A PARTICLE-BED GAS-COOLED FAST REACTOR CORE DESIGN FOR WASTE MINIMISATION

E.A. Hoffman, T.A. Taiwo, W.S. Yang and M. Fatone Reactor Analysis and Engineering Division Argonne National Laboratory, Argonne, IL 60439, USA

Abstract

The issue of waste minimisation in advanced reactor systems has been investigated using the Particle-Bed Gas-cooled Fast Reactor (PB-GCFR) design being developed and funded under the U.S. Department of Energy Nuclear Energy Research Initiative (USDOE NERI) Programme. Results indicate that for the given core power density and constraint on the maximum TRU enrichment allowable, the lowest amount of radiotoxic transuranics to be processed and hence sent to the repository is obtained for long-life core designs. Calculations were additionally done to investigate long-life core designs using LWR spent fuel TRU and recycle TRU, and different feed, matrix and reflector materials. The recycled TRU and LWR spent TRU fuels give similar core behaviours, because of the fast spectrum environment which does not significantly degrade the TRU composition. Using light elements as reflector material was found to be unattractive because of power peaking problems and large reactivity swings. The application of a lead reflector gave the longest cycle length and lowest TRU processing requirement. Materials compatibility and performance issues require additional investigation.

1. Introduction

A solution must be found for the LWR spent nuclear fuel (SNF) that is accumulating in spent-fuel ponds and dry-storage sites, if nuclear is to play a prominent role in future energy generation in the U.S. It is currently planned that the spent fuel would ultimately be stored in a waste repository (Yucca Mountain in the U.S. case). The capacity of this repository has been set by law and there are requirements to determine the need for an additional repository by 2007. Clearly, if nuclear maintains its share of the electric generation capacity or continues to grow, the limit set for Yucca Mountain will be exceeded. This would necessitate additional repositories, unless approaches can be found to increase the effective repository capacity. Given the current difficulties encountered in setting up the first repository, the waste to be sent to the repository needs to be minimised. An approach for this is to recycle and incinerate the radiotoxic transuranics (TRU) in nuclear power reactor systems. Nuclear waste minimisation is one of the key attributes that have been defined for Generation IV reactors. In addition to waste minimisation, the burning of the legacy TRU in advanced systems should contribute to increasing the repository load capacity.

The issue of nuclear waste minimisation is being investigated under a USDOE-sponsored NERI project on the Particle-Bed Gas-cooled Fast Reactor (PB-GCFR) design that is evaluating the impact of different fuel forms and types on core performance. In addition, the other Generation-IV-systems goals of improved safety, enhanced proliferation resistance, and reduced system cost are being pursued in the study. The key requirement for the project is the application of a fast neutron spectrum environment to enhance both the passive safety and transmutation characteristics of advanced pebble-bed reactor designs.

A healthy future nuclear enterprise in a sustainable environment requires that fuel material be efficiently utilised. In this regard, the PB-GCFR study has focused on reactor designs with a high TRU conversion ratio. The intent is to design a TRU self-sustaining core in which for a given initial core load, the continuous recycle and application of the fuel would be obtained without need for external TRU make-up material. As a basis for trade-off studies, a reference compact fast-spectrum core based on the pebble-bed system has been developed. This core is designed for a power rating of 300 MW_{th} (about 50 W/cc) and has no blanket zone, to reduce the production of high-purity fissile material. It was additionally imposed on this design that the TRU component of SNF extracted by a separation process be used as fuel, in order to minimise the TRU sent to the repository. This fuel type would also enhance proliferation resistance because the TRU would be unavailable in a repository that could become a plutonium mine.

The relatively low power and power density of the PB-GFR design are due to the requirements that the system be passively safe and be optionally exportable to international markets having little or no nuclear infrastructure. On this basis, the TRU content of the heavy-metal in the fuel has been limited to 20%, in support of non-proliferation goals of the U.S. government. Additionally, intrinsic proliferation protection is provided by the pebble fuel form. Because the system is for foreign export, it is currently planned that the core would be loaded off-shore and following long-life irradiation (15 to 30 years), the core would be removed and discharged off-shore. If fuel reprocessing is required, this too would be done off-shore.

When TRU is recycled, reprocessing losses only would be sent to the repository. In this case, the amount of TRU lost to the repository is determined by the amount of TRU to be processed and the recovery factor in recycle. Therefore, in order to minimise the TRU sent to the repository, it is desirable to design a reactor fuelled with TRU in such a way that the total amount of TRU fuel required to operate the reactor for its lifetime is minimised. This implies that the initial TRU inventory

and TRU mass flow for the core lifetime should be minimised. Various trade-studies have been performed to investigate the impact of typical fuel management schemes on fuel waste minimisation. Based on the finding of this work, additional studies have been performed to optimise the long-life core, by investigating the impact of fuel materials on the core physics performance.

In Section 2, the analysis approach used for the study is discussed. The reference core design is presented in Section 3. This design is for a long-life core, which intuitively should result in the minimum amount of TRU being sent to the repository, for a given core size. The results of the sensitivity study on various fuel management schemes are presented in Section 4. Typical schemes such as 3-batch schemes with various cycle lengths (1, 3, 10 years), and semi-continuous schemes were evaluated. In Section 5, the additional optimisation studies employing different fuel, matrix and reflector materials are reported. The conclusions from the work are presented in Section 6.

2. Analysis method

Full-core equilibrium cycle calculations have been performed using the REBUS-3 fuel cycle analysis code. [1] Region-dependent 33-group cross-sections were generated with the MC^2 -2 code [2] based on ENDF/B-V nuclear data. Beginning of cycle material compositions and temperatures were used in the MC^2 -2 calculations. An R-Z computational model with homogenised pebbles has been developed for the PB-GCFR core. The flux distributions were obtained using the finite-difference diffusion theory option of the DIF3D code, [3] based on the observation that it gives similar results as S_n transport calculations for the homogenised core model.

From physics considerations, it is expected that such simple homogenisation models should be able to adequately treat the double heterogeneity effect of fuel particles in pebbles, in the fast system that is of interest in this study. This is because the resonance region's (4 to 9 000 eV) contribution to the multiplication factor is small in such systems. Preliminary evaluations of this effect, with higher fidelity (deterministic transport and Monte Carlo) codes confirm this trend. However it is additionally planned to perform a more rigorous investigation of this issue in the future using an MCNP model as reference.

3. Reference design

A reference compact fast-spectrum core based on the pebble-bed system has been developed. This core is designed for a power rating of ~300 MW_{th} (about 50 W/cc) and uses the pebble fuel type. Each pebble consists of a spherical container (pebble) made of matrix material that contains a central zone of coated or particulate fuel dispersed in the matrix material. Since the graphite used as matrix material in the Pebble-Bed Modular Reactor (PBMR) design might not be appropriate for the fast concept, because of its strong neutron moderating properties, alternative matrix materials were used in this study. The requirements for high operating temperatures that give a high thermal efficiency (45-50%) necessitated the use of refractory metals and high melting point materials having favourable fast neutron properties (e.g., low absorption cross-section).

The pebble employed in this study has an outer radius of 6 cm. A dispersion fuel zone diameter of 5.5 cm was used in the study (compare to 5 cm for PBMR), in order to increase the fuel loading in the pebble. The pebbles are packed into a cylindrical core and are assumed to occupy 61% of the volume. Helium gas flows through the pebble-bed to remove the heat generated by the fission process. The helium coolant, being neutronically benign (i.e., low macroscopic absorption cross-section), does not adversely affect the fraction of neutrons available for converting fertile nuclides to fissile nuclides.

Thus, reactivity losses can be effectively compensated by the inclusion of fertile material, which tends to reduce the enrichment and excess reactivity requirements of long-life systems. For the reference design, depleted uranium is employed as the fertile material.

The fuel pebble design used for this study utilises the same matrix and coating material (or uncoated). This design permits a core fuel volume fraction of up to ~30% which ensures a sustained critical mode operation for a 15-30 years fuel irradiation cycle. Currently we have used potentially compatible fuel and matrix forms for this fuel type, mixed uranium-transuranics carbide [(U,TRU)C] fuel in a zirconium carbide (ZrC) matrix. The core volume fractions are 30.5% fuel, 30.5% matrix, and 39% helium (He) coolant. A 50 cm thick SS-316 reflector (20% He coolant) is assumed. Additional fuels material study is however required to ensure that these are feasible fuel-matrix forms in the irradiation and temperature fields of the PB-GCFR.

4. Impact of fuel management scheme on TRU losses

The Generation IV goals of waste minimisation and effective fuel utilisation (sustainability) can be met by designing a TRU breakeven core. For this core, the amount of TRU fissioned is equal to the amount created from uranium conversion. In this case, the TRU charged per cycle is equivalent to the TRU discharged. Based on the constraint of a breakeven core, the total amount of TRU required to operate a reactor for its lifetime was estimated for various fuel management schemes, in order to determine the favourable approach for waste minimisation. A thirty-year core lifetime was used as basis for comparison of the various cases. This implies that the core would be initially loaded, periodically reloaded (depending on the fuel management scheme), and ultimately offloaded at the end of the thirty years. Considerations were therefore given to all the fuel separations and losses resulting from this assumed scenario. The material from the final core is processed and used as fuel for a newer core in the sustainable nuclear enterprise. Because of the fast spectrum and the TRU breakevendesign, the discharge isotopic vector would be similar to the charge vector, and the irradiated fuel should be readily applicable to the new core. If necessary, the discharge fuel will be blended with the legacy LWR transuranics.

The *lifetime TRU processed* is the figure of merit in this study. It is assumed that the (U,TRU)C fuel can be separated to (1) obtain high purity uranium, (2) recover the TRU, and (3) partition the fission products. The uranium would be stored for future use (preferred) or buried in a low-level disposal site. Fission products and TRU losses from the separation stage would be packaged into waste forms and buried at the repository after cooling. The amount of TRU ultimately reaching the repository has an effect on the amount of nuclear waste that can be contained in the repository, as this material constitutes the primary long-term hazard. Waste minimisation and the ability to effectively package the material in the repository is dependent on the lifetime TRU processed. With multi-batch management, additional partial reloads of transuranics are required to start-up the initial core, while later batches utilise the reprocessed TRU. In this regard, it is assumed that there is an interval of five years between fuel discharge and recycle back into the core following fuel separation and fabrication.

The fuel management schemes considered in this study include a single long-life cycle of 30 years, 3-batch schemes with cycle lengths of 1, 3 and 10 years, and semi-continuous schemes (½- and 1-year cycles representing continuous loading of fuel). The results for these cases are summarised in Table 1.

In order to limit the solution search space for this study, the core geometry was fixed with a height-to-diameter ratio of one. The fuel volume fraction and enrichment were searched to satisfy the requirements of the TRU breakeven design and the criticality condition during the cycle. The core

power density for all the cases is 50 W/cc, which is lower than that employed for typical fast reactor systems, but higher by a factor of ~8 than the value for the PBMR. Passive safety requirements might change the final value of this core parameter.

From Table 1, it is seen that the TRU discharge rate (in MT/GW_{th} -yr) is dependent on the burnup. Maximising the burn-up reduces the TRU discharge rate and hence the required processing. For a fixed fuel residence time (30 years), the initial TRU inventory increases slightly with the cycle length and hence the TRU discharge burn-up decreases slightly. As a result, longer cycle length increases the TRU processing per cycle. However, because of less frequent recycling, the lifetime TRU processed decreases considerably with the cycle length.

For a fixed number of batches, the discharge burn-up can be increased by maximising the cycle length or the specific power under the material irradiation and safety constraints. As discussed above, in the PB-GCFR design, the passive safety requirements limit the specific power significantly. Therefore, in order to increase the discharge burn-up and hence to reduce the amount of TRU to be processed, it is desirable to increase the cycle length. This can be confirmed by comparing the results for 3-batch fuel management schemes with different cycle lengths shown in Table 1. In addition, the fuel cycle cost can be reduced with a longer cycle length since the initial inventory increase is marginal. The single-batch core results in the lowest TRU procurement requirement and lowest lifetime TRU processed. This is because in this case, no TRU is discharged until the 30 year interval. Additionally, by virtue of the pebble core design, attaining a critical core with the TRU enrichment limit of 20% is a design requirement that is difficult to meet. As a result, all the cores require about the same TRU enrichment (or content) to satisfy this requirement. If instead of the TRU breakeven core, the design was for a burner core, the single-batch case would be penalised because of the TRU enrichment requirement.

5. TRU breakeven fuel cycles

Because the long-life core appears to be attractive from the waste minimisation viewpoint, we have additionally performed parametric studies of this fuel management scheme for TRU breakeven cores, using different uranium feed, TRU feed, fuel form, and matrix and reflector materials. The description for each case is provided in Table 2. The maximum cycle length that achieves TRU breakeven for each design option was determined for a fixed fuel volume fraction (30.5%). Two different uranium feeds were considered. These are natural uranium (NU) and depleted uranium (DU). The TRU feed options include that coming from the LWR spent nuclear fuel and another coming from recycled fuel. The (U, TRU) C and (U, TRU)N fuel forms were included in the study. For the nitride fuel, it is assumed that the nitride is enriched to 99.9% ¹⁵N. When the nitride fuel is used, only the TiN (enriched in ¹⁵N) matrix type is considered. With the carbide fuel, cases using zirconium carbide (ZrC), titanium (Ti), vanadium (V), and graphite (C) as matrix were evaluated. The reflectors considered are SS-316, graphite, beryllium (Be) and its oxide (BeO), nickel (Ni), lead (Pb), tungsten (W).

Table 1. Impact of fuel management scheme on lifetime TRU processing

			Ch	Charge	Disc	Discharge	Lifet	Lifetime TRU processed	ssed
Fuel management scheme	Fuel volume fraction (%)	Core TRU inventory (MT)	HM mass/Cycle (MT)	TRU enrichment (%)	HM Burn-up (GWd/MT)	TRU rate (MT/GW-yr)	Legacy LWR TRU (MT)	Recycled PB-GCFR TRU (MT)	Total TRU (MT)
1-batch; 30-yr cycle	32.0	4.09	24.82	16.5	132.4	0.45	4.09	0.00	4.09
3-batch; 10-yr cycle	28.1	3.69	7.26	17.0	150.9	0.41	4.93	1.23	6.16
3-batch; 3-yr cycle	23.5	3.46	6.08	19.0	54.0	1.28	4.61	9.23	13.84
3-batch; 1-yr cycle	22.5	3.44	5.82	19.7	18.8	3.83	9.18	27.55	36.73
30-batch; 1-yr cycle	26.9	3.58	0.70	17.2	157.6	0.40	4.18	2.87	7.04
60-batch; ½-yr cycle	26.9	3.58	0.35	17.2	157.7	0.40	4.17	2.92	7.10
NOTE:									

NOTE:

(1) HM means heavy metal.

(2) Legacy LWR TRU is the amount of LWR transuranics required to start-up the system, including initial reloads.

From a waste management point of view, it may be desirable to initially have a net TRU destruction rate to reduce the legacy (accumulated) TRU inventory. Eventually, the equilibrium system would need to operate on a TRU breakeven system or slightly positive TRU production to feed a growing nuclear fleet. To first order, the relative TRU waste merits of a TRU breakeven fuel cycle are proportional to the TRU processing rate on a per unit electrical energy basis. Assuming the same processing systems, the amount of TRU lost to the waste stream is proportional to the rate of processing. For fuel cycles that either produce or consume TRU, other figures of merit need to be evaluated to account for the net change in the TRU inventory.

As can be seen in Table 3, the TRU breakeven cores range from 20 years (W reflector) to 30 years (Pb reflector) with the reference (SS-316 reflector) design being in the middle of this range. The thermalisation of neutrons in the light element reflectors (C, Be, BeO) resulted in very large power peaking factors near the periphery of the core. This seems to be sufficiently problematic to preclude the use of the very light elements in the reflector. In addition, the light element reflectors give slightly larger reactivity swings than SS-316 reflector. Along with the Pb results, it appears that the reflector material should be made of the heaviest elements possible to obtain a low reactivity swing. Lead (Pb) has a low melting point and may not be acceptable, however, as reflector for the PB-GCFR. Other heavy elements with higher melting points will be evaluated.

The reactivity swing for all of these designs is very large, with the lowest having a BOC k_{eff} of 1.064; for all the cases the end of cycle k_{eff} is 1.0. The rate of TRU discharge and consequently recycle is proportional to the TRU charge enrichment and inversely proportional to the cycle length. The long cycle length of the Pb reflector leads to the lowest TRU recycle rate of 0.44 MT per GW_{th}-yr of energy generation. The recycle of TRU, instead of using LWR spent nuclear TRU feed, has a small effect on the performance of the reactor as shown by the equilibrium cycle results.

Case	U Feed	TRU Feed	Fuel Form	Matrix	Reflector
Reference	DU	LWR	С	ZrC	SS-316
NU Feed	NU	LWR	С	ZrC	SS-316
Ti Matrix	DU	LWR	С	Ti	SS-316
V Matrix	DU	LWR	С	V	SS-316
Graphite Matrix	DU	LWR	C	С	SS-316
Nitride Fuel	DU	LWR	15 N	Ti ¹⁵ N	SS-316
Graphite Reflector	DU	LWR	С	ZrC	C
Be Reflector	DU	LWR	С	ZrC	Be
BeO Reflector	DU	LWR	С	ZrC	BeO
Ni Reflector	DU	LWR	С	ZrC	Ni
Pb Reflector	DU	LWR	С	ZrC	Pb
V Reflector	DU	LWR	С	ZrC	V
W Reflector	NU	LWR	C	ZrC	W
Equilibrium (DU)	DU	Recycle	С	ZrC	SS-316
Equilibrium (NU)	NU	Recycle	С	ZrC	SS-316

Table 2.Case descriptions

The core design using nitride fuel and TiN-15 matrix did not perform as well as the core using carbide-fuel, ZrC-matrix and SS-316 reflector. The nitride fuel case requires about the same enrichment. However, because this fuel has a higher heavy metal density, it results in a higher fuel loading in the core, a lower discharge burn-up for the same cycle length, and hence a higher TRU discharge rate, than the ZrC matrix case.

Case	Cycle length (FPY)	BOC k _{eff}	TRU charge enrichment	Discharge burn-up (GW _{th} d/MTHM)	TRU discharge rate (MT/GW _{th} -yr)
Reference	24.9	1.082	17.1%	115	0.54
NU Feed	25.1	1.091	16.8%	116	0.53
Ti Matrix	23.8	1.064	15.3%	110	0.51
V Matrix	23.2	1.069	16.1%	108	0.55
Graphite Matrix	22.7	1.100	17.8%	105	0.62
Nitride Fuel	21.9	1.082	17.3%	97	0.65
Graphite Reflector	25.8	1.090	15.9%	119	0.49
Be Reflector	23.8	1.096	15.7%	110	0.52
BeO Reflector	26.1	1.104	15.8%	121	0.48
Ni Reflector	24.1	1.075	16.8%	111	0.55
Pb Reflector	29.7	1.084	16.8%	138	0.44
V Reflector	22.2	1.069	16.8%	103	0.60
W Reflector	20.3	1.069	17.0%	94	0.66
Equilibrium (DU)	23.7	1.057	16.9%	119	0.52
Equilibrium (NU)	23.7	1.061	16.9%	120	0.51

Table 3. TRU breakeven cycle behaviour

Of the pebble matrix materials considered, titanium appears to be the most favourable. Its low material density and neutron absorption results in the lowest enrichment requirements and hence in the lowest TRU discharge rate, of the different matrix cases. The lower melting temperature of titanium, compared to TiN and ZrC, might negate this advantage.

To reduce the reactivity swing for the long core life, a number of options exist. These include loading multiple fuel enrichments, the average will need to remain approximately the same. Another option is to reduce the fuel volume fraction and operate with a shorter cycle length. The targeted cycle length is 15 to 30 years and this study has shown that a cycle length greater than 20 years can be achieved. The other option is for a core design with a slight net production of TRU to offset the negative reactivity effects from fuel depletion. This option might require a steady growth in the number of TRU breakeven PG-GCFRs or feed to other types of reactors, which is a more complex system with additional assumptions and subjective comparisons.

6. Conclusions and future work

Calculations in support of a reference compact fast-spectrum core based on the pebble-bed design have been done in this work. The impact of the fuel management scheme on TRU waste minimisation was evaluated, using the *lifetime TRU material to be processed* as the figure of merit. A thirty-year period was used in the study. Because of the requirement that all the cores have the same power density and the constraint on the maximum TRU enrichment (20%), the long-life core was found to be the preferable design.

Additional study of the long-life core was pursued by performing parametric studies using different fuel forms (carbide and nitride fuel, depleted and natural uranium base), different matrix and reflector materials, and LWR spent fuel TRU and recycle TRU material. The results have provided indications that titanium is desirable as matrix material, though it might be discarded because of its relatively low melting point. Carbide fuel appears to offer some advantage over nitride fuel. Reflector

studies indicated that lead would be the ideal reflector, but may not be feasible because of its low melting point. As a reflector, SS-316 appears to perform nearly as well as other more exotic and expensive materials. Light elements commonly used in the reflectors of thermal reactors produce prohibitively high peaking factors and high burn-up reactivity swings. By employing a core fuel volume of about 30%, it is possible to obtain a neutronically sustainable long life (15 to 30 years) core operating at a power density of 50 W/cc that has a high fuel discharge burn-up and low TRU discharge rate. Reduction of the fairly high burn-up reactivity swing obtained in this study is an item that is being pursued.

REFERENCES

- [1] B.J. Toppel (1983), A User's Guide to the REBUS-3 Fuel Cycle Analysis Capability, ANL-83-2, Argonne National Laboratory.
- [2] H. Henryson II, B.J. Toppel and C.G. Stenberg (1976), *MC²-2: A Code to Calculate Fast Neutron Spectra and Multigroup Cross-sections*, ANL-8144, Argonne National Laboratory.
- [3] K.L. Derstine (1984), *DIF3D: A Code to Solve One-, Two-, and Three-dimensional Finite-Difference Diffusion Theory Problems*, ANL-82-64, Argonne National Laboratory.