# Effective use of 234U in ThORIUM fuel cycle

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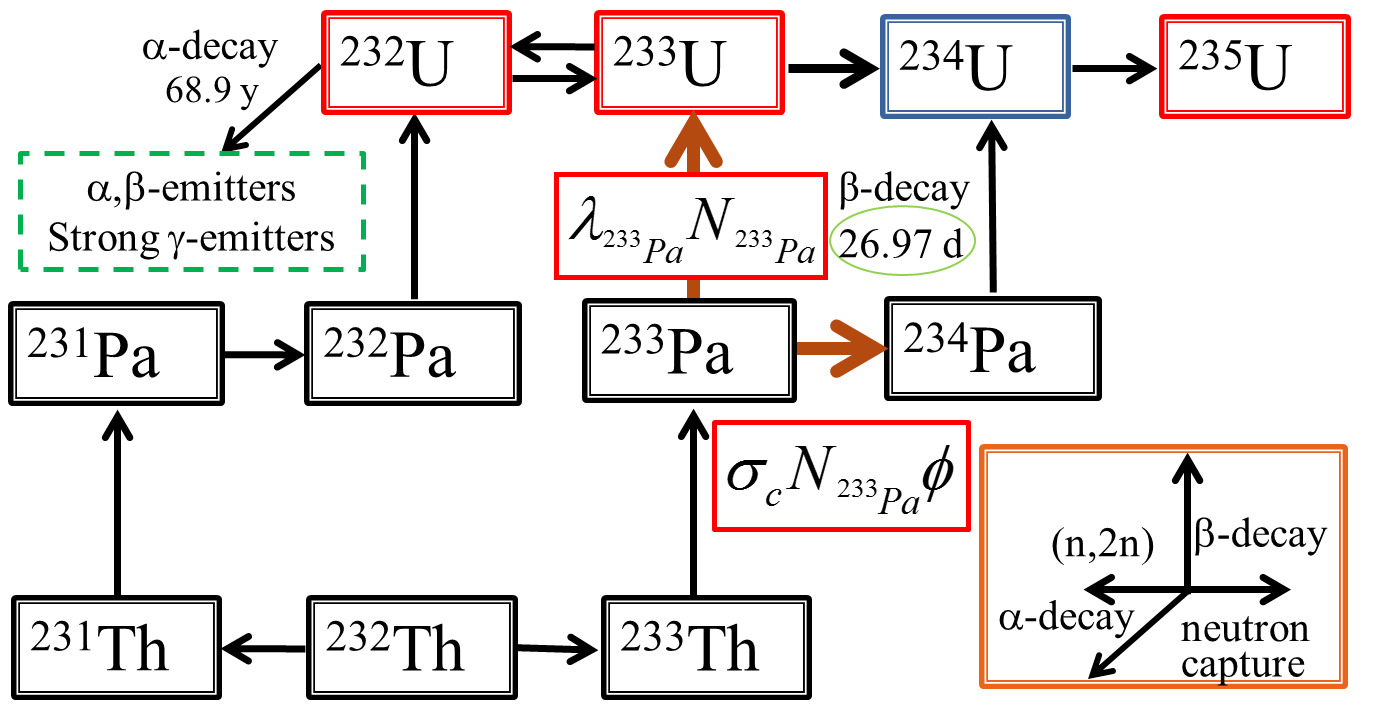
**Abstract**

The Th fuel cycle is attracting interest again globally because of its advantages over the current Pu fuel cycle, such as breeding fissile 233U from fertile 232Th without using a fast reactor, lower minor actinide production and higher Pu burning. However, there are some concerns, such as the small critical mass of the bred 233U. Using 234U, which is not considered an important isotope, may overcome some problems with the Th fuel cycle. In this study, the effect and roles of 234U in the Th fuel cycle were surveyed from the perspectives of proliferation resistance (PR), fuel burn-up, and breeding in single and multiple cycles. Increasing the 234U isotope ratio increases bare critical mass, which in turn increases PR by increasing the heat generation and radiation dose rate from 232U and their daughter nuclei. The effects of the moderator-to-fuel ratio, neutron energy spectrum, and neutron flux (linear power density) on criticality were estimated. 234U was fissile in the faster neutron energy spectrum, which can increase the fuel burn-up under some conditions. A higher fuel burn-up is preferable to increase the 234U isotopic ratio. For multiple cycles, the breeding ability of 234U was higher with a softer neutron energy spectrum (33.3% at the end of the fifth cycle), but the mass balance was worse. When 234U was used with a harder neutron energy spectrum, the 234U isotopic ratio was as high as 23.6%, but the mass balance was better. The role of 234U in Th has not been thoroughly investigated until now, but this study has revealed the importance of 234U, which may lead to the development of a new Th fuel cycle.

## INTRODUCTION

Despite the Fukushima Daiichi nuclear disaster, nuclear energy is still important due to environmental concerns and increasing energy demand. In Japan, we have been developing a U fuel cycle to ensure energy security. However, the Monju fast breeder reactor has been earmarked for decommissioning, meaning that the energy strategy in Japan will have to be rethought. Consequently, the use of Th is attracting interest again. In the Th fuel cycle, fissile 233U is generated from 232Th, and the 233U recovered from spent nuclear fuels can be used to make new fuels. The Th fuel cycle has some advantages over the U cycle and it is still being developed [1-4]. However, the Th fuel cycle also has problems. For example, 233U generated from 232Th has a much smaller critical mass, which leads to greater concerns about nuclear proliferation. Therefore, if Th is used in a closed fuel cycle, some countermeasures will be needed. In this research, the use of 234U, which has not been studied thoroughly in the Th fuel cycle, is investigated from the perspectives of proliferation resistance (PR), higher burn-up, and the controllability of 234U isotopic ratio in single and multiple cycles. These insights will help to advance the development of a new nuclear fuel cycle. The bare critical masses (BCMs) of 233U, 234U, and 235U are 15, 150, and 46 kg, respectively. Hence, increasing the 234U isotopic ratio can make the critical mass larger than that of pure 233U, which increases PR. The change in ratio increases the effects of the daughter nuclei of 232U, such as 208Tl and 208Pb, on the radiation dose rate and heat emission, which makes U fuel less attractive for malicious use. Furthermore, the Rossi-, which indicates the intensity of the fission reaction, can also be decreased because the Rossi- of 234U is 1/10 that of 233U. These observations indicate that the PR should be improved by increasing the ratio of 234U. Fig. 1 shows the chain reaction of Th fuel. Typically, 233U is produced by the chain reaction 232Th→233Th→233Pa→233U. This is similar to 239Pu production in the U-Pu cycle, 238U→239U→239Np→239Pu. The main difference between the two chains is the half-lives of 233Pa and 239Np. 239Np decays to 239Pu with a half-life of 2.35 days. However, the half-life of 233Pa to 233U is 26.97 days. Therefore, especially at higher neutron fluxes, there should be a considerable amount of 233Pa, and then the neutron capture reaction becomes important, and the contribution of the chain reaction 232Th→233Th→233Pa→234Pa→234U becomes more substantial. 234U can absorb neutrons to produce 235U, which may result in higher fuel burn-up under some conditions. The isotopic ratio of 234U can also be controlled by adjusting the neutron energy spectrum with the reactor type, core arrangement, and neutron flux. However, the use of 234U has not been investigated previously. Therefore, the following issues were investigated in this study.

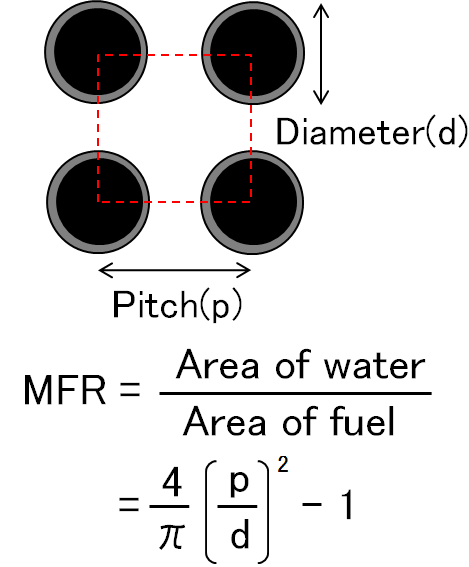
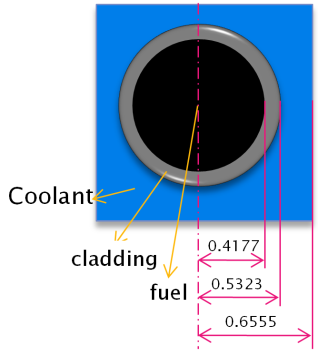
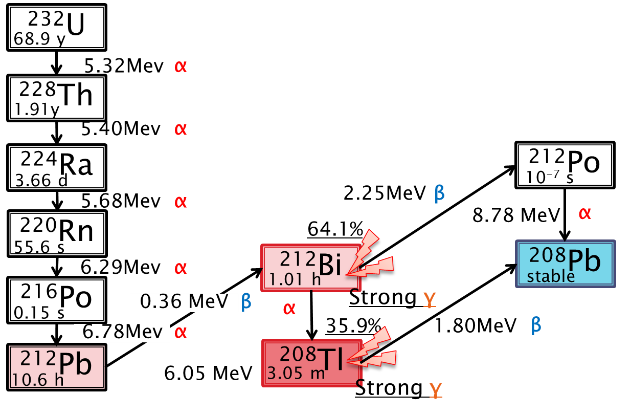
* Effect of 234U on the critical mass, Rossi, and radiation dose rate, which have major effects on PR.
* Controlling the 234U isotopic ratio in spent fuels by adjusting the neutron energy spectrum and neutron flux.
* Feasibility of a multiple Th cycle.



*Fig. 1. Chain reaction in the Th fuel cycle and important reactions in this study.*

## CALCULATION METHOD and conditionS

Fig. 2 shows the pin-cell geometry for the burning calculation. SRAC2006 code was used for the calculations with the collision probability method and JENDL-3.3as nuclear data (107 groups) [5,6]. The neutron energy spectrum was adjusted by changing the moderator-to-fuel ratio (MFR) from 0.5 to 3.0 (Fig. 2). Water was used as a coolant. In some cases, the density of the coolant was artificially decreased by up to 1/100 to reveal the effect of the neutron energy spectrum. Th oxide fuels with natural zirconium cladding were considered.

*Fig. 2. Calculation geometry and MFR.*

*Fig. 3. Decay chain considered in the study.*

MCNPX and FSXLIBJ-3.3based on JENDL-3.3 were used for calculating BCM and Rossi- [7-9]. Equation (1) is the definition of Rossi-

　　 　(1)

where *kinf* is the infinite multiplication factor, ** is the delayed neutron fraction, and *lprompt* is the lifetime of a prompt neutron. Rossi- was normalized with that of 233U to see the effect of the 234U mixture. To calculate the decay heat (*Q*) and radiation dose rate of the generated U, 232U and its corresponding daughter nuclides were considered (Fig. 3). For the heat calculation in equation (2),  and ** decays were considered, and for the dose rate calculation in equation (3),  decay was considered.

　　 　(2)  　　 　(3)

Here, *Nj*is the number of nuclei and *N01* is the initial number of 232U nuclei. Energy from -decay was not considered for heat generation because -rays penetrate the small BCM sphere. The radiation dose rate was estimated based on a literature method [10]. The intensity of the -rays was calculated with JENDL-4.0 [11]. Transport calculations of photons were carried out with MCNPX with CNPLIB02, and the photon fluence 1 m from the metallic sphere’s surface was estimated [12]. The correlation constants of the photon-equivalent dose rate reported in ICRP-21were used to estimate the dose equivalent rate [13]. As reported by the International Atomic Energy Agency (IAEA), when the radiation dose 1 m from the point is above 100 rem/h (for  radiation, the equivalent dose is 1 Sv/h), the material is self-protecting [14]. Therefore, in this paper, the PR is discussed using the IAEA-INPRO classification based on the radiation dose (TABLE 1).

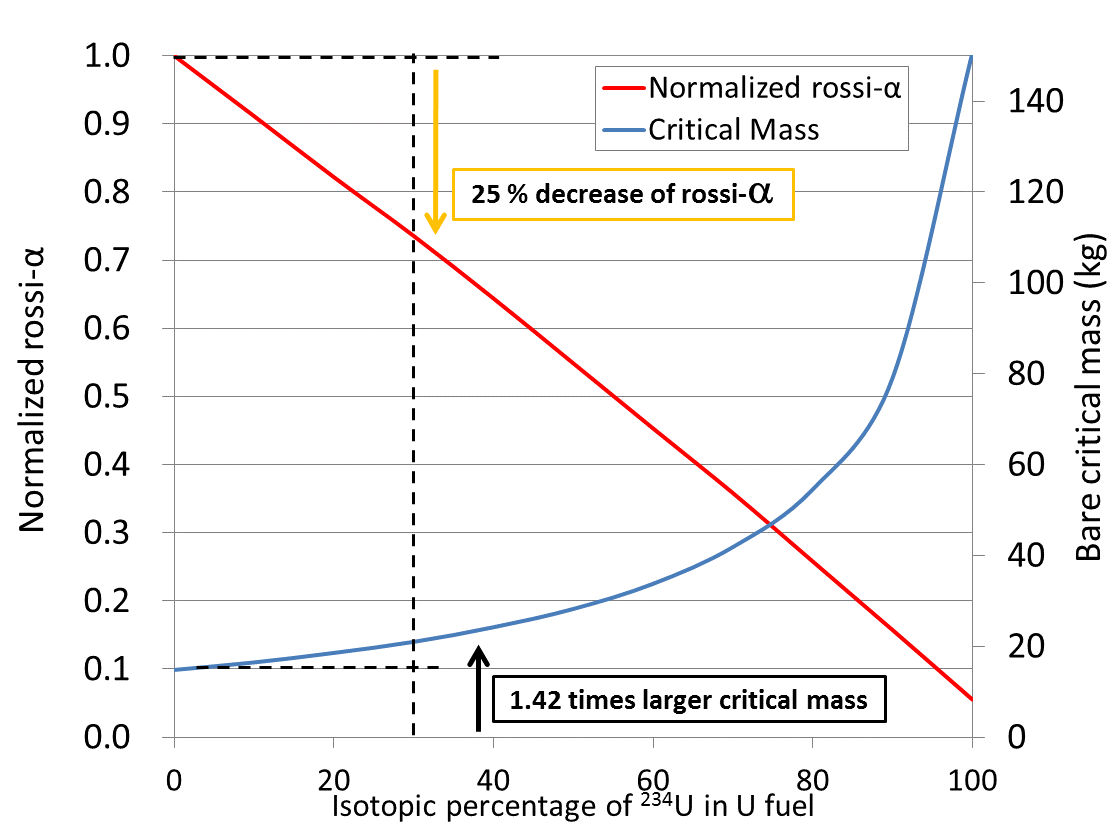
TABLE 1. EVALUATION OF ATTRACTIVENESS OF RADIOACTIVE MATERIAL BY THE IAEA [9]

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| Evaluation  Parameter | Evaluation scale | | | | |
| VERY WEAK | WEAK | MEDIUM | STONG | VERY STRONG |
| 232U conc. in 233U (ppm) | < 400 | 400 – 1,000 | 1,000 – 2,500 | 2,500 – 25,000 | > 25,000 |
| Dose at 1 m far from the surface (mSv/h) | < 150 | 150 – 350 | 350 – 1,000 | 1,000 – 10,000 | > 10,000 |

## ResultS and discussion

### 3.1. Increasing the PR of bred U with 234U

Fig. 4 shows the effect of the 234U isotopic ratio on BCM and Rossi-. The Rossi-was normalized by that of pure 233U. The Rossi- decreased linearly from 1 to 0.15 and BCM increased non-linearly with the increase in 234U content. A 234U isotopic ratio of 30% in U was set as the reference. For the reference case, the Rossi- was 25% smaller and BCM was 1.5 times larger than for pure 233U. The BCM for the reference case was calculated to be only 20 kg, and this indicates that the improvement of PR with respect to BCM was not sufficient.



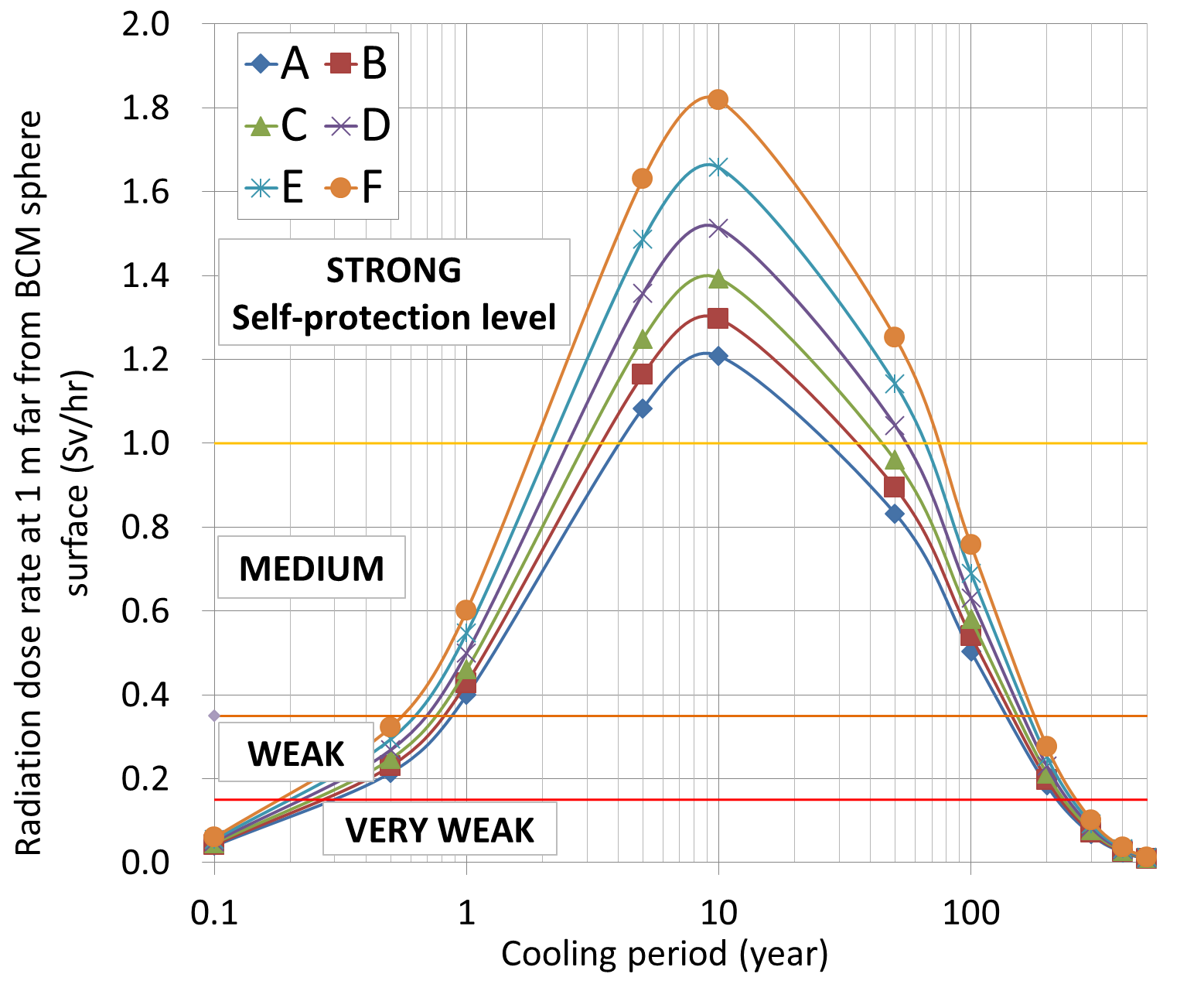
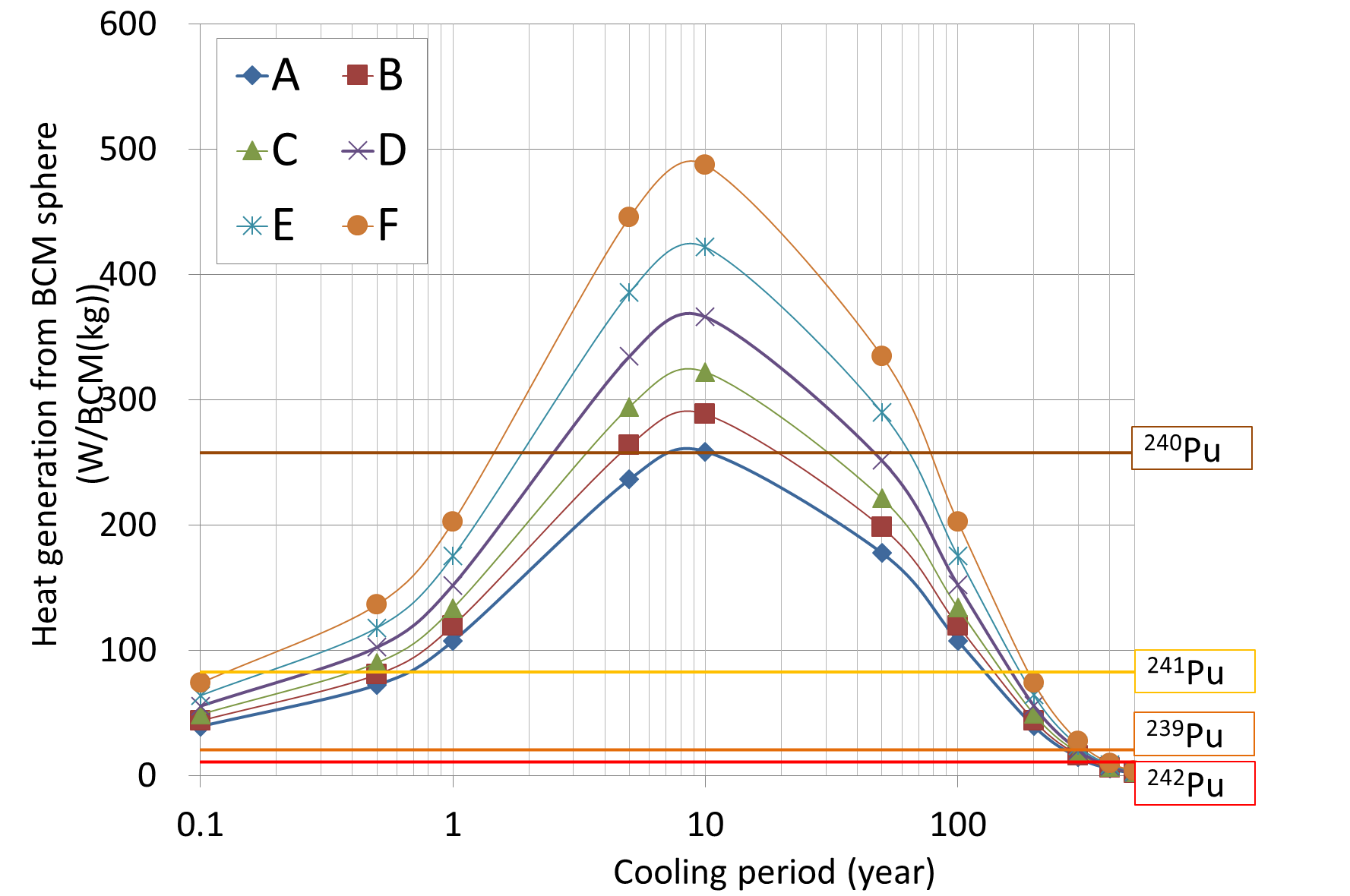
*Fig. 4. Effect of 234U in 233U on BCM and Rossi-*

To estimate the effect of 234U in U fuels on radiation dose and decay heat, the isotopic ratio of 232U was fixed to 0.2% and the ratio of 233U and 234U was varied (TABLE 2). The effect of 232U in spent fuels on radiation dose rate has been reported [10]; however, in this study, we considered the effect of 234U as well.

TABLE 2. ISOTOPIC COMPOSITION FOR BURNING CALCULATION

|  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- |
|  |  | A | B | C | D | E | F |
| Isotopic ratio of 233U and 234U  (232U = 0.2 %) | 233U | 100 | 90 | 80 | 70 | 60 | 50 |
| 234U | 0 | 10 | 20 | 30 | 40 | 50 |

The radiation dose rate 1 m from the surface of the BCM sphere and the heat generation are shown in Figs. 5 (1) and (2), respectively. In Fig. 5 (1), the range of attractiveness for fuels proposed by the IAEA listed in TABLE 1 is also shown. In every case in Fig. 5 (1), the radiation dose rate at the initial point was lower than the IAEA criterion for very weak PR (TABLE 1). As the 234U ratio increased, the time to reach the 1 Sv/h dose 1 m from the surface of the BCM sphere decreased. This is because the increase in 234U increased the BCM; namely, the critical radius increased, which increased the surface ratio over the volume (specific surface). The increase in absolute content of 232U also had an effect. In case D, with 30% 234U (reference case), the self-protection property was sustained for 1.5 to 55 years and radiation dose rate was calculated to be 1.25 times higher than that for pure 233U (case A). The amount of 232U needed to cross the boundary between the medium and strong levels, namely the criteria to exceed the self-protection level of 1 Sv/h, was 80%, which is favorable for PR. The heat generation showed a similar trend to the radiation dose (Fig. 5 (2)); the initial heat generation was small and after around 10 years, it reached a maximum due to the delayed generation of heat-emitting daughter nuclei. For comparison, heat generation from the main isotopes of Pu in BCMs except 238Pu is also shown in Fig. 5 (2). The heat generation per unit weight of 232U is larger than that of 238Pu, but it is difficult to exceed the IAEA level of exemption from safeguards for Pu (238Pu isotopic ratio of 80%), 6300 W/BCM (kg) at such concentrations in spent nuclear fuels. Based on this result, we concluded that decay heat is not sufficient to improve PR, and improving PR via radiation appears to be a more practical strategy.

(2)

(1)

*Fig. 5. Relation between cooling time and (1) radiation dose rate and (2) decay heat for different isotopic ratios of 233U/234U with 0.02% 232U and comparison with Pu isotopes.*

### Contribution of 234U to higher fuel burn-up

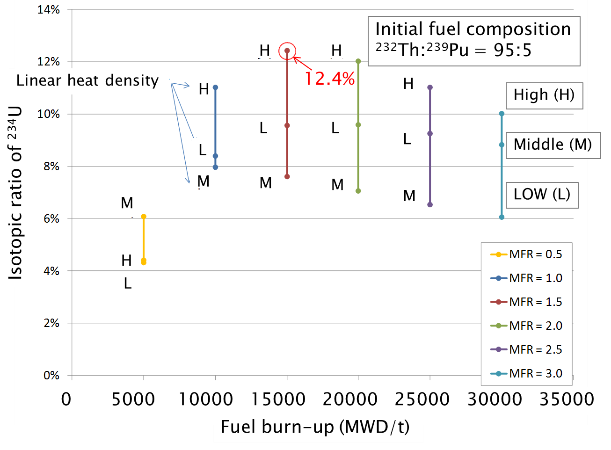
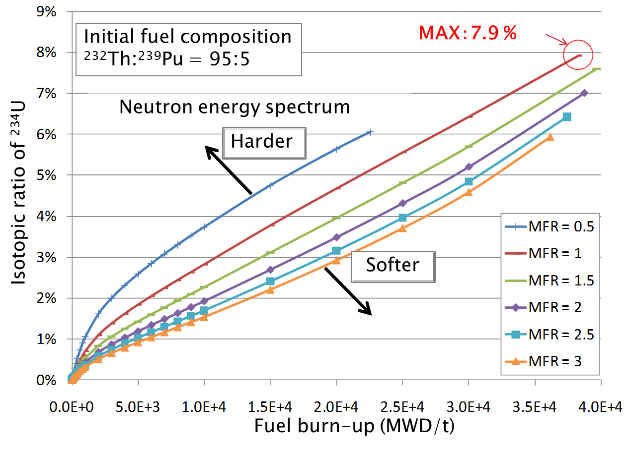
To examine the effect of 234U on criticality and fuel burn-up, three fuel cases were surveyed (TABLE 3). The infinite multiplication factor for the initial () and calculated fuel burn-up with a fast neutron energy spectrum were calculated, and the relationship between fuel burn-up and  is also shown in TABLE 3. When water was used as a coolant in cases A, B, and C, the order of the fuel burn-up was A > C > B, whereas the order of  was A > B > C. For cases a, b, and c, in which water with reduced density was used, the order of the fuel burn-up was c > a > b, whereas the order of  was a > c > b. This is because 234U can be fissile to some extent in a harder neutron energy spectrum. Especially in case c, a mixture of 7% 233U and 3% 234U showed better core performance, indicating that 234U is better for a harder neutron energy spectrum. In a softer neutron energy spectrum, a higher 234U isotopic ratio causes lower fuel burn-up; hence, a harder neutron energy spectrum, namely, a fast reactor, is appropriate to achieve higher fuel burn-up for 234U-containing fuel. More detailed calculations are possible, but the general behavior of 234U-containing fuel is well understood.

TABLE 3. FUEL COMPOSITION AND CONDITIONS FOR CALCULATING THE EFFECT OF 234U CONTENT IN THORIUM FUEL

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
|  | 232Th | 233U | 234U | Density of water |  | Burn-up  (MWD/t) |
| A | 90 | 10 | 0 | 1.00 | 1.588 | 1.199 × 105 |
| B | 93 | 7 | 0 | 1.00 | 1.492 | 8.518×104 |
| C | 90 | 7 | 3 | 1.00 | 1.396 | 8.60×104 |
| a | 90 | 10 | 0 | 0.01 | 1.221 | 3.199×105 |
| b | 93 | 7 | 0 | 0.01 | 0.990 | 3.089×105 |
| c | 90 | 7 | 3 | 0.01 | 1.009 | 3.260×105 |

### Controlling the 234U isotopic ratio by adjusting the neutron flux and neutron energy spectrum

We investigated controlling the 234U isotopic ratio. The linear power density was fixed as 1.79 × 10-4 MW/cm, which is typical for a light water reactor and the effect of MFR is shown in Fig. 7. The smaller MFR made the neutron energy spectrum harder and it promoted the accumulation of 234U, but the larger effect of the 233Pa→234Pa capture reaction due to the high neutron flux made the criticality lower. In contrast, a softer neutron energy spectrum (higher MFR) showed slow breeding of 234U against fuel burn-up, which allowed the decay reaction of 233Pa to 233U to occur more readily. This indicates that the optimum MFR is around 1 and the maximum accumulation ratio of 234U is 7.9%. Fig. 8 summarizes the isotopic ratio of 234U for neutron fluxes of 5.4 × 1011 (low), 5.4 × 1013 (medium), and 5.4 × 1015 1/cm2 s (high) with different MFRs. An MFR of 1.5 and 5.4 × 1015 1/cm2 s, which is 100 times higher neutron flux than a typical light-water reactor, the 234U isotopic ratio reaches a maximum of 12.5%. Therefore, if a higher concentration is needed, multiple fuel cycles will be necessary.



*Fig. 8. Effect of MFR and linear power density (neutron flux) on 234U isotopic ratio.*

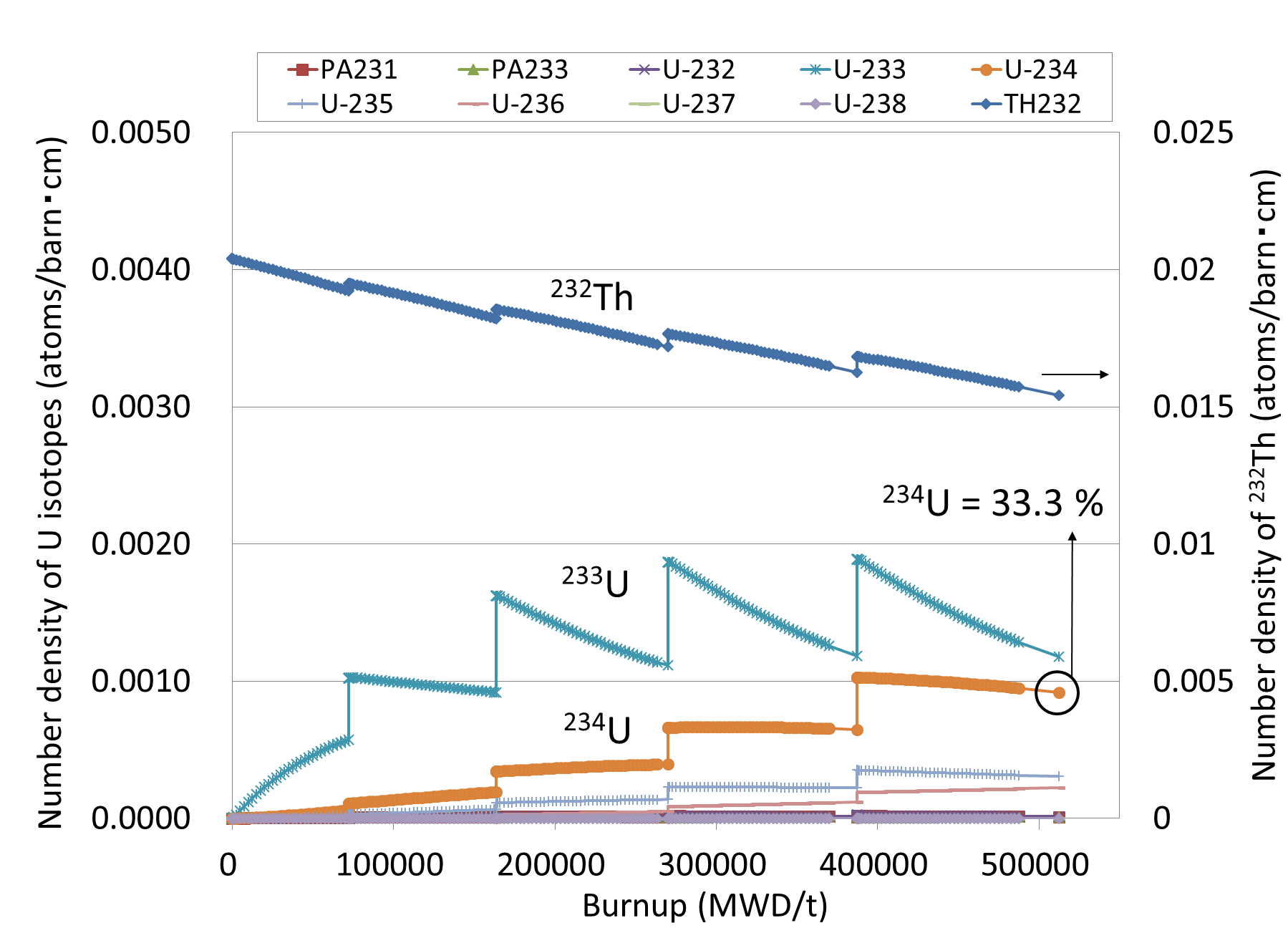
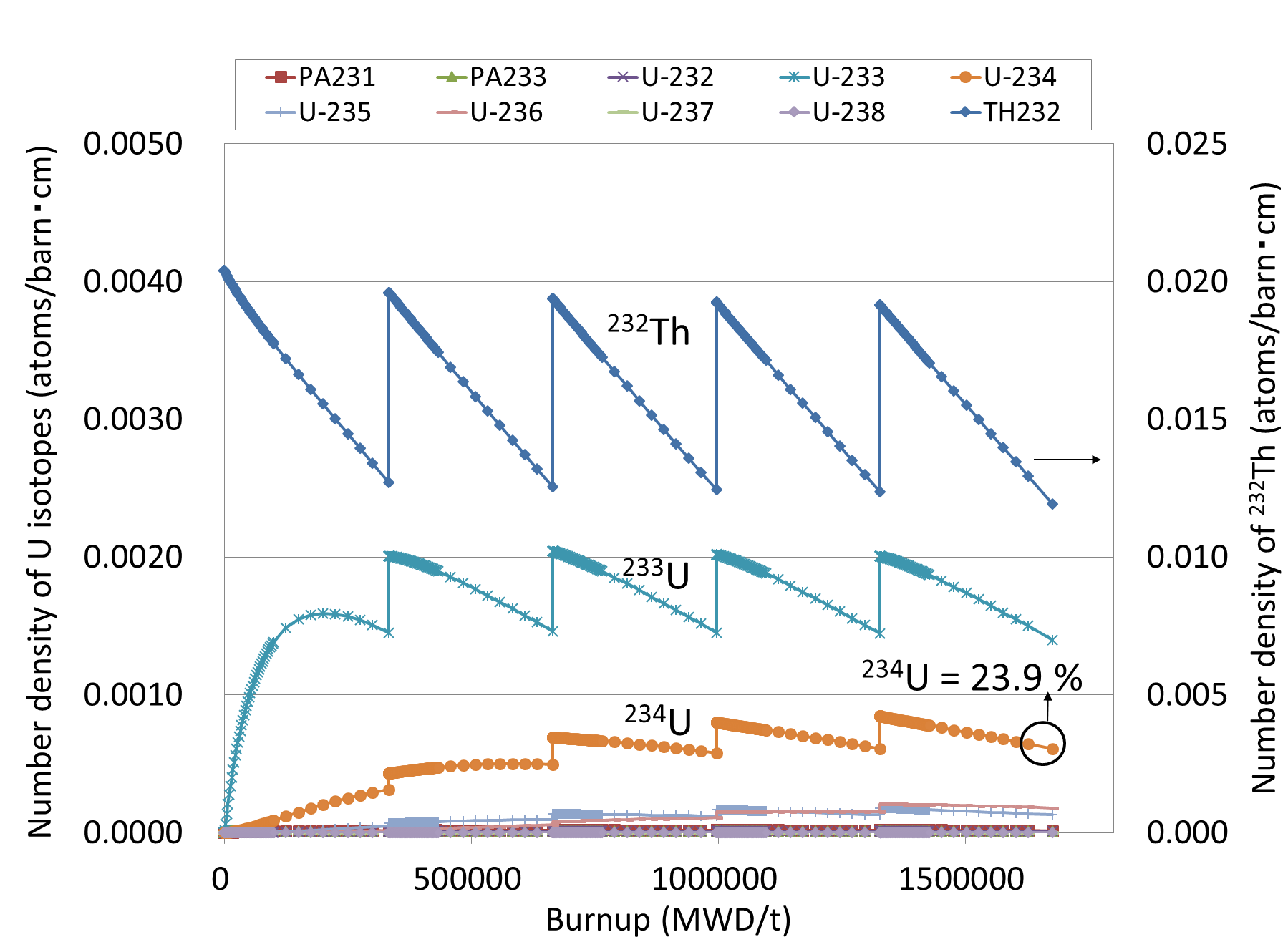
*Fig. 7. Effect of MFR and fuel burn-up on 234U isotopic ratio.*

### Effect of multiple cycles

After the first cycle, all the U isotopes, 233Pa, and remaining Pu isotopes were retrieved and the fissile amount was adjusted to 10%, and then the burn-up calculation was performed four more times. The initial fuel composition in the Th core contained 10% 239Pu and the mass balance was not considered in this study. 233Pa was considered to be 233U and the cooling period and decay of other actinides were not considered because the Th cycle does not generate short-lived actinides in contrast to the Pu cycle, and the cooling time makes no significant difference for fissile except for 233Pa. The calculation conditions and results are summarized in TABLE 4. The relationship between the number density of the U isotope and burn-up in the multiple-cycle operation is shown in Figs. 9 (1) and (2). In both cases, the amount of 234U and 232U increased with the increase in fuel burn-up. Comparison with cases A and B confirmed the effect of the neutron energy spectrum; a softer neutron energy spectrum increased the amount of 234U and 232U, and for fuel burn-up and mass balance, a faster neutron energy spectrum was better. The amount of 232U accumulated in the fuel was also higher with faster neutrons and the bred U fuel had a strong self-protection level (TABLE 1) from the first cycle, although the decay of 232U was not considered. More detailed surveys, such as core layout design and quantitative scenario analysis were beyond the scope of our conceptual study.

TABLE 4. MULTIPLE-CYCLE CALCULATION CONDITIONS AND RESULTS

|  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- |
|  | Initial fuel composition | | Linear Heat Rate (MW/cm) | Coolant density  (g/mL) | Isotopic ratio of 234U at end of 5th cycle (%) | 232U concentration at end of 5th cycle  (ppm) | Total burn-up  (MWd/t) |
| 232Th | 239Pu |
| A  B | 90 | 10 | 1.79×10-4 | 1 | 34.7 | 1.23×104 | 1.22×105 |
| 0.01 | 26.0 | 6.31×103 | 3.31×105 |

(2)

(1)

*Fig. 9. Relationship between fuel burn-up up to the fifth cycle and number density of 232Th and U isotopes for (1) water (softer neutron energy spectrum) and (2) 1/100 water density (harder neutron energy spectrum).*

### Waste management and reprocessing

The back-end technology associated with the Th fuel cycle is less mature, but an extraction method similar to plutonium–uranium redox extraction (PUREX), thorium–uranium extraction (THOREX), has been studied [15,16]. In the THOREX, Acid THOREX, and Interim 23 processes, UO22+ is extracted by tri-*n*-butyl phosphate (TBP) with Al(NO3)3 andHNO3 [17-19]. To replace TBP in the PUREX process, monoamide ligands can separate U/Th either with or without TBP [20–26]. TBP is depleted much faster in the strong radiation environment encountered in Th reprocessing. The depleted TBP can form unwanted complexes with actinides and other elements, which can make separation difficult. Therefore, monoamide ligands were studied because of their ease of synthesis, compliance with the CHON principle, and ability to extract hexavalent actinides. The other difficulty in reprocessing spent Th fuel is the requirement to work at a high dose rate. Either ligands or methods that are more robust against a radiation environment are required; therefore, strong ligands or other methods should be developed. For instance, we are developing a gel/liquid adsorption separation technique to be used in this type of radiation environment [27].

The waste management of spent Th fuel has not been examined much compared with the Pu cycle. The composition of the fission products generated from Th fuel is similar to that of U fuel, but the main difference is that minor actinides, such as Am and Cm. However, the raffinate from reprocessing Th oxide fuels contains fluoride ions and Th ions depending on the fuel cycle scenario and the manufacture of such glasses does not appear to have been reported. In addition, the safety and heat analysis of disposal sites for this type of new glass is needed. In summary, the back-end technology for the Th fuel cycle should be examined to advance the development of the Th fuel cycle. This may be the most important and challenging hurdle in Th fuel cycle development.

## CONCLUSIONS

The use of 234U in Th fuel cycles for higher PR, higher fuel burn-up, and controlling the 234U isotopic ratio were surveyed in this conceptual study. Our conclusions are as follows.

* The characteristics of fuels related to PR can be improved by increasing the 234U isotopic ratio.
* Uranium fuels with 0.3% 232U and 30% 234U had 1.5 times heavier BCM, 25% lower Rossi-, and 41% higher decay heat than 233U alone.
* The 232U isotopic ratio, which the IAEA suggests for fuel self-protection, can be minimized to about 20% by having a 234U isotopic ratio of 30% in U.
* Uranium fuels that contain 234U have increased burn-up with a harder neutron energy spectrum. For multiple cycles with a softer neutron energy spectrum, the 234U isotopic ratio reached 33.3% after the fifth cycle. With a harder neutron energy spectrum, a lower 234U ratio (23.6%) was obtained, but the fuel burn-up was longer. Because the mass balance was not considered, detailed simulations, such as quantitative analysis, are needed.
* The back-end technology for the Th fuel cycle is still not sufficiently mature. Breeding 234U creates more 232U and its daughter nuclei, producing a high-radiation environment during reprocessing and vitrification. Our work highlights the importance of upgrading back-end technology. In discussing both conventional and advanced Th fuel cycles, waste management should not be ignored.

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