Journal of Nuclear and Radiochemical Sciences, Vol. 6, No. 3, pp. 279-282, 2005

Transmutation of Technetium in the Experimental Fast Reactor “JOYO”

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Received: July 31, 2005; In Final Form: September 30, 2005

The present study examines the potential for the demonstration of fission product transmutation in the experimental fast reactor JOYO at JNC’s Oarai Engineering Center. The possibility of creating a highly-efficient transmutation irradiation field of JOYO by loading neutron moderating subassemblies in the reflector region was investigated in a series of scoping calculations. A cluster of reflector subassemblies was replaced with beryllium or zirconium hydride ($ZrH_{1.65}$) moderated subassemblies. These moderated subassemblies surrounded one central test subassembly that would contain ^{99}Tc or ^{129}I target material. With this modification, a high fast neutron flux can be tailored to epithermal or thermal energies. A comparison of reaction rates among the spectra is made illustrating the integral effects on selected target reactions. The ^{99}Tc transmutation rate achieved was 21.0% using $ZrH_{1.65}$ and 27.8% using beryllium as moderator. As a result of this study, basic characteristics of LLFP transmutation in JOYO using relevant moderator materials were clarified and the future feasibility was shown.

1. Introduction

One of the most important roles of Partitioning and Transmutation (P&T) is to reduce the practical risk ascribed to the disposed nuclear waste. Based on the current safety evaluation for a deep geologic repository, the transmutation of Long-Lived Fission Product (LLFP) could be more effective at reducing risk than minor actinide transmutation due to the higher mobility of LLFP in groundwater. In the feasibility study on commercialized fast reactor cycle systems, transmutation technology of LLFP using fast reactors has been studied.[1]

The present study examines the potential to demonstrate the transmutation of fission products such as technetium in the experimental fast reactor JOYO at JNC’s Oarai Engineering Center. ^{99}Tc and ^{129}I were considered as the target LLFP to be transmuted in this study because these nuclides have high fission yield, high dose risk, and a fast reactor is suitable for their transmutation.

2. Description of JOYO

The experimental fast reactor JOYO was constructed as the first step in sodium cooled fast reactor development in Japan. Its current missions include improvements in fast reactor safety and operation, and irradiation testing of advanced fuels and materials. The JOYO reactor attained initial criticality in 1977 with the MK-I breeder core. From 1983 to 2000, JOYO was operated at 100 MWt for thirty five operational cycles with the MK-II irradiation test core.

In 2003 the JOYO reactor upgrade to the 140 MWt MK-III core was completed to increase the irradiation testing capability of JOYO. The MK-III core[2, 3] incorporates significant changes to the core size and arrangement, fuel enrichment, and reactor power level compared to the MK-II core. This upgraded JOYO MK-III has been operated since 2003 with the primary mission of developing fuels and materials considered in the feasibility study on commercialized fast reactor cycle systems.

The main parameters of the MK-III core are tabulated in Table 1. The MK-III core specifications were determined such that the fast neutron flux would be increased and a large number of irradiation rigs could be irradiated simultaneously. The fuel region in the MK-III core is divided into two radial enrichment zones to flatten the neutron flux distribution. The active core is cylindrical and about 80 cm in equivalent diameter and 50 cm in height. There is a radial reflector region of stainless steel surrounding the core that is 25 to 30 cm thick. Shielding subassemblies with B_4C are loaded in the outer two rows of the reactor grid, replacing radial stainless steel reflector subassemblies.

TABLE 1: Main Core Parameters of JOYO MK-III

Specification			Data
Reactor Thermal Power		(MWt)	140
Maximum Number of Driver Fuel Subassemblies*			85
Equivalent Core Diameter		(cm)	80
Core Height		(cm)	50
^{235}U Enrichment		(wt%)	18
Pu Content: Pu/(Pu+U)		(wt%)	23/30**
Fissile Pu Content: (^{239}Pu+^{241}Pu)/(Pu+U)		(wt%)	16/21**
Maximum Linear Heat Rate of Fuel Pin		(W/cm)	420
Maximum Burn-up of Fuel(Pin Average)		(GWd/t)	90
Total Neutron Flux		($n/cm^2 \cdot s$)	5.7×10^{15}
Fast Neutron Flux		($n/cm^2 \cdot s$)	4.0×10^{15}
Number of Control Rod	In 3rd Row		4
	In 5th Row		2
Reflector/Shielding			SUS/B_4C
Primary Coolant Temperature (Inlet/Outlet)		(˚C)	350/500
Operation Period per Cycle		(day)	60
Operation Cycle per Year		(cycle)	5

*Including Number of Irradiation Test Fuel Subassemblies
**Inner Core /Outer Core

As a result, the fast neutron flux is up to 1.3 times higher than that of the MK-II core, with 140 MWt reactor power. Also, the irradiation test field space with a high neutron flux is approximately double that of the MK-II core.

Four neutron environments are available for experiments in the upgraded JOYO MK-III, ranging from the reactor core to the ex-vessel irradiation hole as follows.

(1) Fuel Region: High temperature and high neutron flux
(2) Reflector Region: Softer neutron spectrum and two orders lower neutron intensity than fuel region

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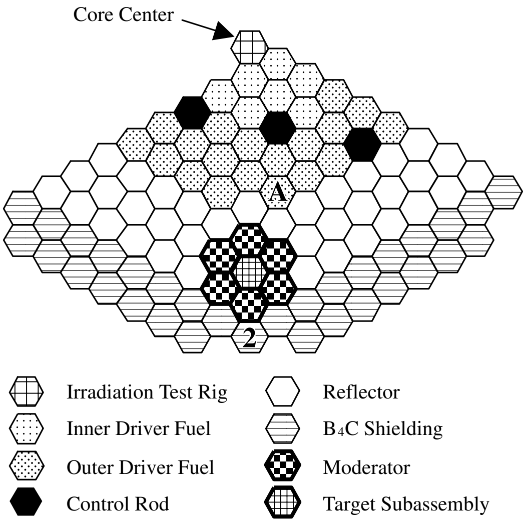

Figure 1. Target locations in JOYO core (1/3rd core model).

(3) Upper Core Region: Higher temperature and softer neutron spectrum than reflector region
(4) Ex-vessel Region: Lower energy neutron dominated

In the fuel region, the total flux is 3 to 5 × 10^{15} n/cm^2·s and the spectrum index ($\phi_{E>0.1MeV}$ /ϕ_{total}) is approximately 0.6 to 0.7. In the ex-vessel irradiation hole, the total neutron flux magnitude is about 10^{12} and the spectrum index is on the order of 0.01.

3. Neutron Flux Tailoring

3.1. Neutron Moderator and Target Subassembly. The possibility of creating a highly-efficient transmutation irradiation field by loading neutron moderating subassemblies in the reflector region was investigated in a series of scoping calculations.

Figure 1 shows a layout of the JOYO core for this study, where a cluster of reflector subassemblies was replaced with new moderator and target subassemblies. Many materials were considered for use as a fast reactor neutron moderator, including hydride materials, B_4C enriched in ^{11}B, graphite, and beryllium (Be).

Among hydride materials, the investigation indicated that $ZrH_{1.65}$ is the most promising candidate from the view point of temperature resistance and small neutron capture cross section.[1] However, hydride materials have the problem of dissociation of hydrogen, so further investigation is required.

On the other hand, beryllium has better transmutation performance than $^{11}B_4C$ and we have experience using beryllium in JOYO as a neutron source material by means of the Be (γ, n) reaction. Beryllium metal or zirconium hydride ($ZrH_{1.65}$) were preliminary selected for the moderator.

Six of the row 7, 8 and 9 reflector subassemblies were replaced with beryllium or $ZrH_{1.65}$ moderated subassemblies. The structure of a moderator subassembly is similar to the other core components. The wrapper tube is made of stainless steel (SUS). The moderator fraction ratio (moderator/(moderator + sodium + SUS)) was varied in the range from 0% to 100%. The moderator subassemblies surrounded one test subassembly that would contain ^{99}Tc or ^{129}I target material. As the purpose of this study was to investigate neutronic feasibility of the LLFP transmutation, the form of the target material was not considered.

This core configuration was based on the fact that the volume of a single subassembly is not large enough to contain sufficient moderator material. Therefore, the concept of separating moderator and target subassembly has a clear advantage for the irradiation tests. The radial reflector between the driver fuel and the moderator subassemblies works as a thermal neutron buffer that splits the thermal neutron region of the moderator subassemblies from the fast neutron region of the nearby driver fuel subassemblies. This avoids the thermal spike that may occur near the boundary between extremely different neutron spectrum regions.

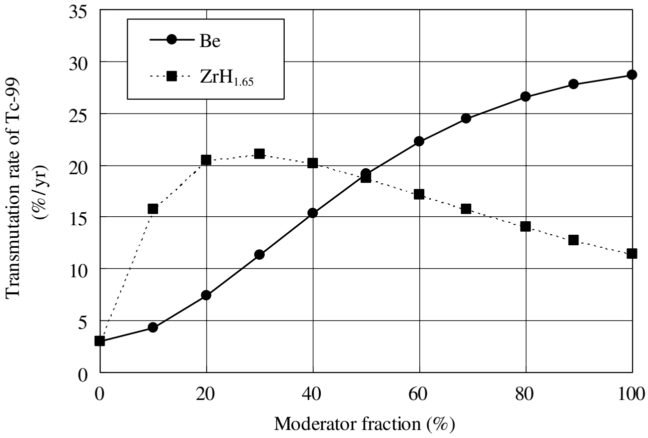

Figure 2. ^{99}Tc transmutation rate as a function of moderator fraction.

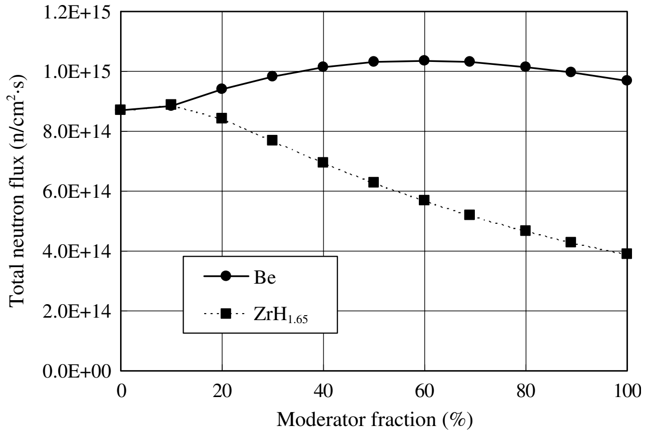

Figure 3. Neutron flux change as a function of moderator fraction.

3.2 Analysis Method. The neutron flux distribution in the core was calculated in 70 energy groups using the three-dimensional diffusion code CITATION[4] in this study. The geometric models used homogeneous representations for all the core component subassemblies. The JFS-3-J3.2R cross section set based on the JENDL-3.2 library[5] was used for the neutron spectrum and transmutation calculation. The absolute value of the neutron flux was determined from the heat balance at the 140 MWt full power condition.

The ORIGEN2 code[6] was used to provide transmutation rates and one group cross sections were collapsed using the calculated 70 group neutron spectrum and the JFS-3-J3.2R cross sections without considering neutron self shielding effects. Five operational cycles per year were assumed in the ORIGEN2 calculation, where each cycle runs 60 effective full power days. The change of neutron flux distribution associated with fuel burn-up and core arrangement change due to refueling is not considered here, however it does not affect the major transmutation characteristics.

4. Results

4.1. Moderator Composition. Figure 2 shows the calculated results for the ^{99}Tc transmutation rate. This could be increased by increasing the moderator fraction of beryllium due to the large resonance in the ^{99}Tc (n, γ) cross section around 6 eV. When $ZrH_{1.65}$ is used for the moderator, however, the transmutation rate has a peak at about 30% fraction caused by a cancellation between the capture reaction increase due to neutron moderation and the neutron shielding effect, as shown in Figure 3.

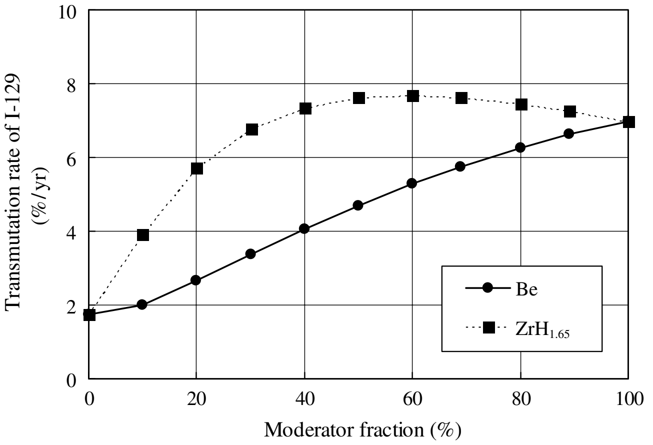

Figure 4. ^{129}I transmutation rate as a function of moderator fraction.

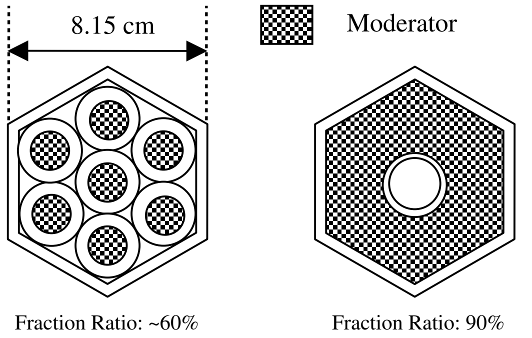

Figure 5. Cross section view of moderator subassemblies.

For irradiating ^{129}I targets, the transmutation rate showed the same tendency as ^{99}Tc. However, using $ZrH_{1.65}$ moderator indicated a peak at around 60% fraction as shown in Figure 4. This is due to the different neutron energy sensitivity to the capture cross section between ^{99}Tc and ^{129}I. Higher ^{129}I transmutation could be achieved with a softer neutron spectrum than ^{99}Tc.

The cross section views of the subassemblies with the optimized moderator fraction are illustrated in Figure 5. Table 2 shows the main design parameters of the moderator subassemblies. The one structure with a fraction under 60% is similar to the B_4C shielding subassembly and the other fraction of 90% is similar to the outer radial stainless steel reflector. Both B_4C shielding subassembly and outer radial reflector are routinely fabricated for use in JOYO.

TABLE 2: Moderator Subassembly Parameters

Subassembly length	2970 mm	
Moderator stack length	650 mm	
Moderator	Be	$ZrH_{1.65}$
Number of element	1	7
Fraction ratio of the moderator material	90%	30, 60%

4.2. Neutron Spectrum and LLFP Transmutation rate. Figure 6 shows the 70 group neutron spectrum and flux values in the target subassembly. The spectra averaged over the 5 cm axial height at core mid-plane were shown and compared with those in the driver fuel and radial reflector regions. In these cases, the moderator fractions in the moderator subassemblies were the optimized results above. The neutron flux in four different energy ranges for the range of cases examined is summarized in Table 3. These data show that the installation of the moderator and target subassemblies could tailor the spectrum to epithermal or thermal energies.

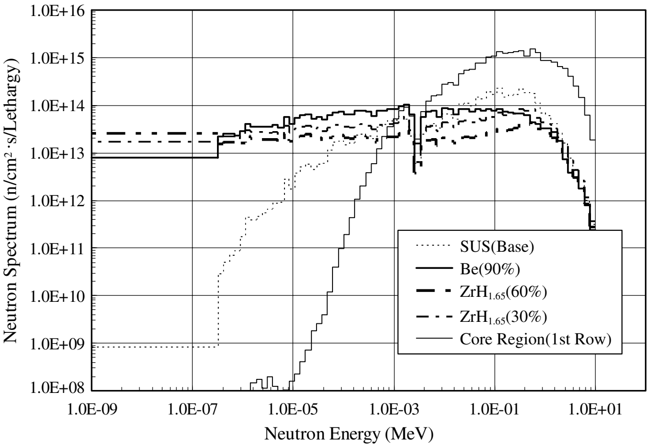

Figure 6. Comparison of neutron spectrum in various locations of JOYO.

TABLE 3: Energy Dependent Neutron Flux

	Neutron Flux (n/cm^2·s)			
	Total	Ratio (%)		
		> 0.1 MeV	< 1 keV	< 0.312 eV
Rad. reflector	8.71×10^{14}	49.7	9.9	0.0
Be (90%)	9.95×10^{14}	16.8	48.6	6.4
$ZrH_{1.65}$ (30%)	7.69×10^{14}	24.5	51.0	18.2
$ZrH_{1.65}$ (60%)	5.68×10^{14}	17.8	65.7	36.8
Core region	5.53×10^{15}	70.6	0.4	0.0

TABLE 4: Transmutation Rate at the Core Mid Plane

	Fractional transmutation rate (%/yr)	
	^{99}Tc	^{129}I
Rad. reflector	3.0	1.7
Be (90%)	27.8	6.6
$ZrH_{1.65}$ (30%)	21.0	6.8
$ZrH_{1.65}$ (60%)	17.1	7.7
Core region	5.4	3.4

Table 4 compares the corresponding transmutation rates at the core mid-plane. The ^{99}Tc transmutation rate was 21.0% using $ZrH_{1.65}$ and 27.8% using beryllium as moderator.

4.3. Moderator Effect on Core Components. The effect of adding moderation extends to the neighboring core components of driver fuel and B_4C shielding subassemblies. Moderated neutrons leaking from the moderator subassembly can reach and be absorbed in the adjacent fuel pins. This causes fuel pin power to increase since the ^{239}Pu fission cross section is higher at thermal energies than at fast energies.

Figure 7 shows the ratio of ^{239}Pu fission rate in the fuel pins of the driver fuel subassembly shown as “A” in Figure 1 to that of the original radial reflector subassembly. In this calculation, one subassembly was divided into 6 triangular meshes. When using beryllium the fission rate at the mesh nearest the moderator (No. 5) is ~19% higher than the reference value for the reflector case. However, the maximum fission rate is only 7% higher than the original maximum value at mesh No. 2.

Considering the radial fuel power distribution, this is not a problem. The driver fuel subassemblies in the 4th and 5th rows generally have adequate margin to the peak power limits during rated 140 MWt operation. This fuel power increase can be countered by replacing fresh fuel with burned fuel or rotating the fuel subassembly. On the other hand, the fission rates when using $ZrH_{1.65}$ moderator were decreased in all meshes due to the large neutron flux depression.

Next, the influence on the B_4C shielding subassemblies was evaluated and the results are shown in Figure 8. The ^{10}B(n, α) reaction rate decreased in all cases due to neutron flux depression.

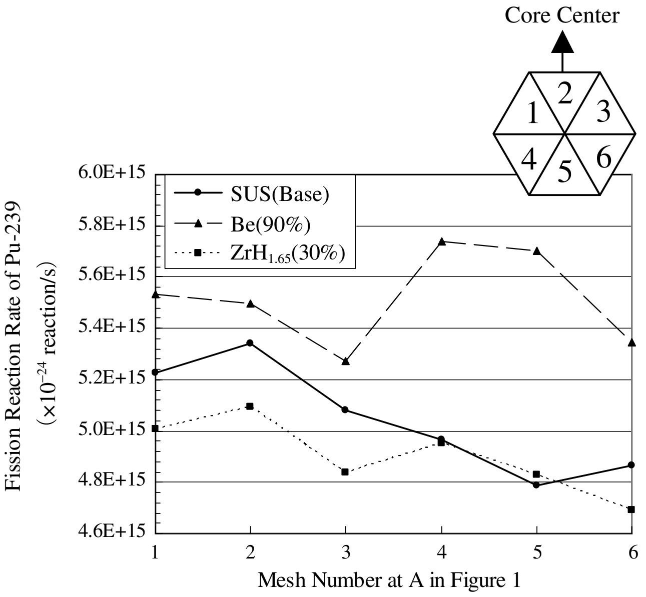

Figure 7. Pu fission rate at driver fuel A with and without moderator.

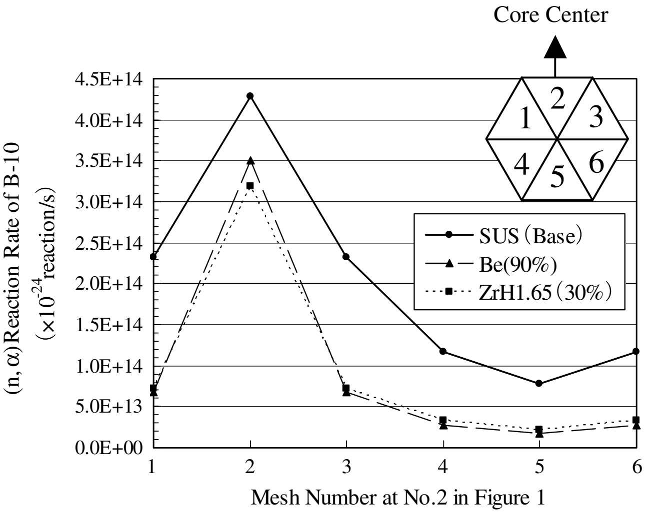

Figure 8. ^{10}B (n, α) reaction rate at shielding subassembly No. 2 with and without moderator.

5. Concluding Remarks

These studies demonstrated that the JOYO reactor has the flexibility to tailor the neutron spectrum in the reflector region to accelerate LLFP transmutation experiments and also to meet a wide variety of irradiation test missions. The irradiation space and excess neutrons inherent in the JOYO design allows this spectral tailoring for LLFP transmutation and other purpose such as isotope production with minimal impact on other simultaneous irradiation missions.

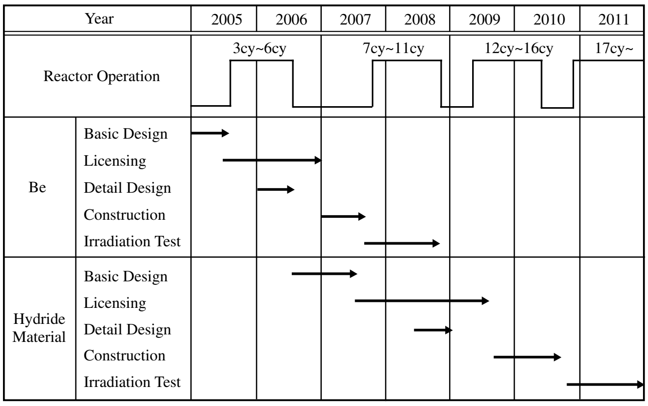

Figure 9. Schedule of LLFP transmutation test in JOYO.

Optimum spectral tailoring depends on the target LLFP to be transmuted. Just the thermal portion of the neutron flux achievable in JOYO can be comparable to many thermal experimental reactors. While the introduction of the moderated subassemblies into the reflector region affects the adjacent driver fuel subassemblies, the effects are within the design margin or can be easily mitigated by fuel management strategies.

Further investigations, such as detailed pin by pin model calculations using the MCNP code will be required to precisely evaluate the effect of neutron moderators on fuel subassemblies. Neutron self shielding effect on the transmutation ratio of the target nuclide will be calculated and the material concentration in the test pin needs to be optimized. Thermal-hydraulic and structural calculations need to be carried out for the moderator subassembly design.

The planning schedule to install the neutron moderator into JOYO is shown in Figure 9. Beryllium will be adopted first based on the previous experience in JOYO. The next step will use hydride materials to achieve higher transmutation rates.

Acknowledgements. The authors would like to note the contribution of Mr. T. Ito of Information Technology Japan Inc. for the calculation using CITATION. We also would like to appreciate Mr. D. W. Wootan, the JNC international fellow, for his cooperation and valuable comments.

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