ACTINIDE BURNING IN A LEAD-BISMUTH-COOLED CRITICAL FAST REACTOR WITH ECONOMIC ELECTRICITY GENERATION*

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Abstract

A Lead Bismuth-cooled fast reactor concept for actinide burning and low-cost electricity production and excellent safety characteristics is proposed. The concept employs metallic thorium-based fuel (Th-U-Pu-MA-Zr) in a 700 MWth core. The transuranics destruction rate per MWth-yr is 66% that of an accelerator-driven system using the same initial transuranics composition in fertile-free fuel. Coolant void reactivity mitigation is achieved through the use of streaming fuel assemblies and both the Doppler and fuel thermal expansion feedback are negative. The small negative fuel temperature and coolant temperature reactivity coefficients and negative core radial expansion coefficient provide self-regulating characteristics, so that the reactor is capable of inherent shutdown in major transients without scram, as in the Integral Fast Reactor (IFR).

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Objective

The purpose of this joint Idaho National Engineering and Environmental Laboratory (INEEL) and Massachusetts Institute of Technology (MIT) project is to investigate the suitability of Lead Bismuth-cooled fast reactors for actinide burning while producing low-cost electricity. The goal is to identify and analyse the key technical issues in core neutronics, materials, thermal-hydraulics, fuels, and economics associated with the development of this reactor concept. In this paper, the focus is on the effect of introduction of fertile fuels on reactor safety and actinide burning.

Background and introduction

Lead based coolants, because of their characteristic hard neutron spectrum, are being considered as possible candidates for actinide burning accelerator-driven sub-critical systems, such as the "Energy Amplifier" technology for destruction of transuranics and fission products proposed by the CERN group [17] and the accelerator transmutation of waste (ATW) facility proposed by LANL. [5] Sub-criticality of the ATW facilitates tasks that would be difficult or inefficient to do in critical systems. The key advantage of such a system is that it does not rely on delayed neutrons for reactor control. Moreover, the reactivity feedback, when there is sufficient sub-criticality margin, have a lower safety importance, and these systems can burn a wider variety of materials in the fuel with little concern for their neutronic behaviour. On the other hand, the sub-critical systems are still vulnerable to overheating during loss of coolant, loss of flow and loss of heat sink accidents and from accelerator power spikes. Further, the high cost of the high-current, high-energy accelerator, the lack of demonstrated high accelerator availability, and material concerns about the proton beam window and spallation target are also important considerations when comparing alternative means for the transmutation mission. Because lead-alloy critical reactors, with their hard spectrum, offer certain favourable neutronics characteristics beyond their sodium-cooled counterparts, and provide high neutron efficiency for actinide burning, it is desirable to investigate their potential for transuranics destruction.

MIT explored the potential for a critical, Lead-Bismuth-cooled actinide burning reactor (ABR) design with fertile free fuel. [8] It has been shown that such a reactor is feasible having negative coolant void worth and acceptable fuel temperature feedback. However, a large number of control rods had to be employed to compensate for the substantial reactivity swing of the fertile-free core, resulting in a reduction of effective delayed neutron fraction. The reactivity swing can be substantially reduced, and the need for control rods reduced if the reactor is optimised for minor actinide (i.e. TRUs of atomic number higher than Pu) burning. Hejzlar *et al.,* [9] have shown that a very small reactivity swing can be achieved in a fertile-free core utilising the attractive properties of ^{237}Np and ^{241}Am, whose progeny exhibit significantly higher fission-to-capture cross-sections than the original nuclides, and thus can exhibit reactivity behaviour like the fertile cores. However, these reactors lack sufficient negative Doppler feedback and exhibit a small delayed neutron fraction, making the reliance on inherent reactivity feedback very difficult. Relaxing the requirement of maximising the actinide destruction rate allows for the introduction of fertile nuclides (^{238}U, ^{232}Th) into the core and hence an improvement in the safety characteristics. This paper will investigate the potential of a critical actinide burner utilising thorium-based fuel to burn actinides with the focus on core safety characteristics and the actinide destruction rate.

While the two goals outlined above are the primary thrust in the development of a conceptual design, proliferation resistance and safety are also important aspects to be incorporated in the design process. These goals, together with achievement of Gen IV goals, once translated into actual design

concepts, result in a number of competing requirements, which makes it impossible to achieve each of the goals to the maximum extent possible. Therefore, a delicate balance among individual objectives is necessary. The top-level issues affecting general design choices to address these goals, as well as the design constraints are discussed in detail in [10] and [14] and will not be repeated here. Rather, we will focus on the core design and its neutronic characteristics.

Fuel and structural material selection

Fertile-free actinide or minor actinide critical burners suffer from small delayed neutron fraction and lack of fuel temperature feedback. Both the Doppler coefficient and delayed neutron fraction can be improved by adding fertile materials, but this negatively impacts the net TRU destruction rate – one of the targets set for the development of this reactor design. Therefore, a compromise must be made. To minimise generation of plutonium, and thus to reduce the degradation of actinide destruction rate, thorium was selected as the major fertile material. The benefits of the thorium-based fuel are the following:

1. It reduces generation of plutonium, because the amount of uranium in the fuel can be significantly decreased (only a small amount sufficient to denature bred-in ^{233}U is needed).
2. It improves the coolant density reactivity coefficient in comparison with ^{238}U based fuels. This is due to a smaller increase of fission to capture cross-section ratio (compared to ^{238}U) from spectrum hardening under the conditions of an LBE-cooled reactor. Also, the fission cross-section of ^{232}Th is four times smaller than that of ^{238}U, hence the threshold effect from increased fission rate in fertile material is smaller than in case of ^{238}U.
3. It provides breeding of ^{233}U, hence significantly reducing the reactivity swing and requirements for control rods, thereby improving safety.

Metallic fuel (Pu-MA-Th-U-Zr) has been selected as a primary candidate because of the good EBR-II experience database with this type of fuel (albeit without MAs and Th). The fuel composition used in the calculations is given in Table 1. The fuel considered is a metallic alloy of zirconium (10wt%), thorium, uranium, plutonium and minor actinides, arranged in a square pitch. The isotopic composition of plutonium and minor actinides (MA) and plutonium/MA ratio correspond to those of PWR spent fuel 50 years after discharge. The ratio of U to (U+Th) of 30wt% shown in Table 1 was selected to satisfy proliferation resistance requirements for uranium in spent fuel by denaturing of the inbred ^{233}U.

The cladding is assumed to be made of a special stainless steel, designated ^{823}EP, developed by the Russians for their Lead-Bismuth-cooled reactors. The same steel was used for other core structural materials. The gap between the cladding and fuel requires a bonding material to enhance thermal conductivity. The proven bonding material from the IFR programme is sodium. However, to avoid having a different material than coolant in the core, lead-bismuth-tin alloy (33wt%Pb-33wt%Sn-33wt%Bi) was selected as a bond between the fuel pellets and stainless steel cladding. This material was found to be compatible with Zircaloy-4, [23] although for LWR conditions. Therefore, a research programme will be required to confirm its compatibility with Zr-based fuel and stainless steel cladding. Sodium bonding as in EBR-II and IFR is an alternative in case the lead-bismuth-tin alloy does not provide satisfactory performance. In any event, the amount of bonding agent is not large enough to significantly affect neutronics, which is the focus of the present study.

Table 1. **Reference fuel composition**

Isotope	Weight percent
Zr	10%
Heavy metal total	90%
Pu	18.4%
MAs	4.6%
Pu+Ma (TRU)	21.7%*
Th+U	78.3%*
U	23.49% (30%wt% in ThU mixture)
Pu composition (50 years after discharge)	**100%=18.4% total**
^{238}Pu	1%
^{239}Pu	63%
^{240}Pu	29%
^{241}Pu	1%
^{242}Pu	6%
MA composition (50 years after discharge)	**100%=4.6%total**
^{237}Np	30%
^{241}Am	64.5%
^{242}Am	0%
^{243}Am	5.1%
^{244}Cm	0.4%

* Variable values depending on core design to reach criticality.

Table 2. **Design parameters for large core option**

Design parameter	Value
Pin outer diameter (clad outer)	7.52 mm*
Pitch	9.776 mm
P/D	1.3
Heated core length	1.3 m
Gas plenum height	2.47 m*
Number of fuel assemblies	157
Control rod material	B_4C (60% B10)
Coolant	Pb-Bi
Core-average coolant temperature and density	521°C; 10.09g/cm^3
Temperature of Pb-Bi coolant in the chimney	580°C
Temperature of Pb-Bi coolant in lower plenum	462°C

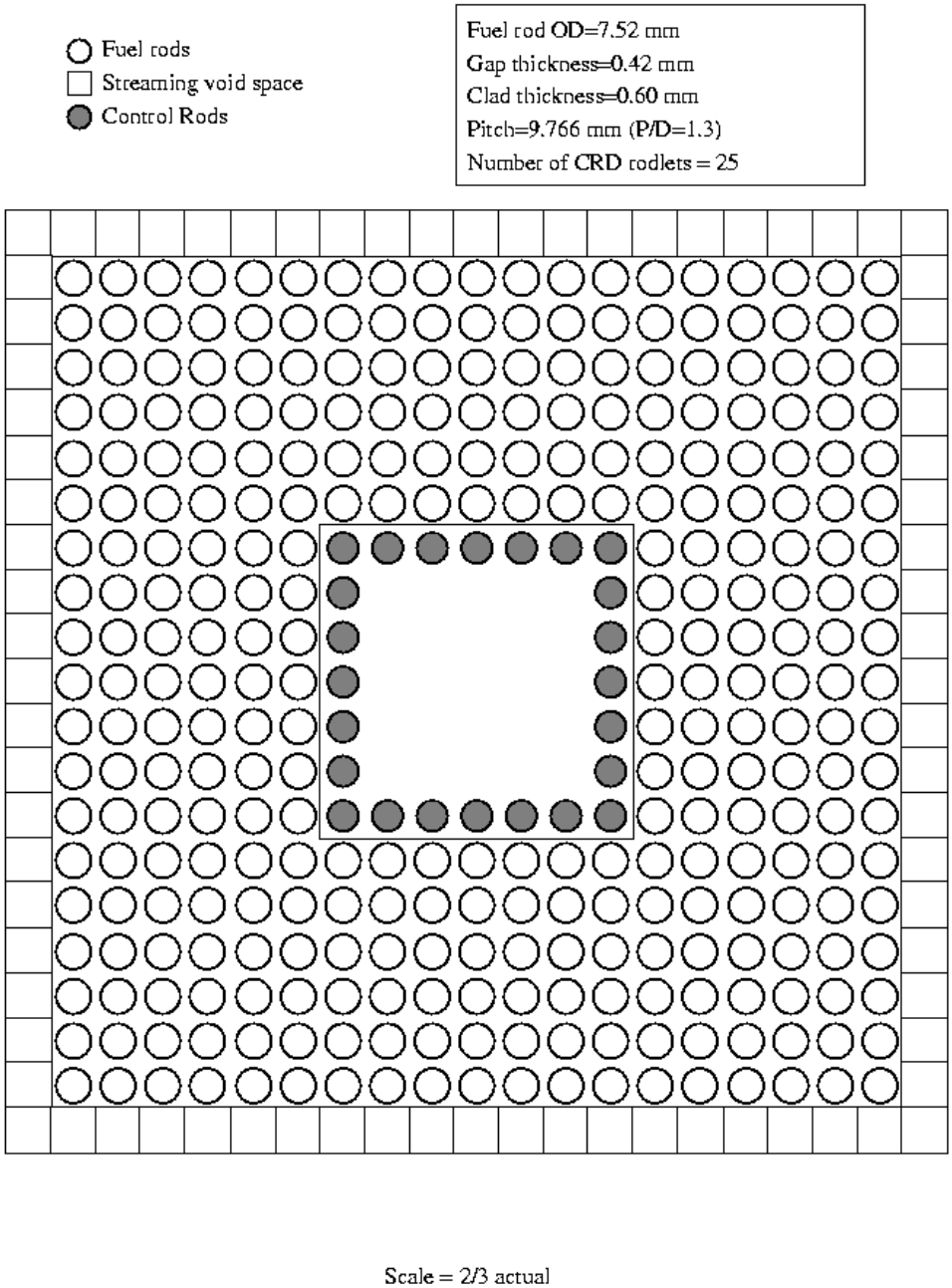
* Based on INEEL cladding stress and strain evaluations. [14]

Core with streaming fuel assemblies

A large positive coolant void worth is one the major problems for fast reactors loaded heavily with transuranics. To overcome this challenge, special streaming fuel assemblies that allow neutron streaming in both the radial and axial directions were proposed. [7] The streaming assembly used for actinide burner core is shown on Figure 1. It employs large central streaming channel occupying 7×7 positions and one row of small peripheral streaming channels. Streaming channels are filled with gas and sealed. Control rods are inserted into the central streaming channel of selected assemblies. A

core layout is drawn on Figure 2. The core has 157 fuel assemblies and was designed to fit within the equivalent diameter of 3.2 m, which is the largest diameter allowed by the constraint imposed by vessel transportability and passive decay heat removal requirements. The key design parameters are listed in Table 2.

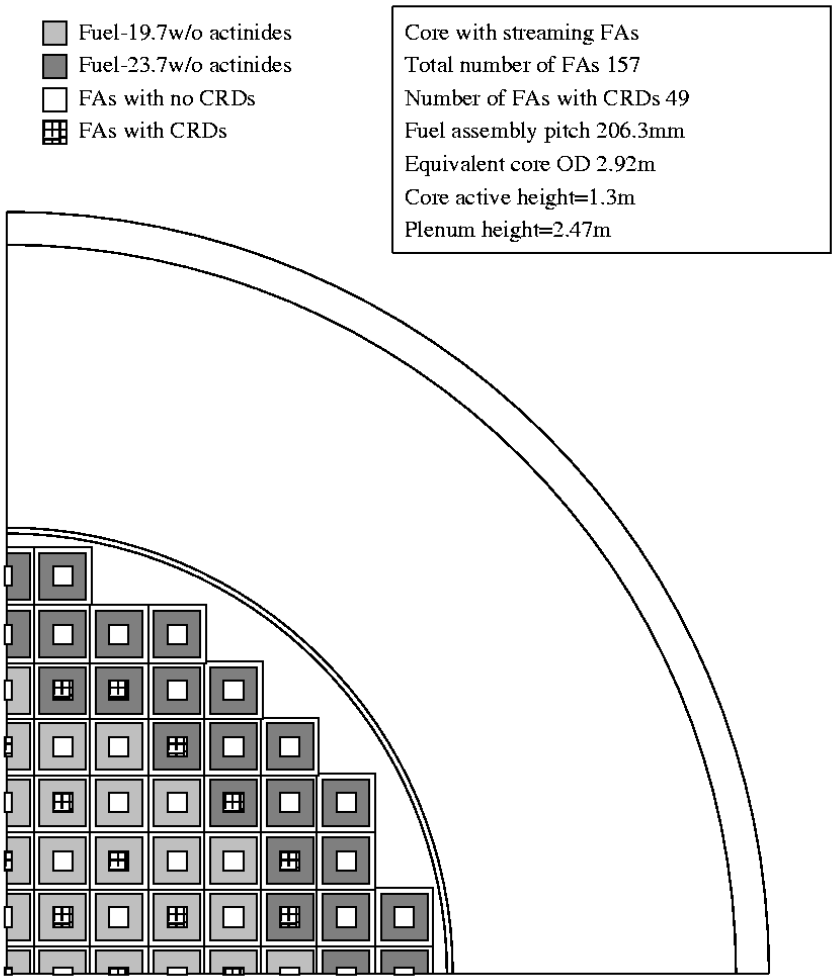
Figure 1. **Fuel assembly with 1 row of peripheral streaming channels**



Analysis tools

All calculations were performed using the Monte Carlo Neutron Photon Transport (MCNP) code MCNP4C, [2] developed at Los Alamos National Laboratory. To perform the burn-up calculations, the ORIGEN2.1 code, [3] developed at Oak Ridge National Laboratory (ORNL), has been chosen. The coupling between MCNP and ORIGEN2.1 has been attained through the newly MIT-developed code MCODE. [24] Other MCNP-ORIGEN2.1 codes, such as INEEL-developed MOCUP [15] and LANL-developed MONTEBURNS 1.0 [16] were also used for comparison.

Figure 2. **Core layout for the reference design**



Power distribution and peak fast fluence

The core with streaming fuel assemblies exhibits very small both radial (<1.2) and axial (<1.2) power peaking (in spite of relatively large core size) due to an increased neutron migration area due to streaming channels. The benefit of small power peaking is lower peak cladding temperature and reduced maximum fluence on cladding. Peak fast flux on cladding is 1.92×10^{15} n/cm^2-s (E>0.1MeV), yielding after 7 years of irradiation a peak fluence of 3.7×10^{23} (E>0.1MeV) n/cm^2. This is below the limiting value of 4.0×10^{23} (E>0.1MeV) n/cm^2 for HT-9 steel. Therefore, operation without refuelling for about 7 years seems feasible based on the fluence limit on core structures.

Burn-up performance

Figure 3 shows the trace of k_{eff} as a function of irradiation time obtained by MCODE. A relatively large number of neutron histories was chosen (600 000) to attain a small MCNP statistical error. It can be observed that reactivity-limited irradiation time is about 6.3 EFPYs, or 7 years assuming 90% capacity factor. The maximum reactivity excess of 1.7%Δk/k, or USD 6 is reached at ~2 EFPYs. For 49 control rods (see Figure 1), the reactivity vested in the maximum-worth control rod at that time is USD 0.13. This is a sufficiently low value to satisfy the requirements on fuel and cladding temperature limits for any unprotected overpower transient (withdrawal of one control rod without scram). The discharge burn-up in the inner core region is 120 MWd/kgHM, which is well within the established experience for metallic U-Pu-Zr fuels. [12]

The consumption rate of individual transuranic isotopes per EFPY is 0.23kg/EFPY. In comparison with the fertile-free actinide burner investigated earlier during this project, [8] the TRU destruction rate per MWth is about 36% less. Hence, the penalty of introducing fertile ^{238}U with respect to the effectiveness of actinide burning is acceptable and high TRU burning rates can still be achieved. It is instructive to compare this TRU consumption rate with that of ATW. ATW designers report destruction of 220 kgTRU/y per 840 MWt in ATW, or 0.262 kg/y per MWt. [1] This compares with 0.23kg/EFPY, or 0.21 kg/y assuming 90% capacity factor for this ABR design. Thus, the destruction rate per year is 80% of that of ATW. This is relatively high destruction rate considering the presence of fertile materials and comparing the simplicity of the reactor to the complex accelerator-reactor tandem configuration. Further improvement is possible because a capacity factor of 95% should be attainable in a reactor refuelling once in 7 years. A higher capacity factor is an expected advantage of a critical reactor over an accelerator-driven system. This is because: (1) the capacity factor of two systems, the accelerator and the sub-critical target, which must concurrently maintain operation is less than the capacity factor of one system and (2) current large accelerators experience frequent trips. The latter reason is a key issue to be resolved in further accelerator development because in current high-power accelerators the time between failures is measured in hours or tens of hours. [6]

Figure 3. **Core eigenvalue change with irradiation time**

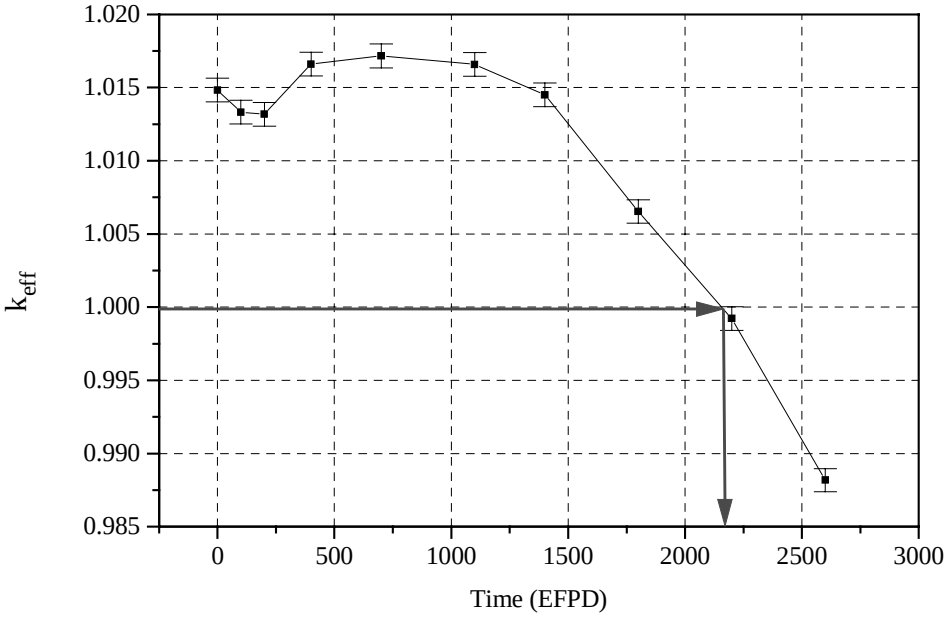


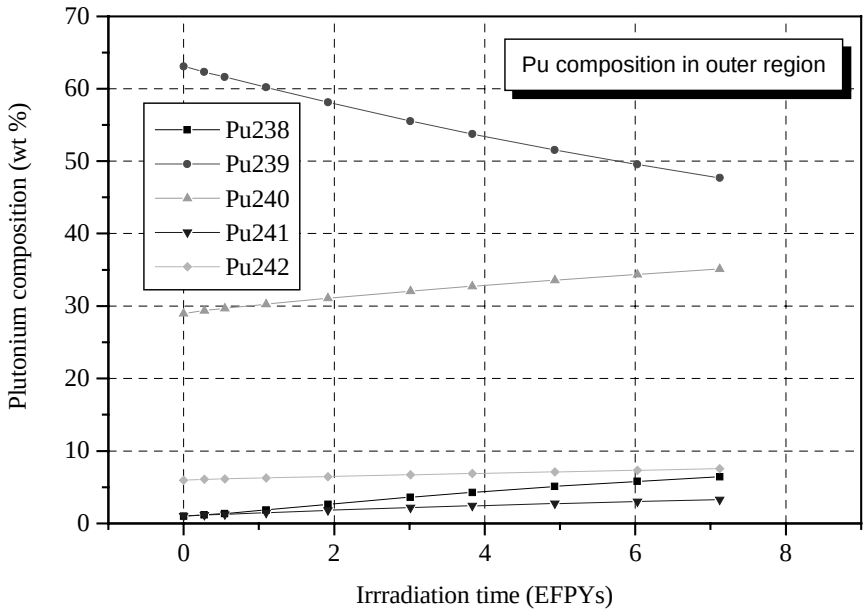
Figure 4 shows that plutonium isotopics are significantly degraded from that of the PWR spent fuel vector making it virtually weapons unusable. Especially noteworthy is the very high content of ^{238}Pu (~7wt%) – a result of β^- decay of ^{238}Np, which is generated upon neutron capture in ^{237}Np. Also, the fractions of other even isotopes are very high, i.e., 35% and 7wt% for ^{240}Pu and ^{242}Pu, respectively. Also, the ^{233}U proliferation index (^{233}U+0.6^{235}U)/Total U ≤0.12, is satisfied.

Reactivity feedback

Reactivity response to coolant density changes

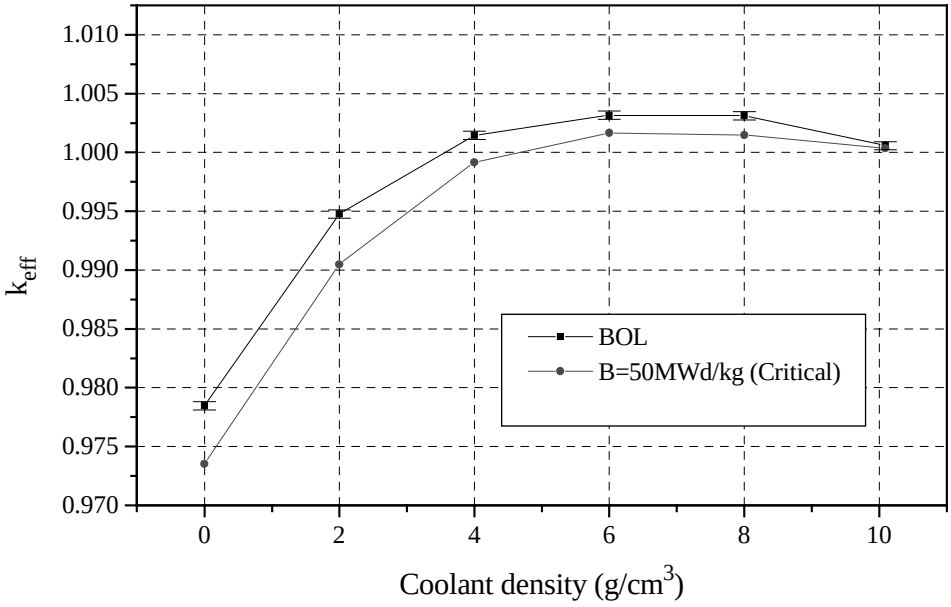
Reactivity changes with coolant density at BOL and at 50 MWd/kg are plotted in Figure 5. Coolant void reactivity worth (the case of voiding all coolant in the core – see the zero-density point on Figure 5 – while maintaining coolant outside the core at reference density) is negative. If coolant density is decreased homogeneously throughout the core, reactivity is slightly increased for densities down to 6g/cc. The maximum reactivity insertion remains less than 1 USD (0.9 USD) for delayed neutron fraction of 0.003.

Figure 4. **Plutonium vector in the outer core region for fuel with 30w/o U**



Further, it is to be noted that the density of 6g/cc cannot be effectively reached by coolant heat up, because the coolant density at boiling (1 670°C) is 8.7g/cc, which corresponds to a reactivity insertion of +0.46 USD. But even this high temperature cannot be reached because the core structural material and fuel melt at lower temperatures. Assuming that the maximum allowable coolant temperature is equal to the transient cladding temperature limit, i.e., 725°C, the minimum density from coolant temperature changes is 9.85 g/cc corresponding to a very small reactivity insertion of 0.08 USD, or 0.04¢/°C. This is almost 5 times less than the 18¢/°C value cited for a sodium-cooled IFR core. For higher burn-ups, coolant temperature reactivity coefficient is even lower.

Figure 5. **Reactivity response to coolant density changes**



Doppler coefficient and fuel thermal expansion effect

The Doppler coefficient was calculated using the MCNP4C code with the JEF temperature dependent cross-section libraries. The results at BOL are shown in Figure 6. Because of the limited number of temperatures available in the library set, only two points could be obtained. MCNP-calculated points are linearised average values between two temperatures and assume β_{eff}=0.0028, as calculated in the next section.

In typical LMFBRs, the Doppler coefficient is described by a law of the type

$$T\frac{dK_D}{dT} = AT^{-\alpha}, \qquad (4)$$

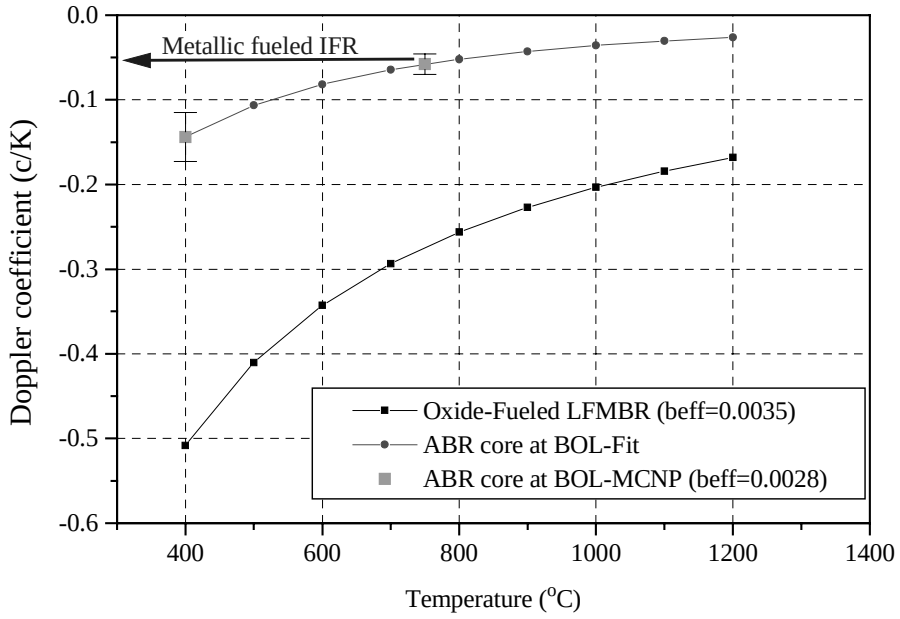
where A and α are constants. [18] In practice the Doppler coefficient in a 1 200 MWe LMFBR with PuO_2/UO_2 fuel varies from ~-1.2pcm/°C at 400°C to ~-0.5pcm/°C at 1 200°C. [18] Substituting these values into Eq. (3), the unknown constants can be calculated to match Doppler coefficients at temperatures of 400°C and 1 200°C. This procedure yields the constants. *A* = -0.01737 and α= 0.11765. Using Eq.(3) with these constants and an LMFBR delayed neutron fraction of 0.0035 yields Doppler coefficients shown in Figure 6 on curve "Typical Oxide-Fueled LMFBR". A similar fit to MCNP – calculated values was performed for actinide burner core (curve "ABR core at BOL-Fit").

The Doppler coefficient for the ABR fuel is negative and about 3 times smaller than that for the oxide core. Smaller magnitude of the Doppler coefficient is consistent with metallic-fueled cores, such as IFR for plutonium burning where Hill *et al.*, [11] reported the value of the Doppler coefficient of ~-0.05¢/K. Because the criteria for self-controllability favour small negative Doppler feedback to mitigate the power rise from cooldown scenarios, the small negative Doppler coefficient is preferable to provide, in combination with the negative prompt fuel thermal expansion coefficient and negative coolant void worth, inherent reactor shutdown in the whole spectrum of unprotected accidents. Fuel thermal expansion coefficient was calculated using MCNP to yield -0.06±0.02c/K, which effectively doubles the fuel temperature feedback.

Delayed neutron fraction

The effective delayed neutron fraction was calculated using the MCNP4C model of the full core at BOL to yield 0.0030 ±0.0002. This value compares with 0.003 for CDFR (UK), [22] 0.0034 for conventional IFR core, [11] or 0.002 for pure Pu burner core with Metallic Pu-28Zr + Hf-26Zr fuel. [11] The optimised core exhibits a slightly smaller delayed neutron fraction than in a conventional IFR. On the other hand, significant improvement in comparison to a pure plutonium burner or an actinide burner, can be observed. Thus, the effect of fertile isotopes proved to be beneficial to control and safety characteristics of this transmutation oriented design.

Figure 6. **Doppler coefficient**



Self-controllability characteristics

Neutronics safety constraints are primarily the reactivity feedback that control passive shutdown characteristics in compliance with the top-level requirement for self-controllability of a reactor imposed by the objective of excellent safety, comparable to that of IFR. Self-controllability requires that, in a quasi-static reactivity balance sense, the reactor is inherently shut down to a safe state under the most restricting anticipated transients without scram. Wade and Hill [21] derived criteria for passive self-regulation in the IFR from the neutronics viewpoint. Following their approach, the same criteria (called here S-criteria) can be derived for Lead Bismuth-cooled reactor as follows: [14]

- S1: A/B ≤ 1, where A is the net power reactivity coefficient in cents and B represents the power/flow coefficient of reactivity in cents/100% power/flow, which controls the asymptotic temperature rise in an unprotected loss of flow transient. In the case of natural circulation at full power, this criterion can be relaxed.

- S2: $1 \le \frac{C\Delta T_c}{B} \le 1.8$, where C characterises the inlet temperature coefficient of reactivity in cents/°C and ΔT_c is full-power, steady-state coolant temperature rise. The term ensures inherent balanced response to an unprotected loss of heat sink transient and coolant inlet freezing.

- S3: $\frac{\Delta\rho_{TOP}}{|B|} \leq 1.25$, where $\Delta\beta_{TOP}$ is the reactivity vested in a single control rod.

Table 3. **Self-controllability characteristics for Na-cooled IFR and LBE-cooled ABR**

	Doppler	**Fuel expansion**	**Coolant density**	**Rod driveline**	**Radial expansion**	**Coolant ΔT (K)**	**Fuel ΔT(K)**
Expressions for reactivity coefficients [19]							
A (¢) =	(α_D	+ α_e)					x ΔT_f
B (¢) =	α_D	+ α_e	+ α_{Co}	+ $2\alpha_{RD}$	+ 2 α_R)	x $\Delta T_c/2$	
C (¢/K) =	[α_D	+ α_e	+ α_{Co}		+ α_R)]		
Typical IFR values for1800MWth core [20]							
	-0.12	-0.09	+0.18	~0	-0.22 (α_R)	150	150
Calculated reactivity coefficients for IFR: A=-31¢, B=-35¢, C=-0.25¢/K **Calculated criteria for IFR A/B = 0.88<1, CΔT_c/B = 1.1** $\in \langle 1;2 \rangle$							
Values for optimised ABR with fertile fuel (700MWth core)							
Optimised ABR	-0.07	-0.06	+0.04	~0	-0.20	92	90
Calculated reactivity coefficients ratios for optimised ABR: A= -12¢, B= -25¢, C= -0.32¢/K **Calculated criteria for optimised ABR A/B = 0.48<1.25, CΔT_c/B = 1.18** $\in \langle 1.0;1.8 \rangle$							

Individual reactivity coefficients and coefficients ratios for the ABR are compared against those of metallic fuelled IFR in Table 3. Both S1 and S2 values fell well within the required range, as for the IFR design. As mentioned earlier, the maximum control rod worth is 0.13 USD = 13¢. Thus the third criterion,

$$\frac{\Delta\rho_{TOP}}{|B|} = \frac{13}{25} = 0.52 < 1.25$$

is also satisfied and the large margin suggests that the number of control rods can be reduced. Therefore, it can be concluded that the above set of reactivity coefficients having small negative fuel temperature feedback together with a small coolant temperature reactivity coefficient and negative core radial expansion coefficient yields self-regulating characteristics, capable of inherent shutdown in major transients without scram. This has been proved by the analyses of a number of unprotected transients using ATHENA code. [4]

Conclusions

The concept of a Pb-Bi-cooled critical reactor that can destroy actinides from spent LWR fuel while maintaining excellent safety has been proposed and evaluated from the neutronics viewpoint. The analyses were performed for metallic thorium-based fuel (Th-U-Pu-MA-Zr) in a once-through cycle assuming that the discharged fuel from the ABR remains in temporary storage before multi-recycling is introduced. The major conclusions can be summarised as follows:

- Using thorium as a major fertile material is an effective means to reduce the large reactivity swing occurring in fertile-free cores while still allowing high actinide destruction rate per MWth. In addition, thorium use increases the Doppler feedback in comparison with fertile-free fuels and reduces the coolant density reactivity coefficient.

- The ABR destruction rate of actinides per MWth-yr is 60-65% of that of the fertile-free rate of destruction (in ATW or ABR). This is a very appealing number considering the simplicity of the proposed reactor versus the more complex accelerator plus reactor system.
- High coolant void worth typical for minor actinide fuels can be effectively mitigated by the employment of streaming fuel assemblies. Using streaming fuel assemblies with one row of peripheral streaming channels and a central streaming region results in negative coolant void worth and very small positive coolant temperature coefficient, which provides very good potential for a design with self-regulation characteristics, similar to the IFR. Streaming assemblies also reduce power and neutron flux peaking, and thus peak fast fluence.
- The discharged fuel from the ABR satisfies proliferation constraints for both the plutonium and uranium compositions. Plutonium isotopics are significantly degraded from that of the PWR spent fuel vector making it virtually weapons unusable. The 12% proliferation limit on fissile uranium with ^{233}U content is satisfied if depleted uranium is mixed with thorium with ~30wt% of uranium in the U+Th mixture.
- Both the Doppler and fuel thermal expansion feedback are negative and their values are comparable to those for IFR fuel.
- The combination of reactivity coefficients satisfies requirements of self-control so that in combination with passive decay heat removal design all transients without scram lead to inherent shutdown without exceeding safe fuel and structural temperature limits.

In summary, the proposed design of the 7-year life core for burning transuranics from spent LWR fuel appears to be very promising and deserving of future refined analyses and optimisation because it offers high consumption of actinides, excellent safety characteristics and has the potential to have low electricity generation cost due to its modularity, simplicity and high capacity factor. Finally, current evaluations did not assume reprocessing of spent ABR fuel and recycling it back through the ABR to reach equilibrium mass flows. Future analyses will have to be extended to include multi-recycling to exploit the full potential of TRU inventory reduction. To achieve good economy with multiple reprocessing will require less expensive reprocessing technologies than those practised or proven today. [13]

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