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Core design study on rock-like oxide fuel light water reactor and improvements of core characteristics

H. Akie *, H. Takano, Y. Anoda

Japan Atomic Energy Research Institute, Tokai-mura, Naka-gun, Ibaraki-ken, 319-11, Japan

Abstract

A rock-like oxide (ROX) fuel – LWR burning system has been studied for efficient plutonium transmutation. A zirconia based ROX (Zr-ROX) core has problems such as a small negative Doppler coefficient and a large power peaking factor, which causes severe transients in accidents and high fuel temperature even under nominal condition. For the improvement of these characteristics, two approaches were considered: the additives UO_2, ThO_2 and Er_2O_3, or a heterogeneous core with Zr-ROX and UO_2 assemblies. As a result, the combination of the additives UO_2 and Er_2O_3 is found to sufficiently improve the accident behavior, while a further power peaking reduction may be necessary for the Zr-ROX + UO_2 heterogeneous core. The plutonium transmutation rate is extremely high in Zr-ROX assemblies in the heterogeneous core, to be more than 85% and 70%, respectively for weapons- and reactor-grade plutonium. The plutonium transmutation rate becomes smaller in the full-ROX core with the UO_2 or ThO_2 additive, but the annual transmutation amount of plutonium is large, in comparison with the full-MOX fuel core.

1. Introduction

A rock-like oxide (ROX) fuel in a LWR burning system has been studied at the Japan Atomic Energy Research Institute (JAERI) for plutonium annihilation, because of its high plutonium transmutation rate. This type of U-free matrix fuel has nuclear characteristics different from UO_2 and MOX fuels. Particularly, stabilized zirconia (ZrO_2) type ROX (Zr-ROX: PuO_2–$(Zr,Y,Gd)O_2$–$MgAl_2O_4$) fuel contains no fertile nuclide such as ^{238}U or ^{232}Th, and therefore has a smaller reactivity coefficient and a larger burnup reactivity swing than UO_2 and MOX. Furthermore, Zr-ROX is a multiphase type fuel and has a low melting point. Due to these characteristics, some problems arise in ROX–LWR core design study [1]: The small Doppler coefficient results in extremely severe transients, especially in a reactivity insertion accident (RIA). The large burnup reactivity swing causes a large power peaking factor, which should be avoided for a moderate loss of coolant accident (LOCA) and other thermal-hydraulic behavior. Even in the nominal operating condition, the fuel temperature can easily become as high as the melting point. The large excess reactivity, together with the hard neutron spectrum, also results in an excess reactivity control problem [2].

In this study, emphasis is put on the improvements of these Zr-ROX core characteristics. First, the effect of additives such as Th and U is examined in the Zr-ROX fuel. Furthermore for the power distribution flattening, Er is considered as a burnable poison to reduce the content of Gd in stabilized ZrO_2. Second, a heterogeneous core concept is studied, where the Zr-ROX fuel assemblies are partially charged in a UO_2 fuel core. The results of accidents analyses and fuel temperature calculations are presented.

2. Improvement of Zr-ROX core physics characteristics

For the improvement of the Doppler coefficient, resonant isotopes ^{232}Th and ^{238}U are very effective. These nuclides are to be mixed in the ROX fuel in oxide form, ThO_2 or UO_2. Thoria (ThO_2) is one of the

* Corresponding author. Tel.: +81-29 282 6939; fax: +81-29 282 6805; e-mail: akie@mike.tokai.jaeri.go.jp

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candidate matrices of ROX (Th-ROX) fuel, and can be expected not to degrade the ROX fuel stability. From the phase relation study, it appears that $(Zr,Y,Gd)O_2$ and UO_2 form an ideal solid solution, at least up to 40 mol% UO_2 [3]. On the other hand, Er as an additive was also considered for power distribution flattening. Erbium also shows the Doppler effect, but an amount of Er to sufficiently increase the negative Doppler reactivity causes an substantial decrease in the excess reactivity. The objective of the Er additive here is not the Doppler effect, but to reduce the Gd content. In Zr-ROX fuel, Gd_2O_3 stabilizes the fluorite type phase of ZrO_2, and at the same time suppresses the excess reactivity at the beginning of burnup. Without burnable poison, the excess reactivity of the Zr-ROX core becomes extremely large. As shown in Fig. 1, however, Gd causes a large reactivity swing during burnup, and leads to a large power level mismatch between fuel assemblies at different burnup stages. This leads to a large power peaking in the Zr-ROX core. By using Er and reducing Gd content, it is possible to make the burnup reactivity swing much more moderate. Erbium is added to Zr-ROX in the form of Er_2O_3, which also stabilizes the fluorite type phase of ZrO_2 in the same way as Gd_2O_3.

For a sufficient RIA behavior improvement, it was found necessary to increase the Doppler coefficient by about five times [1]. It was decided for such an improvement, that the amount of additives are to be 24 mol% of ThO_2 or 15 mol% of UO_2 in the weapons plutonium case, and 18 mol% ThO_2 or 8 mol% UO_2 in the reactor-grade plutonium case. These additives also decrease the burnup reactivity change, which is helpful for power peaking reduction. In the Zr-ROX–UO_2 case, about 0.3 mol% Er_2O_3 is considered for a further power distribution flattening. Consequently, the Gd_2O_3 content can be reduced by less than 1/10 (about 0.02 mol%), and the burnup reactivity swing becomes very much moderate (Fig. 1). For a heterogeneous core with Zr-ROX and UO_2 fuels, 1/3 Zr-ROX + 2/3 UO_2 was also considered.

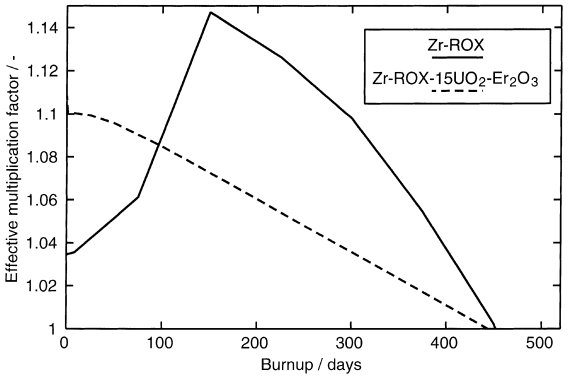

Fig. 1. Burnup reactivity change in PWR of Zr-ROX fuel and the effect of UO_2 and Er_2O_3 additives (weapons-Pu).

Table 1
Doppler reactivity ($\Delta k\ k^{-1}$/%) and power peaking factor in a Zr-ROX PWR core (BOC)

	Doppler reactivity (900→1200 K)	Peaking factor
Weapons-Pu		
Zr-ROX	−0.098	2.7
Zr-ROX(Er)–15UO_2	−0.61	2.1
Zr-ROX–24ThO_2	−0.56	2.4
1/3Zr-ROX+2/3UO_2	−0.48	2.8
Reactor-Pu		
Zr-ROX(Er)–8UO_2	−0.59	2.2
Zr-ROX(Er)–18ThO_2	−0.64	–
UO_2	−0.75	2.0

The physics characteristics of the original and the modified Zr-ROX PWR cores are compared in Table 1. These were evaluated with two-dimensional core burnup calculations based on the diffusion method by using the SRAC95 code system [4]. As shown in the Table, the Doppler reactivity of the original Zr-ROX core is far smaller than that of UO_2, and the power peaking factor is as high as 2.7. The Doppler reactivities of the modified cores are all improved. With ThO_2 or UO_2, the Doppler reactivity can be increased by a factor of more than 5, and by a smaller factor in the 1/3 Zr-ROX core. Power distribution flattening becomes much easier with ThO_2, and is very well achieved with UO_2 + Er_2O_3. In the heterogeneous core with U and Pu fuel assemblies, it was difficult to reduce the power peaking to less than 2.8.

3. Accident analyses

The reactivity coefficients and the power profile estimated in the previous chapter were used in the analyses of RIA and LOCA. It should be noted that the fuel properties such as melting point and thermal conductivity are dependent on the fuel composition, especially on the content of spinel ($MgAl_2O_4$). In these analyses, the fuel properties of Zr-ROX in Table 2 were used. The

Table 2
Fuel properties of Zr-ROX evaluated at around 800 K

Specific heat capacity ($J\ kg^{-1}K^{-1}$)	0.71
Thermal conductivity ($W\ m^{-1}K^{-1}$)	6 (400 K) ∼ 3 (1600 K)
Specific gravity ($10^3\ kg\ m^{-3}$)	5.5
Melting temperature (K)	∼ 2200

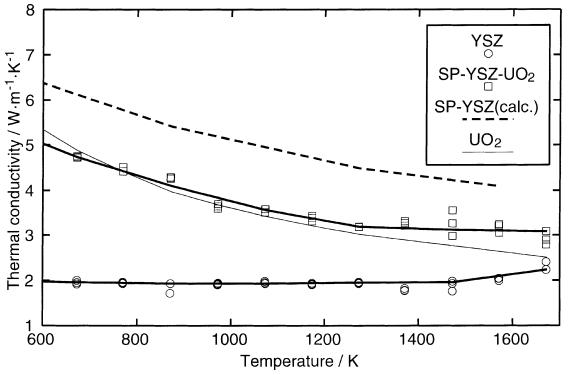

Fig. 2. Thermal conductivity of stabilized zirconia (YSZ), Zr-ROX and UO_2.

temperature dependence of the thermal conductivity of Zr-ROX [5] is shown in Fig. 2, in comparison with yttria stabilized zirconia (YSZ) [5] and UO_2 [6]. In this figure, 'SP–YSZ–UO_2' denotes the conductivity value of Zr-ROX measured for a 37 mol% spinel – 49 mol% YSZ – 14 mol% UO_2 composition. The line 'SP–YSZ(calc.)' is the estimated thermal conductivity of higher spinel content (56 mol%) [3], based on the other measurements.

3.1. Reactivity insertion accident

The reactivity insertion accident (RIA) analysis of Zr-ROX PWR with the EUREKA-2 code [7] showed that a reactivity insertion under hot zero power condition at EOC leads to a maximum fuel enthalpy far larger than the limiting value of about 960 kJ kg^{-1} (230 kcal kg^{-1}) in the conventional UO_2 PWR. When the Doppler reactivity is five times larger, the fuel enthalpy becomes less than 960 kJ kg^{-1}.

The RIA analysis results for improved Zr-ROX cores are compared with those for conventional UO_2 fuel PWR in Table 3. It is shown in this table that ThO_2 additive can reduce the maximum fuel enthalpy (H) to less than 960 kJ kg^{-1}, and the maximum fuel temperature (T_{FC}) to less than 2200 K. The fuel enthalpy is still two times as large as that in the UO_2 PWR case. Considering the density of Zr-ROX fuel, i.e. less than 6×10^3 kg m^{-3}, the energy release of 960 kJ kg^{-1} in Zr-ROX fuel might be equivalent to about 500 kJ kg^{-1} in UO_2 fuel. The energy release per unit volume is also shown in Table 3, and the value of Zr-ROX–ThO_2 fuel approaches that of UO_2 fuel. The actual enthalpy condition for Zr-ROX pin failure under RIA is not yet known, and is now being studied in the Nuclear Safety Research Reactor (NSRR) at JAERI by pulse irradiation experiments [8]. Both from the viewpoints of enthalpy (per unit volume) and temperature, the RIA behavior in the Zr-ROX(Er)–UO_2 fuel core is comparable to that of the UO_2 fuel core. In 1/3 ROX fuel core, the fuel enthalpy slightly exceeds 960 kJ kg^{-1}, and the fuel temperature rises as high as the melting point. Further improvement will be necessary for this core.

Table 3
Maximum fuel enthalpy (H) and fuel temperature (T_{FC}) in Zr-ROX PWR RIA event

	H (kJ kg^{-1})	H (10^6 kJ m^{-3})	T_{FC} (K)
Weapons-Pu			
Zr-ROX	$\gg$960		$\gg$220
Zr-ROX(Er)–15UO_2	810	4.5	1700
Zr-ROX–24ThO_2	940	5.2	1950
1/3Zr-ROX+2/3UO_2	1020	5.7	2100
Reactor-Pu			
Zr-ROX(Er)–8UO_2	800	4.5	1700
UO_2	390	4.3	2080

3.2. Loss of coolant accident

The loss of coolant accident (LOCA) in a Zr-ROX fueled PWR was analyzed by using the RETRAN2 code [9]. The analyzed system and event are four-loop type 1100 MWe class PWR and a cold-leg large break LOCA event at BOC and EOC, respectively. Similarly to the RIA case, the severer result in the original Zr-ROX case was obtained at EOC, and the peak cladding temperature (T_{PC}) rises to more than 1500 K, higher than the limit of 1470 K (1200°C). The results of a sensitivity analysis showed that the T_{PC} is sensitive to the thermal conductivity of the fuel and to the power peaking in the core [1]. The power peaking factor has the largest effect on T_{PC}. By decreasing the power peaking by 0.85 times smaller than the original ($2.7 \times 0.85 = 2.3$), it is possible to reduce the T_{PC} by about 400 K, and thus well below 1470 K.

The LOCA analysis results for the modified Zr-ROX fuel cores are shown in Table 4, in comparison with the conventional UO_2 PWR results. In all of the modified

Table 4
Peak cladding temperature (T_{PC}) during LOCA of a a Zr-ROX fueled PWR

	T_{PC} (K)
Weapons-Pu	
Zr-ROX	>1470
Zr-ROX(Er)–15UO_2	1090
Zr-ROX–24ThO_2	1240
1/3Zr-ROX+2/3UO_2	1240
Reactor-Pu	
Zr-ROX(Er)–8UO_2	1090
UO_2	1080

Zr-ROX fuel cores, the T_{PC} is successfully reduced to less than 1470 K. In the Zr-ROX(Er)–UO_2 fuel core, the cladding temperature is lower than in the other cores mainly because of the smaller power peaking factor. In this case, T_{PC} becomes as low as that of the UO_2 fuel case. For improving the temperature behavior of fuel and cladding in both RIA and LOCA, UO_2 with Er_2O_3 additive is considerably effective. In the other two modified cores, T_{PC}s are not as low as that in the conventional UO_2 core. Better LOCA behavior may be preferable in these cores by further reducing the power peaking factor.

4. Fuel temperature under operation condition

The melting temperature of Zr-ROX fuel is much lower than those of UO_2 and MOX fuels, and the large power peaking may lead to a severe problem with fuel temperature even under nominal operation conditions.As shown in Table 1, the power peaking factor of Zr-ROX fuel core can be improved to 2.1 by considering UO_2 as an additive in Zr-ROX fuel. The fuel temperature within the Zr-ROX fuel pellet was estimated by using the GAPCON-THERMAL2-HC code [10] for a single channel model. Fig. 3 shows the fuel temperature radial distribution in the fuel pin at the peak power position. The temperature was calculated with the thermal conductivity of SP–YSZ–UO_2 shown in Fig. 2. The average linear heat rate of 18 kW m^{-1} is assumed, which corresponds to the condition of a conventional PWR. As can be seen in this figure, the peaking factor of 2.1 gives a maximum fuel temperature less than the melting point.

It is possible to further reduce the fuel temperature by improving the peaking factor. When the peaking factor is 1.7, the maximum fuel temperature becomes about 1700 K and is less than the melting point by about

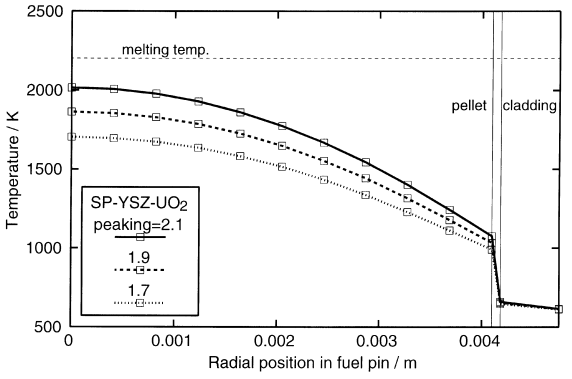

Fig. 3. Fuel temperature at peak power position calculated with different power peaking factors at the beginning of life.

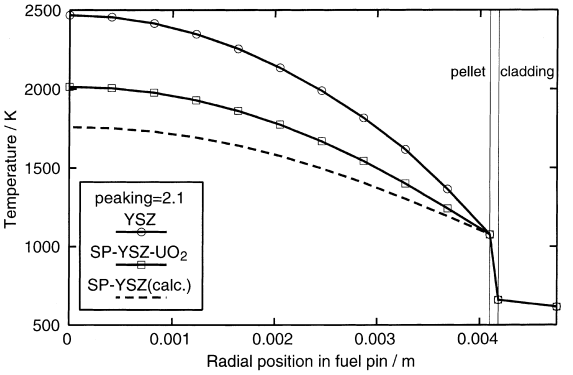

Fig. 4. Fuel temperature at peak power position calculated with different fuel thermal conductivities at the beginning of life.

500 K. But such a peaking factor may be difficult to realize in reactor core design. Another approach to fuel temperature reduction is to increase the fuel thermal conductivity. Fig. 4 compares the fuel temperatures calculated with different fuel thermal conductivities. In this figure, power peaking factor is 2.1 and the average linear heat rate is 18 kW m^{-1}. When the conductivity of Zr-ROX with higher spinel content SP–YSZ(calc.) is considered, the maximum fuel temperature becomes about 1700 K, the same level as the peaking factor 1.7 case in Fig. 3. In Fig. 4, the fuel temperature estimated for stabilized zirconia YSZ thermal conductivity is also shown. The temperature rises to about 2500 K. However, the melting point of stabilized zirconia is nearly 3000 K, so the fuel temperature seems sufficiently lower than the melting point. To suppress the fission gas release, however, the fuel temperature should be still lower. From this point of view, further studies seem necessary to reduce the Zr-ROX fuel temperature.

5. Plutonium transmutation

The additives ThO_2 and UO_2 effectively increase the negative Doppler coefficient of Zr-ROX fuel. With these additives, however, the plutonium transmutation rate decreases, because both ^{232}Th and ^{238}U produce fissile nuclides ^{233}U and ^{239}Pu, respectively.

The amount of plutonium transmuted (the difference between input and output amount) and the transmutation rate (the ratio of transmuted/input amount) per 1 GWe reactor power per 300 d of equivalent full power operation is shown in Table 5 for Zr-ROX and MOX fueled PWR cores. In all cases, the assumed discharge burnup is about 1170 d with 18 kW m^{-1} average linear heat rate, corresponding to about 45 GWd t^{-1} in the MOX case. In this table, ROX fuel data were obtained with 2-dimensional core burnup calculations, and the

Table 5
Input and transmuted amount (t GWe^{-1} per 300 d of full power operation) of plutonium in ROX and MOX fueled cores (1170 d discharge burnup, corresponding to 45GW $d{\cdot}t^{-1}$ in MOX core)

		Weapons-Pu			Reactor-Pu		
		Input	Transmuted		Input	Transmuted	
		(t)	(t)	(%)	(t)	(t)	(%)
1/3Zr-ROX+2/3UO_2	Pu-239	0.34	0.25	75	0.27	0.18	67
	Total Pu	0.36	0.16	43	0.46	0.19	41
(in 1/3ROX only)	Pu-239	0.34	0.33	99	0.27	0.26	98
	Total Pu	0.36	0.31	86	0.46	0.34	74
Zr-ROX(Er)–UO_2	Pu-239	0.93	0.85	92	0.80	0.70	88
	Total Pu	0.99	0.69	69	1.36	0.81	60
Zr-ROX–ThO_2	Pu-239	0.97	0.94	97	0.80	0.75	93
	Total Pu	1.03	0.82	79	1.37	0.90	66
MOX (once-through)	Pu-239	0.88	0.56	63	0.99	0.44	45
	Total Pu	0.94	0.30	32	1.69	0.41	25
(recycle once)	Pu-239	0.76	0.54	71	0.76	0.44	58
	Total Pu	0.81	0.35	43	1.30	0.48	37
(recycle three times)	Pu-239	0.66	0.52	79	0.61	0.42	69
	Total Pu	0.70	0.40	57	1.05	0.53	50

ROX results with two-dimensional core calculations, and MOX results with cell calculations.

MOX data with cell burnup calculations. Though resulting from different methods, these data are still useful to compare the plutonium transmutation capability of the fuels. With the condition of three-batch-refueling, the plutonium enrichment was adjusted in the core burnup calculations to obtain the effective multiplication factor (k_{eff}) of 1.0 at the end of refueling cycle, i.e. at 390 d. In the cell burnup calculations, the plutonium enrichment was determined by assuming the core averaged k_{eff} at EOC (k_{EOC}). From the k_{eff} values obtained by the cell burnup calculation with the geometrical buckling, k_{EOC} is defined as the average of k_{eff}s at 390, 780 and 1170 d burnup time. The plutonium enrichment of the MOX PWR cell is then adjusted so that k_{EOC} becomes 1.035. Here, the value 1.035 was decided by comparing the results of the cell and the core calculations.

For MOX fuel, plutonium recycling is considered. Here, core characteristics such as the void reactivity coefficient are not estimated, and this table does not mean that plutonium can be recycled in MOX fueled LWRs without any difficulties up to three times. In the recycling calculation, as schematically described in Fig. 5, it is assumed that all the output amount of plutonium of ith cycle, O_i, is recycled to the next cycle $i+1$, and to keep $k_{\text{EOC},i+1} = 1.035$, the necessary amount of 'fresh' plutonium, I_{i+1}, is added. Thus, the net input amount of plutonium from 1st to Nth cycles becomes, $\Sigma_{i=1}^{N} I_i$ and the net output amount of plutonium after N cycles is O_N. In Table 5, there are shown the input amount per cycle, $\Sigma_{i=1}^{N} I_i/N$, and the transmuted amount per cycle, $\left(\Sigma_{i=1}^{N} I_i - O_N\right)/N$. From this table, it can be seen the plutonium transmutation rate is increasing with increasing the number of recyclings. But it seems there is a saturating value in the transmutation rate.

In Zr-ROX fuel assemblies in the 1/3 Zr-ROX + 2/3 UO_2 core, the plutonium transmutation rate is extremely high. This is the advantage of this heterogeneous core. The plutonium transmutation rates in the full core Zr-ROX cases are also larger than in the full-MOX core, but the rate decreases with the addition of ThO_2 or UO_2. Particularly with UO_2 additive, ^{239}Pu is newly produced from ^{238}U. As a result, the difference in the plutonium transmutation rate becomes smaller between the UO_2-added Zr-ROX and the plutonium recycled MOX cases. The full-ROX fueled cores, however, can transmute much larger amount of plutonium annually, i.e. 1.5–2.0 times as much as the MOX core, and about three times larger than in the 1/3 Zr-ROX core. From the viewpoints

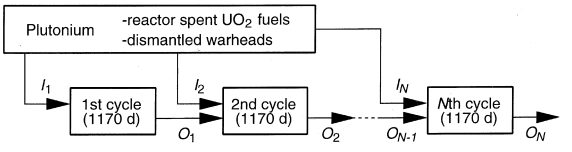

Fig. 5. Plutonium recycling in the MOX PWR in Table 5 (I_i: input Pu amount of ith cycle, O_i: output plutonium amount of ith cycle).

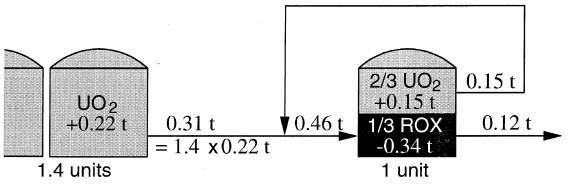

Fig. 6. Reactor-grade plutonium balance in the UO_2 PWR and 1/3 ROX+2/3 UO_2 PWR system (values in t per 1 GWe reactor power per 300 d of effective full power operation, '+' means Pu production and '−' Pu transmutation).

of both transmutation rate and annual transmutation amount, it is better to use ThO_2 additive than UO_2. It should be noted, however, that there is produced another fissile isotope ^{233}U. In Zr-ROX-24ThO_2 case, about 0.1 t of ^{233}U is produced per 1 GWe per 300 d.

The total plutonium amount loaded in the 1/3 Zr-ROX + 2/3 UO_2 core is only about 1/3 compared to the full core ROX or MOX cases. For the reactor-grade plutonium burning case, 0.46 t plutonium can be input in the 1/3 Zr-ROX + 2/3UO_2 core every 300 d per 1 GWe reactor power. At the same time in 2/3 UO_2 assemblies, 0.15 t plutonium is newly produced. When the plutonium in spent UO_2 fuels in this core is recycled to the Zr-ROX fuel assembly in the same core, there is still $0.46 - 0.15 = 0.31$ t of acceptable amount of plutonium from conventional UO_2 cores. The production rate of plutonium in the conventional UO_2 PWR spent fuels of 45 GWd t^{-1} burnup is calculated to be about 0.22 t GWe^{-1} per 300 d. Thus, it can be estimated that one unit of 1/3 Zr-ROX + 2/3 UO_2 PWR can burnup discharged plutonium from $0.31/0.22 = 1.4$ units of UO_2 PWR, while producing $0.46 - 0.34 = 0.12$ t surplus Pu in the 1/3 Zr-ROX assemblies (Fig. 6). On the other hand, one unit of both Zr-ROX–UO_2 and Zr-ROX–ThO_2 fueled PWR can accept plutonium from about $1.36/0.22 = 6.2$ units UO_2 PWR's spent fuels, leaving behind 0.55 and 0.47 t Pu, respectively.

6. Conclusion

In Zr-ROX–LWR cores, different characteristics from UO_2 core have caused several problems under accidents conditions and even in the nominal operation condition. Some improvements of the Zr-ROX core characteristics were discussed here.

For the improvement of the small negative Doppler reactivity coefficient, the additives such as UO_2 or ThO_2 were considered, while a Zr-ROX + UO_2 heterogeneous core was also discussed. The large burnup reactivity swing of Zr-ROX core can be moderated with Th or U additives, and it becomes easier for power distribution flattening. For power peaking reduction, Er_2O_3 additive was also found to be effective.

Reactivity insertion accident (RIA) and loss of coolant accident (LOCA) analyses showed the effectiveness of these improvements. Especially with the combination of UO_2 (about 15 mol% in weapons plutonium and 8 mol% in reactor plutonium) and Er_2O_3 (about 0.3 mol%) additives, it is proved possible to achieve transient behaviors comparable to the UO_2 core. In both accidents, 1/3 Zr-ROX + 2/3 UO_2 core shows only insufficient progress in transient performance, due to the large power peaking factor. This could be reduced by considering Er_2O_3 additive in Zr-ROX, which have a pin-by-pin density distribution within Zr-ROX assembly. A pin-by-pin Pu enrichment distribution may also be necessary. The fuel temperature in nominal operation condition is lower than the melting temperature of Zr-ROX. To further decrease the fuel temperature, a reduction of power peaking factor is important, and at the same time an increase of fuel thermal conductivity by adjusting the fuel composition.

In the 1/3 Zr-ROX core, the plutonium transmutation rate in Zr-ROX assembly is very high, which is a very important factor for plutonium once-through disposal. By considering UO_2 or ThO_2 in the fuel, the transmutation rate is deteriorated slightly. The advantage of the full-ROX core with these additives is the larger amount of annual transmutation. For rapid annihilation of plutonium, full-ROX cores have an advantage.

In these studies, a feasibility of the Zr-ROX LWR was shown. In future studies, an optimization of the fuel composition will be necessary from several view points of fuel temperature, reactor safety and plutonium transmutation, based on the results of fuel studies, pulse irradiation experiments, etc.

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