Feasibility study on small and medium modular light water reactors with inherent nuclear safety and security features using U_3Si_2 fuel and MA

Natsumi Mitsuboshi[1], Hiroshi Sagara[1]

[1]Tokyo Institute of Technology, Tokyo, Japan

Abstract

Small modular reactors (SMRs) have been paid much attention owing to their various potential applications and flexibility to various social demands. However, it is overloaded in safety and security, if the same grade regulation with large scale reactors are required for SMRs. Therefore, to introduce graded approach is important for reasonable and feasible regulation in SMRs. Uranium silicide (U_3Si_2) fuel has the higher heavy metal density, the thermal conductivity and chemical stability than U oxide (UO_2) fuel, and it is expected to enhance the inherent safety and security features. ^{238}Pu is produced from ^{237}Np and ^{241}Am by the nuclear transmutations, and its large decay heat and spontaneous fission neutron emission rates adverse effects of nuclear explosive devices (NEDs).

The objectives of the present paper are to reveal the fundamental neutronics, nuclear safety and security features of small and medium PWR (SMPWR) by utilizing uranium silicide fuel and minor actinides (MAs). In fundamental neutronics and safety analysis, neutron transport and burn-up calculation were performed by MVP and MVP-BURN code with JENDL-4.0 cross section library. The results of the fundamental neutronics showed the U enrichment required to provide the same burn-up days in U_3Si_2 fuel was smaller than in UO_2 fuel. In addition, due to the large neutron capture cross-sections of ^{237}Np and ^{241}Am, the initial reactivity and burn-up reactivity change of MA doped fuel was reduced. In the safety analysis, the temperature gradient inside U_3Si_2 fuel was smaller than in UO_2 fuel due to the higher thermal conductivity of U_3Si_2 fuel. Material Attractiveness evaluations were conducted to evaluate the relative utility of nuclear materials to assemble NEDs for non-state actors in each phase of “acquisition”, “processing”, and “utilization”. The degree of influence in weapon utility was divided in 4 grades. The attractiveness of fresh and spent U_3Si_2 and UO_2 fuel assemblies were equivalent. However, Pu separation in U_3Si_2 fuel was more complex and impractical compared with UO_2 fuel. The decay heat from Pu in spent MA doped U_3Si_2 and UO_2 fuel assemblies were enhanced due to higher ratio of ^{238}Pu in MA doped assemblies.

1. Introduction

SMRs are expected as distributed and stable energy source responding to various social demands. To realize the flexibility, introduction of graded approach is necessary for reasonable and feasible regulation in SMRs.

Accident Tolerant Fuel (ATF) has been actively conducted with growing interest in nuclear safety. ATFs are nuclear fuels that does not progress to a severe accident such as a core meltdown or significantly delays the progress to a severe accident even in the event of a severe situation such as the loss of all power as a result of the physical properties of the fuel themselves. In addition, ATFs can perform equal to or higher than that of previous fuels. Uranium silicide (U_3Si_2) fuel is one of the ATF candidates. The density of U_3Si_2 fuel is 16% higher than U oxide (UO_2) fuel, and the thermal conductivity of U_3Si_2 fuel is twice as much as UO_2 fuel [1]. Additionally, U_3Si_2 fuel have chemical stability because Si residue remains in the nitric acid solution during the U_3Si_2 fuel reprocessing process [2]. Minor actinides (MAs) such as ^{237}Np and ^{241}Am produce ^{238}Pu by the nuclear transmutations. ^{238}Pu has large decay heat and spontaneous fission neutron emission rates, which will give adverse effects on nuclear explosive devices (NEDs). The nuclear security features of nuclear materials are assessed with the material attractiveness to investigate the impacts of nuclear material properties to nuclear security features. The material attractiveness is defined as "the relative utility of nuclear material for an adversary to assemble a NED", based on the physical properties of nuclear materials [3]. The fundamental neutronics and nuclear safety features of U_3Si_2 fuels have been reported in *Mitsuboshi et al.,* [4].

The objectives of the present paper are to reveal the nuclear security features based on the material attractiveness of small and medium PWR (SMPWR) by utilizing ^{237}Np/^{241}Am doped U_3Si_2 fuel in comparison to UO_2 fuel and U_3Si_2 fuel.

2. Nuclear security features of ^{237}Np/^{241}Am doped U_3Si_2 fuel

2.1. Evaluation method

The evaluations of material attractiveness were conducted for the inherent material properties of the ^{237}Np/^{241}Am doped U_3Si_2 fuel. The attractiveness represents the relative utility of nuclear materials for non-state actors to assemble a NED [3].

In the evaluation, the physical properties of important nuclear materials were evaluated for non-state actors such as terrorists in each phase of "acquisition," "processing," and "utilization". The degree of influence on weapon utility was divided into four stages, as shown in **Table 1**. **Table 2** shows the scale for the categorization.

The evaluation targets were fresh and spent fuel assemblies of UO_2 fuel, U_3Si_2 fuel, 0.5 wt.% of ^{237}Np doped U_3Si_2 fuel, and 0.5 wt.% of ^{241}Am doped U_3Si_2 fuel. The final target material in this evaluation is U metal and Pu metal. The isotopic ratios of spent fuels were obtained by burn-up calculations which were performed by MVP and MVP-BURN code [5] with JENDL-4.0 cross section library [6]. The calculation model is two-dimensional pin cell model. The calculation condition is shown in **Table 3**. Net weight, acquisition time, and radiation dose are indicators of the acquisition phase. Processing time and complexity are indicators of the processing phase. Bare critical mass (BCM) and heat content (HC) are indicators of the utilization phase. U_3Si_2 fuel was produced by mixing U metal powder and Si powder, using arc melting, and sintering [7]. The U_3Si_2 fuel was dissolved in nitric acid solution. The reprocessing of U_3Si_2 fuel has been studied in La Hague reprocessing plant. According to their experimental results, over 98% of the Si in the nitric acid solution was amorphous [2]. A step was added to separate the solid Si, which is amorphous, from U, Pu, and FPs in the nitric acid solution using a centrifuge that is used in the clarification process, which is a conventional reprocessing process. However, it is considered that there is insufficient processing time and complexity to reduce the attractiveness.

Table 1 Definition of attractiveness Level Bins [3]

Weapon Utility	Category
Preferred	1
Potentially usable, but not preferred	2
Impractical, but not impossible	3
Impossible	4

Table 2 Scale of categorization for attractiveness for non-state actors [3]

Category	Acquisition phase			Processing phase	Utilization phase	
	Net weight [kg]	Acquisition time [min]	Dose rate at 1 m [Gy/h]	Processing time and complexity	BCM [kg]	Heat content[3] [W/BCM]
1	< 50	< 15	< 0.1	Direct conversion in one step (metal form)	< 80	< 1292
2	50 – 3000	15 – 60	0.1 -1	Conversion in two or more steps(compounds)	80 – 800	1292 – 6274
3	3000 – 6500	60 – 240	1 – 10	Conversion with relative difficult purification step (irradiation material)	800 - 4000	6274 <
4	6500 <	240 <	10 <	Conversion requiring either irradiation or enrichment	4000 <	

[3] Heat content is an index calculated, assuming the δ phase **[8]**.

Table 3 The specifications of fuel assemblies

	UO_2 fuel	U_3Si_2 fuel	0.5 wt.% ^{237}Np doped U_3Si_2 fuel	0.5wt.% ^{241}Am doped U_3Si_2 fuel
Smear density [%T.D.]	95	←	←	←
^{235}U enrichment [wt.%]	4.11	←	←	←
Density of heave metal [g/cm^3]	10.96[1]	12.2[1]	-	-
Assembly design	17×17	←	←	←
Length of assembly [m]	2.0	←	←	←
Atomic ratio				
^{234}U [%]	0.04	0.04	0.04	0.04
^{235}U [%]	4.16	4.16	4.14	4.14
^{238}U [%]	95.80	95.85	95.28	95.29
^{237}Np [%]	-	-	0.54	-
^{241}Am [%]	-	-	-	0.53
Mass of materials in a fresh fuel assembly				
Total [kg]	308	351	349	349
U [kg]	247	289	287	287
Pu [kg]	-	-	-	-
Np [kg]	-	-	1.56	-
Am [kg]	-	-	-	1.56
Mass of materials in a spent fuel assembly (48 GWd/t, ten-years cooling)				
Total [kg]	296	336	336	336
U [kg]	232	271	270	270
Pu [kg]	2.58	3.19	4.12	4.29
Np [kg]	-	-	0.65	-
Am [kg]	-	-	-	0.37

2.2. Evaluation result

Table 4 shows the evaluation results for the attractiveness of the new/spent fuel assemblies. A finite critical amount of metallic U spheres cannot be produced for the fresh fuels. Furthermore, because of requirement for the enrichment process, the attractiveness of fresh fuels was also downgraded. Since BCM of fresh fuels are infinite in the utilization phase, it was not possible to evaluate HC and acquisition phase of fresh fuels. Therefore, the attractiveness of fresh U_3Si_2 fuel and UO_2 fuel were found to be Category 4, which require an enrichment process for non-state actors.

The attractiveness of spent fuel assemblies of UO_2 fuel, U_3Si_2 fuel, $^{237}Np/^{241}Am$ doped U_3Si_2 fuel were Category 4. In processing phase, Si separation process are additionally required for mid- and long-term storage spent $^{237}Np/^{241}Am$ doped U_3Si_2 fuel assemblies. Moreover, in the utilization phase, the categories of decay heat in spent $^{237}Np/^{241}Am$ doped U_3Si_2 fuel assemblies were one level lower than that of the non-doped spent U_3Si_2 fuel assembly. This is because the isotopic ratios of ^{238}Pu in $^{237}Np/^{241}Am$ doped spent fuels are higher than in non-doped spent fuels.

Therefore, not only Pu separation in $^{237}Np/^{241}Am$ doped U_3Si_2 fuel was more complex and impractical than UO_2 fuel, but also the categories of decay heat in spent fuel assemblies were downgraded by doping MAs.

Table 4 Measures of the material attractiveness assessment

Theft target	Final Target Material	Acquisition Phase			Processing Phase	Utilization Phase	
		Net weight [kg]	Acquisition time[min]	Dose rate at 1m [Gy/h]	Procession time and complexity	BCM [kg]	HC [W/BCM]
Fresh fuel assembly							
UO_2 fuel [3]	U metal	N/A	N/A	N/A	Conversion requiring enrichment (4)	∞(4)	N/A
U_3Si_2 fuel [3]	U metal	N/A	N/A	N/A	Si separation + Conversion requiring enrichment (4)	∞(4)	N/A
Spent fuel assembly (48 GWd/t, ten-years cooling)							
UO_2 fuel [3]	Pu metal	296(2) 1assembly	~20(2)	~30(4)	Conversion with relative difficult purification step (3)	22(1)	424(1)
U_3Si_2 fuel [3]	Pu metal	336(2) 1assembly	~20(2)	~30(4)	Si separation + Conversion with relative difficult purification step (3)	22(1)	458(1)
U_3Si_2 fuel (^{237}Np 0.5 wt.%)	Pu metal	336 (2) 1assembly	~20(2)	~30(4)	Si separation + Conversion with relative difficult purification step (3)	20(1)	2099(2)
U_3Si_2 fuel (^{241}Am 0.5 wt.%)	Pu metal	336 (2) 1assembly	~20(2)	~30(4)	Si separation + Conversion with relative difficult purification step (3)	20(1)	1994(2)

[4] The number in () indicates the category.

3. Conclusion

The effects of $^{237}Np/^{241}Am$ doped U_3Si_2 fuel on nuclear security of SMPWR were evaluated by comparing it with UO_2 fuel and U_3Si_2 fuel.

Attractiveness was evaluated for non-state actors aiming for NED manufacturing. The attractiveness of fresh and spent $^{237}Np/^{241}Am$ doped U_3Si_2 fuel was equivalent to that of the fresh and spent UO_2 fuel in the processing phase, although the complexity of Pu separation in spent $^{237}Np/^{241}Am$ doped U_3Si_2 fuel was higher than that in UO_2 fuel. By 0.5 wt.% $^{237}Np/^{241}Am$ doping, the decay heat of the separated Pu from the spent fuel was enhanced to degrade by one level in the utilization phase.

In conclusion, the present study demonstrated that $^{237}Np/^{241}Am$ doped U_3Si_2 fuel are superior to UO_2 fuel and U_3Si_2 fuel in nuclear security features. As a future study, researchers should consider the material attractiveness of the cases which final target material is MA metals, reactor safety analysis of the SMPWR and acquisition/diversion pathway analysis for SMPWR.

Reference

[1] Johnson, K.D, et al., Fabrication and microstructural analysis of UN-U_3Si_2 composites for accident tolerant fuel applications. J. Nucl. Mater. 477, 18-23, (2016).

[2] Valery, J. F., et al., Status on Silicide Fuel Reprocessing at AREVA La Hague, (2015).

[3] Bathke, C. G., Sagara H, et al., Summary of a Joint US-Japan Study of Potential Approaches to Reduce the Attractiveness of Various Nuclear Material for Use in a Nuclear Explosive Device by a Terrorist Group, Global 2013, United States.

[4] Mitsuboshi, N., Sagara, H., "Effects of U_3Si_2 fuel and minor actinide doping on fundamental neutronics, nuclear safety, and security of small and medium PWRs in comparison to conventional UO_2 fuel", Ann. Nucl. Energy 153, (2021).

[5] Okumura, K., Mori, T., et al., Validation of a Continuous-Energy Monte Carlo Burn-up Code MVP-BURN and Its Application to Analysis of Post Irradiation Experiment. J. Nucl. Sci. Technol. 37 (2), 128-138, (2000).

[6] Shibata, K., Iwamoto, O., et al., JENDL-4.0: a new library for nuclear science and engineering. J. Nucl. Sci. Technol. 48 (1), 1-30, (2010).

[7] Harp, J. M., et al., Uranium Silicide Pellet Fabrication by Powder Metallurgy for Accident Tolerant Fuel Evaluation and irradiation, J. Nucl. Mater. 466,728-738, (2015).

[8] Aoki, T., Sagara, H., Han, C.Y., Material attractiveness evaluation of inert fuel for nuclear security and non-proliferation. Ann. Nucl. Energy 126, 427–433(2019).