Nuclear Transmutation of Long-Lived Fission Product I-129 in Radial Blanket of Sodium-Cooled Fast Reactor

Peng Hong Liem [1,2,*], Donny Hartanto[3], Yoshihisa Tahara[1] and Naoyuki Takaki[1]

[1)] Tokyo City University (TCU),
1-28-1 Tamazutsumi, Setagaya, Tokyo, Japan
[2)] Nippon Advanced Information Service (NAIS Co., Inc.),
416 Muramatsu, Tokai-mura, Naka-gun Ibaraki, Japan
[3)] University of Sharjah,
P.O. BOX 27272, Sharjah, United Arab Emirates
*) Corresponding author (liemph@nais.ne.jp)

Abstract

An investigation on the nuclear transmutation of elemental long-lived fission product (LLFP) in a fast reactor is being conducted focusing on the I-129 LLFP (half-life 15.7 million years) to reduce the environmental burden. The LLFP assembly is loaded into the radial blanket region of a Japanese MONJU class sodium-cooled fast reactor (710 MWth, 148 days/cycle). The iodine element containing I-129 LLFP (without isotope separation) is mixed with YD_2 and/or YH_2 moderator material to enhance the nuclear transmutation rate. We studied the optimal moderator volume fraction to maximize the transmutation rate (TR, %/year) and the support factor (SF is defined as the ratio of transmuted to produced LLFP). We also investigate the effect of LLFP assembly loading against the fast reactor core characteristics and severe power peak appeared at the fuel assembly adjacent to the LLFP assembly.

Keywords

Nuclear transmutation, long-lived fission product, I-129, radial blanket, sodium-cooled fast reactor

I. INTRODUCTION

Transmutation of long-lived fission products (LLFPs: Se-79, Zr-93, Tc-99, Pd-107, I-129, and Cs-135) into short-lived or stable nuclides by fast neutron spectrum reactors is being revisited in Japan (Chiba et al., 2017). The geological disposal of high-level radioactive wastes (HLRW) as the nuclear fuel cycle by-products raises public concern in Japan and many other countries since even after thousands of years, minor actinides (MAs) and some LLFPs remain and become the main contributors of the radioactive hazard. Under the partitioning and transmutation (P&T) strategy, research and development efforts are being conducted to reduce the radioactive waste to be stored and the above-mentioned long-term hazard for future generations (IAEA, 1982). The P&T strategy in general is implemented first by elemental separation of MAs and LLFPs from HLRW using chemical processes (partitioning) and then followed by nuclear reactions of the MAs and LLFPs into short-lived or stable nuclides (transmutation). The transmutation can be done in a nuclear reactor or in an accelerator-driven transmutation system (ADS).

In this present study, we focus on the transmutation of one LLFP, i.e. I-129 (half-life 15.7 million years) using a sodium-cooled fast reactor, in particular in the radial blanket region.

I-129 is selected in the present study since the chemical compound and manufacturing of this LLFP have been previously investigated (Osaka et al., 2007). The investigation concluded that BaI_2 is the suitable chemical compound for this particular LLFP from the points of view of chemical stability and easy manufacturing process. As mentioned before, the iodine from the partitioning process does not experience isotope separation process that is the LLFP iodine element consists of approximately 76 at % of I-129 and the rest I-127, if we assume that the LLFP iodine originated from a spent mixed oxide (MOX) fuel of a fast breeder reactor core at 80 GWd/t (two years of irradiation). The I-129 LLFP irradiated in a sodium-cooled fast reactor will transmute into I-130 by neutron capture reaction and then decay (half-life 15.5 second) into stable Xe-130.

A sodium-cooled fast reactor is selected in the present study because of its high excess neutrons and high neutron flux available for the transmutation. Radial blanket regions of a fast reactor can be used for the proposed LLFP transmutation since they provide large space (volume) while the core characteristics of the fast reactor are not significantly affected. One important fast reactor characteristic which is expected to be affected is the reactor breeding capability. By replacing radial blanket sub-assemblies (S/As) containing depleted uranium with LLFP S/As containing BaI_2, the reactor breeding capability will be lower. However, for Japan, the accumulated surplus of plutonium may be one argument for not pursuing a high breeding ratio. Although Japan has decided to close

MONJU fast reactor in December 2016, scientific knowledge, design, engineering and technical experiences as well as high quality measurement data are well accumulated and documented. Therefore, in this study we use MONJU as the fast reactor system for the LLFP transmutation (readers are referred to JNC TN 4520 2004-001 document for detail descriptions of MONJU reactor). Findings and conclusions from this study should be also valid in other similar fast reactor systems.

II. TRANSMUTATION OF I-129 LLFP

The MONJU reactor core layout for the proposed I-129 LLFP transmutation is shown in Figure 1. The reactor has a nominal power of 710 MWth and under this nominal power it is typically operated in 148 days per core cycle. From Figure 1, one can observe that the reactor core consists of inner and outer core regions, and the core is surrounded by a radial blanket region. The radial blanket region is occupied by three layers of S/As.

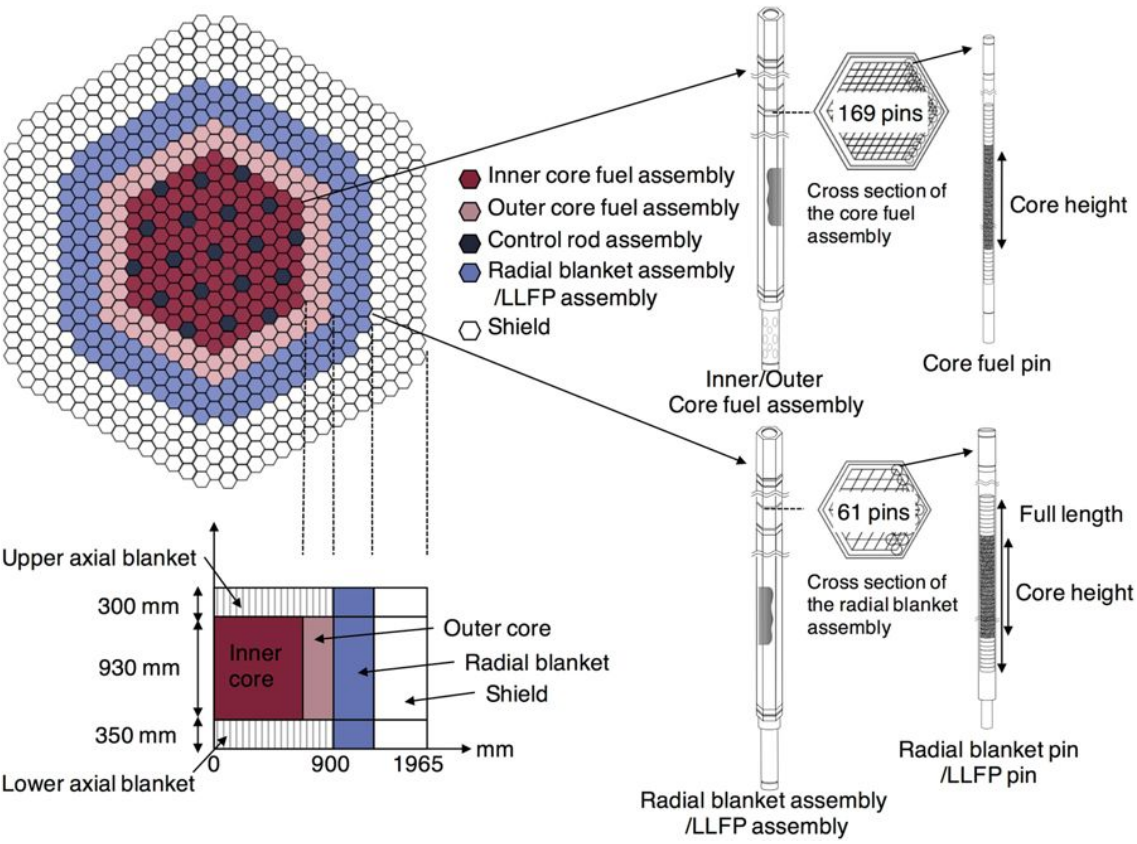

Figure 1. Japanese MONJU reactor configuration and dimension (left), and fuel and radial blanket S/A (right). A radial blanket S/A may be replaced by the same dimension of a LLFP S/A.

It is obvious that the first layer of the radial blanket will have the highest neutron flux and the hardest neutron spectrum. However, since the mean free path of fast neutrons is comparable to the radial blanket S/A dimension, still moderately high neutron flux can be expected in the second and the last layers. In this study, the performance of I-129 transmutation is investigated by loading the I-129 LLFP S/As in the first, second or third layers, as well as in several combinations of layers.

The effectiveness of LLFP (including I-129) nuclides depends on both the neutron flux level and LLFP capture cross sections, namely the product of the two parameters. A sodium-cooled fast reactor can provide a high neutron flux level however in general the LLFP neutron capture cross sections are low in fast spectrum regimes as shown in Figure 2. For I-129, the neutron capture cross section shows 1/v behavior in the thermal energy region, followed by many resolved resonances from several tens eV up to several kilos eV. The neutron capture cross section at 0.0253 eV and resonance integral of I-129 are approximately 30 and 33 barns (Shibata et al., 2011). These data suggest that softer neutron spectra than the one of a typical sodium-cooled fast reactor will enhance the neutron capture and increase the transmutation rate of I-129.

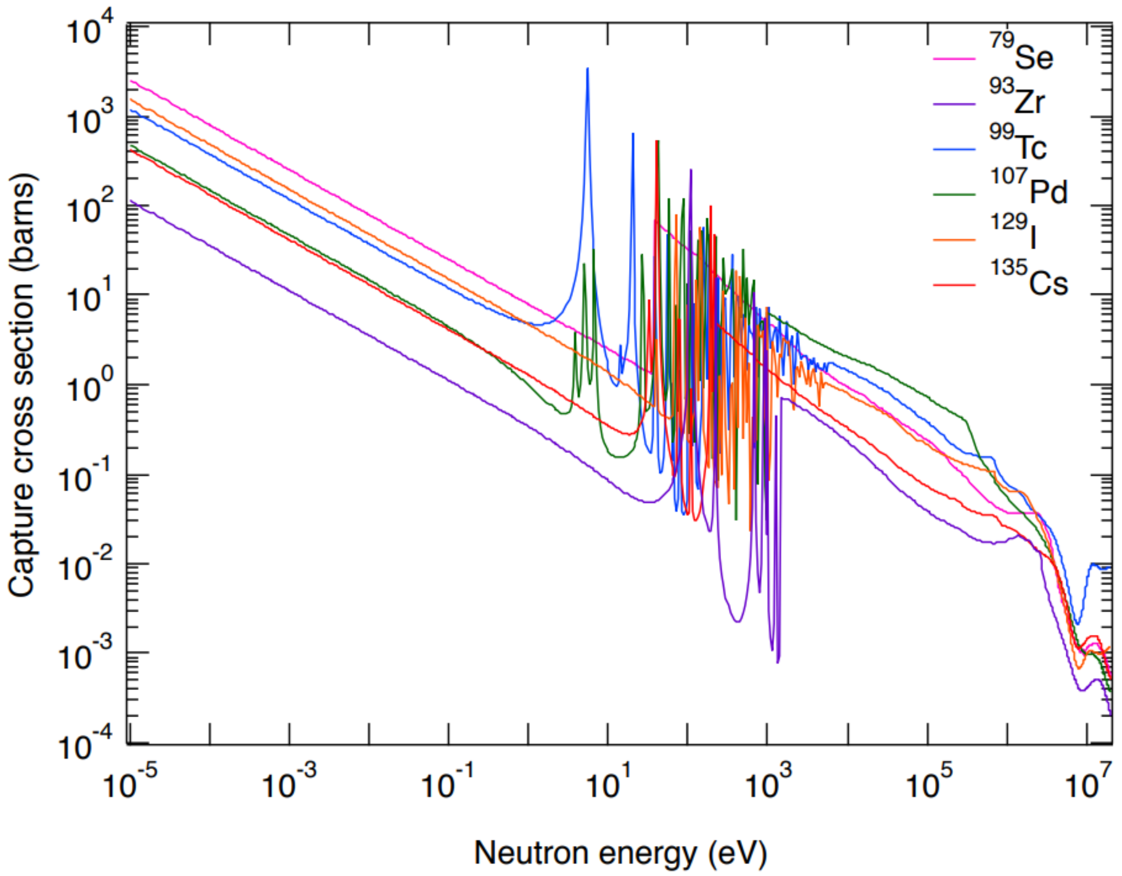

Figure 2. Important LLFP nuclides capture cross sections (JENDL-4.0).

In the present study, yttrium deuteride (YD_2) and yttrium hydride (YH_2) moderator materials are adopted for softening the neutron spectrum in the LLFP S/A. Yttrium is superior due to its low absorption cross section and does not produce long-lived daughters during irradiation. For enhancing the I-129 transmutation, the volume fraction between BaI_2 and moderator material will be optimized. The use of a moderator material in the LLFP S/A is expected to increase the power peaking factor (PPF) of fuel pins adjacent to the LLFP S/A. Therefore, in the present study we estimate the PPF of the fuel pins of the most outer fuel S/As in the outer core region.

To evaluate quantitatively the I-129 transmutation effectiveness in a fast reactor, the following performance indices are defined:

$$\text{Transmutation Rate (TR)} \equiv \frac{\mathrm{N(0) - N(T)}}{\mathrm{T \times N(0)}} \tag{1}$$

$$\text{Support Factor (SF)} \equiv \frac{\mathrm{N(0) - N(T)}}{\gamma \times \mathrm{F \times T}} \tag{2}$$

where N(0) and T are the number of initial atoms of I-129 in a LLFP S/A and the irradiation period, respectively, while γ and F are the I-129 yield per fission and the total fission rate of the core, respectively. From the definition, it is clear that TR is the ratio of the initially loaded I-129 to the amount of the transmuted I-129 in the LLFP S/A per unit time. On the other hand, SF indicates the ratio of the amount of transmuted I-129 to the amount of I-129 produced in the core over the same period of time.

As mentioned above, TR is strongly dependent on the product of I-129 effective microscopic capture cross section and the neutron flux level. The performance index SR is needed because during the I-129 transmutation in the blanket region, the reactor core also produces I-129. If the condition of SR>1.0 can be achieved, then the self-produced I-129 can be transmuted (eliminated) during electric power generation.

III. ANALYSIS AND RESULTS

Analytical Tools, Library and Calculation Model

The continuous energy Monte Carlo SERPENT2 code (Leppänen et al., 2015) with JENDL-4.0 library (Shibata et al., 2011) was used for the neutronics and burnup analysis. The MONJU reactor was modeled in 2-D R-Z geometry to reduce the overall calculation time. In the axial (Z) direction, a fuel S/A was divided into many zones with the height of approximately 5 cm for burnup calculation. The LLFP S/A was modeled as a homogeneous mixture of BaI_2 and moderator material. For each predictor-corrector burnup step, the effective and skip numbers of generations (10,000 histories per generation) were 100 and 20, respectively. The core characteristics and transmutation performance were evaluated at the beginning of cycle (BOC, 0 day), middle of cycle (MOC, 74 days) and end of cycle (EOC, 148 days). The reactor is assumed to be operated at its nominal power (710 MWth) with all control rods fully withdrawn.

Analysis Results and Discussions

The performance of I-129 transmutation in the radial blanket region of MONJU reactor is summarized in Tables 1 and 2 for YD_2 and YH_2 moderator material, respectively. The I-129 LLFP S/As are loaded into the first, second and third layer of the radial blanket region, or the combination of them. The BaI_2 volume fraction is varied from 100 % volume (no moderator material) to 10 %.

First the I-129 transmutation performance of LLFP S/A with YD_2 moderator material is discussed. For cases where all three layers are loaded with LLFP S/As, increasing the moderator volume fraction (or equivalently decreasing the BaI_2 volume fraction) softens the neutron spectrum and enhances the TR values, but decreases the SF values. When a single LLFP S/A loaded location is varied from layer 1, 2 and 3, both the TR and SF values decrease because of lower neutron flux levels. Similar trends are observed for varied location of a pair of LLFP S/As. The largest TR value (3.83) is found for a pair of LLFP S/As located in the first and second layers with BaI_2 volume fraction of 10 %, while the largest SF value (14.0) is found for the case of LLFP S/As loaded in all layers with BaI_2 volume fraction of 70 %. Concerning the PPF, severe PPF values appear if a LLFP S/A is located in the first layer, in particular for cases with a high volume fraction of moderator material. For the case with the highest volume fraction of moderator material (90 %), the PPF is approximately 1.50.

Secondly the transmutation performance of LLFP S/A with YH_2 moderator material is discussed. Compared to deuterium, hydrogen mass is half therefore it is expected that with the same nuclide density hydrogen moderates significantly better than the deuterium does. However, at the same time the use of hydrogen is expected to increase the PPF value. From Table 2 one can observe many similar trends appeared in the YD_2 cases related to moderator material volume fraction, LLFP S/A location etc. Nevertheless, severe PPF values already appear at a low moderator material volume fraction of 30 %. The largest TR value (5.12) is found for a pair of LLFP S/As located in the first and second layer with volume fraction of 10 %, while the largest SF value (17.4) is found for the case of

LLFP S/As loaded in all layers with BaI_2 volume fraction of 70 %. These largest TR and SF values of YH_2 are higher than the ones of YD_2, with the penalty of severer PPF values.

One possible solution for decreasing the high PPF value is by using a combination of YD_2 and YH_2 moderator materials as shown in the last case of Table 2. In this particular case the moderator material volume fraction is fixed to 30 % for all layers but the first layer uses YD_2 instead of YH_2. The TR and SF values decrease slightly from 1.46 to 1.37 and 17.4 to 16.3, respectively, but the PPF can be improved from 1.41 to 0.96.

Table 1. Performance of I-129 transmutation in LLFP S/A using YD_2 moderator material.

BaI_2 [%v]	**Mod [%v]**	**Layer 1**	**Layer 2**	**Layer 3**	**TR [%/y]**	**SF**	**PPF**
100	0	YD_2	YD_2	YD_2	0.74	12.6	0.89
70	30	YD_2	YD_2	YD_2	1.17	14.0	0.93
40	60	YD_2	YD_2	YD_2	1.77	12.1	1.13
10	90	YD_2	YD_2	YD_2	3.37	5.8	1.50
70	30	YD_2			1.60	6.0	0.94
			YD_2		1.04	4.4	
				YD_2	0.68	2.9	
		YD_2	YD_2		1.36	10.7	0.94
			YD_2	YD_2	0.91	7.6	
40	60	YD_2			2.32	5.0	1.10
			YD_2		1.47	3.5	
				YD_2	0.97	2.3	
		YD_2	YD_2		2.06	9.3	1.13
			YD_2	YD_2	1.35	6.5	
10	90	YD_2			3.80	2.0	1.37
			YD_2		2.38	1.4	
				YD_2	1.56	0.9	
		YD_2	YD_2		3.83	4.3	1.50
			YD_2	YD_2	2.43	2.9	

Table 2. Performance of I-129 transmutation in LLFP S/A using YH_2 moderator material.

BaI_2 [%v]	Mod [%v]	Layer 1	Layer 2	Layer 3	TR [%/y]	SF	PPF
100	0	YH_2	YH_2	YH_2	0.74	12.6	0.89
70	30	YH_2	YH_2	YH_2	1.46	17.4	1.41
40	60	YH_2	YH_2	YH_2	2.26	15.4	2.03
10	90	YH_2	YH_2	YH_2	3.58	6.1	2.74
70	30	YH_2			2.44	9.2	1.55
			YH_2		1.55	6.5	
				YH_2	0.96	4.1	
		YH_2	YH_2		1.90	15.0	1.43
			YH_2	YH_2	1.20	10.0	
40	60	YH_2			4.38	9.5	2.18
			YH_2		2.68	6.5	
				YH_2	1.57	3.8	
		YH_2	YH_2		3.16	14.2	2.04
			YH_2	YH_2	1.89	9.1	
10	90	YH_2			7.44	4.0	2.84
			YH_2		4.35	2.7	
				YH_2	2.44	1.5	
		YH_2	YH_2		5.12	5.8	2.74
			YH_2	YH_2	2.92	3.5	
70	30	YD_2	YH_2	YH_2	1.37	16.3	0.96

In order to have a deeper insight of the above mentioned results, the effective microscopic cross sections of I-129, neutron flux levels and I-129 neutron capture reaction rates of LLFP S/A are processed and shown in Figures 3, 4 and 5, respectively. The figures are for the case of YD_2 moderator material with 30 % volume fraction (or equivalently BaI_2 volume fraction of 70 %).

When a single LLFP S/A loaded location is varied from layer 1, 2 and 3, the effective capture cross section of I-129 increases slightly (cf. Figure 3) since the neutron spectra become softer, while as shown in Figure 4 the neutron flux levels decrease significantly. As a final result, the I-129 capture rates (proportional to the TR values) decrease. The same trends can be observed for a pair of LLFP

S/As loaded in layers 1 & 2 and 2 & 3.

The softest neutron spectrum amongst all cases listed in Tables 1 and 2 is shown in Figure 6, namely the case of YH_2 moderator material with volume fraction of 90 %. A pronounced thermal peak can be observed below 1 eV which is the reason for the high PPF values.

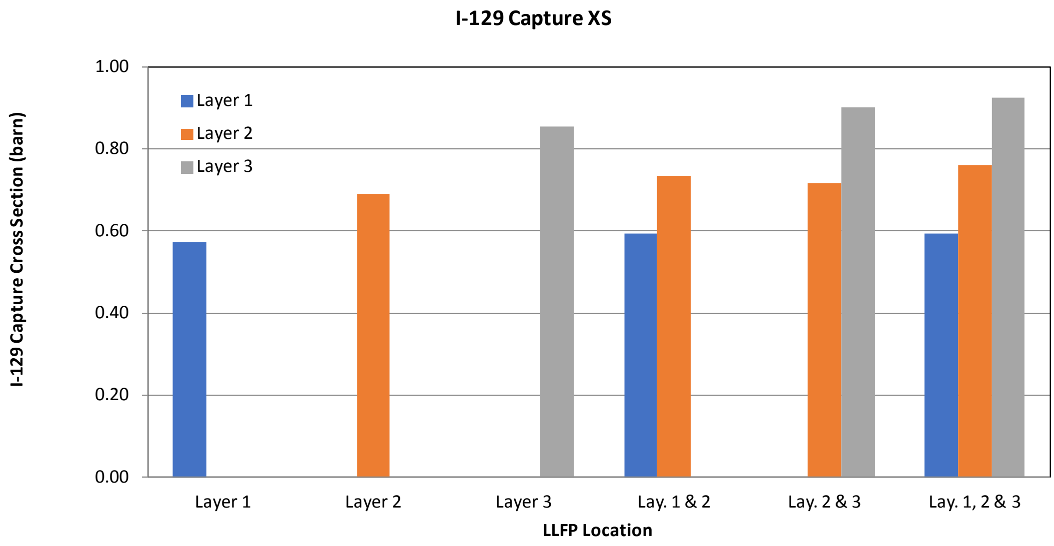

Figure 3. I-129 effective microscopic capture cross section for the case of BaI_2 volume fraction of 70 % (YD_2 moderator material).

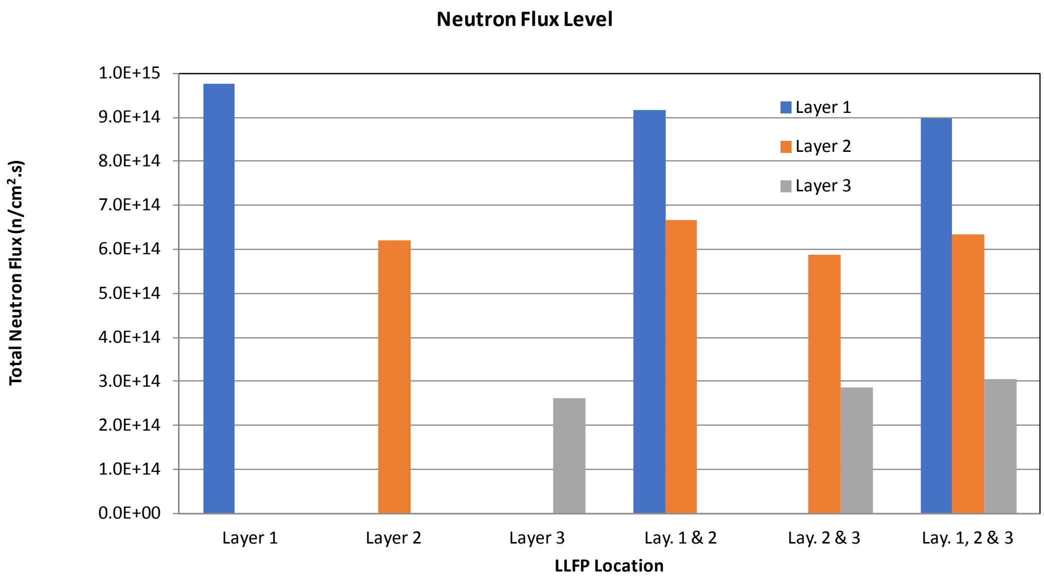

Figure 4. Total neutron flux levels for the case of BaI_2 volume fraction of 70 % (YD_2 moderator material).

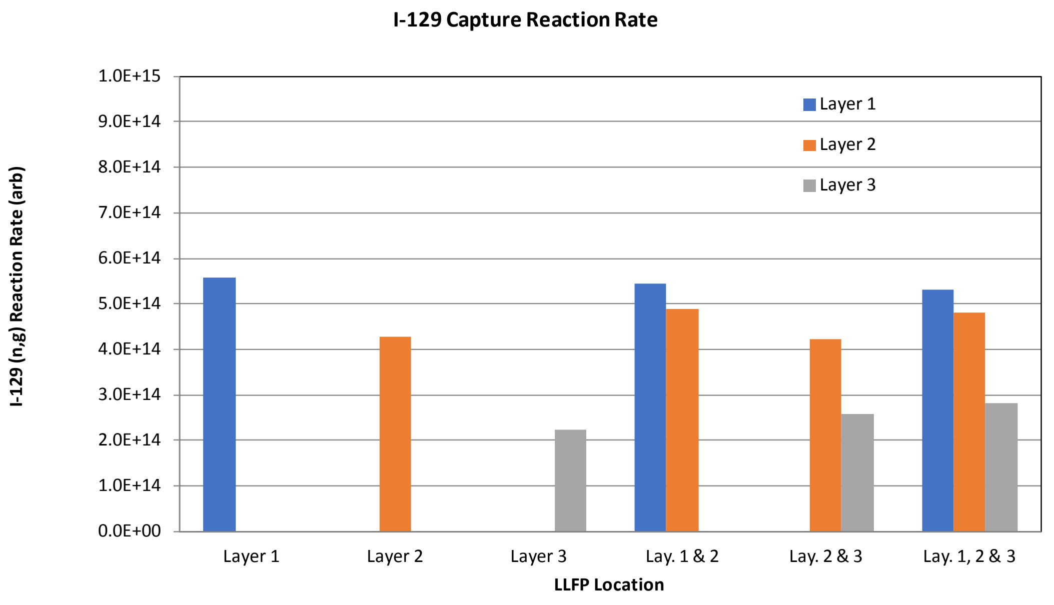

Figure 5. I-129 neutron capture reaction rates for the case of BaI_2 volume fraction of 70 % (YD_2 moderator material).

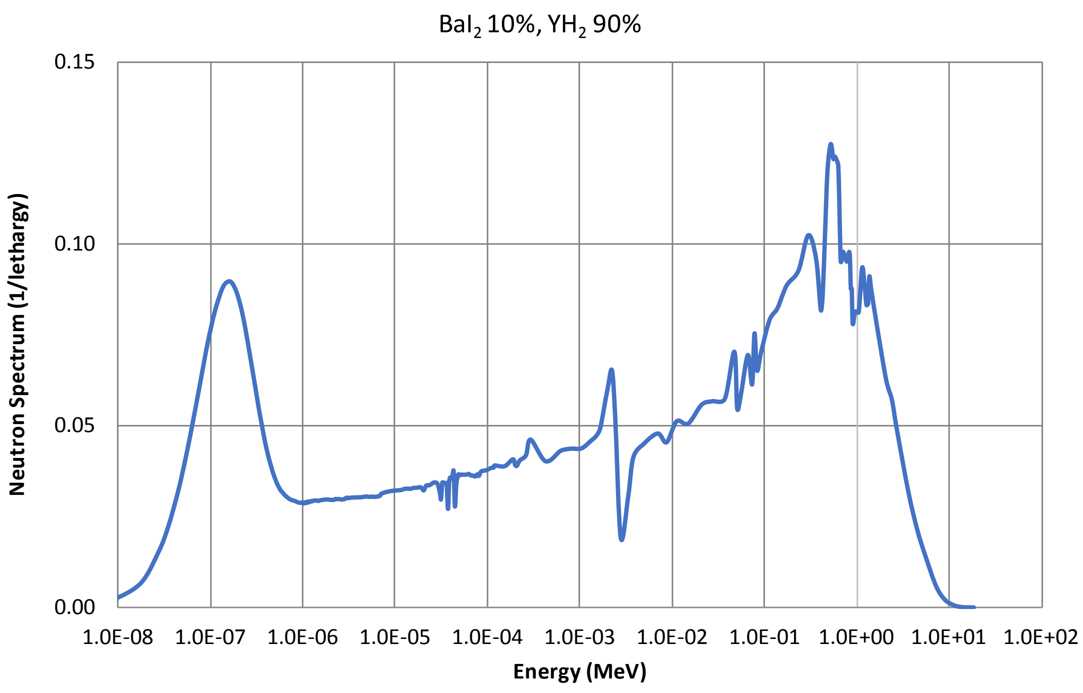

Figure 5. Neutron spectrum of the I-129 LLFP S/A loaded in the first layer for the case of BaI_2 volume fraction of 10 % (YH_2 moderator material).

IV. CONCLUDING REMARKS

The performance of elemental I-129 (chemical form BaI_2) LLFP nuclear transmutation in the radial blanket region of the Japanese MONJU fast reactor was investigated. Several important parameters such as the LLFP S/A position, moderator material type (YD_2 or YH_2) and its volume fraction are taken into account for optimizing the transmutation effectiveness guided by using two performance indices, i.e. the transmutation rate (TR, %/year) and support factor (SF). The largest TR values (3.83 and 5.12) are found for a pair of LLFP S/A loaded into the first and second layer with BaI_2 volume fraction of 10 %, while the largest SF values (14.0 and 17.4) are found for the case of 3 LLFP S/As with BaI_2 volume fraction of 70 % for YD_2 and YH_2 moderator material, respectively. YD_2 moderator material shows better PPF values and should be used in a LLFP S/A loaded in the first layer of the blanket region.

ACKNOWLEDGMENTS

The work is partly supported by a contract from Tokyo City University to NAIS Co., Inc. in the fiscal year of 2018. Tables and figures were carefully prepared by Mrs. Suzuko Ikegami.

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