PARTITIONING AND TRANSMUTATION OF SPENT NUCLEAR FUEL BY PEACER

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Abstract

Challenges of managing long-living spent nuclear fuel wastes are to be technically overcome by a partitioning and transmutation system named as PEACER that is an abbreviation of its design goals: Proliferation-resistant, Environment-friendly, Accident-tolerant, Continuable and Economical Reactor. PEACER has been developed to manage spent nuclear fuels by uniquely combining Pb-Bi cooled critical reactor and pyrochemical partitioning process. The PEACER design with thermal traps can transmute long-living transuranic (TRU) elements and fission products including ^{99}Tc and ^{129}I at least two times the rate they are produced from an LWR having the same electric power rating. Based on asymptotic core outlet temperature estimated using Wade, et al's method, the safety characteristics of PEACER was evaluated for loss of heat sink accident and loss of flow accident without scram. Result shows that PEACER has an equivalent or better safety characteristics compared with the sodium cooled ALMR burner design. The pyrochemical partitioning process has been conceptually designed so that the transmutation of spent LWR fuels in PEACER can produce mainly low-level waste (U.S. Class C waste) for near-surface burial. Judged from a system analysis, an overall decontamination factor for transuranic elements has to be greater than 2×10^4 in order to assure the economy. Under the condition, the volume of the final low-level waste is estimated to be small enough to make PEACER concept viable for densely populated countries.

Introduction

The spent nuclear fuel of current nuclear reactors is one of challenging issues for the continuous utilisation of nuclear power. As a technical solution of this problem, deep geological disposal has been suggested and studied for decades. This approach requires highly qualified sites that can tolerate great uncertainties in the scientific predictions of geological behaviour over 10 000 years. As an alternative approach, the partitioning-and-transmutation (P&T) has been studied in U. S. A., Europe, and Japan since 1990s. [1] P&T options assume that transuranic elements (TRU) are first partitioned from the spent nuclear fuel and then transmuted in dedicated nuclear fission systems. Final wastes are disposed of at the repository. In a geological repository, technetium and iodine are known as the most hazardous nuclides because of their high solubility in ground water. [2] Therefore, in most P&T options, they are also recycled for transmutation into stable nuclides.

In order to realise P&T option of spent nuclear fuel in Korea, we proposed a transmutation concept designated as PEACER that is acronym of Proliferation-resistant, Environment-friendly, Accident-tolerant, Continuable and Economical Reactor. [3] The PEACER concept has been developed on the basis of two key technologies that are pyroprocess-based partitioning system and lead-bismuth cooled transmutation reactor. [3] In order to make this concept more attractive, it is hoped to convert all the final waste into the class of low-level waste (LLW) that is defined in NRC 10CFR61 [4] or IAEA Safety Series No. 111-G-1.1. [5] This paper describes the present status of the research on PEACER.

PEACER conceptual design

The conceptual design of PEACER has been developed by combining the Integral Fast Reactor (IFR) approach with the heavy liquid metal cooled reactor technology. As its basic core design, an LWR-type square-lattice is employed with metallic fuel elements having high pitch-to-diameter ratio in order to accommodate the viscosity nature of lead-bismuth coolant. Both uranium and TRU were used as fuel materials. To maximise the transmutation rate of TRU elements, the production of TRU in the core by neutron capture reaction of ^{238}U must be minimised. PEACER selected leakage-enhanced core geometry by the sparse lattice and low core height-to-diameter ratio so that the excess neutrons do not contribute to TRU production. Parameters of a reference core characteristics are listed in Table 1. [3] The reference core layout of PEACER is shown in Figure 1.

Plant layout of PEACER is shown in Figure 2. [6] Intermediate heat exchanger was removed from primary heat transfer system because of non-reactive Pb-Bi coolant. Inlet coolant temperature of PEACER has been chosen as 300 C in order to enhance material compatibility. Total mass flow rate of Pb-Bi is 106.12 ton/sec. A balance-of-plant (BOP) of PEACER has been chosen to be similar to PWR's BOP system so that development cost of BOP is greatly reduced.

A reference pyrochemical process for PEACER is based on LiCl-KCl molten-salt and liquid cadmium cathode as shown in Figure 3. [7] LiCl-KCl based pyroprocess has been developed at ANL [8,9] and recently has been redesigned for ATW system. [10] Key processes of Pyrochemical partitioning process are electrolysis process for TRU recovery that is electrorefining or electrowinning and reductive extraction process for waste treatment. Decontamination of TRU in LLW is subject to the combination of electrolysis and reductive extraction process. Decontamination factor (DF) is introduced for indication of process performance. Overall DF in pyrochemical partitioning is defined as the ratio of mass of TRU loaded into the process to TRU lost into waste stream. DF is a function of TRU loss fractions in electrorefining process and reductive extraction process. Asymptotically, DF is expressed as a reciprocal of a product of TRU loss fractions in electrorefining process and in reductive extraction process. According to IFR process, [8] 99% of the TRU is recovered by electrorefining process. Of the 1% that is discharged from the electrorefiner, more than 90% is recovered in the reductive extraction process. As a result of IFR process, more than 99.9% of discharged TRU is recovered. Hence, it could be possible to obtain 1% of TRU loss fraction in the electrorefining process. CRIEPI has evaluate distribution coefficient of actinides and lanthanides in LiCl-KCl/Bi and LiCl-KCl/Cd. CRIEPI's results show that more than 99.9% of TRU are recovered using 5 stages counter current extraction in LiCl-KCl/Bi system. [11] Therefore, it could be possible to obtain 0.1% of TRU loss fraction. In PEACER pyrochemical partitioning process, it is assumed that 1% of the TRU is discharged from the electrorefiner into reductive extraction process and 0.1% of the TRU is lost into waste stream in the reductive extraction process. Then overall DF becomes 100 000. Based on LiCl-KCl based pyroprocess and 100 000 of DF, PEACER pyrochemical partitioning flowsheet is conceptually designed as shown in Figure 3.

A PEACER park, as shown in Figure 4, consists of four Pb-Bi cooled reactors and two pyroprocessers. Each reactor has a refueling shutdown for 35 days every year. One pyroprocesser can treat spent fuels from all four reactors. The second pyroprocesser is either on standby or on maintenance outage.

Table 1. **Reference core parameters of PEACER**

Thermal power		1 560 MWt
Electric power		550 MWe
Thermal efficiency		35.3%
Coolant		Liquid Pb-Bi eutectic alloy
Core diameter		489.6 cm
Core active height		50.1 cm
Scram system		B_4C control assembly
Effective full power day		330 days
Cycle length		365 days
Capacity factor		90%
No. of enrichment zone		2
Average power density		204.5 MW/m^3
Fuel composition	Inner core	U-TRU-Zr (59.2-30.0-10.8)
	Outer core	U-TRU-Zr (55.2-33.9-10.9)
Fuel cladding material		HT-9
Fuel smeared density		67% (< 73%)
Fuel rod pitch/Assembly pitch		1.46 cm / 20.4 cm
Fuel pin per assembly		196 (14×14 square cell)
Number of fuel assembly	Inner core fuel	184
	Outer core fuel	176
	Inner radial reflector	32
	Outer radial reflector	100
	Radial shield	192
	Axial shield	360
	Control/shutdown	20

Figure 1. **Reference core model of PEACER**

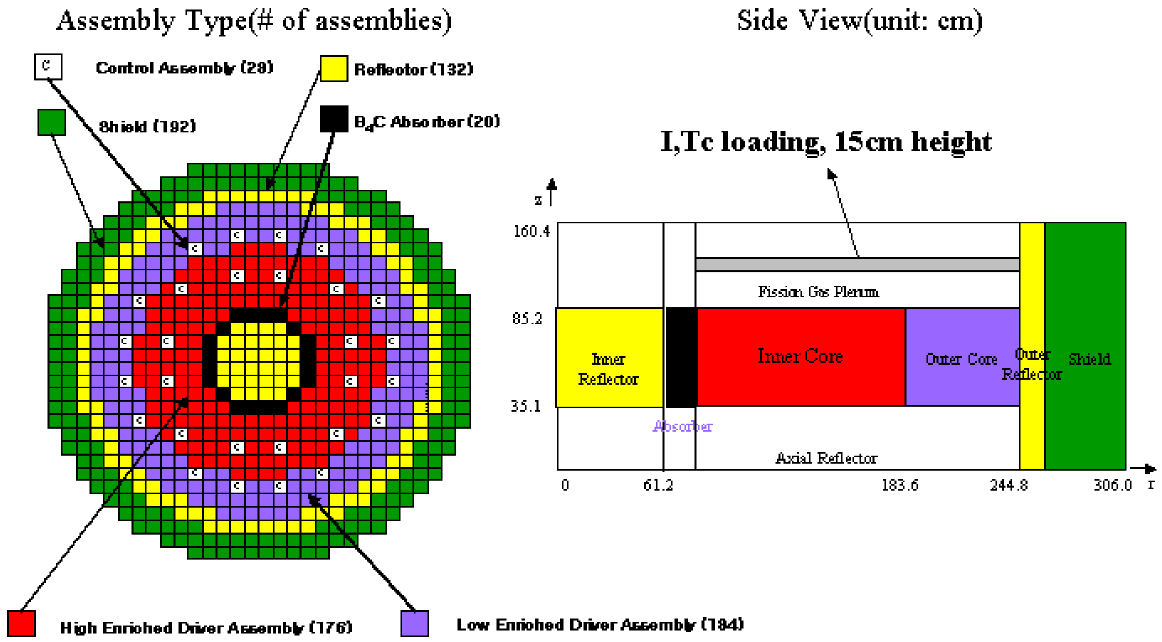


Figure 2. **Plant schematics of PEACER**

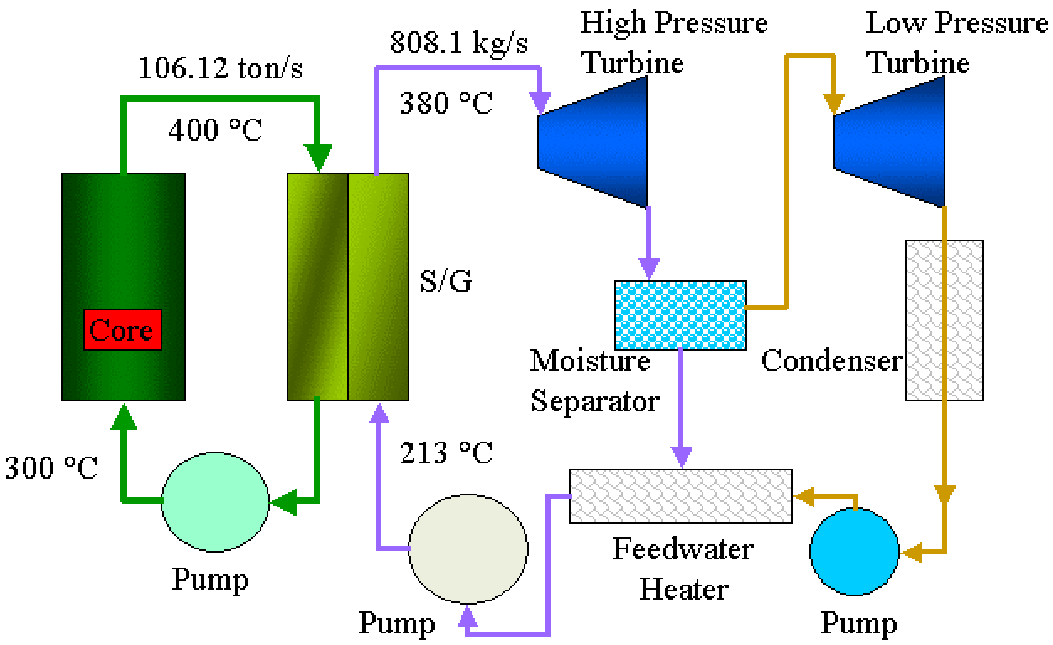


Figure 3. **Concept of pyrochemical separation process for PEACER**

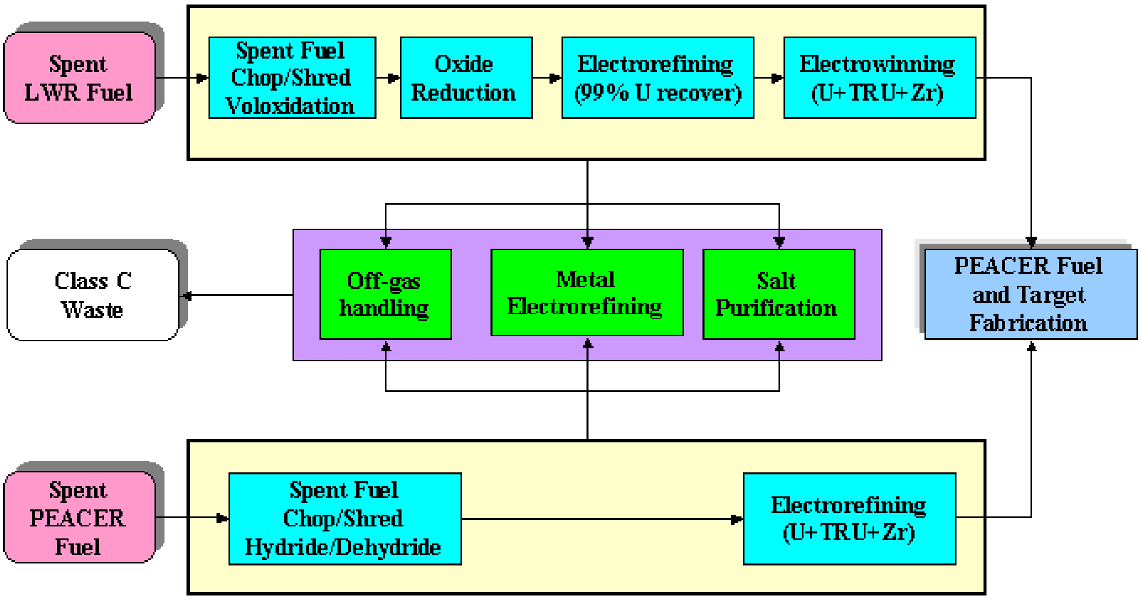
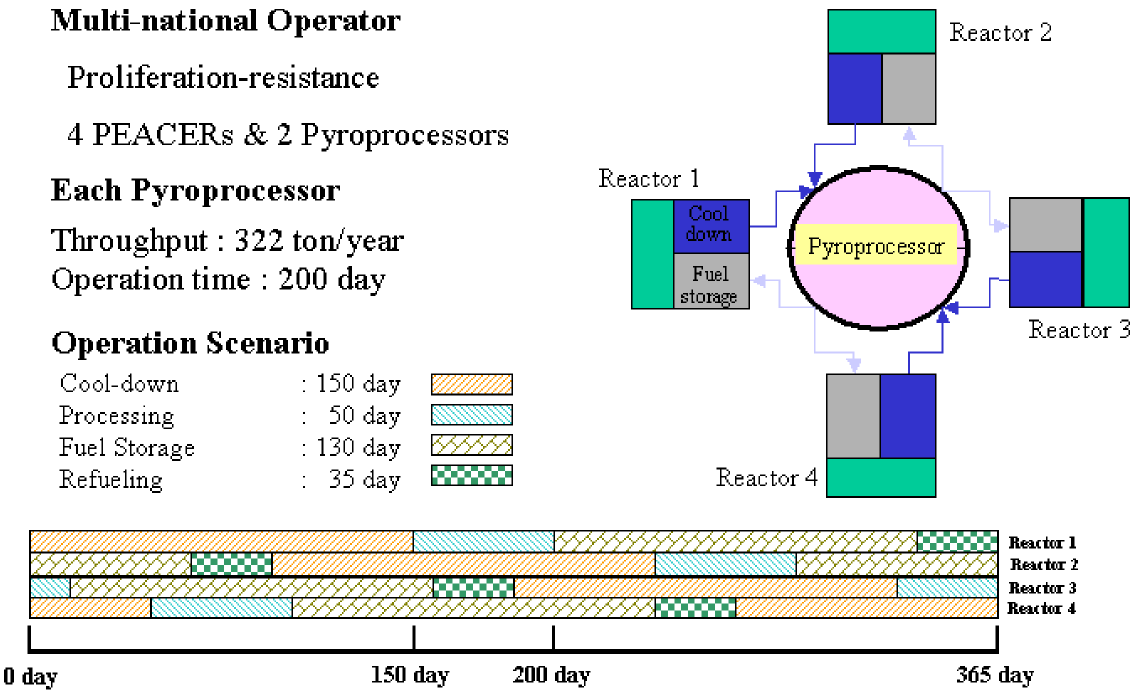


Figure 4. **Concept of PEACER transmutation park**



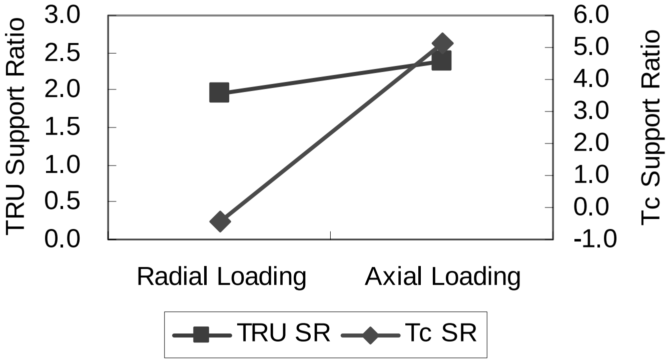
Results of design analysis

Proliferation-resistance: The proliferation-resistance was embedded by both material and institutional barriers. The performance parameter for material barrier was odd number isotope contents in Pu after discharge from the PEACER core (Pu Odd Ratio, OR). The target value for OR is set at 0.5

for a fuel discharge condition. Such a low OR would make the material unfavourable for theft and diversion by any low technology group. In PEACER reference core, OR of Pu decreases down to about 0.5 at EOEC. [4] This is much lower than that of LWR spent fuel, 0.7. The difference between two OR values at the beginning of equilibrium cycle (BOEC) and ending of equilibrium cycle(EOEC) is fairly constant at about 0.04 for the range of this study. [3] A PEACER transmutation park is proposed to be operated by a multinational collaboration as this will serve as a dependable institutional barrier, as shown in Figure 4.

Environmental-friendliness: The environment-friendliness is measured by Support Ratio (SR). SR is defined by the number of LWRs with its spent fuel TRU elements transmuted by a PEACER at the same electric power rating. The target value for SR is set as 2.0 for an equilibrium core. The leakage-enhanced sparse core design of PEACER also helped increase SR and meet the goal. The environment-friendliness has been concerned with TRU. Long-living fission products, especially ^{99}Tc, ^{129}I, ^{137}Cs and ^{90}Sr are identified as important isotopes because a very large stabilisation volume is required to meet the low-level waste criteria if they are included in the final waste form. In order to transmute fission products ^{99}Tc and ^{129}I, a pan-cake shaped thermal neutron trap was introduced between the top of fuel elements and the bottom of axial reflector as shown in Figure 1. Metal hydroxides are assumed to constitute a suitable medium to thermalise neutrons while Tc and I loaded in the region. PEACER with the axial thermal trap shows sufficient transmutation rate: SR of Tc reaching 5 and SR of TRU reaching 2.2, as shown in Figure 5. An alternative thermal trap design is also considered. The thermal trap can be applied to the inner reflector region in order to utilise some of leakage neutron for transmutation. However the radial loadings resulted in a negative value for SR, making the option un-useful. If we wish to utilise radial loading option, pan-caked core design must be significantly modified.

Figure 5. **Support ratio of PEACER reference core**



In PEACER transmutation park, waste volume after transmutation has been estimated with simple analysis model and scenario that is 20 LWRs with 40 years life time and 3 PEACER transmutation park with 60 years life time, as shown in Figure 6. [7] The stabilisation volume required for disposal site was estimated as shown in Figure 7. [7] The volume requirement of the repository is 1 651,333 m^3 with TRU decontamination factor of 10^4 for the transmutation of spent fuels produced from 20 LWR's for 40 years. The volume is reduced to 145 493 m^3 when Cs and Sr are separated for 200 years of interim cooling and the decontamination factor is increased to 10^5. The latter scheme is expected to be more practical for partitioning and transmutation using PEACER.

Figure 6. **Preliminary analysis of waste volume produced by PEACER transmutation park**

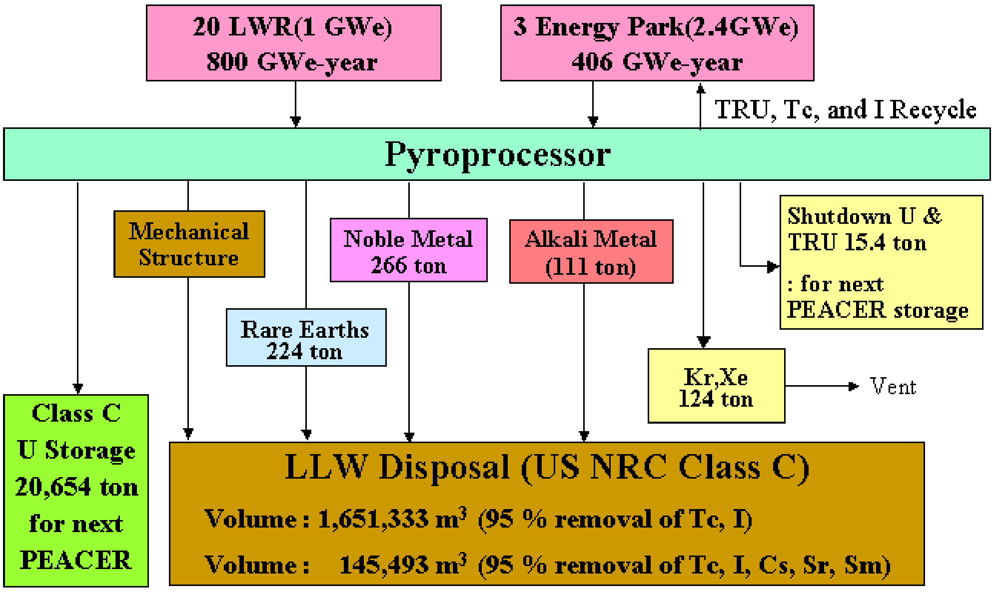
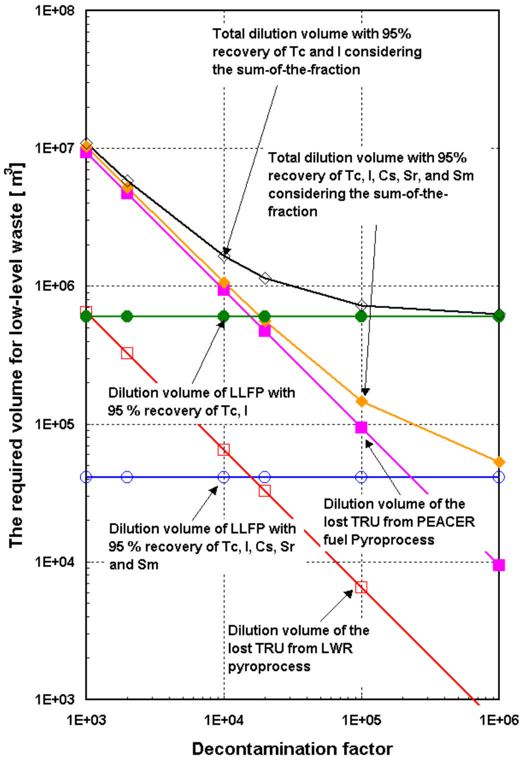


Figure 7. **Volume requirement for LLW stabilisation**



Accident-tolerance: A preliminary calculation for the natural convection ability of lead-bismuth coolant also showed favourable characteristics. [6] Safety characteristics of the conceptual design have been examined for two types of design basis accidents. Various reactivity coefficients and asymptotic coolant outlet temperature were calculated for LOFWS (Loss of Flow Without Scram) and LOHSWS

(Loss of Heat Sink Without Scram). Wade, *et al.* method was adopted to examine the core self-regulation characteristics. [8,13] In this method, A, B, C are integral reactivity parameters that can be determined from the thermal-hydraulic condition and reactivity coefficients. Theses are defined as follows:

$$A = \left[\frac{\partial \delta\rho}{\partial P}\right]_{(P/F, T_{in}\ \text{Constant})} = (\alpha_D + \alpha_H)\Delta T_f \qquad (1)$$

$$B = \left[\frac{\partial \delta\rho}{\partial (P/F)}\right]_{(P, T_{in}\ \text{Constant})} = (\alpha_D + \alpha_H + \alpha_C + 2\alpha_R)\frac{\Delta T_f}{2} \qquad (2)$$

$$C = \left[\frac{\partial \delta\rho}{\partial T_{in}}\right]_{(P, F\ \text{Constant})} = (\alpha_D + \alpha_H + \alpha_C + \alpha_R) \qquad (3)$$

where, $\delta\rho$ is reactivity difference, A is net power reactivity increment, B is power/flow coefficient of reactivity, C is inlet temperature coefficient of reactivity, and P and F are normalised power and flow, respectively. As reactivity coefficients, α_D, α_H, α_C , and α_R indicate effects of Doppler, axial fuel expansion, coolant density, and radial expansion, respectively. T_f ($=T_{in}+\Delta T_c/2+\Delta T_f$) is average fuel temperature, ΔT_f is difference between average fuel temperature and average coolant temperature, T_{in} is coolant inlet temperature, T_{out} ($=T_{in}+\Delta T_c$) is coolant outlet temperature, and ΔT_c is coolant temperature rise through the core.

Asymptotic core outlet temperatures during the postulated accidents can be determined from A, B and C. The calculated results are shown in Table 2. The outlet temperature of PEACER in LOHSWS and LOFWS is much lower than that of a sodium-cooled burner design, ALMR. [9] As showed in Table 2, outlet temperatures of PEACER in both LOHSWS and LOFWS are sufficiently below 970 K that corresponds to the temperature of strong fuel-cladding chemical interaction. In order to closely compare the safety characteristics, PEACER reference core having exactly the same thermal-hydraulic condition as the ALMR burner was also analysed as shown in Table 1. Core outlet temperatures of PEACER are shown to be lower than corresponding values for ALMR under the accidents. Wade, et al proposed five safety criteria required on the integral parameters. [9] Three of five passive reactivity shutdown performance requirements can be determined at the current stage of PEACER design. They are 1) A, B, C; all numbers must be negative, 2) $A/B < 1$, and $1 < C\ \Delta T_c/B < 2$. All these criteria are met with the conceptual design of PEACER, as shown in Table 2. Therefore the safety characteristics are considered to be satisfactory for the conceptual design.

Summary and conclusions

PEACER design concept was reviewed and recent results of design analysis were reported. The earlier PEACER design was focused on TRU elements for transmutation. Incorporation of thermal traps into PEACER core can transmute long-living fission product including ^{99}Tc and ^{129}I at sufficiently high support ratio. This feature, if combined with a highly efficient partitioning process, can lead to the capability of qualifying the final waste form as the low-level waste class. Total volume of the low-level waste was estimated to be 145 493 m^3 for the case of 20 LWR's operated for 40 years when Cs and Sr are separated for 200 years interim cooling and DF on TRU's becomes 10^5. Judged from the analysis, an overall decontamination factor for transuranic elements has to be greater than 2×10^4 in order to assure the economy. Under the condition, the volume of the final low-level waste is estimated to be small enough to make PEACER concept viable for densely populated countries. Three PEACER park, each with four reactors and two pyroprocessers are required to operate for 60 years to treat the spent fuel. Core safety characteristics were determined based on the reactivity coefficients and thermal-hydraulic analysis. Both integral parameters (A, B, C) and core outlet temperature under two types of design basis accidents showed that the safety characteristics of PEACER using Pb-Bi as coolant can be better than that of a sodium-cooled burner reactor.

Table 2. **Comparison of reactivity coefficients, integral parameters and core outlet temperature during the design basis accidents for PEACER and ALMR**

Parameters	PEACER reference core	ALMR metal fuel burner	PEACER reference core with ALMR thermal condition
T_f [K]	770	774	774
ΔT_f [K]	120	80	80
ΔT_c [K]	100	129	129
T_{in} [K]	600	629	629
T_{out} [K]	700	758	758
α_D [¢/K]	-0.05009	-0.114	-0.05009
α_H [¢/K]	-0.03740	-0.117	-0.03740
α_R [¢/K]	-0.28857	-0.291	-0.28857
α_C [¢/K]	0.004704	0.0363	0.004704
A [¢]	-6.011	-9.1	-4.007
B [¢]	-33.00	-50.1	-42.57
C [¢/K]	-0.3714	-0.486	-0.3714
A/B	0.18215	0.18	0.09413
$C\Delta T_c/B$	1.125455	1.2514	1.125455
T_{out} in LOFWS [K]	722	781	770.1
T_{out} in LOHSWS [K]	705	751	704

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