AN INVESTIGATION OF TRU RECYCLING WITH VARIOUS NEUTRON SPECTRUMS

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Abstract

This study is intended to evaluate the dependency of TRU recycling characteristics on the neutron spectrum shift in a Pb-Bi cooled core. Considering two Pb-Bi cooled cores with the soft and the hard spectrum, respectively, various characteristics of the recycled core are carefully examined and compared with each other. Assuming very simplified fuel cycle management with the homogeneous and single batch fuel loading, the burn-up calculations are performed until the recycled core reached to the (quasi-) equilibrium state. The mechanism of TRU recycling toward the equilibrium is analysed in terms of burn-up reactivity and the isotopic compositions of TRU fuel. In the comparative analyses, the difference in the recycling behaviour between the two cores is clarified. In addition, the basic safety characteristics of the recycled core are also discussed in terms of the Doppler coefficient, the coolant loss reactivity coefficient, and the effective delayed neutron fraction.

Introduction

In many countries and organisations, the various concepts are having been proposed for TRU (Transuranics) transmutation. The fast neutron spectrum is being regarded as superior to the thermal spectrum due to its own attributes of the destruction capability through direct fission. [1] Therefore, development of TRU transmutation system is directed toward the liquid metal cooled reactor.

The research and development work are being performed in many design concepts, necessary for the utilisation of the excellent performances of fast reactor core. In the pervious study, the sensitivities of neutron spectrum and core performances to fuel pin cell configuration of Pb-Bi cooled core were investigated. [2] The results revealed that core performances are highly dependent on the neutron spectrum shift caused by changes of various core design options. From this result, we can consider a question still open. How much hardened spectrum meets the best performance to incinerate TRU safely and efficiently? To say in other words, even though the liquid metal cooled core has been selected for a fast neutron spectrum as an optimal option, another decision must be given how much hardened spectrum should be employed. In order to achieve the optimal design for the best core performances, this question is required to be evaluated quantitatively from the viewpoint of not only the core neutronic characteristics but also the fuel cycle management.

This study investigated how the recycling of TRU is influenced by the neutron spectrum shift in the Pb-Bi cooled core. Considering two Pb-Bi cooled cores with the soft and the hard spectrum, respectively, the various characteristics of the recycled core were compared with each other. TRU was recycled homogeneously in both cores until the recycled core reached to the equilibrium cycle. The attention was given to variation of the isotopic composition of the TRU fuel loaded in to the recycled core as well as burn-up reactivity with progress in multi-recycling. In addition, the basic safety characteristics of the recycled core were also discussed in terms of the Doppler coefficient, the coolant loss reactivity coefficient, and the effective delayed neutron fraction.

Calculation methods and conditions

The burn-up calculations were carried out using the burn-up code, REBUS3. [3] The nuclear characteristics of TRU-recycled cores were calculated by the diffusion calculations. Cross sections used in this study are based on the JEF-2.2. In the burn-up calculations, 9 group cross sections were used. Reactivity coefficients such as Doppler coefficient and coolant loss reactivity coefficients were calculated using 25 group data. Doppler coefficients were estimated by assuming that the temperature in the active core region is increased by 300K, from 980K to 1 280K. In order to evaluate reactivity feedback upon coolant loss, it is assumed that the Pb-Bi coolant is expelled by 5% volume uniformly from the whole core including not only the active fuel region but also the other regions (axial and radial reflector).

Core modelling

For the comparative analyses, two initial cores at the 1st cycle were modelled with the soft and the hard spectrum core, respectively. The HYPER (HYbrid Power Extraction Reactor) system was selected as a reference core, which is a 1 000 MW(thermal) class sub-critical Pb-Bi cooled reactor under development for TRU transmutation at KAERI (Korea Atomic Energy Research Institute). [4]

Soft spectrum core

Based on the design concept of HYPER, a critical core with the power level of 1 000 MW (thermal) was firstly modelled as the soft spectrum core. The active core height and the effective core diameters are 1.2 m and around 3 m, respectively. The thicknesses of the axial reflector below and above the active core are assumed to be equivalent to the active core height. The homogeneous core was considered, fueled with the chemical form of *x*TRU+(1-*x*)Zr throughout the active core, as shown in Figure 1. The weight fraction of TRU to Zr was adjusted to the excess reactivity for the cycle length of 365 EFPDs (Effective Full Power Days). At this time, the fuel composition was estimated as 23.7%TRU+73.6%Zr and the effective multiplication factor at the BOC (Beginning of Cycle) as 1.1907. The mean neutron energy in the soft spectrum core was calculated as 97.3 keV. The TRU fuel was assumed to be discharged from the standard PWR after burn-up of 35 GWD/MTH.

Hard spectrum core

In order to diminish the neutron moderation by the core material and attain the hard spectrum core, the P/D (Pitch-to-Diameter) of the fuel pin cell is reduced from 1.5 to 1.2. In addition, all the geometrical dimensions such as the core height and the assembly pitch were scaled down to the half size of the soft core. The main core design parameters are listed in Table 1, compared with the soft core. The TRU concentration and the cycle length were adjusted to the design criteria consistent with the soft core. From the standpoint of the thermal-hydraulic and material limitations, the average linear power of the fuel rods and the average burn-up of TRU are kept equal to the value of the soft core, as listed in Table 1. The fuel composition was estimated as 39%TRU+61%Zr and the cycle length as 600EFPDs, respectively. After all, because the volume of the active fuel region was reduced to the one-eighth, the reactor power was scaled down to 125 MW(thermal), which is very small and similar to that of the Pb-Bi-cooled long-life nuclear power reactor by Sekimoto and Su'ud. [5] The neutron spectrum was hardened considerably, depicted comparatively with the soft spectrum in Figure 2 and mean neutron energy was calculated as 271.8keV.

Figure 1. **Geometric configuration of the soft spectrum core**

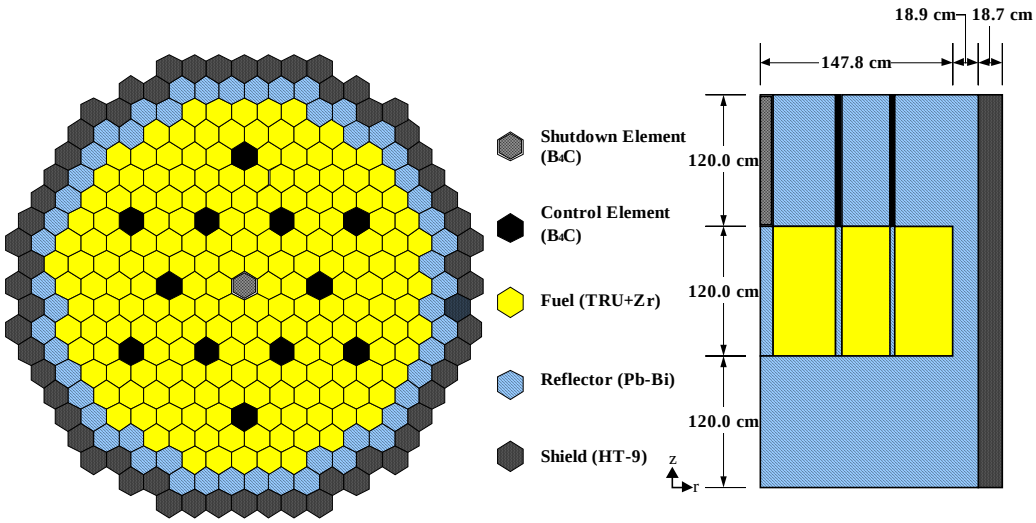


Table 1. **Comparison of design properties between the soft and the hard spectrum core**

Parameters	Soft core	Hard core
Reactor power [MW(thermal)]	1,000	125
Cycle length [EFPDs]	365	600
Active core height [cm]	120	60
Effective core diameter [cm]	295.7	118.3
Reflector thickness [cm] (top/bottom)	120/120	60/60
Assembly pitch [cm]	19.96	7.984
Pitch-to-diameter of fuel pin cell	1.5	1.2
Fuel composition	23.7%TRU+73.6%Zr	39%TRU+61%Zr
Effective Multiplication Factor at the BOC	1.1907	1.1613
Average linear power [W/cm]	251.6	251.6
Average fuel burn-up [GWD/MTH]	125.6	125.2

TRU recycling methods

At the 1st cycle, the TRU produced by the standard PWR, abbreviated as FTRU (fresh TRU) in this study, was loaded into the initial core. At the BOC of the next cycle from the 2nd cycle and on, all the TRU that have been irradiated during the previous cycle but not fissioned, abbreviated as ITRU (irradiated TRU), were reloaded into the recycled core. Simultaneously, a certain amount of FTRU was loaded additionally, necessary to compensate for reactivity loss due to fuel depletion during the previous cycle. Figure 3 shows the schematic diagram describing the recycling strategy.

Figure 2. **Comparison of the neutron spectrum**

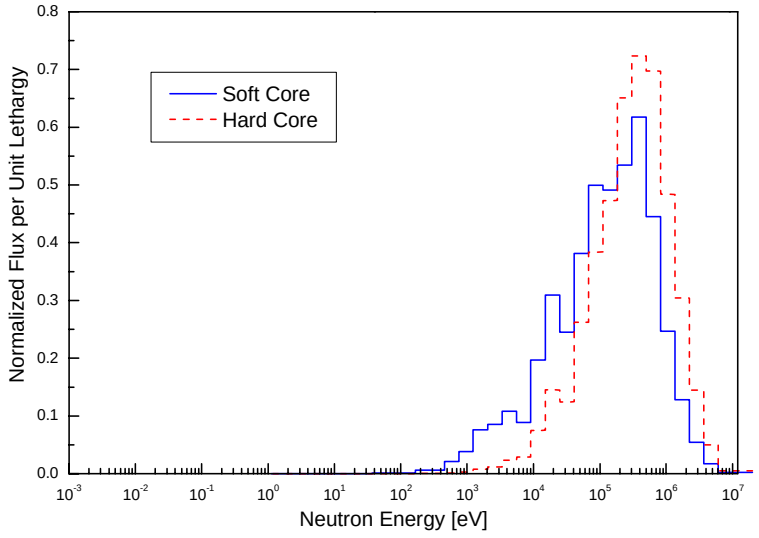
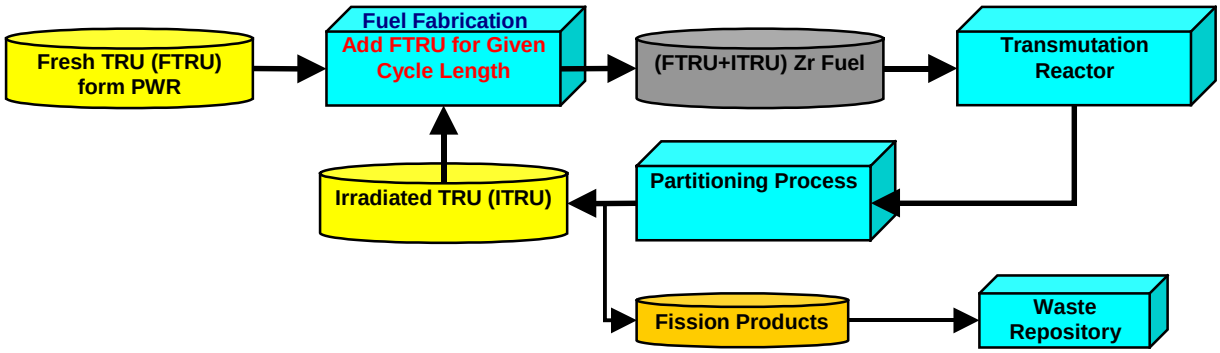


Figure 3. **TRU recycling strategy**



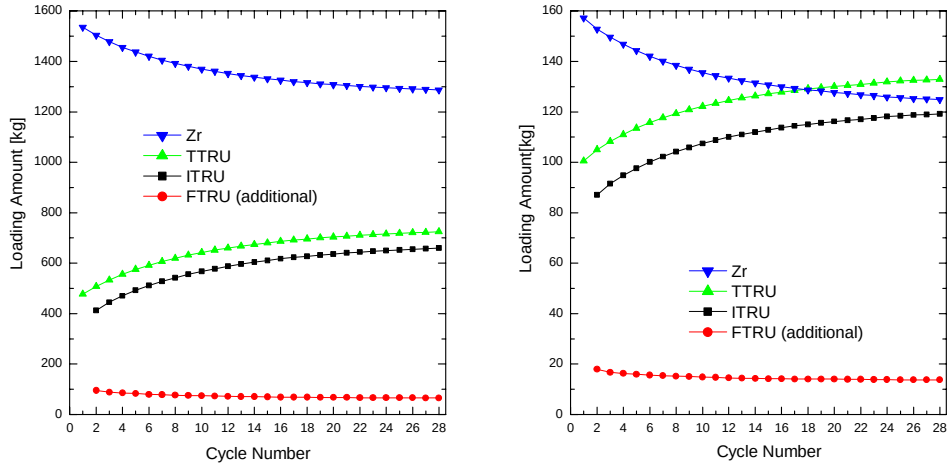
Even though TRU is loaded homogeneously throughout the active core at the beginning of each cycle, the isotopic composition of ITRU at the end of each cycle in a certain assembly is very different from that in the other assemblies, since the local burn-up after irradiation is highly dependent on the position in the core. In this study, after TRU was irradiated during the given cycle length and extracted from the core, all the ITRU in the various regions were assumed to be mixed with altogether. When this ITRU was reloaded into the recycled core mixed with the additional FTRU, the fuel isotopes were homogeneously redistributed throughout the active core. Any kind of cooling and/or reprocessing between in-core cycles was not considered.

Equilibrium search

At the First, we investigated how the recycled cores reach to the equilibrium in terms of the mass flow of fuel and the burn-up reactivity. In the recycled core, after TRU is irradiated during the given cycle length, the neutronic characteristics of ITRU is deteriorated in comparison with FTRU from the PWR. In other words, when the loading amount of FTRU, mixed with ITRU at the BOC, is equivalent to TRU destruction, difference in mass of TRU between at the BOC and the EOC (End of Cycle) of the previous cycle, the excess reactivity is not sufficient to maintain the given cycle length. Therefore, more FTRU than TRU destruction is required to be loaded additionally at the BOC of the next cycle, in order to keep the cycle length constant. At this time, Zr as a diluents material was removed from the fuel rod by the amount corresponding to increment in the loading amount of TTRU (Total TRU; ITRU+additional FTRU) and the density of fuel rod was kept constant. After all, the TTRU loading into the core increases gradually and Zr loading decreases on the contrary as TRU fuel is recycled in the core, as shown at Figure 4. In both cores the soft and the hard spectrum core, the variations of TRU composition to Zr are not different from each other but the TRU composition in the hard spectrum core becomes eventually larger than the Zr nuclide after the sufficient recycling over the 18^{th}.

The additional loading of FTRU at the BOC of each cycle improves the neutronic characteristics of ITRU at the EOC. Therefore, the required amount of additional FTRU at the BOC of the next cycle decreases on with progress in multi-recycling and it eventually approaches to the amount equivalent to TRU destruction. Accordingly, the increment in the total loading of TTRU is gradually diminished and, therefore, the loading amount of Zr as well as TTRU converges to a certain point, i.e., the recycled core goes toward the equilibrium, as shown in Figure 4.

Figure 4. **Comparison of fuel loading**



(a) Soft Spectrum Core (b) Hard Spectrum Core

In order to evaluate quantitatively the recycling behaviour toward the equilibrium, a convergence index was defined as *CF* (Convergence Factor).

$$CF\ (\%) \equiv \frac{FTRU(n) - \Delta TRU(n-1)}{\Delta TRU(n-1)} \times 100,$$

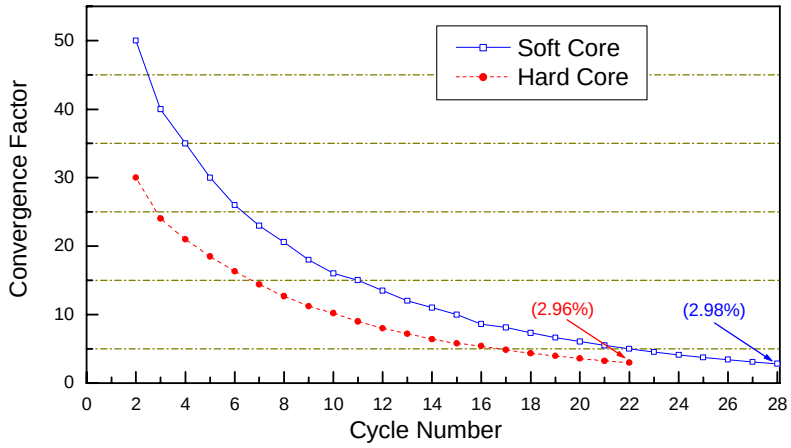
where

$FTRU(n) =$ loading amount of FTRU at the BOC in the n^{th} cycle (1)

$\Delta TRU(n-1) =$ difference between the total loading of TTRU at the BOC and the rsdidual of ITRU at the EOC in the $n-1^{th}$ cycle.

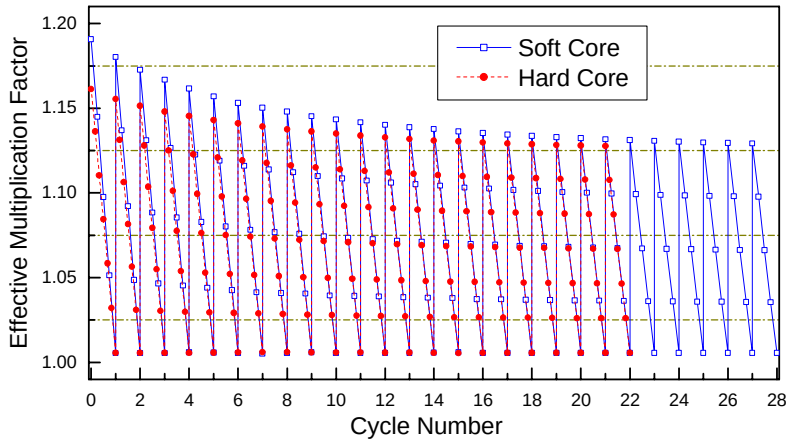
The calculation results are compared between the two cores as shown in Figure 5. It is indicated that, in the soft spectrum core, the amount of FTRU required to be loaded at the BOC of the 2nd cycle is 50% larger than TRU destruction in the 1st cycle. On the other hand, in the hard spectrum core, the value is much smaller (30%), because the core characteristic is not very different from the equilibrium core in comparison with the soft spectrum core, as described later. In the result, the hard spectrum core converges more rapidly to the equilibrium than the soft core. In this study, the core with the convergence factor smaller than 3% was considered as the (quasi-)equilibrium. The soft spectrum core reached to the equilibrium after the 28th cycle and the hard core after the 22nd cycle.

Figure 5. **Comparison of the convergence factors**



In the results shown in Figure 6, the effective multiplication factors were calculated with all control rods out. In the 1st cycle, the hard spectrum core requires smaller excess reactivity at the BOC in order to irradiate the fuel up to the same average burn-up to the case of soft spectrum core. It is attributed to the fact that the hard spectrum core has larger conversion ratio by virtue of higher η-value. In this figure an interesting result is noticed that regardless of whether the spectrum is harder or softer, the excessive reactivity eventually approaches to the same point in the result of multi-recycling. Thus, the excess reactivity converges more rapidly than the case of soft spectrum, consistent with the result of the convergence factor. Judging from the intuition, the equilibrium point of the excess reactivity is governed by only the average fuel burn-up, regardless of how much hardened spectrum is chosen.

Figure 6. **Comparison of the burn-up reactivity**



Fuel composition in core inventory

Figures 7 and 8 give the comparison of the isotopic compositions of TRU fuel in the recycled core. FTRU produced from PWR has the large amount of ^{239}Pu; its weight fraction in FTRU is about 52%. When it is loaded additionally, mixed with ITRU at the BOC of each cycle, the large fraction is disappeared through direct fission but the considerable amount is transferred into ^{240}Pu through the

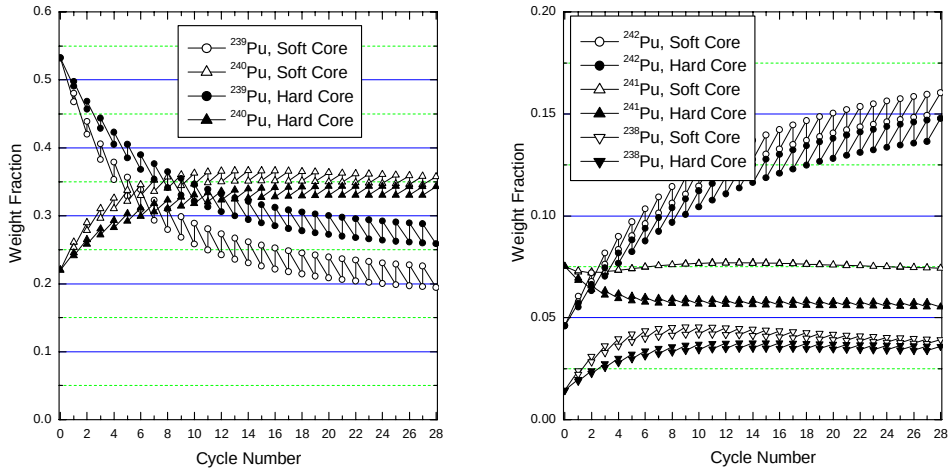
neutron capture during the given cycle length. In the result, its weight fraction in the TRU fuel decreases on whereas that of ^{239}Pu increases gradually as the recycling is iterated, as shown in Figure 7(a). Eventually, the fraction of ^{240}Pu becomes larger than that of ^{239}Pu after the considerable recycling.

In the both cores, the variation tendencies of the weight fraction of these nuclides (^{239}Pu and ^{240}Pu) are not very different from each other but they converge toward the very different point in the equilibrium. The fraction of ^{239}Pu decreases more in the soft core in comparison with the hard core since the soft spectrum has higher α–value (capture-to-fission ratio), as was expected. Here, the result discussed in the previous section needs to be reminded that the excess reactivity converges to the same point regardless of the neutron spectrum. It should be also noted that the spectrum effect on the variation of the isotopic composition in the fuel is not small but considerable and this variation gives rise to the not negligible change in the safety performance, especially the delayed neutron fraction.

Another noticeable finding in Figure7(b) is that the weight fraction of ^{241}Pu in the soft spectrum core remains nearly constant whereas decreases considerably in the hard core. It is attributed to the fact that the considerable amount of ^{241}Pu is produced from the neutron capture of ^{240}Pu in the soft core. This fissile nuclide cancels out the reactivity drop due to the decrease in the weight fraction of ^{239}Pu and assists to keep the excess reactivity same to the case of hard spectrum core in the equilibrium.

In Figure 8, the weight fraction of ^{241}Am decreases on in the soft spectrum core whereas increases slightly at the early stage in the hard core. The behaviours of the other minor actinides are also very different between the two cores; for an instance, the higher actinides such as ^{243}Am and ^{244}Cm is accumulated in the soft spectrum core much more and their weight fraction becomes eventually larger than the other minor actinides such as ^{237}Np and ^{241}Am. However, their contribution to the core characteristics is not considerable because their cross section as well as the weight fraction is small.

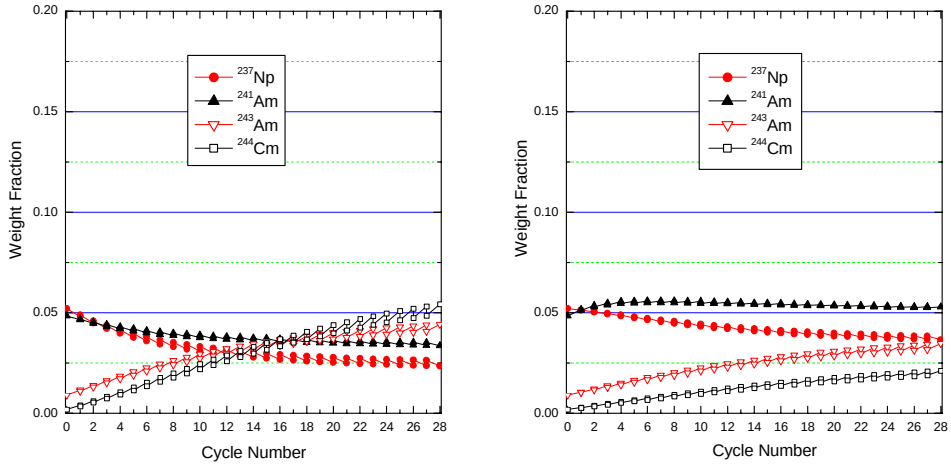
Figure 7. **Comparison of the Pu isotopes compositions**



(a) ^{239}Pu and ^{240}Pu

(b) ^{238}Pu, ^{241}Pu and ^{242}Pu

Figure 8. **Comparison of the minor actinides isotopes compositions**



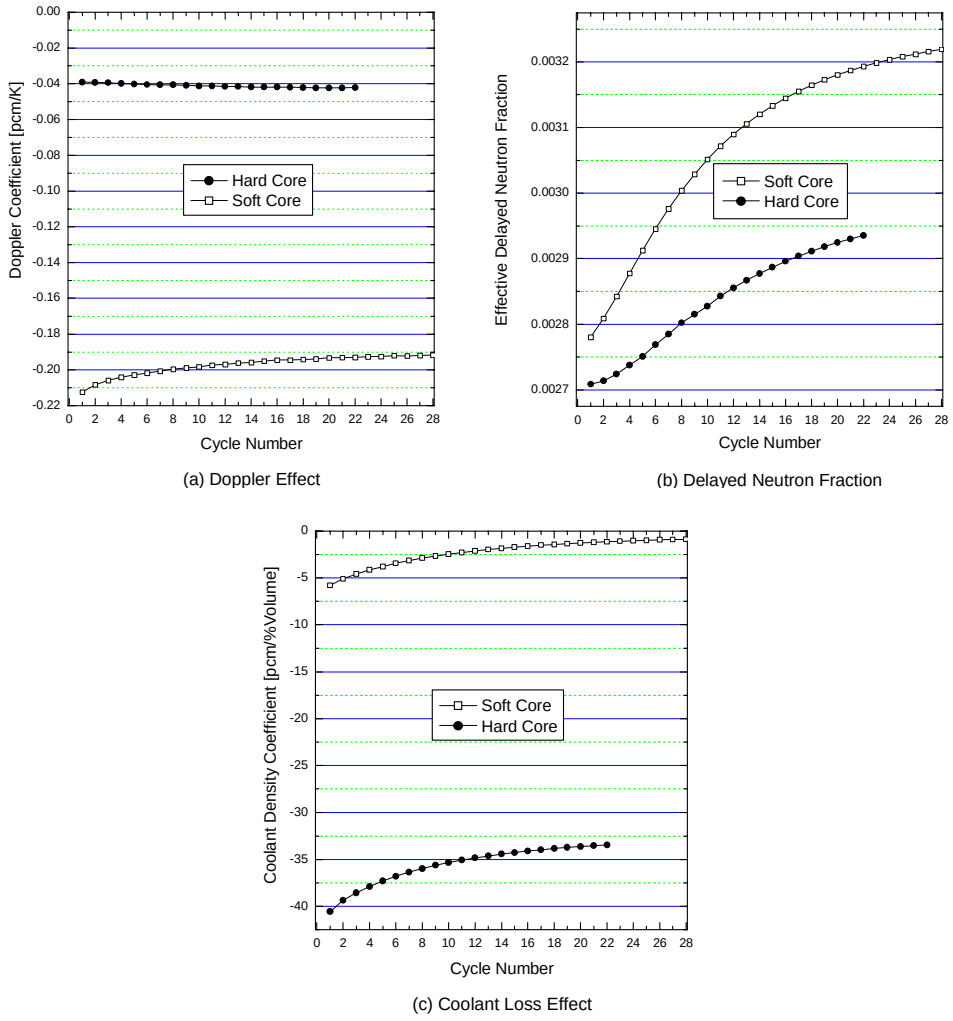
Safety performances

Figure 9 shows the comparison of some safety parameters. The result indicates that the negative reactivity effect of Doppler broadening is not improved even though the relative contribution of ^{240}Pu to ^{239}Pu increases considerably. In Figure 9(a), the Doppler coefficient in the hard spectrum core becomes slightly more negative but remains almost constant throughout the multi-recycling. Moreover, in the soft core, the Doppler broadening effect decreases on and the reactivity coefficient goes toward less negative on the contrary.

The fission by ^{240}Pu produce more delayed neutrons in comparison with ^{239}Pu. At the BOC of the soft spectrum core in the 1st cycle, the effective delayed neutron fraction by ^{239}Pu and ^{240}Pu were calculated as 0.0022 and 0.0031, respectively. Therefore, the effective delayed neutron fractions in the both cores increases on, as shown in Figure 9(b), since the contribution of ^{240}Pu is increased. Simultaneously, the considerable accumulation of ^{242}Pu in the soft core, which has the large value of delayed neutron fraction (estimated as 0.0066), assists to increase the fraction of delayed neutron over the contribution of ^{240}Pu. On the other hand, in the hard spectrum core, the weight fraction of ^{239}Pu is less decreased in comparison with the soft core and it gives still negative contribution to the production of delayed neutron. Moreover, the considerable decrease in the weight fraction of ^{241}Pu, which has the large contribution (0.0054 in the hard spectrum core), cancels out the positive contribution of ^{240}Pu and ^{242}Pu.

When the coolant is expelled from the core, the fission cross sections of the fertile nuclides such as ^{240}Pu and ^{242}Pu are increased much more than those of the fissile nuclides (^{239}Pu and ^{241}Pu). Therefore, as the TRU fuel is recycled on, the reactivity feedback upon the coolant loss event becomes less negative, as shown in Figure 9(c). The relative compositions of these fertile nuclides increase more in the soft spectrum core than in the hard core, as described in the previous section. However, the coolant density coefficient goes toward less negative more sensitively in the hard spectrum core because the fission cross sections of the fertile nuclides is increased more steeply in the higher neutron energy region.

Figure 9. **Comparison of the safety parameters**



Summary and conclusions

As stated in the first section, this study was intended to evaluate the dependency of TRU recycling characteristics on the neutron spectrum shift in a Pb-Bi cooled core. The results can be summarised as follows:

1. The excessive reactivity converges toward the same point regardless of whether the soft spectrum is chosen or the hard spectrum, despite its starting point in the initial core of the 1st cycle are different from each other.
2. The reactivity variation is smaller in the hard spectrum not only during the burn-up period at each cycle but also throughout the entire periods of multi-recycling to the equilibrium.
3. As TRU is recycled, the effective delayed neutron is significantly increased especially in the soft spectrum core but the negative reactivity effect of the Doppler broadening is decreased considerably. The coolant loss reactivity also goes toward less negative considerably.

In conclusions, from the view point of safety concerning an accidental positive reactivity insertion in the critical core, the hard spectrum is superior to the soft spectrum. When an ADS (Accelerator-driven sub-critical system) for TRU incineration is considered, the harder spectrum is still favourable over the soft spectrum, as far as the requirement for minimisation of the beam current throughout the recycling is concerned. In the soft spectrum core, competition of increase in the effective delayed neutron fraction with decrease in the negative reactivity effect such as Doppler broadening and the coolant loss effect should be evaluated carefully by means of dynamics analyses.

Future work will take into account the transmutation capability and the advanced core design characteristics (e.g., utilisation of the burnable absorber for minimisation of the burn-up reactivity swing and maximisation of the external source multiplication in an ADS, etc.)

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REFERENCES

[1] W.S. Park *et al.* (1999), *TRU Incineration Characteristics of Thermal and Fast Sub-critical Reactors*, Annals of Nuclear Energy, 26, pp.1497-1508.

[2] Y.N. Kim *et al.* (2002), *A Study on the Neutron Spectrum Optimization for TRU Transmutation Reactor*, Transactions of the American Nuclear Society, 86, p.428.

[3] B.J. Toppel (1983), *A User's Guide for the REBUS-3 Fuel Cycle Analysis Capability*, ANL-83-2, Argonne National Laboratory, USA.

[4] W.S. Park *et al.* (1999), *Design Concept of HYPER (HYbrid Power Extraction Reactor)*, *Proceedings* of the 3rd International Conference on Accelerator-driven Technologies and Applications (ADTTA'99), Praha, Czech Republic, June.

[5] H. Sekimoto and Z. Su'ud (1995), *Design Study of Lead- and Lead-Bismuth-cooled Small Long-life Nuclear Power Reactors Using Metallic and Nitride Fuel*, Nuclear Technology, 109, pp.307-313.