NUCET NUCLEAR ENERGY AND TECHNOLOGY

Nuclear Energy and Technology 8(2): 91–96
DOI 10.3897/nucet.8.78428

Research Article

Evaluation of transmutation rate of some LLFP in experimental fast reactor JOYO

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Academic editor: Georgy Tikhomirov ♦ **Received** 24 November 2021 ♦ **Accepted** 10 May 2022 ♦ **Published** 27 June 2022

Citation: Amrani N, Boucenna A, Galahom AA (2022) Evaluation of transmutation rate of some LLFP in experimental fast reactor JOYO. Nuclear Energy and Technology 8(2): 91–96. https://doi.org/10.3897/nucet.8.78428

Abstract

A transmutation process of three long-lived fission products (^{79}Se, ^{99}Tc and ^{107}Pd) in the experimental fast reactor JOYO is postulated. The possibility of increasing the transmutation rate utilizing the high neutron flux present in the JOYO reactor by loading neutron-moderating subassemblies in the reflector zone has been investigated. 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 the three long-lived fission products (LLFP) simultaneous and without isotopic separation. ChainSolver 2.34 code is used to calculate the transmutation rates. In this study, the new characteristics of LLFP transmutation in a fast reactor using moderator materials were shown for future applications.

Keywords

Experimental fast reactor JOYO, ChainSolver 2.34 code, transmutation of LLFP

Introduction

One of the major problems in the application of nuclear energy is the presence of a significant amount of minor actinides MA and long-lived fission products (LLFP) in spent nuclear fuel. The composition of a 1 ton spent nuclear fuel from a pressurized water reactor (operating at 33 GWd/t and after 10 years of cooling) is 0.9% Pu, 0.1% minor actinides (MA) and 0.2% long-lived fission products of the total (Kailas et al. 2015). Different means of reducing the radiotoxicity of the LLFP are under investigation as some nuclides such as ^{99}Tc and ^{129}I have a relatively higher solubility in water and may leak into the environment. The LLFP can be converted into short-lived isotopes or stable atoms by using the excess neutrons in the nuclear reactor. The transmutation offers the possibility to reduce the storage volume, radioactivity and long-term radiotoxicity due to the higher mobility of LLFPs in groundwater. From this viewpoint, nuclear reactors provide a suitable means for the transmutation of LLFP. Fast reactor is more appropriate than thermal reactor for transmuting the LLFP, as the thermal neutron absorption cross-section of LLFP is small (Gunsing et al. 2000; Wakabayashi 2002; Kora et al. 2016).

Numerous studies on LLFP transmutation using nuclear reactors have been carried out (Salvatores et al. 1994;

Tommasi et al. 1995; Wakabayashi and Higano 1998; OECD-NEA 1999; Aoyama et al. 2005; Salvatores 2005; Arie et al. 2007; Tachi et al. 2009). Accelerator drive system (ADS) and thermal reactors can be used to transmute the LLFPs (Setiawan and Kitamoto 2011). Different fuel types were investigated in high temperature gas-cooled reactors (HTGRs) for the transmutation of LLFPs (Kora et al. 2016). The performance of the supercritical water-cooled fast reactor (SCWR) for the transmutation LLFPs was evaluated (Lu et al. 2011). The core concept of simultaneous transmutation of six LLFP (^{79}Se, ^{93}Zr, ^{99}Tc, ^{107}Pd, ^{129}I, and ^{135}Cs) was investigated using MONJU fast reactor (Wakabayashi 2019a). The main LLFPs and their half-lives present in spent nuclear fuel are ^{79}Se: 327,000 years, ^{93}Zr: 1,570,000 years, ^{99}Tc: 211,000 years, ^{107}Pd: 6.5 million years, ^{129}I: 15.7 million years and ^{135}Cs: 2.3 million years (Wakabayachi et al. 2019b).

The objective of this study is to evaluate the transmutation rate for three major long-lived fission products: ^{99}Tc, ^{79}Se and ^{107}Pd using a neutron moderating subassembly in the experimental JOYO fast reactor without performing isotopic separation. The purpose of moderator assembly is to moderate the fast neutrons present in the JOYO reactor and use them to improve LLFP transmutation rates. The three long-lived fission products used in this study (^{79}Se, ^{99}Tc and ^{107}Pd) are important from the point of view of the environmental impact reduction that should be carried out.

Description of the experimental fast reactor JOYO

JOYO is the first sodium-cooled fast reactor with plutonium-uranium mixed oxide (MOX) fuel in Japan's development program. The JOYO reactor attained initial criticality in 1977 with the MK-I breeder core. From 1983 to 2000, JOYO was operated at 100 MW_{th} for thirty-five operational cycles with the MK-II irradiation test core. In 2003, up-gradation of the JOYO reactor to the 140 MW_{th} MK-III core was completed to increase the irradiation testing capability. The Main parameters of the MK-III core are given in Table 1 (Maeda et al. 2011).

The active core is approximately 80 cm in equivalent diameter and 50 cm in height. There is a 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 (Maeda et al. 2012). The fuel region is divided into two radial enrichment zones to flatten the power. The MK-III driver fuel is MOX with about 18 wt% enriched U. The fissile Pu content (^{239}Pu + ^{241}Pu)/(U + Pu) is about 16 wt% in the inner core fuel and about 21 wt% in the outer core fuel. An example of the core configuration during an operational duty cycle is presented in Fig. 1 (Aoyama et al. 2007).

Table 1. Main core parameters of JOYO MK-III

Specification	Data
Rector thermal power (MWt)	140
Maximum number of driver fuel subassembly *	85
Equivalent core diameter (cm)	80
Core high (cm)	50
^{235}U enrichment (wt%)	18
Pu content: Pu/(Pu+U) (wt%)	23/30**
Fissile Plutonium 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.s)	5.7×10^{15}
Fast neutron flux (n/cm^2.s)	4.0×10^{15}
Number of control road 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
Operating Cycle per year (Cycle)	5

* Including Number of irradiation test fuel assemblies;
** Inner Core / Outer Core.

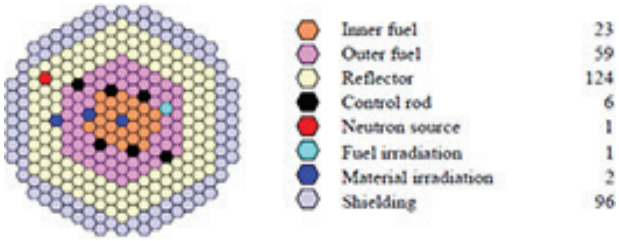

Figure 1. Example of JOYO MKIII Core configuration.

Evaluation method

In this study, we evaluate the transmutation rate for three long-lived fission products: ^{79}Se, ^{99}Tc and ^{107}Pd using ChainSolver 2.34 code (Romanov 2003). The transmutation schemes of the three LLFP are shown in Fig. 2. We explore the loading of a neutron moderator and target subassemblies in the reflector region with the objective of obtaining a high transmutation rate. The JOYO core design adopted for this study is shown in Fig. 3. The cluster of reflector subassemblies is replaced with a new moderator and target subassembly. The beryllium metal or zirconium hydride ($ZrH_{1.65}$) was selected as moderator material. The fraction ratio of moderator material in the target subassembly is 90% and 60% for beryllium and zirconium hydride respectively.

Six of row 7, 8 and 9 reflector subassemblies were replaced with beryllium or $ZrH_{1.65}$ moderated subassemblies. The structure of the moderator subassembly is similar to the other core components. The moderator subassembly surrounded one test assembly that contain the LLFP.

The chemical form for LLFP adopted in this study is the metallic form because it has a high melting point and the space volume for loading can be minimized (Chiba 2017) The idea of separating the moderator and the target subassembly has an advantage for the LLFP irradiation

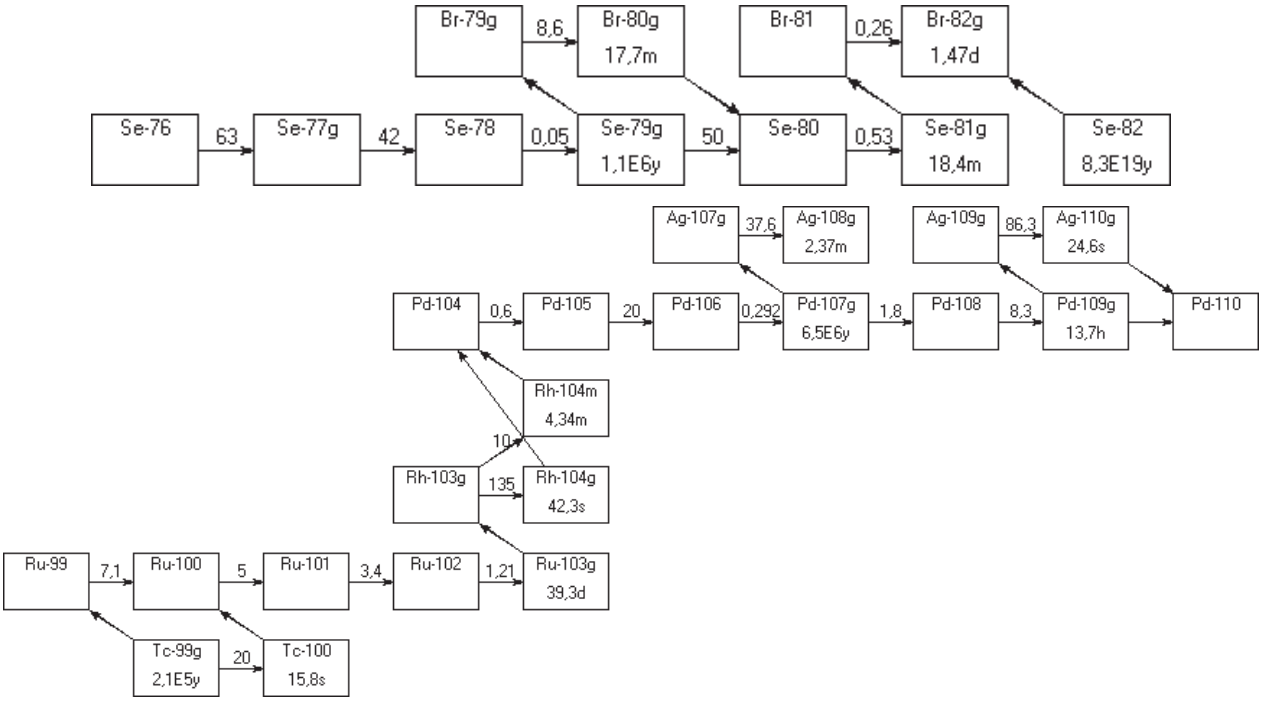

Figure 2. ^{79}Se ^{99}Tc and ^{107}Pd transmutation chain under neutrons irradiation.

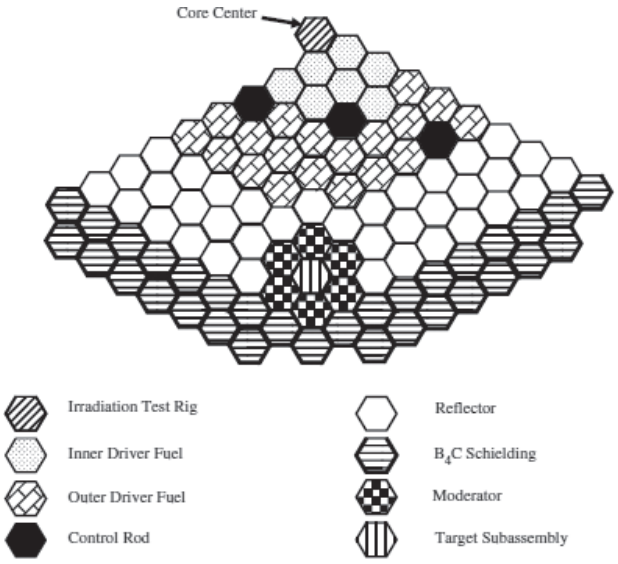

Figure 3. Target assembly location (1/3rd core configuration).

process. The moderator subassembly acts as a buffer that separates the thermal neutron region from the fast neutron region of the fuel subassembly and creates a considerable epithermal region. This prevents the thermal peak that can occur near the region that separates the two neutron spectra. The neutron flux in the radial reflector region and in the target subassemblies proposed with different moderators was calculated in 70 energy groups using the three-dimensional diffusion code CITATION. The absolute value of the neutron flux was determined from the heat balance at 140 MW full power condition (Ayoama 2005).

The neutrons flux dependence on energy in the target and moderator subassemblies shows that the thermal and

Table 2. Neutron flux dependence on energy

Core position	Neutron Flux (n/cm^2.s)			
	Total	Fast (E>0.1 MeV)	Epithermal (E<1 KeV)	Thermal (E<0.312 eV)
Radial reflector	8.71×10^{14}	4.33×10^{14}	8.6×10^{13}	0.0
Be (90%)	9.95×10^{14}	1.67×10^{14}	4.84×10^{14}	6.34×10^{13}
$ZrH_{1.65}$(30%)	7.69×10^{14}	1.88×10^{14}	3.92×10^{14}	1.39×10^{14}
Core region	5.53×10^{15}	3.90×10^{15}	2.21×10^{13}	0.0

epithermal fluxes increase considerably by using beryllium as moderator more than for the $ZrH_{1.65}$ (Table 2).

The LLFP transmutation rates in the different regions and in moderator-target subassemblies are evaluated with ChainSolver 2.34 code used for transmutation calculations (Amrani et al. 2007). The ChainSolver 2.34 code is intended for fast transmutation simulation of samples during irradiation in nuclear reactors. The code calculates a nuclide density time evolution with burn up, decay and build-up. The depression of a thermal neutrons flux, resonance self-shielding of isotopes during irradiation, and the irradiation schedule (the schedule of the reactor work and rearrangement of an irradiated target in various positions) are taken into account. These calculations are extremely tedious because at each stage of irradiation the fast, epithermal and thermal fluxes, cross sections of reactions, time of irradiation and structure of initial product should all be taken in consideration. Such calculations allow one to define the chosen mode of irradiation and to calculate the expected outputs for both products and inevitable impurities. The main approximation used in the calculation of transmutations is the assumption that the influence of changes in the irradiated material structure on characteristics of a reactor as a neutron source is insignificant (Amrani et al. 2011).

Results and discussion

In this study, the transmutation rates of three LLFP nuclides (^{79}Se, ^{99}Tc, ^{107}Pd) are evaluated to improve the transmutation possibilities of LLFP in the experimental fast reactor JOYO. The choice of these three LLFP is due to their high neutron absorption cross section in the thermal and epithermal regions as illustrated in Fig. 4.

The isotopic composition of loaded LLFP is given in Table 3 (Wakabaychi 2019b). The LLFP are loaded in transmutation assemblies without isotopic separation in the core region in radial reflector and in target subassembly with a moderator. We used beryllium and zirconium hydride as neutron moderators to investigate the impact of moderator type on the transmutation of LLFP performances.

Table 3. Isotopic abundance of loaded LLFP

LLFP Elements	Decay mode	Iγ	Abundance %
^{76}Se	Stable	–	0.027
^{77}Se	Stable	–	2.786
^{78}Se	Stable	–	5.587
^{79}Se	β-	96	13.32
^{80}Se	Stable	–	22.75
^{82}Se	Stable	–	55.52
^{99}Tc	β-	141	100.00
^{104}Pd	Stable	–	2.93
^{105}Pd	Stable	–	35.14
^{106}Pd	Stable	–	17.86
^{107}Pd	β-	214	21.72
^{108}Pd	Stable	–	17.12
^{110}Pd	β-	189	5.24

The LLFP mass considered for transmutation is about 200 g. For 110 effective full power days, the transmutation rate TR (%/day) is calculated as follows:

$$TR = \frac{M_i - M_f}{tM_i}$$

Where, M_i and M_f are the initial and the final mass of LLFP loaded in target subassembly respectively and the t is the irradiation period. The initial and final mass for the three LLFPs in different JOYO reactor core region and with the beryllium and zirconium hydride neutron moderator is calculated using Chainsolver 2.34 code. The ^{79}Se, ^{99}Tc and ^{107}Pd mass evolution under neutron irradiation are given in Table 4.

The transmutation rate for the three LLFPs in different zones is illustrated in Table 5. The evolution of transmutation rates as a function of irradiation time for ^{79}Se, ^{99}Tc and ^{107}Pd are illustrated in Fig. 5. Based on the obtained transmutation rates for LLFP, we consider that the use of target subassembly and the beryllium as the moderator modify the spectrum of thermal and epithermal energy and increase considerably the transmutation more than for the zirconium hydride material. In general, the introductions of the moderated subassembly influence considerably the transmutation rate of LLPF in the fast spectrum.

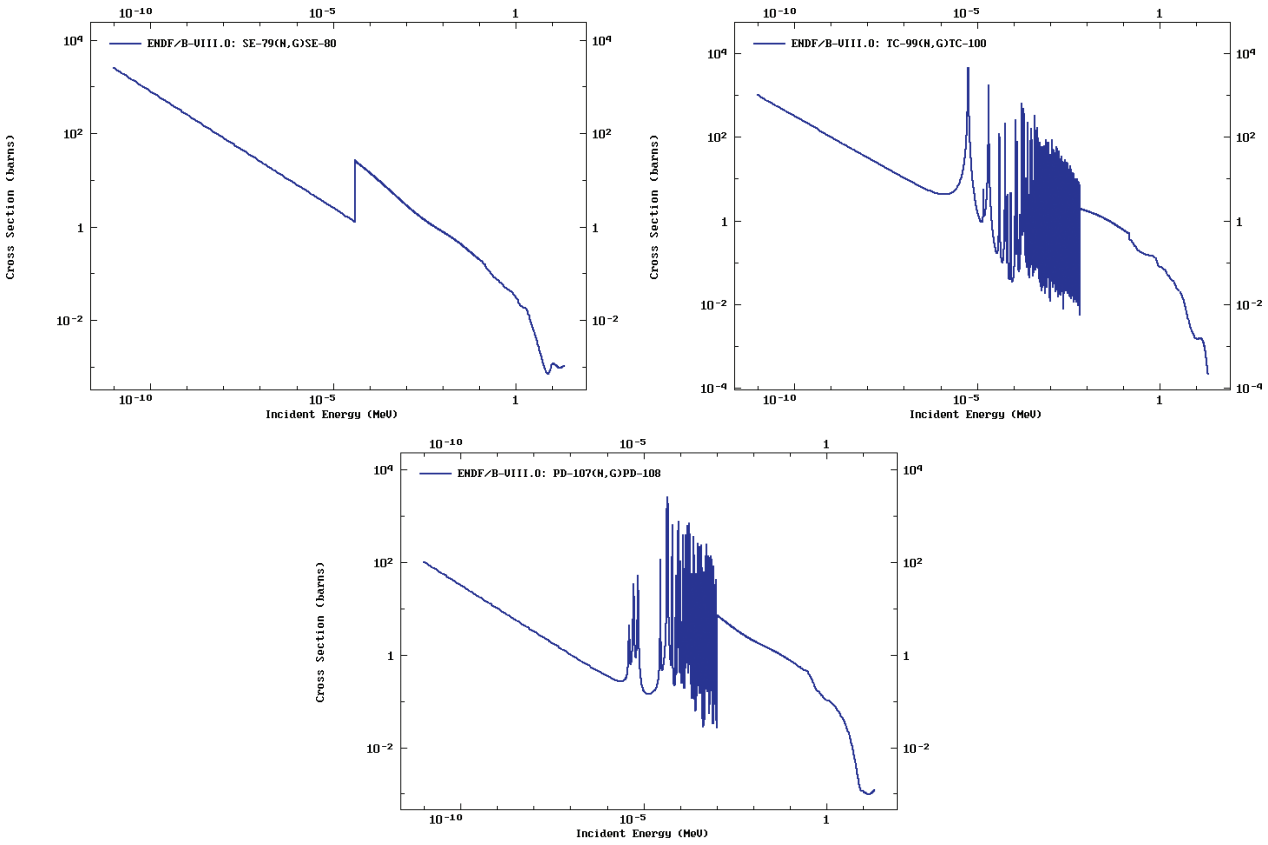

Figure 4. Neutron absorption cress section from ENDF file, (a) ^{79}Se, (b) ^{99}Tc, (c) ^{107}Pd.

Table 4. ^{79}Se, ^{99}Tc and ^{107}Pd mass evolution for 110 days irradiation period

Element	isotope	Initial mass (g)	Final mass (g)			
			Core	Radial reflector	Be (90%) moderator	$ZrH_{1.65}$ (60%) moderator
Se	^{76}Se	0,0135	0.013377	0.013062	0.0077572	0.010528
	^{77}Se	1,393	1.3827	1.3592	0.97525	1.1878
	^{78}Se	2,7935	2.7982	2.8191	3.1633	2.962
	^{79}Se	6,66	6.6509	6.659	5.1137	6.2848
	^{80}Se	11,375	11.367	11.356	12.791	11.66
	^{82}Se	27,76	27.748	27.758	27.748	27.754
Tc	^{99}Tc	100	92.805	75.71	18.82	27.529
Pd	^{104}Pd	1.465	1.4559	1.4455	1.3566	1.3791
	^{105}Pd	17.75	17.347	16.718	11.956	13.693
	^{106}Pd	8.93	9.1247	9.7631	14.366	12.693
	^{107}Pd	10.86	10.623	10.155	7.4992	8.0526
	^{108}Pd	8.56	8.3077	7.6902	4.7863	5.3792
	^{110}Pd	2.71	2.7052	2.7028	2.6683	2.678

Table 5. Transmutation rate for ^{79}Se, ^{99}Tc and ^{107}Pd

Isotope	Half-life (year)	Transmutation Rate (%/day)			
		Core	Radial reflector	Beryllium moderator	Zirconium hydride moderator
^{79}Se	3.27×10^{5}	1.23×10^{-3}	1.36×10^{-4}	0.211	0.0512
^{99}Tc	2.11×10^{5}	0.065	0.22	0.73	0.658
^{107}Pd	6.5×10^{6}	0.0198	0.059	0.281	0.235

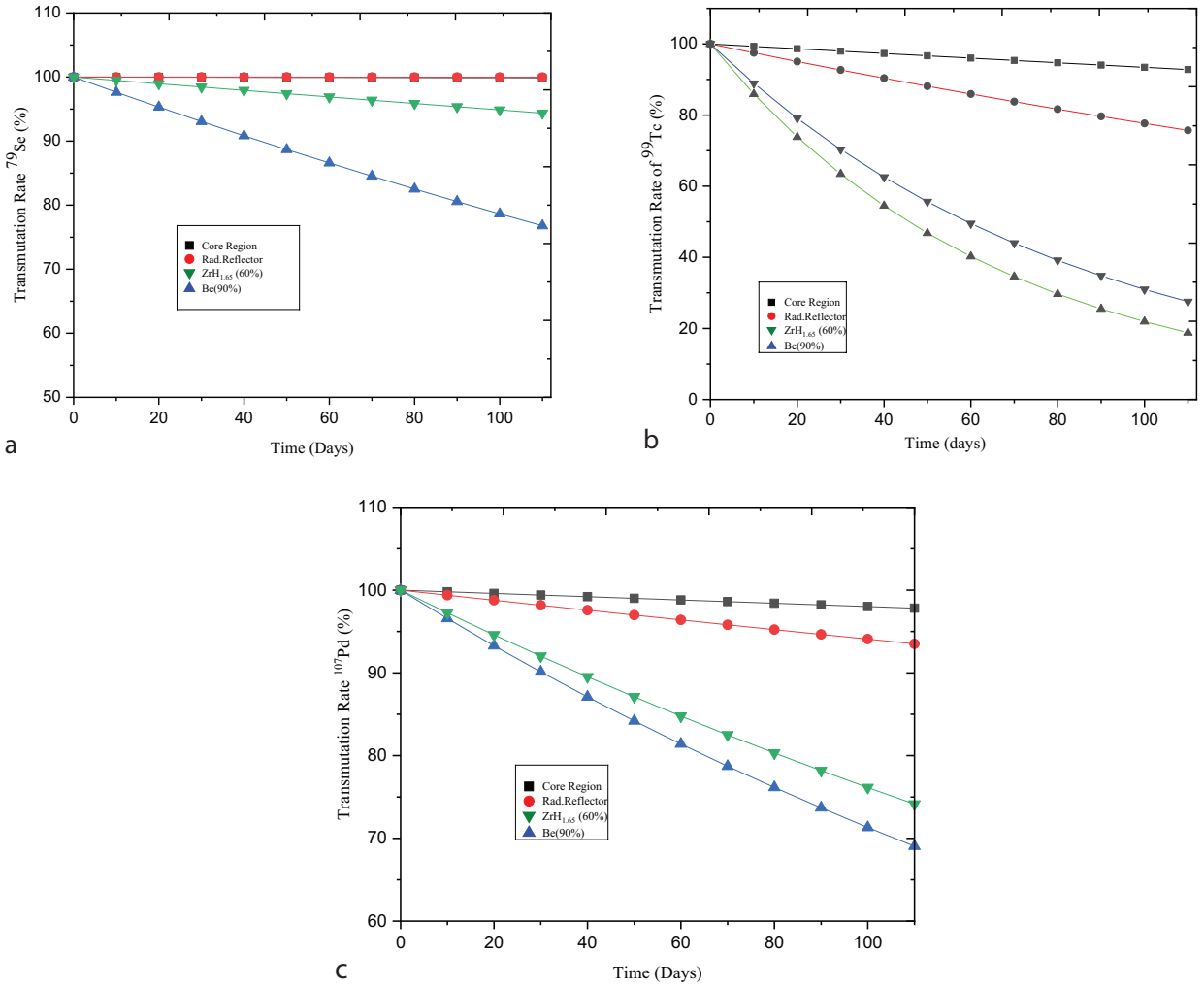

Figure 5. Transmutation rate evolution for ^{79}Se, ^{99}Tc and ^{107}Pd.

Conclusion

In this paper, the effectiveness of three LLFP (^{79}Se, ^{99}Tc and ^{107}Pd) transmutation processes in the experimental Fast Reactor JOYO was studied. The cluster reflector subassembly of the experimental fast reactor "JOYO" was replaced with a target subassembly with beryllium and zirconium hydride neutron moderator. The "JOYO" reactor can now modify the neutron spectrum in the reflector region to speed up the LLFP transmutation process with this modification. The beryllium metal as moderator provides good performance for the LLFP transmutation process by increasing epithermal flux in the target assembly region.

The calculation of LLFP mass change under irradiation was performed using ChainSolver 2.34 code. The use of beryllium as a moderator significantly increases the transmutation rate of LLFP. Further investigations, is to study the impact of placing a permanent moderator and target subassemblies on fast reactors core performances and the use of other moderators to increase the transmutation rate using MCNP code.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

The authors thanks the general direction of scientific research and technological development (DGRSDT) of the Algerian higher education and scientific research ministry.

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