rewritten as follows in a two-group form:

$$\frac{1}{k_{eff}} = \left(\Sigma_{a2}^{d} \frac{V_d}{V_c} \right) \frac{\varphi_2^d}{k_\infty (\Sigma_{a1}^c \varphi_1^c + \Sigma_{a2}^c \varphi_2^c)} + \frac{(D_1 B^2 \varphi_1^c + D_2 B^2 \varphi_2^c)}{k_\infty (\Sigma_{a1}^c \varphi_1^c + \Sigma_{a2}^c \varphi_2^c)} + \frac{1}{k_\infty} . \quad (4)$$

The thermal flux of the device (φ_2^d) can be replaced by the edge flux of the lattice cell. The effective thermal absorption cross-section of the device $\left(\Sigma_e^d = \Sigma_{a2}^d \, V_d/V_c\right)$ can be estimated from the reference core calculation. When the discharge burnup of the natural uranium fuel is 7,300 MWd/t, the estimated effective thermal absorption cross-section of the device is 1.096×10^{-3} cm^{-1}. If the fuel is different from that of the natural uranium, the lattice properties can be fully incorporated into the above equation as well as the effective device cross-section obtained from the reference core, and the discharge burnup of that specific fuel can be estimated without the need of a full-core fuel management simulation.

III. Recycling Simulation of ThO_2/UO_2 Fuel

One of the important features of the closed fuel cycle is to maintain the material balance. In this study, a transport code WIMS-AECL [Ref. 5] was used for the lattice calculation and material balance analysis. For the physics analysis of the closed fuel cycle, the analysis model and assumptions were made as follows:

- The CANDU-6 reactor was used as the reference core.
- The 43-element fuel bundle design was chosen.
- The enrichment of uranium feed is 20 wt%.
- By the dry reprocess, all the actinides are recycled, while the fission products are removed. The fuel mass is kept constant by adding a thorium and uranium feed. The ratio of uranium/thorium mass is determined to keep the fuel discharge burnup.
- The fuel material is the mixture of ThO_2 and UO_2.

Based on the analysis model and assumptions described above, a series of parametric calculations were performed on the uranium fraction, ^{235}U enrichment of the fresh fuel and the fission product contamination effect of the recycled fuel. The results were produced for the basic physics parameters and material balance of the recycled fuel.

III.A Effect of UO_2 Volume Fraction

The volume fractions of UO_2 considered in this study are 9%, 10% and 11% with an initial ^{235}U enrichment of 20 wt%. The estimated discharge burnups are 14000, 26000 and 36000 MWd/t for the ThO_2-9%UO_2, ThO_2-10%UO_2, and ThO_2-11%UO_2 fuels, respectively. The variations of the infinite multiplication factor (k_{inf}) are shown in Fig. 1. It can be seen that the neutronic property represented by the k_{inf} converges immediately after the first recycle. For the 9%UO_2 case, the amount of uranium feed is 0.152 kg/bundle/recycle, which corresponds to 4.28 kg of the natural

uranium as far as the fissile content is concerned. For 10%UO_2 and 11%UO_2, the equivalent natural uranium feeds are 11.21 and 17.35 kg/bundle/recycle, respectively. Considering that the discharge burnup of the ThO_2-UO_2 fuel is higher than that of the natural uranium fuel, the natural uranium consumption will be 2.23, 3.15 and 3.52 kg/bundle for 9%UO_2, 10%UO_2 and 11%UO_2, respectively, when normalized to the discharge burnup of natural uranium.

Table I shows the results of the uranium utilization for the three cases. The results indicate that the recycling of thorium/uranium fuel in a CANDU-6 reactor is feasible as far as the mass balance is concerned under the assumptions described above. The uranium consumption of the recycled ThO_2-UO_2 fuel considered in this study is much less than that of the conventional natural uranium fuel. Specifically, the natural uranium utilization (defined as the ratio of natural uranium consumption) of the ThO_2-UO_2 fuel is at least five times better than that of the standard natural uranium fuel. The high burnup fuel consumes more uranium resources compared to the low burnup fuel, because it burns more ^{235}U rather than utilizing the ^{233}U bred from ^{232}Th.

Figure 2 shows the fissile content change of the recycled ThO_2-UO_2 fuel as a function of irradiation. In the equilibrium thorium/uranium fuel cycle, the fissile isotope such as ^{235}U is continuously supplied in order to maintain the excess reactivity. In general, the fissile content of the fuel is proportional to the burnup reactivity swing and, therefore, the ThO_2-9%UO_2 fuel has the smallest change of the fissile content among three fuel types. Furthermore, because the separation of fissile material is inherently prohibited in the dry process, the thorium/uranium fuel can have an incentive for proliferation-resistance. The neutron capture of ^{233}Pa is important in the thorium fuel reactor because it enables the neutron capture to be effectively competed with the beta decay of ^{233}Th. As a result, the flux-dependent production of ^{233}U also competes with the burning of the ^{235}U and eventually reduces the burnup reactivity swing to a certain extent.

The safety-related parameters were calculated for the three cases (ThO_2-9%UO_2, ThO_2-10%UO_2 and ThO_2-11%UO_2) and the results are also summarized in Table I. The results were obtained for the middle burnup of one recycling period after a sufficient number of recycles (defined as the equilibrium burnup of the recycled fuel in this study). Therefore the accumulated burnup of the recycled fuel is ~200,000 MWd/t. The safety-related parameters of the ThO_2-UO_2 are also compared to those of the natural uranium fuel of the equilibrium (middle) burnup state.

The fuel temperature coefficient (FTC) of the natural uranium fuel is near zero at the equilibrium burnup, while the FTC of the thorium/uranium fuel is slightly negative as shown in Fig. 3. It is understood that the ^{239}Pu fission reaction is mitigated to a certain extent in the thorium/uranium fuel owing to the preferential fission reaction by ^{233}U, which can reduce the FTC even though the ^{238}U content is low. For the thorium/uranium fuel, most of the fission reactions are from ^{233}U (72%) while the fission reaction by ^{239}Pu is 44% for the natural uranium fuel. As the fuel temperature increases, the renormalized fission reactions by the fissile uranium isotopes tend to decrease while those by the fissile plutonium isotopes tend to increase. As a result, the FTC of the thorium/uranium fuel is smaller than that of the natural uranium fuel. However the FTC's of both fuels are very small.

The coolant temperature coefficients (CTC) are similar for both the natural uranium and thorium/uranium fuel. The CTC is positive at the beginning and increases slightly with recycling. The coolant temperature change has a positive effect on the reactivity due to the reduced heavy water density that decreases the upscattering of the thermal neutrons and enables more penetration of thermal neutrons into the fuel cluster. This phenomenon results

in a slight rise of both the fission and capture reaction rates of uranium and plutonium. However the integrated effect results in a positive reactivity.

The moderator temperature coefficient (MTC) is appreciably reduced for the thorium/ uranium fuel. The thermal spectrum of the thorium/uranium fuel is much less than that of the natural uranium fuel. If the moderator temperature increases, the neutron spectrum of the thermal energy region increases more for the natural uranium fuel compared to the thorium/uranium fuel at the equilibrium state, which results in a larger MTC value for the natural uranium fuel. ^{232}Th has a slightly larger value of the absorption cross section than ^{238}U around the thermal energy regions. But the total capture rates are similar and the fission reaction rate of ^{239}Pu is significant for the natural uranium fuel, which comes from the capture reaction of ^{238}U.

The void reactivity is one of the most important physics parameters of the CANDU fuel because it is positive and determines the magnitude of the power pulse upon the loss of coolant accident. When the coolant channel is voided, more thermal neutrons travel deep into the fuel bundle center as was seen for the CTC. If the fuel has a more excess reactivity, the void reactivity will increase more. For the cases considered here, however, it turned out that the void reactivity is similar for both the natural uranium and thorium/uranium fuels.

III.B Effect of ^{235}U Enrichment

The parametric calculation was also performed for the initial enrichment of the thorium/uranium fuel. The purpose of this calculation is to see how much the uranium volume fraction can be tolerated in the fuel mixture without deteriorating the recycling capability. Because it is known that the OREOX process may not be applicable to pure thorium-dioxide fuel which is chemically stable, it may be necessary to have a larger fraction of UO_2 in the fuel mixture so that the fuel mixture is voloxidized by the aid of uranium oxidation.

The initial ^{235}U enrichments considered were 5, 10 and 15 wt%; and the estimated discharge burnup was fixed to 16,000 MWd/t, which is nearly twice that of the natural uranium fuel. In order to obtain the target discharge burnup, the initial UO_2 volume fraction was investigated by a trial-and-error method. The final values of the volume fraction were 32.5, 18 and 12% for the initial enrichment of 5, 10 and 15 wt%, respectively. The natural uranium utilization of these fuels are summarized in Table II and compared to the results of the natural uranium and ThO_2-9%UO_2 fuels. The results indicate that the recycling of thorium/uranium fuel with a higher uranium fraction is feasible considering the natural uranium utilization. For the cases considered here, it was possible to obtain a natural uranium utilization greater than 5. The natural uranium utilization is higher when the uranium fraction is smaller. In other words, the natural uranium utilization increases if the thorium fraction increases even though the initial uranium enrichment is high.

Table II also shows the safety-related parameters of the fuels with different initial ^{235}U enrichments. They also provide similar behaviors compared with a 20wt% initial enriched uranium fuel. The only difference is the initial uranium loading and it is found that the enrichment of uranium can be reduced to around 5 wt% to obtain the thorium/uranium multiple recycles with a sufficient burnup. As the enrichment decreases, the moderator and the coolant temperature coefficients increase and the void reactivity decreases. This effect can be interpreted by the behavior of the enriched uranium fuel which is described in the previous section. The recycled fuel contains actinides and fission products, which may well provide a

quite different irradiation behavior compared to the fresh fuel.

III.C Effect of Fission Products Contamination

Fission product is one of the most important factors in the OREOX process for the uranium/thorium recycle and the effect of a fission product must be considered. Therefore parametric calculations were performed for a 9% UO_2 volume fraction with different fission product recovery rates.

The fission product contamination rates selected in this study were 5, 10 and 20% for the discharge burnup of 14,000 MWd/t. In the case a fission product contamination over 20%, the mass balance of multiple thorium recycle is not maintained due to an increased uranium feed. The natural uranium utilization of these fuels are summarized in Table III and compared to the results of the natural uranium fuel. The results showed that the recycling of thorium/uranium fuel with a certain amount of fission products is feasible from the viewpoint of natural uranium utilization. For the cases considered in this study, the natural uranium utilization is greater than 5 and becomes smaller when the fission product contamination increases.

The safety-related parameters of the 9% UO_2 volume fraction fuel with a different fission product contamination are also given in Table III. They also show similar behaviors for the various fission product contamination rates. As the amount of fission products in the recycled fuel increases, the safety-related parameters gradually degrade from the viewpoint of safety. However the results showed that it is possible to recycle the thorium/uranium fuel with a certain amount of fission products which are not completely removed in the OREOX process.

IV. Summary and Conclusion

The material balance calculations showed that it is feasible to construct a closed fuel cycle using the thorium/uranium fuel in a CANDU-6 reactor. The physics calculation also showed that the safety-related parameters of the thorium/uranium fuel are not deteriorated compared to those of the natural uranium fuel. The parametric calculations on the fuel composition showed that the fission products can be accommodated for with the recycled fuel from the physics viewpoint. It was also found that the uranium fraction in the thorium/uranium mixture fuel can be increased to facilitate the application of the OREOX process, even though an experimental verification will be required in the future.

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Table I. Comparison of safety-related parameters for the equilibrium state with different uranium fraction

Parameter	Natural uranium	ThO_2-9%UO_2	ThO_2-10%UO_2	ThO_2-11%UO_2
Natural uranium resource (kg/bundle)*	18.6	2.23	3.15	3.52
Fuel temperature coefficient (μk/K)	0.221	-9.998	-9.828	-9.679
Coolant temperature coefficient (μk/K)	57.21	46.36	47.55	48.68
Moderator temperature coefficient (μk/K)	3.508	5.538	4.027	3.796
Void reactivity (mk)	14.963	15.258	13.435	16.224

* (uranium feed) × (0.2 / 0.0071) ×(7300/ discharge burnup)

Table II. Comparison of safety-related parameters for the equilibrium state with different initial uranium enrichment

Parameter	Natural uranium	ThO_2-9%UO_2	ThO_2-12%UO_2	ThO_2-18%UO_2	ThO_2-32.5%UO_2
Natural uranium resource (kg/bundle)	18.6	2.23	2.56	2.83	3.38
Fuel temperature coefficient (μk/K)	0.221	-9.998	-9.894	-9.584	-8.511
Coolant temperature coefficient (μk/K)	57.21	46.36	46.55	48.15	52.58
Moderator temperature coefficient (μk/K)	35.08	5.538	6.550	7.523	10.32
Void reactivity (mk)	14.963	15.258	15.276	15.699	16.380

Table III. Comparison of safety-related parameters for the equilibrium state with different fission product contamination rate (ThO_2-9%UO_2 fuel)

Parameter	Natural uranium	0%	5%	10%	20%
Natural uranium resource (kg/bundle)	18.6	2.23	2.59	2.66	2.86
Fuel temperature coefficient (μk/K)	0.221	-9.998	-9.974	-9.985	-9.933
Coolant temperature coefficient (μk/K)	57.21	46.36	0.046493	0.046202	46.35
Moderator temperature coefficient (μk/K)	35.08	5.538	0.005145	0.004556	3.385
Void reactivity (mk)	14.963	15.258	15.279	15.315	15.361

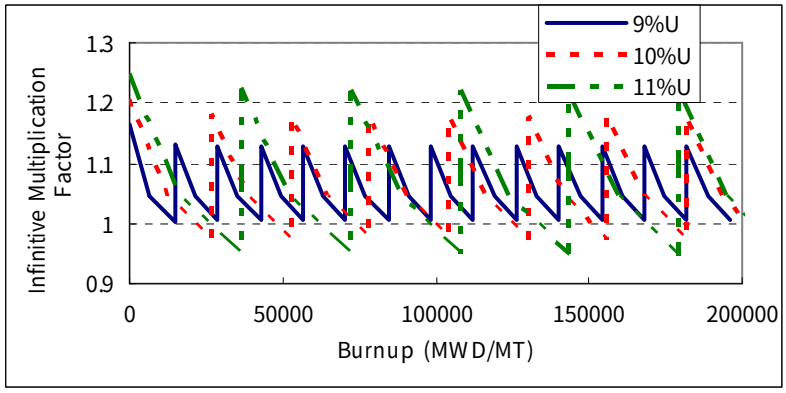

Figure 1. Infinite multiplication factor of recycled ThO_2-UO_2 fuel

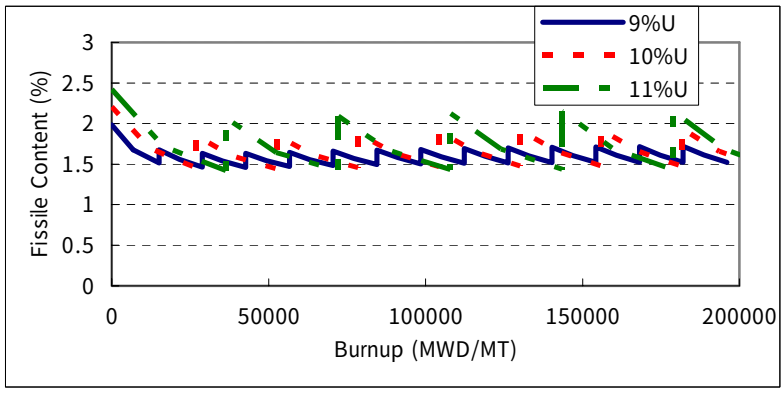

Figure 2. Fissile content of recycled ThO_2-UO_2 fuel