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Feasibility Study on Long-Lived Fission Products Transmutation in Equilibrium Fuel Cycles of PWR

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Abstract

A study on long-lived fission products (LLFP) transmutation in equilibrium fuel cycles of pressurized water reactors (PWR) has been performed. The seven important LLFP, i.e., ^{79}Se, ^{93}Zr, ^{99}Tc, ^{107}Pd, ^{126}Sn, ^{129}I, and ^{135}Cs were considered in this study. In order to get a comprehensive perspective, single isotope/element transmutations and mixed isotopes/elements transmutations for both isotopic separation and elemental separation were employed. The calculation results showed that the transmutation of the all seven LLFP together with isotopic separation are feasible by increasing the required enrichment of loaded uranium fuel up to 7.1wt% for standard PWR. Isotopic separation is not necessary for mixed technetium and iodine transmutation. The transmutation of the all seven LLFP together with elemental separation may not possible since zirconium transmutation in PWR is a formidable task.

Keywords: *long-lived fission products, transmutation, isotopic separation, elemental separation, equilibrium fuel cycle, importance, PWR*

1. Introduction

One of the most important concerns on nuclear power utilization is radioactive wastes management especially high-level wastes (HLW) management[1)]. There are two main policies in dealing with HLW from nuclear reactor: Once through cycle, in which after single use HLW directly dispose to a final deep geological repository site that has to be designed and managed to provide barriers to the release of hazardous materials for thousands of years is the first strategy. Recycling or reprocessing of the HLW prior to the final geological disposal is the second strategy. The latter strategy is very attractive since its can extract up to more than 70 times more energy from the fuel, while at the same time reducing the volume and radioactive lifetime of the wastes that have to be stored in the repository[2)].

Recycling, or reprocessing, essentially involves dissolving spent nuclear fuel and separating out the uranium, plutonium and certain other materials such as minor actinides (MA) for re-use in the reactor. The remaining fission product (FP) wastes, which remain radioactive but much less than those of uranium and plutonium then are dissolved in melted glass for disposal and long-term storage in stainless steel canisters.

Concerning the fission products wastes, severe problem comes from long-lived fission products (LLFP). Although, after some centuries, the potential beta toxicity of FP will remain far below the alpha toxicity of actinides, LLFP contribute significantly to the risk during storage where leaching and transport by water are considered. This is due to their high mobility in these geological environments, which results in that they can migrate easily away from underground repositories and hence eventually reach the biosphere[3,4)]. This fact motivates the author of the present paper to study the feasibility of LLFP transmutation.

Since almost all of commercialized nuclear power plants come from light water reactors (LWR), especially pressurized water reactor (PWR) and isotopic separation is very important for transmutation of LLFP[5)], the present paper is aimed to study the feasibility of LLFP transmutation both with isotopic and elemental separations in the equilibrium fuel cycles of PWR.

This work continues the series of study on characteristics of the equilibrium fuel cycles of PWR. We have reported the numerical results of study on the recycling of plutonium and MA in equilibrium fuel cycles of PWR[6,7)]. These results show the merits of the recycling of plutonium and MA in the equilibrium fuel cycles of PWR on the reduction of the required enrichment of uranium fuel, the required natural uranium supply, and the toxicity of heavy metals in spent fuel. The basic physics characteristics of the core of the studied PWR are shown in Table 1.

2. Long-lived Fission Products: An Overview

Among thousands of FP, a few of them have the half-life of more than some hundred years, which are categorized as LLFP. Among these long-lived fission products, there are seven important LLFP, i.e., ^{79}Se, ^{93}Zr, ^{99}Tc, ^{107}Pd, ^{126}Sn, ^{129}I, and ^{135}Cs. These fission products not only have long half-life but also dominate the total radio-toxicity and total amount of fission products in the

nuclear wastes. The latter fact is shown in Table 2 with relatively large value of the cumulative yields of these nuclides for ^{235}U fissile nuclide. Table 2 also shows the stable isotopes of these elements.

The other LLFP such as ^{92}Nb (3.2 x 10^7 y), ^{94}Nb (2.0 x 10^4 y), ^{93}Mo (3.0 x 10^3 y), ^{97}Tc (2.6 x 10^6 y), and ^{98}Tc (2.14 x 10^5 y) are not so important because their number densities in the reactor core are four to ten orders smaller than the former seven LLFP.

2.1 Long-lived Fission Products Separation

Selenium has six stable isotopes, which dominate the total nuclide number density of Se in the reactor core[8]. Three of these stable isotopes (^{74}Se (51.8b), ^{76}Se (85 b), ^{77}Se (42b)) also have thermal capture cross-sections roughly same as of ^{79}Se (50 b) itself[9]. Therefore, isotopic separation of Se isotopes is necessary if ^{79}Se is to be transmuted in PWR in order to avoid large neutron absorption by stable isotopes.

There are five stable isotopes of zirconium element. Even though they have capture cross-sections in thermal region smaller than that of ^{93}Zr (2.2 b)[9], their nuclide number densities are much larger than that of ^{93}Zr. For this reason they will absorb many neutrons if they are transmuted in PWR. Therefore isotopic separation of Zr isotopes is very important when ^{93}Zr is to be transmuted in PWR.

^{99}Tc with the half-life is 2.13 x 10^5 y is the only one long-lived technetium isotope in reprocessed spent fuel[5]. ^{99}Tc will transmute to ^{100}Tc (~16 s) via (n,γ) reaction, and then ^{100}Tc will decay to become ^{100}Ru (stable nuclide). Moreover, ^{99}Tc has quite large thermal capture cross-section of about 19.6 barns[9]. It may not necessary to undertake the isotopic separation ^{99}Tc before transmute it in PWR. This fact would be confirmed from the results of the present study.

Palladium has six stable isotopes. Three of them (^{104}Pd, ^{105}Pd, and ^{106}Pd) have the nuclide number density slightly larger than that of ^{107}Pd. The thermal capture cross-sections of three stable isotopes (^{102}Pd (3.4b), ^{105}Pd (20.3b), and^{108}Pd (8.5b)) are larger than that of ^{107}Pd (2b)[9]. Consequently, isotopic separation before transmutation is necessary.

According to references[8,10] there are ten stable isotopes of tin, but ^{112}Sn was not employed in this study. ^{126}Sn has very small thermal capture cross-section. Except ^{115}Sn (30 b) other stable isotopes of tin have thermal capture cross-sections around 2 barns and bellows but still larger than that of ^{126}Sn (90 mb). The nuclide number densities of the stable isotopes are comparable with that of ^{126}Sn, so that isotopic separation is important if ^{126}Sn is going to be transmuted in PWR.

There is only one stable isotope of iodine that is ^{127}I. The nuclide number density of ^{127}I in spent fuel is almost same as that of ^{129}I, but its thermal capture cross-section much smaller than that of ^{129}I (27 b). Therefore isotopic separation may not necessary for iodine transmutation in PWR. As for ^{99}Tc, ^{129}I can be transmuted by single neutron capture into stable isotope. ^{129}I will transmute to ^{130}I (12.36 h) via (n,γ) reaction, and then ^{130}I will decay to become the noble gas ^{130}Xe.

Cesium has only one stable isotope that is ^{133}Cs with the cumulative yield of neutron -induced fission of ^{235}U about 6.7%[11]. The thermal capture cross-section of ^{133}Cs is 29 barns. Cesium 135 has the thermal capture cross-section of 8.7 barns and the cumulative yield of neutron -induced fission of ^{235}U about 6.5%. The nuclide number density of ^{133}Cs is spent fuel is higher than that of ^{135}Cs. Therefore, when ^{135}Cs is to be transmuted in PWR we should separate ^{133}Cs from ^{135}Cs. Without isotopic separation, many neutrons will be absorbed by (n, γ) reaction of ^{133}Cs and by consecutive (n, γ) reactions more ^{135}Cs will be produced rather than destroying it.

2.2 Thermodynamic properties of Long-lived Fission Products

Technetium is a transition metal, which is rather stable in its elemental form. Thus pure metallic technetium is a good choice as a target material for transmutation.

The simplest compound for iodine transmutation is elemental iodine. Elemental Iodine (I_2) has a high iodine density, which is favorable for transmutation purposes. However, iodine has a very low melting point (387 K) and, in addition, it is a very corrosive material. Therefore, elemental iodine may not favorable as a target for iodine transmutation. Then binary compounds of iodine are considered for transmutation targets since they have high melting point and less corrosive. Among them are CeI_3, MgI_2, ZnI_2, YI_3, CaI_2, PbI_2, NaI. CeI_3 and YI_3 have a relatively low thermal neutron capture cross-section and high thermodynamic stability, but are difficult to obtain in pure form. PbI_2 and ZnI_2 are the least stable in terms of thermal dissociation but relatively easy to be handled. NaI, CaI_2, and MgI_2 have intermediate characteristics, but they are easier for handling than CeI_3 and YI_3 [12].

Pure elemental cesium has low melting point (302 K) like iodine. Only ternary and quaternary compounds have reliable thermodynamic stability. Cs_2CrO_4, Cs_3CrO_4, and Cs_2SO_4 have relatively high melting points (1223-1273 K) and high Cs mass fraction (>70%). $CsAlSi_2O_6$ and $CsAlSiO_4$ have low Cs mass fraction (<55%) but higher melting points (1873-1973 K)[12].

In the present paper, only pure substances (isotopes or elements) were employed since this scenario is the best fit with the equilibrium fuel cycle requirement of a continuous refueling process and the material targets selection is not the aim of this study. The other reason for employing only pure substances is because the present study is a general introductory study.

3. Methodology

Two characteristics, namely: required uranium enrichment for criticality and the required amount of natural uranium supply for the LLFP transmutation in the equilibrium fuel cycles of PWR are evaluated in the present paper. In this study, the following fuel cycles are investigated, where in standard condition all FP and final products of heavy metals (HM) natural decay chain

(tellurium – francium) are discharged from the reactor at a standard rate (33%/year)[4]:

Case 1: All HM are discharged from the reactor with the standard rate.

Case 2: All HM except plutonium are discharged from the reactor with the standard rate. Plutonium is discharged at the rate of one-half of the standard rate.

Case 3: All HM except plutonium are discharged from the reactor with the standard rate. Plutonium is confined in the reactor.

Case 4: All HM except uranium are confined in the reactor. Uranium is discharged from the reactor with the standard rate.

In the nuclear equilibrium-fuel cycle the number density of each nuclide in reactor is consider to be constant and refueling process is performed continuously. The governing burnup equation of the equilibrium-fuel cycle can be expressed by the following equation[6,7]:

$$\frac{dn_i}{dt} = -(\lambda_i + \phi\sigma_{a,i} + r_i)n_i + \sum_j \lambda_{j\to i} n_j + \phi\sum_j \sigma_{j\to i} n_j + s_i = 0 \quad , \qquad (1)$$

where n_i : number density of i-th nuclide
ϕ : neutron flux,
λ_i : decay constant of *i*-th nuclide,
r_i : discharge constant of *i*-th nuclide,
$\lambda_{j\to i}$: decay constant of *j*-th nuclide to produce *i*-th nuclide,
$\sigma_{j\to i}$: microscopic transmutation cross-section of *j*-th nuclide to produce *i*-th nuclide,
s_i : supply rate of *i*-th nuclide,
σ_{aj}: microscopic absorption cross-section of *i*-th nuclide.

Here, fission cross-section, capture cross-section, and other nuclear transmutation cross-sections, such as (n, 2n) and (n, 3n) cross-sections are all included in the absorption cross-section.

The selection whether some certain HM and LLFP will be recycled in or taken out from the core is done by setting the discharged constant of those nuclides to be zero or 1.046×10^{-8} (0.33/year), respectively.

The generated fission products can be estimated by using $\sigma_{j\to i}$: or $\lambda_{j\to i}$:given by the following equations:

$$\sigma_{j\to i} = \sigma_{f,j}\gamma_{j\to i} \quad , \qquad (2)$$

for neutron induced fission, and

$$\lambda_{j\to i} = \lambda_{f,j}\gamma_{s,j\to i} \qquad (3)$$

where
$\sigma_{f,j}$: microscopic fission cross-section of *j*-th nuclide,
$\gamma_{j\to i}$: yield of *i*-th nuclide from *j*-th fissile nuclide.
$\gamma_{f,j}$: spontaneous fission decay constant of *j*-th nuclide
$\gamma_{s,j\to i}$: $\gamma_{s,j\to i}$: yield of *i*-th nuclide from *j*-th fissile nuclide spontaneous fission.

In a matrix representation, the governing burnup equation can be expressed as follows[6,7]:

$$\mathbf{Mn} = \mathbf{s} \quad , \qquad (4)$$

where the elements of matrix **M** include all the transmutation parameters of all nuclides such as microscopic transmutation cross-sections and natural decay constants. The **n** and **s** are the vectors of the number density of nuclides in the reactor core and the supply rate of fuel nuclides (uranium), respectively. The one-group microscopic cross-sections for Eq. (1) are calculated by using the collision probability method (PIJ) of SRAC95[13] code with nuclear data library from JENDL-3.2.

The performances of the investigated fuel cycles are evaluated by using the nuclide importance values. The nuclide importance vectors **f** and **a** can be calculated from the following adjoint equations:

$$\mathbf{M}^t\mathbf{f} = \phi\boldsymbol{\nu}\boldsymbol{\sigma}_\mathbf{f}, \quad \mathbf{M}^t\mathbf{a} = \phi\boldsymbol{\sigma}_\mathbf{a}, \qquad (5)$$

where $\mathbf{M}^t$ is the adjoint matrix of **M**. ϕ is neutron flux. $\boldsymbol{\sigma}_\mathbf{f}$ and $\boldsymbol{\sigma}_\mathbf{a}$ are the vectors of microscopic fission cross-sections and microscopic absorption cross-sections, correspondingly. The $\boldsymbol{\nu}$ represents the number of neutrons produced in each fission reaction. We have called **f** and **a** as fission neutron importance and absorbed neutron importance, respectively. The fission neutron importance represents the number of neutrons produced from fission of one nucleus of the studied nuclide and its family members (reaction products) during its existence in the reactor, while the absorbed neutron importance represents the number of neutrons absorbed by one nucleus of the studied nuclide and its family members during its presence in the reactor[7].

By employing the nuclide importance vectors, the infinite multiplication factor, *k*, can be expressed simply as the following equation.

$$k = \frac{(\boldsymbol{\nu}\boldsymbol{\sigma}_\mathbf{f}, \mathbf{n})}{\alpha(\boldsymbol{\sigma}_\mathbf{a}, \mathbf{n})} = \frac{(\mathbf{f}, \mathbf{s})}{\alpha(\mathbf{a}, \mathbf{s})}, \qquad (6)$$

where α is a correction parameter for estimating neutron absorption of non-fuel nuclides such as coolant and structural materials. The actual calculation for *k* is performed by SRAC95 code.

The neutron leakage from the system should be evaluated for criticality judgment of the system. For current PWR, the neutron leakage is estimated about 2% of produced neutrons. Based on this fact the following condition is employed for the criticality condition in the present study.

$$k = 1.02 \equiv k_c \qquad (7)$$

The uranium enrichment to satisfy the criticality condition for each case is calculated as follows. The equilibrium burnup calculation is performed to determine the flux level and the number density of each nuclide in the fuel pellet. This calculation is coupled with SRAC95 cell calculation code in order to get the neutron spectrum and the one-group microscopic cross-sections of each investigated case. From this coupling calculation procedure we evaluate the value of the infinite

multiplication factor, *k*. If *k* equal to k_c, then we choose the initial enrichment input as the required uranium enrichment for the criticality of the investigated case. In case *k* differs from k_c, the uranium enrichment is determined by using Eq. (6) and the following ones.

$$s_{24} + s_{25} + s_{28} = 100 \ , \tag{8}$$

$$100s_{24} - 0.9937s_{25} = -0.1925 \ , \tag{9}$$

where s_x is an atomic percent of uranium isotopes (^{234}U, ^{235}U and ^{238}U) in the supplied fuel. The Eq. (6) is given by enrichment condition.

The required amount of natural uranium, S_0, which given by the following Eq. (10) is calculated by means of two different enrichment processes where the concentration of ^{235}U in the tail was chosen to be 0.3 $^{w}/_{0}$ and 0.1 $^{w}/_{0}$, respectively[6].

$$S_0 = \frac{(e_1 - e_2)}{(e_0 - e_2)} S_1 \ , \tag{10}$$

where S_1 is the amount of required enriched uranium. Here e_0, e_1, and e_2 are the ^{235}U abundance in natural uranium (0.711 $^{w}/_{0}$), the concentration of ^{235}U in the produced enriched uranium and the concentration of ^{235}U in the tail of enrichment plant, correspondingly.

Detail explanation of the calculations procedure to determine the required uranium enrichment for criticality and the required amount of natural uranium supply can be found in our previous works[7].

4. Calculated Results and Discussion

4.1 LLFP Transmutation with Isotopic Separation

The required uranium enrichment for the criticality of the system dedicated for LLFP transmutation with isotopic separation of all cases is shown in Figure 1. In this figure, *std* means the standard condition of the reactor operation without recycling of any LLFP isotope, so that there are four standard conditions according to the four investigated fuel cycles. The combination of ^{99}Tc, ^{129}I, and ^{126}Sn recycling, and ^{99}Tc, ^{129}I, ^{126}Sn, and ^{79}Se recycling are denoted by *3llfp* and *4llfp*, respectively. The *5llfp* and *6llfp* represent the transmutation of mixing of ^{99}Tc, ^{129}I, ^{126}Sn, ^{79}Se, and ^{135}Cs isotopes, and the transmutation of mixing of ^{99}Tc, ^{129}I, ^{126}Sn, ^{79}Se, ^{135}Cs, and ^{107}Pd isotopes, correspondingly. The transmutation of all seven LLFP isotopes is denoted by *7llfp*. As can be seen from Figure 1, for the transmutation of single ^{79}Se isotope with isotopic separation, no additional enrichment is required. To transmute the single ^{129}I isotope the required additional enrichment from the standard cases is only about 0.1w%. Additional enrichment from the standard case of about 0.5 w% is required for transmutation of the single ^{99}Tc isotope. For single transmutation with isotopic separation, ^{93}Zr transmutation needs the highest enrichment of loaded uranium fuel for all investigated fuel cycle cases, from 5.12w% for case1 to 4.9w% for case4. The rank of required enrichment of LLFP for single isotopes transmutation from the highest one to the lowest one is ^{93}Zr, ^{126}Sn, ^{99}Tc, ^{135}Cs, ^{107}Pd, ^{129}I, and ^{79}Se, correspondingly. For single isotope transmutations, the required enrichment for criticality of the system decrease with increasing number of confined HM.

In case of mixed isotopes transmutation, the transmutation of ^{99}Tc together with ^{129}I requires the additional enrichment about 0.5w% for the fuel cycle case 1. This required additional enrichment increases slightly for case 2 to case 4, even though the total enrichment decreases monotonically from case 1 to case 4. The *3llfp* and *4llfp* mixed isotopes transmutation give the same results on the required enrichment for criticality due to the zero additional enrichment for single ^{79}Se isotope transmutation with isotopic separation. Along the change of cases from 1 to 4, the investigated fuel cycle case 2 gives a minimum required enrichment for criticality of the *3llfp*, *4llfp*, *5llfp* and *6llfp* mixed isotopes transmutation. For the seven LLFP transmutation with isotopic separation, the required enrichment increase with increasing number of confined HM nuclides in the reactor core, from 7.13w% for case 1 to 8.2w% for case 4.

The annual amount of the required natural uranium supply for 0.1w% tail of enrichment process of all investigated fuel cycle cases are also shown Figure 1. The annual amount of the required natural uranium supply for 0.3w% tail of enrichment process is shown in Table 4 together with the annual amount of loaded uranium fuel, the burnup, and the neutron flux. The required natural uranium supply shows quite similar trend as that of the required enrichment. Except for *5llfp*, *6llfp*, and *7llfp* mixed isotopes transmutation the annual amount of the required natural uranium supply decrease with increasing number of confined HM in the reactor. The annual amount of loaded uranium fuel decreases with increasing number of recycled HM for both the single isotope transmutation and the mixed isotopes transmutation scenarios. The burnup is inversely proportional to the annual amount of loaded uranium fuel. The neutron flux decreases with increasing number of confined HM for both the single isotope transmutation and the mixed isotopes transmutation scenarios.

The number density of LLFP, the total number density of FP, and the total number density of HM in the reactor core for the investigated fuel cycle case1 are shown in Figure 2. For the single isotope transmutation, the rank of the number density of LLFP from the highest one to the lowest one is ^{93}Zr, ^{126}Sn, ^{135}Cs, ^{99}Tc, ^{129}I, ^{107}Pd, and ^{79}Se, correspondingly. When ^{93}Zr is transmuted in PWR the number density of ^{93}Zr in the core not only dominates the total number density of FP (about 73% of the total number density of FP) but also increase the order of the total number density of FP itself to become more than 3.5 times of that of the standard one. The total number density of FP for this single ^{93}Zr isotope transmutation is even larger than that of the *4llfp* (^{99}Tc, ^{129}I, ^{126}Sn, and ^{79}Se) mixed isotopes transmutation. When ^{126}Sn is transmuted in PWR, 70 percent of the total number density of FP comes from ^{126}Sn. For the mixed isotopes transmutation, the total number density of FP in the core increases with increasing number of transmuted LLFP. By recycling the all seven LLFP the total number

density of FP in the core become 74% of the total number density of HM due to the huge amount of the number density of ^{93}Zr in the core. As a matter of perspective, the total number density of FP in the standard (*std*) one is less than 10% of the total number density of HM. The other investigated fuel cycle cases give the same tendency as for case1, though the absolute number of the number density of each nuclide changes considerably.

The number density of selected actinides in the reactor core for the investigated fuel cycle case1 is shown in Figure 3. The number densities of actinides change considerably for both the single isotope transmutation and the mixed isotopes transmutation. It should be noted that, in the present calculation the total number density of nuclides in the fuel pellet was fixed to 93% of the theoretical density of UO_2. Therefore, an increasing of the total number density of FP results in a reduction of the total number density of HM.

Figure 4 shows the neutron spectrum for the single LLFP isotope transmutation and for the mixed isotopes transmutation of the fuel cycle case1 with isotopic separation. Apparently, for the single LLFP isotope transmutation, the change of the neutron spectrum did not have the homogeneous pattern. The single ^{93}Zr or ^{126}Sn isotope transmutation result in the softening of the neutron spectrum compared to standard case. The single ^{79}Se isotope transmutation results in no significant change of the neutron spectrum. The neutron spectrum hardening was observed on the other single isotope transmutation. The neutron spectrum hardening also happens on the mixed isotopes transmutation, but the order of hardening seems interesting since apparently they have no regular pattern. The most hardening neutron spectrum is for the *6llfp* mixed isotopes transmutation. The *3llfp* and *4llfp* mixed isotopes transmutations produce similar neutron spectrum each other. The order of the hardening of the neutron spectrum from the softest one to the hardest one is *std*, *3llfp* and *4llfp*, *7llfp*, *5llfp*, *Tc-I, and 6llfp,* correspondingly.

The one-group absorption cross-section of the seven LLFP for fuel cycle case1 is shown in Figure 5. In general, the one-group absorption cross-section each LLFP decreases when it is transmuted in the reactor. The one-group absorption cross-section of ^{93}Zr decreases to become less than 45% of that of the *std* case during recycled in the reactor. When ^{107}Pd is transmuted in the reactor, its one-group absorption cross-section becomes 73% of that of the *std* case. There is no considerable change of the one-group absorption cross-section of ^{129}I when it is transmuted in the reactor. In case of ^{79}Se and ^{126}Sn, there are no absorption cross-section changes since these two nuclides are not included in the fission products chains of SRAC95 code. In other words, the same one-group absorption cross-sections of ^{79}Se (3.73b) and ^{126}Sn (0.015b) have been employed for all investigated cases.

The absorbed neutron importance is the measure to evaluate the strength of neutron absorption of the recycled LLFP. Figure 6 shows the absorbed neutron importance of the seven LLFP for the investigated fuel cycle case1. Except for ^{107}Pd, the absorbed neutron importance of each LLFP when it is confined in PWR becomes about unity. The absorbed neutron importance of ^{107}Pd becomes 1.3 when it is confined in PWR. The study about the absorbed neutron importance the confinement of LLFP in the all actinides confinement fuel cycle has been carried out by one of the authors of the present paper in reference[14]. The present paper shows the similar tendency on the absorbed neutron importance of confined LLFP compared with that of Ref. 14.

4.2 LLFP Transmutation with Elemental Separation

The required uranium enrichment for the criticality of the reactor as a function of the recycled LLFP with elemental separation is shown in Figure 7. As predicted, the isotopic separation process is not necessity to transmute the single ^{99}Tc isotope since the transmutation with isotopic separation and elemental separation require the exactly same additional enrichment that is only about 0.5 w% of additional enrichment from the standard condition of all investigated fuel cycles. The single isotope transmutation of ^{129} I may be possible without isotopic separation process. For single isotope transmutations with elemental separation (later: single element transmutation), except for ^{107}Pd and ^{135}Cs, the required enrichment decrease with increasing number of confined HM in the reactor. In case of Pd, the required enrichment increases with increasing number of confined HM in the reactor. The investigated fuel cycle Case 2 gives a minimum requirement of the uranium enrichment for the single element transmutation of Cs. According to Figure 7, Cs and Pd have considerable influences on the increment of the required enrichment with increasing number of confined HM in the reactor for the *5llfp* and *6llfp* mixed isotopes transmutations with elemental separation (later mixed elements transmutation). The order of required enrichment for LLFP transmutations with elemental separation from the highest one to the lowest one is *6llfp*, *5llfp*, ^{107}Pd, *4llfp*, ^{135}Cs, *3llfp*, ^{126}Sn, ^{79}Se, *Tc-I*, ^{99}Tc, and ^{129}I, respectively. The single element transmutation of Zr and the *7llfp* mixed elements transmutation are not shown in Figure 7, since ^{93}Zr transmutation with elemental separation may not possible in PWR. The detail explanation for this fact will be given later.

Figure 7 also shows the annual amount of the required natural uranium supply for 0.1w% tail of enrichment process of all investigated fuel cycle cases. The annual amount of the required natural uranium supply for 0.3w% tail of enrichment process is shown in Table 5 together with the annual amount of loaded uranium fuel, the burnup, and the neutron flux. For the single isotope transmutation, the required natural uranium supply shows the same trend as that of the required enrichment (except for Cesium). For the mixed isotope transmutation, only the required natural uranium supply for *Tc-I* and *6llfp* show the same trend as that of their required enrichment. The annual amount of loaded enriched uranium fuel and the neutron flux decrease with increasing number of confined HM for both the single element transmutations and the mixed elements transmutations scenarios. By

contrast, the burnup increases with increasing number of confined HM in the reactor.

The number density of LLFP and their associated stable isotopes members, the total number density of FP, and the total number density of HM in the reactor core for the investigated fuel cycle case1 are shown in Figure 8. From results in Figure 8 it obvious that the associated stable isotopes members of LLFP play an important role in increasing the total number density of FP in the core, even more dominant than LLFP itself. For single element transmutation of Se, the number density of ^{82}Se in the core becomes 200 times of the number density of ^{79}Se and occupies 47% of the total number density of FP. The single element transmutation of Pd results in an increasing of the nuclide number density of ^{106}Pd to become one order larger than that of ^{107}Pd and dominates about 58% of the total number density of FP. This number density of ^{106}Pd even is comparable with the total number density of FP for the single element transmutation of Se. As noted for the single isotope transmutation, when single tin element is transmuted in PWR the number density of ^{126}Sn still also dominates the total number density of FP but only about 56% since other associated stable isotope such as ^{120}Sn. The total number density of FP for the single Pd element transmutation is even larger than that of the *3llfp* (Tc, I, and Sn) mixed elements transmutation. For the mixed elements transmutations, the total number density of FP in the core increases with increasing number of transmuted LLFP. The total number density of FP for the *4llfp* is comparable with the total number density of HM in the core. The total number density of FP for the *5llfp* and *6llfp* are larger than that of HM. Even worse, the total number density of FP for the *6llfp* is twice larger than that of HM. The other investigated fuel cycle cases give the same tendency as for case1, though the absolute number of the number density of each nuclide changes considerably.

The number density of selected actinides in the reactor core for the investigated fuel cycle case1 is shown in Figure 9. For both the single element transmutations and the mixed elements transmutations, the number densities of actinides change considerably. For the mixed elements transmutations, the total number density of HM in the core decreases with increasing number of transmuted LLFP, since the total number density of FP increases significantly with increasing number of transmuted LLFP.

Figure 10 shows the neutron spectrum for the single and mixed elements transmutations of LLFP for the investigated fuel cycle Case 1. In general, the change of the neutron spectrum for elemental transmutations of LLFP did not follow the pattern of the change of the neutron spectrum for isotopic transmutations one. However, the single tin element transmutation still results in the softest neutron spectrum compared to the others even with the *Std* case. The single selenium element and the *4llfp* mixed elements transmutations also result in the softening of the neutron spectrum. The neutron spectrum hardening was observed on the other elemental transmutations of LLFP. The sequence of the hardening of the neutron spectrum in a series to the hardest one is *Std*, I, *3llfp*, *Tc*, *Tc-I*, *5llfp*, *Cs*, *Pd*, *and 6llfp*, correspondingly. The results for the other fuel cycle cases are could be found in the Appendix and will be mentioned briefly. The same order of the hardening of spectrum was also observed for the fuel cycle Case 2 with elemental separation of LLFP. The elemental transmutations of LLFP for the fuel cycle Case 1 and Case 2 showed a slightly different order of neutron spectrum hardening. The order of the hardening of the neutron spectrum in a series to the hardest one for fuel cycle Case 3 is Sn, *4llfp*, Se, *3llfp*, I, *5llfp*, Tc, *Tc-I*, *Std*, *6llfp*, Cs, *and Pd*, respectively. While the sequence of the hardening of the neutron spectrum for fuel cycle Case 4 in a series from the softest one to the hardest one is Sn, *4llfp*, Se, *3llfp*, *Std*, I, *5llfp*, Tc, *Tc-I*, *6llfp*, Cs, *and Pd*, respectively. Along the change of fuel cycle cases from 1 through 4, the neutron spectrum become harder gradually due to increasing number of confined HM in reactor as was mentioned in reference[7].

The one-group absorption cross-section of selected fission products for elemental LLFP transmutation of fuel cycle Case 1 is shown in Figur 11. As was mentioned before, in case of transmutation of selenium and tin elements there are no absorption cross-section changes, since these two elements are not included in the fission products chains of SRAC95 code. In the broad view, the transmutation of any FP results in the decrement of its one-group absorption cross-section. The one-group absorption cross-section of some nuclides, such as ^{108}Pd and ^{133}Cs decrease in some extent drastically when they were recycled in the reactor. If ^{99}Tc and ^{133}Cs are transmuted in the *6llfp* condition, their one-group absorption cross-section became much smaller than that of their single element transmutation. Along the change of fuel cycle cases from 1 through 4, the one-group absorption cross-section of LLFP and their associated stable isotopes decrease gradually due to the hardening of the neutron spectrum.

Figure 12 shows the absorbed neutron importance of selected FP for the investigated fuel cycle Case 1 with LLFP elemental transmutation. The absorbed neutron importance of ^{99}Tc, ^{129}I, and ^{135}Cs become about unity when they are transmuted in PWR core. Qualitatively, the absorbed neutron importance of other LLFP and their associated stable isotopes become much larger than unity if they are recycled in PWR. Along the change of fuel cycle cases from 1 through 4, the absorbed neutron importance of LLFP and their associated stable isotopes decrease gradually.

In dealing with the transmutation of ^{93}Zr with elemental separation, some difficulties were found due to huge accumulation of stable zirconium isotopes in the core. We have tried to calculate several cases of the single elemental transmutation of Zr, and the results are summarized in Table 5. Here *Zr-unstable* means the transmutation of Zr element where, all stable isotopes are discharged from the core. The results are similar with the single ^{93}Zr isotope transmutation with isotopic separation. The four cases studied for of the single elemental

transmutation of Zr indicated by Zr-test1, Zr-test2, Zr-test3, and Zr-test4. These studied cases correspond to the initial enrichment of uranium of 10 %, 20%, 30%, and 40%, respectively. The k-infinity decreases with increasing of enrichment. The number density of stable Zr isotopes increases with increasing of enrichment, since the cumulative yield of thermal neutron induced fission of ^{235}U for these stable isotopes are very high. The total number density of these stable isotopes even become two order of magnitude larger compared with the total number density of HM in the core. In other words, the total number density of all fission products occupy huge volume fraction of fuel rod and by all means reduce the volume fraction of the uranium fuel and other actinides. In such conditions increasing the initial even worsen the criticality of the system.

5. Conclusions

The study on feasibility of the long-lived fission products (LLFP) transmutations, both with isotopic separation and elemental separation in the equilibrium fuel cycles of PWR have been conducted. The seven important LLFP, i.e., ^{79}Se, ^{93}Zr, ^{99}Tc, ^{107}Pd, ^{126}Sn, ^{129}I, and ^{135}Cs were employed.

For single transmutation with isotopic separation, ^{93}Zr transmutation needs the highest enrichment of loaded uranium fuel for all investigated fuel cycle cases, from 5.12w% for fuel cycle Case 1 to 4.9w% for Case 4.

For the seven LLFP transmutation with isotopic separation, the required enrichment increase with increasing number of confined HM nuclides in the reactor core, from 7.13w% for Case 1 to 8.2w% for Case 4.

The isotopic separation process is not necessity to transmute technetium element, and about 0.5 w% of additional enrichment from the standard condition of all investigated fuel cycles is enough. The single elemental transmutation of pure iodine may also possible. Therefore, the mixed elemental transmutation of pure technetium and iodine is possible in the equilibrium fuel cycles of PWR with slightly added enrichment (less than 1 w%).

Even though, for single ^{93}Zr isotope transmutation requires the highest enrichment, ^{99}Tc transmutation needs the largest amount of the required natural uranium supply.

The single elemental transmutation of zirconium may impossible in PWR systems since the total number density of its stable isotopes will occupy very large volume fraction of fuel rod and by all means reduce the volume fraction of the uranium fuel. Increasing the volume of fuel may overcome this problem, but its need new fuel design.

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Table 1. Basic physics characteristics of studied PWR

Thermal power Output	3000 MW
Fuel pellet average power density	280 Wcm^{-3}
Fuel pellet diameter (= inner pin diameter)	0.80 cm
Fuel pin outer diameter	0.96 cm
Pin pitch	1.18 cm
Materials	
Fuel type	Oxide
Cladding	Zircaloy-4
Coolant	Light water
Volume fraction (%)	
Fuel pellet	36
Cladding	16
Coolant	48

Table 2**.** Half-life and yields (U-235) of LLFP

Nuclide	Half-life (year)	Cumulative Yield (%)[1]	Stable Isotopes[2]
^{79}Se	6.5×10^4	0.04	^{74}Se, ^{76}Se, ^{77}Se, ^{78}Se, ^{80}Se, ^{82}Se
^{93}Zr	1.53×10^6	6.4	^{90}Zr, ^{91}Zr, ^{92}Zr, ^{94}Zr, ^{96}Zr
^{99}Tc	2.13×10^5	6.1	none
^{107}Pd	6.5×10^6	0.14	^{102}Pd, ^{104}Pd, ^{105}Pd, ^{106}Pd, ^{108}Pd, ^{110}Pd.
^{126}Sn	1.0×10^5	0.05	^{112}Sn, ^{114}Sn, ^{115}Sn, ^{116}Sn, ^{117}Sn, ^{118}Sn, ^{119}Sn, ^{120}Sn, ^{122}Sn, ^{124}Sn
^{129}I	1.57×10^7	0.72	^{127}I
^{135}Cs	3.0×10^6	6.5	^{133}Cs

[1] From JAERI 1320 Report, [2]Table of Isotopes (Lederer, C. M., et.al, 1996)

Table 3. Thermodynamic properties of LLFP

No.	LLFP Isotope	Compounds	Density (gcm^{-3})	Melting point (K)	Boiling point (K)
1	^{79}Se	pure	4.81	494	958
		SnSe	6.179	1153	…
2	^{93}Zr	pure	6.52	2125	4777
		ZrN	7.3	3225	….
3	^{99}Tc	pure	11.487	2523	4840
4	^{107}Pd	pure	12.023	1823	3213
5	^{126}Sn	pure	7.28	505	2896
		SnO_2	6.95	1903	2173*
		SnSe	6.179	1153	…
6	^{129}I	Pure, I_2	4.66	386.6	457.2
		CsI	4.51	894	1553
		NaI	3.667	933	1577
		Pb I_2	6.16	683	1105
		Ce I_3	….	1039	1673
7	^{135}Cs	pure	1.879	301.8	951.5
		CsI	4.51	894	1553
		Cs_2SO_4	4.243	1268	….

* Sublimation

Table 4. Annual loaded uranium fuel, burnup and neutron flux for isotopic separation

	Case	Std^{0}	^{79}Se	^{93}Zr	^{99}Tc	^{107}Pd	^{126}Sn	^{129}I	^{135}Cs	Tc_I	3llfp^{1}	4llfp^{2}	5llfp^{3}	6llfp^{4}	7llfp^{5}
Loaded Uranium fuel (t/y)	1	29.0	29.0	25.8	28.8	28.9	26.2	28.9	28.8	28.8	26.0	26.1	25.8	25.7	22.3
	2	28.8	28.8	25.5	28.6	28.7	25.9	28.7	28.5	28.5	25.7	25.7	25.4	25.4	21.9
	3	28.1	28.1	24.7	27.9	28.0	25.2	28.0	27.7	27.8	24.9	24.9	24.5	24.5	20.8
	4	27.6	27.5	24.2	27.4	27.4	24.7	27.5	27.1	27.3	24.3	24.3	23.9	23.8	20.2
Burnup (GWd/tHM)	1	37.8	37.8	42.4	38.0	37.9	41.8	37.9	38.1	38.1	42.1	42.1	42.5	42.7	49.1
	2	38.1	38.1	42.9	38.3	38.2	42.3	38.2	38.4	38.4	42.6	42.7	43.2	43.2	50.0
	3	39.0	39.0	44.3	39.2	39.1	43.5	39.1	39.6	39.4	44.0	44.0	44.8	44.8	52.6
	4	39.7	39.8	45.3	40.0	40.0	44.3	39.8	40.4	40.2	45.1	45.1	45.8	46.1	54.3
Required Natural U (t/y) for 0.3 w% tail	1	276.3	276.4	302.9	305.6	285.0	290.9	281.1	289.9	310.7	323.8	324.6	338.6	346.3	371.3
	2	261.4	261.4	290.7	293.0	271.9	278.0	266.4	277.7	298.4	313.4	313.0	330.3	341.8	367.6
	3	237.9	238.0	277.7	274.6	252.3	262.8	243.3	262.4	280.0	303.6	303.6	329.0	345.7	377.4
	4	222.8	222.6	270.9	262.2	239.6	253.8	228.9	253.3	268.0	296.8	296.9	329.5	347.9	387.6
Neutron Flux (x10^{14}/cm2/s)	1	3.83	3.83	3.83	3.78	3.81	3.85	3.82	3.81	3.77	3.79	3.79	3.77	3.75	3.76
	2	3.80	3.81	3.81	3.75	3.79	3.83	3.80	3.78	3.74	3.77	3.77	3.74	3.72	3.74
	3	3.73	3.73	3.74	3.68	3.71	3.76	3.72	3.70	3.67	3.70	3.70	3.67	3.65	3.67
	4	3.71	3.71	3.71	3.66	3.68	3.73	3.70	3.67	3.65	3.67	3.67	3.64	3.61	3.63

Std^{0} = standard fuel cycle without llfp recycling.

3llfp^{1} = ^{99}Tc+^{129}I+^{126}Sn recycling

4llfp^{2} = ^{99}Tc+^{129}I+^{126}Sn+^{79}Se recycling

5llfp^{3} = ^{99}Tc+^{129}I+^{126}Sn+^{79}Se+^{135}Cs recycling

6llfp^{4} = ^{99}Tc+^{129}I+^{126}Sn+^{79}Se+^{135}Cs+^{107}Pd recycling

7llfp^{5} = ^{99}Tc+^{129}I+^{126}Sn+^{79}Se+^{135}Cs+^{107}Pd+^{93}Zr recycling

Table 5. Annual loaded uranium fuel, burnup, and neutron flux for elemental separation

	Case	Std^0	^{79}Se	^{99}Tc	^{107}Pd	^{126}Sn	^{129}I	^{135}Cs	Tc_I	3llfp^1	4llfp^2	5llfp^3	6llfp^4
Loaded fuel (t/y)	1	29.0	25.9	28.8	23.7	25.2	28.9	27.7	28.7	25.0	21.8	20.2	15.9
	2	28.8	25.8	28.6	23.0	24.9	28.7	27.4	28.5	24.6	21.6	19.9	15.2
	3	28.1	25.4	27.9	21.5	24.0	27.9	26.5	27.8	23.8	21.0	19.1	13.9
	4	27.6	25.0	27.4	20.6	23.5	27.4	25.8	27.3	23.2	20.6	18.6	13.1
Burnup (GWd/t HM)	1	37.8	42.3	38.0	46.3	43.5	37.9	39.6	38.1	43.9	50.3	54.3	68.9
	2	38.1	42.5	38.3	47.6	44.0	38.2	40.0	38.4	44.5	50.6	55.0	71.9
	3	39.0	43.2	39.2	50.9	45.6	39.2	41.4	39.4	46.1	52.2	57.4	78.9
	4	39.7	43.9	40.0	53.3	46.6	40.0	42.5	40.2	47.1	53.3	59.0	83.4
Required Natural U (t/y) for 0.3 w% tail	1	276.3	293.3	305.6	433.1	295.9	284.6	372.0	314.5	333.2	346.3	443.7	587.3
	2	261.4	279.5	293.0	445.4	283.7	270.2	364.2	302.6	323.9	339.2	442.4	604.6
	3	237.9	261.9	274.6	490.1	270.6	247.2	357.7	285.1	317.6	335.3	457.0	665.6
	4	221.0	251.0	262.2	518.1	263.4	233.0	356.0	273.8	314.8	336.4	472.7	711.8
Neutron Flux (x10^{14}/cm2/s)	1	3.83	3.85	3.78	3.62	3.86	3.82	3.68	3.76	3.79	3.82	3.67	3.52
	2	3.80	3.83	3.75	3.58	3.84	3.79	3.66	3.74	3.77	3.79	3.65	3.50
	3	3.73	3.75	3.68	3.5	3.77	3.72	3.59	3.67	3.70	3.73	3.60	3.44
	4	3.71	3.73	3.66	3.46	3.74	3.69	3.57	3.64	3.67	3.70	3.56	3.40

Std^0 = standard fuel cycle without llfp recycling.

3llfp^1 = ^{99}Tc+^{129}I+^{126}Sn recycling

4llfp^2 = ^{99}Tc+^{129}I+^{126}Sn+^{79}Se recycling
5llfp^3 = ^{99}Tc+^{129}I+^{126}Sn+^{79}Se+^{135}Cs recycling
6llfp^4 = ^{99}Tc+^{129}I+^{126}Sn+^{79}Se+^{135}Cs+^{107}Pd recycling

Table 5. Calculation results for Zr elemental transmutation testing

	Zr-unstable[a]	Zr-test1	Zr-test2	Zr-test3	Zr-test4
^{235}U enrichment (w%)	5.1	9.9	19.8	29.7	39.7
k-infinity	1.020	0.261	0.246	0.234	0.224
Number density in core (Cumulative yield (%)) [b]					
^{90}Zr (5.9045)	2.74E+18	7.68E+20	8.34E+20	8.82E+20	9.21E+20
^{91}Zr (5.9187)	3.40E+19	3.98E+20	4.19E+20	4.33E+20	4.44E+20
^{92}Zr (5.9738)	4.16E+19	5.61E+21	5.81E+21	5.94E+21	6.05E+21
^{93}Zr (6.3902)	4.49E+21	5.93E+20	6.02E+20	6.07E+20	6.10E+20
^{94}Zr (6.4395)	9.09E+19	2.81E+22	2.85E+22	2.88E+22	2.90E+22
^{96}Zr (6.2641)	3.99E+19	2.34E+21	2.21E+21	2.12E+21	2.05E+21
Number density in core					
^{234}U	5.80E+18	1.90E+17	3.04E+17	3.83E+17	4.46E+17
^{235}U	4.36E+20	4.94E+18	7.71E+18	9.62E+18	1.11E+19
^{238}U	1.69E+22	9.24E+20	6.31E+20	4.49E+20	3.24E+20
Total FP	6.16E+21	3.95E+22	4.01E+22	4.05E+22	4.07E+22
Total HM	1.77E+22	9.78E+20	6.88E+20	5.07E+20	3.84E+20

[a]: the single elemental transmutation of Zr where stables isotopes are discharged from the core.

[b] : for ^{235}U chain yield induced by thermal neutrons (JAERI-M 89-204)

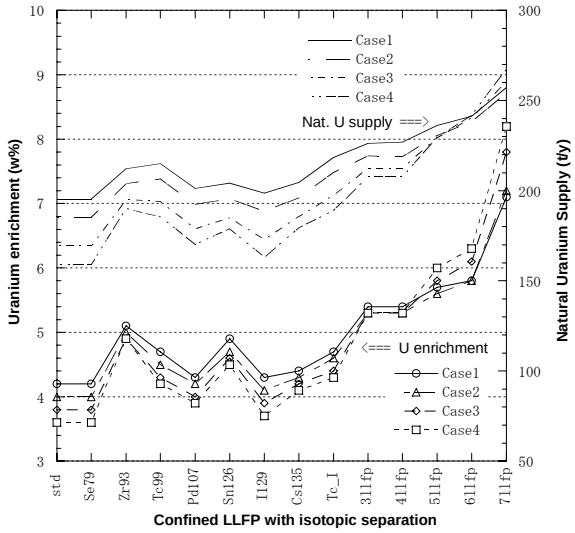

Figure 1. Required enrichment and natural uranium supply for 0.1% tail

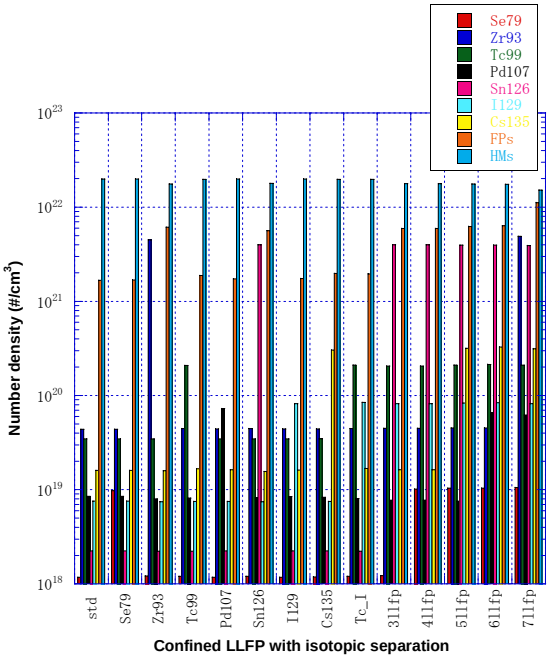

Figure 2. Number density of LLFP in reactor for fuel cycle case1

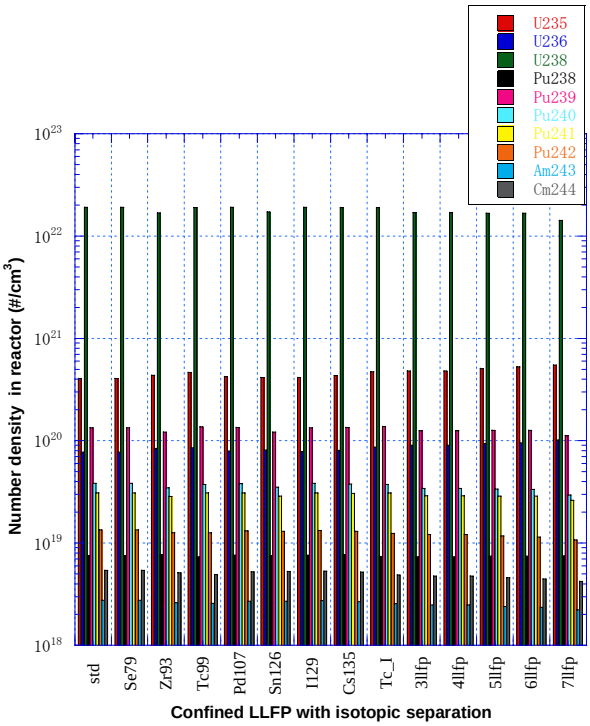

Figure 3. Number density of selected actinides in reactor for fuel cycle case1

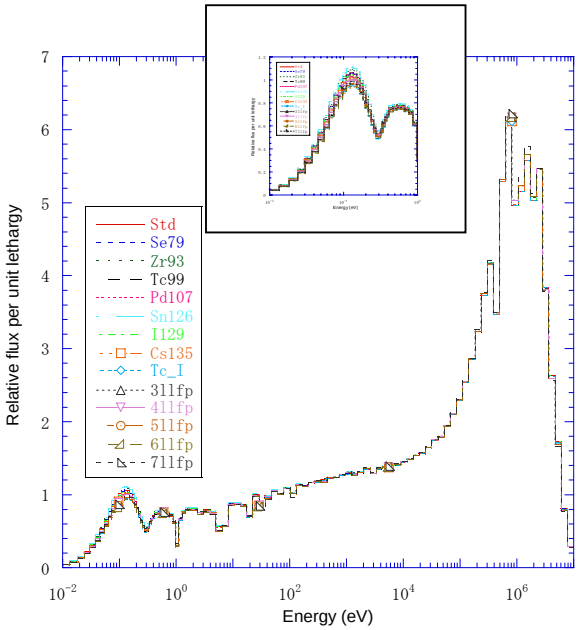

Figure 4. Neutron spectrum for LLFP transmutation of Case1 with isotopic separation

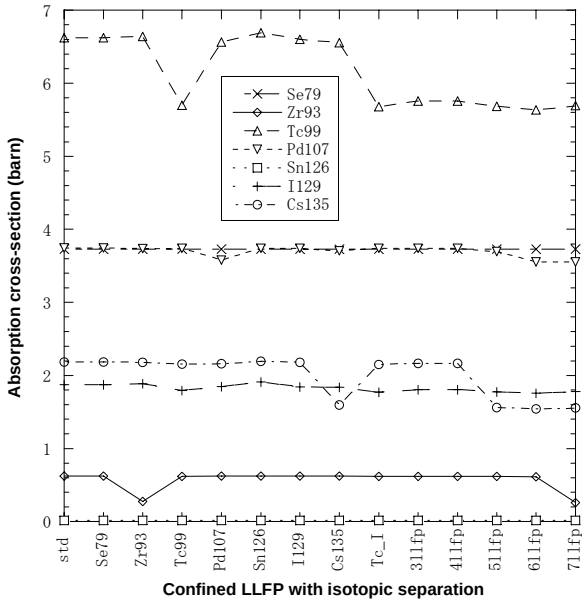

Figure 5. One-group capture cross-section of LLFP for fuel cycle case1

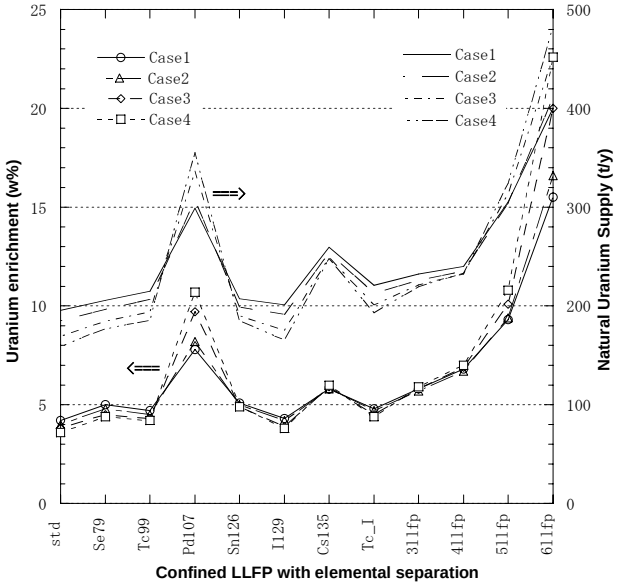

Figure 7. Required enrichment and natural uranium supply for 0.1% tail

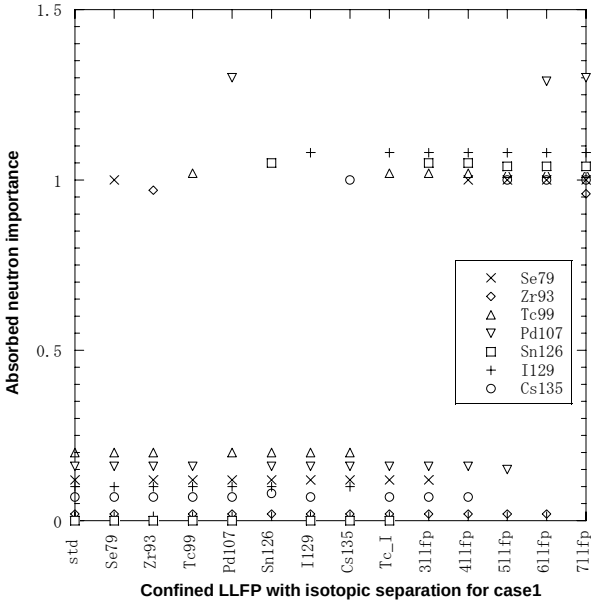

Figure 6. Absorbed neutron importance of LLFP for fuel cycle case1

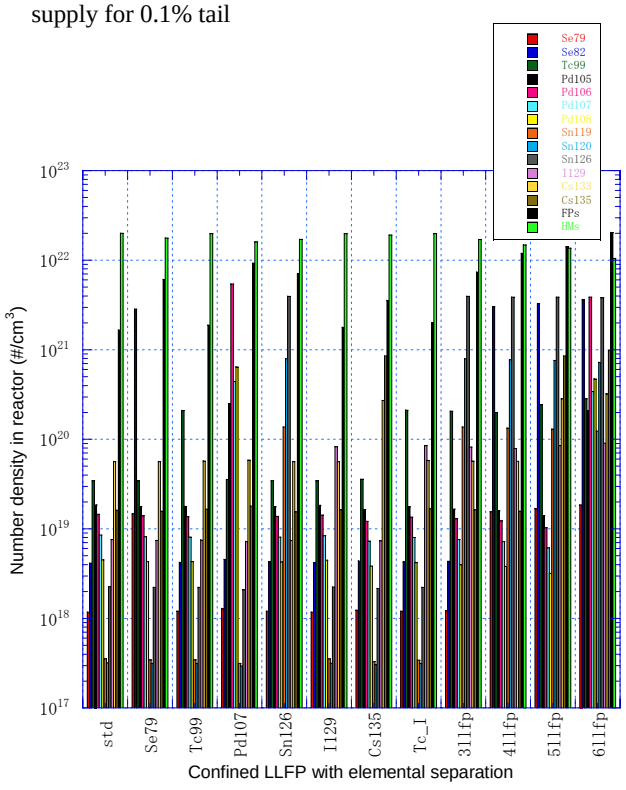

Figure 8. Number density of confined FP with elemental separation for Case 1

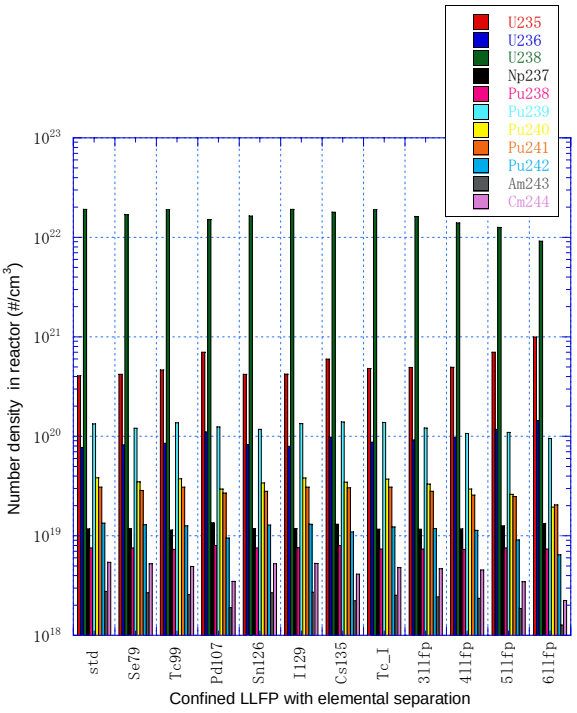

Figure 9. Number density of selected actinides for Case1 with elemental separation

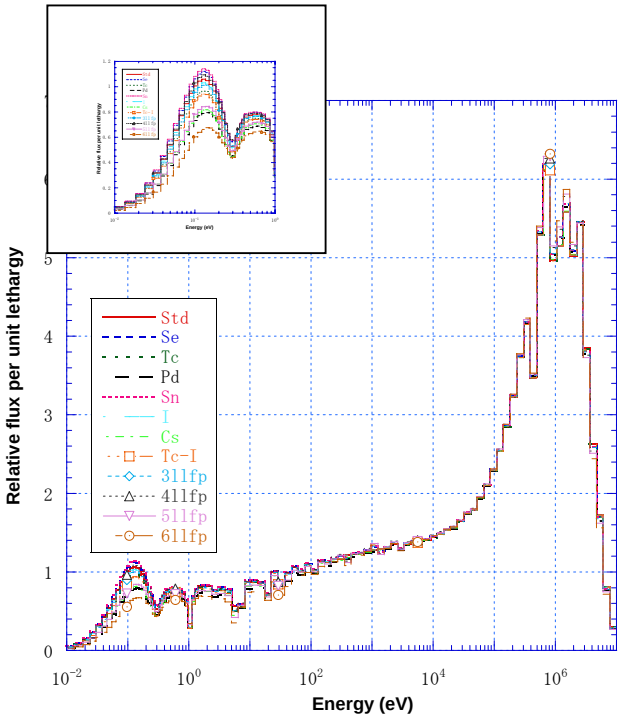

Figure 10. Neutron Spectrum for fuel cycle Case1 with elemental separation

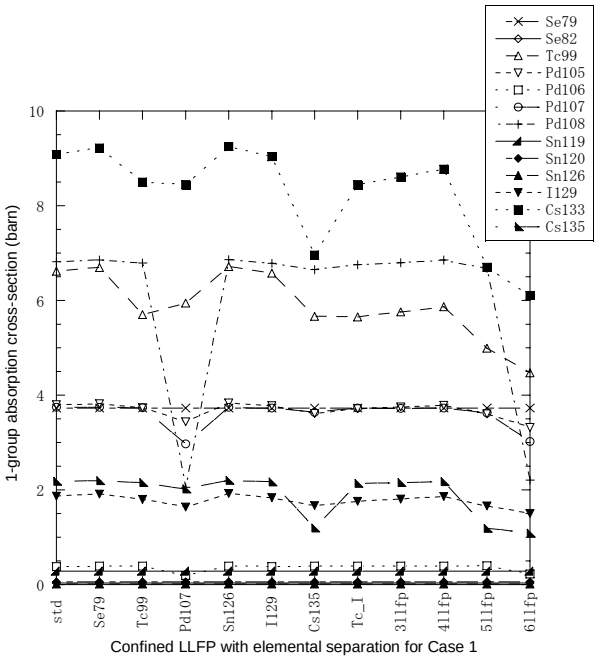

Figure 11. One-group absorption cross-section of selected FP for fuel cycle Case 1 with elemental separation

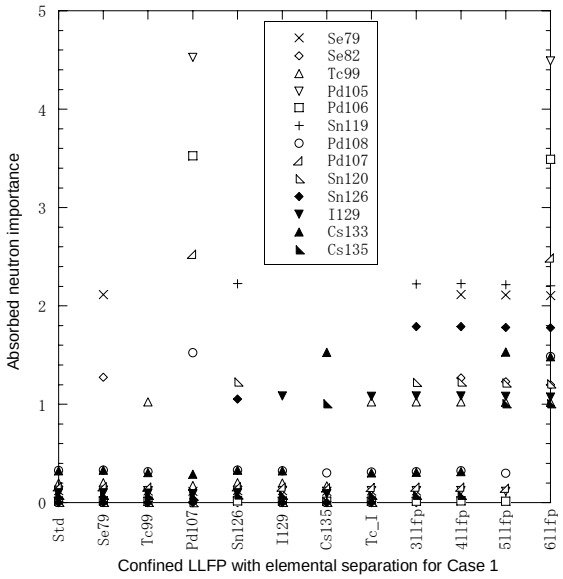

Figure 12. Absorbed neutron importance of selected FP for fuel cycle Case 1 with elemental separation