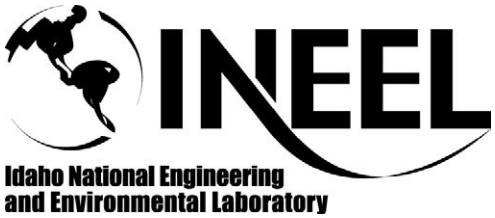

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A COMPARISON OF LONG-LIVED, PROLIFERATION RESISTANT FAST REACTORS

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ABSTRACT

Various methods have been proposed to transmute and thus consume the current inventory of trans-uranic waste that exists in spent light-water-reactor fuel. These methods include both critical and sub-critical systems. The neutronics of metallic and nitride fuels loaded with 20-30wt% light-water-reactor plutonium plus minor actinides for use in a lead-bismuth and sodium cooled fast reactor are discussed, with an emphasis on the fuel cycle life and isotopic content. Calculations show that core life can extend beyond 20 years, and the average actinide burn rate is similar for both the sodium and lead-bismuth cooled cases ranging from 0.5 to 0.9 g/MWd.

Keywords: liquid-metal-cooled, transmutation, fast reactors, proliferation resistance

1 - INTRODUCTION

Nuclear power is expected to play a significant role in meeting future electricity needs, and in significantly reducing emissions compared to fossil-fueled power plants. However, the next generation of nuclear power plants will be expected to demonstrate significant advancements in economics, safety, waste disposal, and proliferation resistance. In an effort to address these issues, systems capable of transmuting (i.e., fissioning) the long-lived higher actinides using a fast neutron spectrum have been proposed. These include both sub-critical (accelerator) and critical systems. The Los Alamos National Laboratory (LANL) has proposed an accelerator-driven, sub-critical actinide burner for the consumption of actinides and long-lived fission products from spent light-water-reactor (LWR) fuel. The LANL system has been labeled Accelerator-driven Transmutation of Waste (ATW). Fast reactors have also been suggested in the disposition of weapons plutonium [1] and minor actinides. The work in this paper focuses on the attributes of fast reactor systems.

While considerable design work has been done in the United States, Europe, and Japan on fast reactors, including actinide burners, most of the work has been done for sodium cooled reactors. The choice of coolant will affect the TRU destruction rate, and the reactivity swing of the reactor. However, there are other considerations in choosing the reactor coolant, such as material compatibility and neutron fluence limits. Lead-bismuth can be corrosive to structural materials at the temperatures of interest, but the advantages of lead-bismuth over sodium as a coolant are related to the higher boiling temperature and high heat of vaporization (a boiling temperature of 1725°C versus 892°C for sodium), making it practically impossible to create a major void in the core due to coolant overheating. It also allows for a simpler containment structure due to the lack of chemical reactions with air and water, and a small volume change upon solidification. With regard to the neutronics, lead-bismuth offers a harder neutron spectrum and, therefore, improved neutron economy, especially when burning actinides. Also, lead-bismuth has better reflective properties, making it possible to get breeding even without blankets.

On the other hand, sodium technology is well developed and has been proven. The Integral Fast Reactor (IFR) project is the most notable accomplishment, where the integrated fuel cycle reduces the TRU discharge significantly, and its safety performance was exceptional. Nevertheless, the main goal of the concept presented here is to maximize the TRU destruction rate while keeping the economic costs low, implying that the fuel remain in the reactor for relatively long periods.

Today, spent fuel contains approximately 165,000 tonnes of heavy metal, of which 1500 tonnes is plutonium. That inventory of plutonium and minor actinides can be used and burned in a fast reactor, thus reducing (or practically eliminating) the amount of TRU to be sent to a repository. The choice of lead or lead-alloy for the reactor coolant in an actinide burning fast reactor has its challenges, but also offers enhanced safety and reliability, as was described above. The Russians adopted lead-bismuth coolant for use in their most advanced nuclear submarines, the so-called “Alpha” class submarines, which are the fastest in the world. The Russians

have built and operated seven lead-bismuth-cooled reactors in submarines and two on-shore prototypes. More recently they have studied the design of a variety of lead and lead-bismuth cooled reactors for electric power generation, with the most recent using a fertile nitride fuel. Greenspan, et al. [2] at UC Berkeley have developed a long-life, once through, modular lead-bismuth cooled reactor. The reactor is cooled by natural convection, is similar to a PRISM design, and is designed as 100 MW(th) modules. The goal of the work presented in this paper was to compare the TRU destruction capabilities of lead-bismuth and sodium cooled reactors, with a special emphasis on the change in reactivity with burnup and minor actinide concentrations.

2 - TECHNICAL CHALLENGES

Striking a balance between actinide destruction, long core life, passive safety, proliferation resistance, and competitive economics are a difficult task. There are other disadvantages in using lead and lead-alloys for cooling a fast reactor in addition to the material compatibility problems. These include high material costs for certain eutectics like lead-bismuth, higher melting temperatures (327°C for lead and 125°C for lead-bismuth, compared to 98°C for sodium), and in the case of lead-bismuth, the production of Po-210 (a radiological hazard).

Fluence limits are also a concern, where the harder spectrum found in lead-based systems will reduce the burnup capability unless they are operated at lower power densities. Although important, this is a materials issue, which is discussed in more detail by Hill et al. [3].

Several different fuel types can be used depending on the scope and purpose of the reactor. These fuel types can be lumped into two general categories: fertile and non-fertile fuels. Each type has its own set of challenges, although most of the challenges are common to both. Previous work has dealt with both the design [4] and performance of different fuel types [5]. In the remainder of this paper, we will limit our discussion to the performance of fertile fuel in lead-bismuth and sodium cooled fast reactors.

To effectively transmute plutonium and minor actinides from spent LWR fuel and attain long fuel cycles, it is desirable to minimize the loss of neutrons in order to retain a large surplus for transmutation and conversion. Metallic fuels based on a zirconium matrix provide large excess reactivities due to the low parasitic absorption cross-section of zirconium, and due to the hard neutron energy spectrum achievable because the fuel does not contain any moderating isotopes. Nitride fuels may also be suitable.

To maximize the *actinide transmutation* capability of the system, breeding of new fissile material must be minimized, making the presence of fertile isotopes (i.e., ^{238}U and ^{232}Th) undesirable. The choice of fuel composition for maximum actinide transmutation is then restrained to the LWR generated plutonium and minor actinides (20 to 30wt%) and to the zirconium matrix (70 to 80wt%), constituting the structural component of the fuel rods. The larger weight fraction of zirconium relative to the heavy metals makes this non-fertile fuel significantly different from the metallic fuel developed by Argonne National Laboratory (ANL) for the Integral Fast Reactor (IFR) project.

However, to minimize the cost of electricity produced, it is desirable to have relatively long refueling cycles so that the plant capacity factors are high and the fuel fabrication costs remain low. Long refueling cycles require relatively constant reactivity and, therefore, the use of some fertile material in the fuel. Thorium, with enough depleted uranium to denature any U-233 produced, is the material of choice because the end product is not easily separable, and there are fewer actinides produced than with an all-uranium fuel. Again, a dispersion type metallic fuel with coated plutonium-thorium-uranium particles in a zirconium matrix is envisioned. Nitride fuel forms were also considered for the fertile fuel.

The fuel choice raises three major neutronic challenges:

- large positive coolant void reactivity coefficient,
- small Doppler feedback, and
- large rate of reactivity loss with burnup (i.e. the reactivity swing).

Each of these will be addressed separately.

Void Reactivity Coefficient. Although positive, void reactivity in sodium cooled reactors has not been an issue due to the strong negative fuel expansion coefficient, and the somewhat smaller but still negative Doppler coefficient. However, a negative void coefficient in a fast reactor would be seen as a definite advantage; especially if the other reactivity coefficients remain negative.

In contrast to sodium-cooled cores, and depending on the core configuration, voiding of an entire lead-bismuth cooled fast reactor core can produce a negative void coefficient. However, local voiding will produce a positive coefficient. The sign of the coolant void coefficient in fast reactors is the combined result of three conflicting effects upon coolant voiding:

- neutron leakage is increased resulting in a reactivity reduction,
- neutron scattering decreases and the spectrum hardens resulting in larger fission-to-capture ratio hence increasing reactivity, and
- parasitic captures in the coolant decrease, leading to a reactivity increase.

The net outcome is typically a strong reactivity increase due to the latter two effects unless leakage is enhanced enough to offset them.

Doppler Reactivity Coefficient. The hard spectrum leads to a decrease of absorption rate in the resonance peaks, which results in a very small Doppler feedback. While negative, the magnitude of the Doppler coefficient can be a challenge, and strong negative feedback from other coefficients will be necessary.

Reactivity Swing with Burnup. For long-lived cores, the excess reactivity must be sufficient to sustain criticality throughout the life of the reactor. However, it is also desirable that the reactivity swing be kept small. These differing requirements seem to point to a high conversion core, but this conflicts with the wish to obtain a high transmutation rate.

3 - NEUTRONIC ANALYSIS

Analysis Tools and Fuel Parameters. The current work uses the MOCUP code [6] to analyze the reactivity characteristics and isotopic concentrations of unit fuel pins/cells, with 38 actinides and 50 fission products being tracked through the MCNP portion of the analysis. MCNP [7] is a well-known Monte Carlo code capable of calculating fluxes, reaction rates, and eigenvalues in general, 3-D geometry using continuous cross-section data. ORIGEN [8] uses a matrix exponential method to calculate the generation and depletion of isotopes, or elements, in a given neutron flux. MOCUP takes specific output data (including cross-section data, fluxes, and reaction rates) from MCNP and passes it to ORIGEN, where new isotopic information is generated and passed back to MCNP for the next calculation. This gives time dependent information about the reactivity swing and isotopics for the specified problem.

The fuels studied were metallic and nitride fuel containing fertile material, had a constant pitch to diameter (P/D) ratio of 1.6 using a square pitch, and an initial actinide loading (i.e., Pu and minor actinides) of 20-30wt%. Table 1 gives a summary of the TRU composition in the fuel.

Table 1 : BOL TRU composition.

Isotope	wt%
Pu-238	0.32%
Pu-239	9.28%
Pu-240	4.16%
Pu-241	1.60%
Pu-242	0.64%
Plutonium Total	**16%**
Np-237	1.72%
Am-241	1.80%
Am-243	0.36%
Cm-244	0.12%
Minor Actinide Total	**4%**

The reason for using a large, square pitch is based on the small thermal-hydraulic resistance in the core, which is an important consideration for natural convective cooling.

The next consideration in using this fuel is the reactor power level (power density) or linear power, where we used two different linear powers in our analysis at 367.5 W/cm and 86 W/cm. These two linear powers

represent upper and lower bounds. The power level becomes important for long-lived cores because the amount of fissile material that will be needed to sustain a critical reactor will have to be optimized. The fissile components used for the current analysis are Pu-239 and Pu-241, and were kept constant at a combined weight percent of 10.88%. The current analysis has been limited to IFR type fuel, i.e., cylindrical pins placed in assemblies. The parameters of the pin can be seen in Table 2.

Table 2 : parameters of the cylindrical fuel pins.

Design Parameter	Value
Fuel OD	0.864 cm
Gap Thickness	0.02 cm
Cladding Thickness	0.063 cm
Cladding OD	1.03 cm
P/D	1.6
Active Fuel Height	120 cm
Gas Plenum Height (metallic fuel only)	90 cm
Average Coolant Density	10.25 g/cm^3

Local Reactivity Void Coefficient. While total and partial voiding of the core due to coolant overheating is nearly impossible because of the high boiling temperature of lead and lead-alloys, local voiding could be a possibility. A hypothetical situation could be a steam bubble passing through the core due to a steam tube rupture. Previous work [5, 9] has shown that the local void reactivity worth is ~ \$3 - \$5 worth of positive reactivity. This is a significant insertion of reactivity that could result in cladding failure due to overheating from a super-prompt-critical excursion, if other reactivity coefficients such as fuel expansion can not compensate. Relying solely on void worth for a negative reactivity insertion requires modification of the core or assemblies either passively or mechanically. Hejzlar et al. [10] devised a passive "streaming" assembly that allows for neutron leakage in the axial and radial directions. The design compensates for the reactivity increase, and creates a negative coefficient due to local voiding.

Doppler Reactivity Coefficient. Work done at MIT [11] has shown that, although small, the Doppler coefficient is negative for the fuels we studied. It is important to note that the computational tools used in this analysis (MCNP) take an enormous amount of computer time to calculate this coefficient. Other tools may be more efficient in calculating the Doppler coefficient, and will be sought for future work.

Reactivity Swing with Burnup. The burnup time steps taken in MOCUP for all cases were one-year steps with no outages. The results for the non-fertile fuel at linear power of 367.5 W/cm are shown in Figs. 1a through 1d. Note that the TRU loading was increased from 20wt% to 30wt% in the ThZr based fuel due to the fact that the 20wt% loading was not critical.

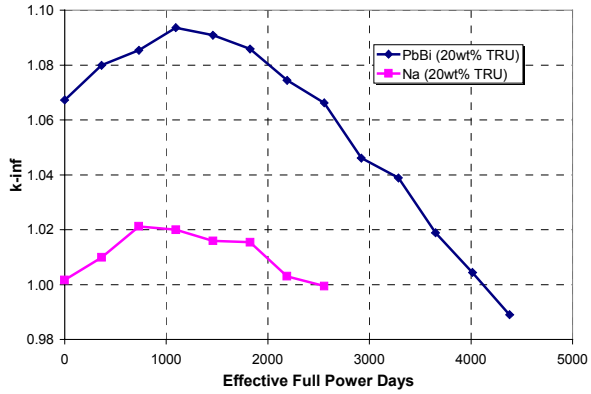

Figure 1a : reactivity comparison at 367.5 W/cm for ThN based fuel.

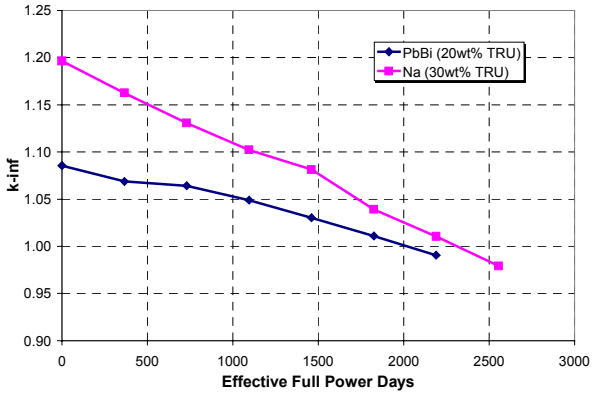

Figure 1b : reactivity comparison at 367.5 W/cm for ThZr based fuel.

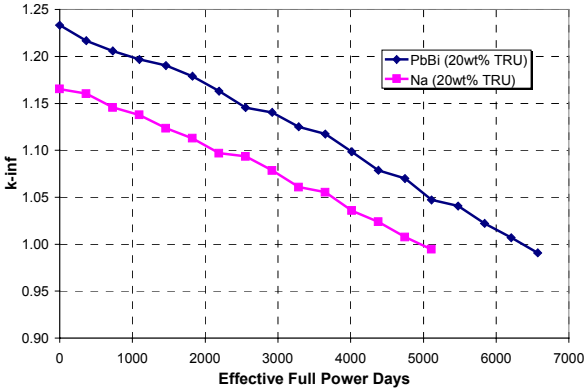

Figure 1c : reactivity comparison at 367.5 W/cm for UN based fuel.

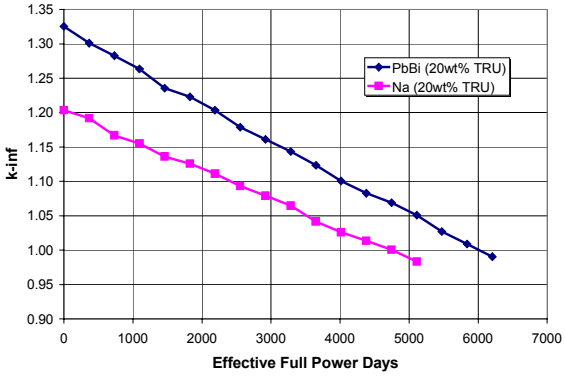

Figure 1d : reactivity comparison at 367.5 W/cm for UZr based fuel.

As can be seen in Figs. 1a through 1d, for similar TRU loadings the lead-bismuth cooled cases have lifetimes that are 26-65% longer than the sodium cooled cases. Of particular interest is the reactivity increase for the ThN based fuel. This is a result of the high conversion of ^{232}Th to ^{233}U.

Figures 2a through 2d show the results for a comparison of a derated linear power at 86 W/cm.

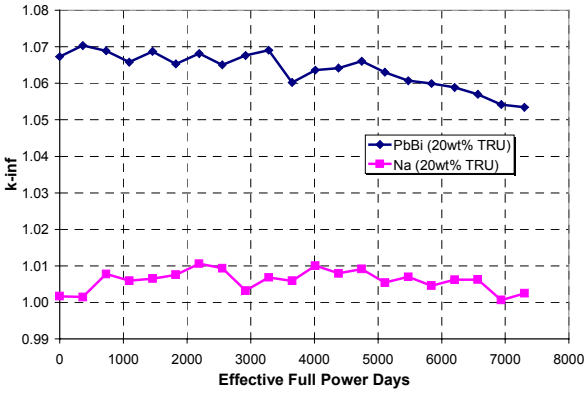

Figure 2a : reactivity comparison at 86 W/cm for ThN based fuel.

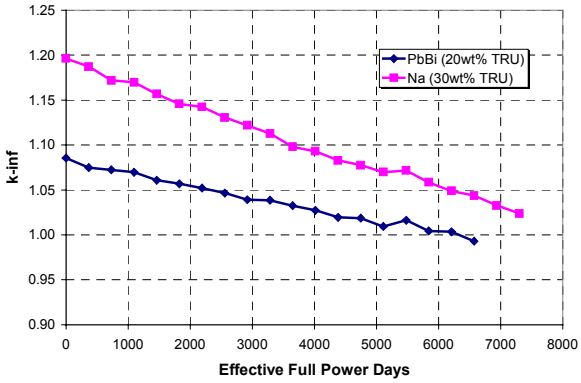

Figure 2b : reactivity comparison at 86 W/cm for ThZr based fuel.

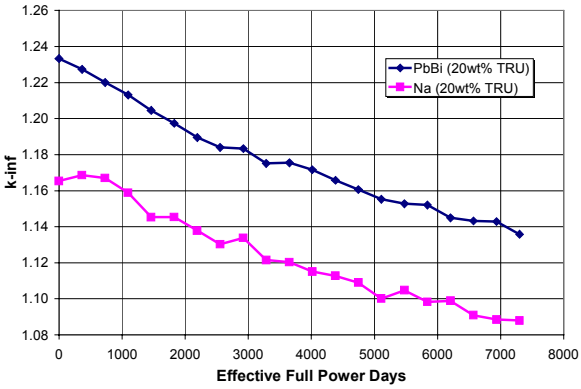

Figure 2c : reactivity comparison at 86 W/cm for UN based fuel.

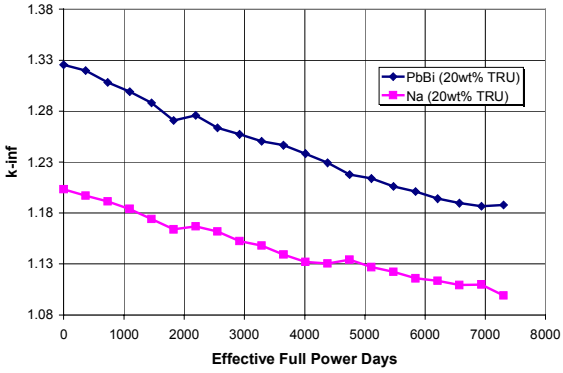

Figure 2d : reactivity comparison at 86 W/cm for UZr based fuel.

Again, the lead-bismuth cooled cases all have longer cycle lengths based on the reactivity versus effective-full-power-day (EFPD) curves. It is important to note that because these calculations were based on a pin cell with axial leakage only, the radial leakage of these cells was not accounted for and is significant in this type of reactor. Thus the effective-full-power-days that were calculated are an overestimation. However, the semi-infinite lattice parameters assumed in these calculations were kept constant in all cases and thus normalized the results presented.

4 - ISOTOPIC ANALYSIS

Actinide Destruction and Generation Rates. The net destruction rates of individual isotopes can be seen in Figs. 3a through 3d.

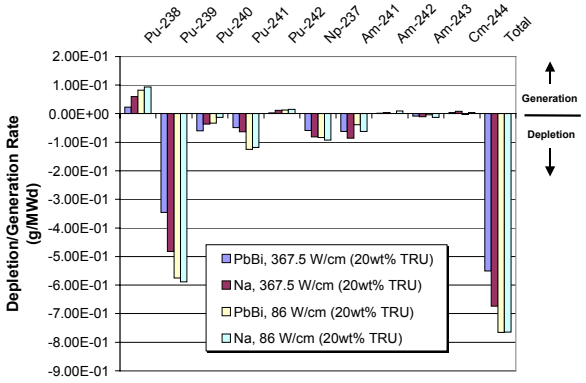

Figure 3a : average isotopic burnup rate for ThN based fuel.

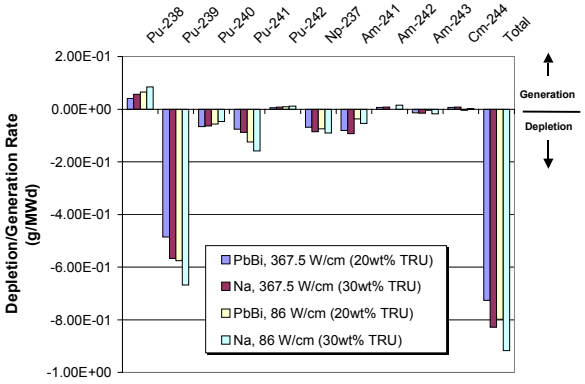

Figure 3b : average isotopic burnup rate for ThZr based fuel.

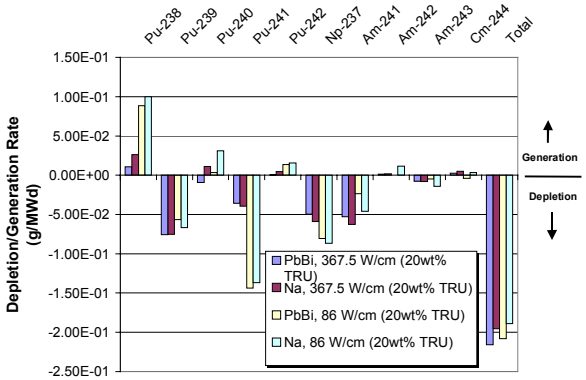

Figure 3c : average isotopic burnup rate for UN based fuel.

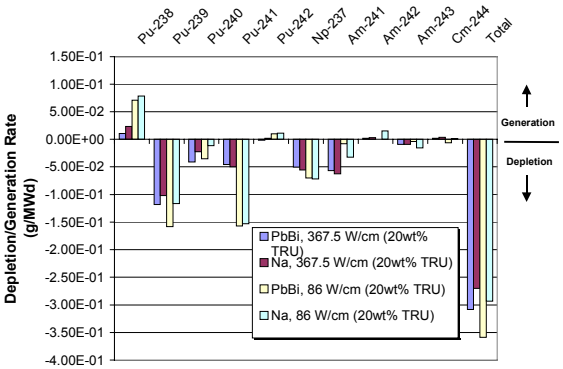

Figure 3d : average isotopic burnup rate for UZr based fuel.

The depletion rates are approximately the same for both coolants at similar linear powers. However, as was seen previously, the shorter fuel life of the sodium cooled cases with similar TRU loadings will not allow these particular cases to "burn" enough of the plutonium to be competitive with the lead-bismuth.

The overall actinide destruction rate for a 1-batch core can be seen in Figs. 4a through 4b, which compares the total burnup capability based on burnup rates and total core life.

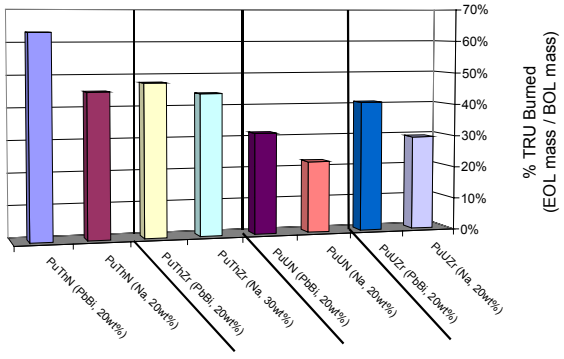

Figure 4a : 1-batch core actinide burnup comparison at a linear power of 367.5 W/cm.

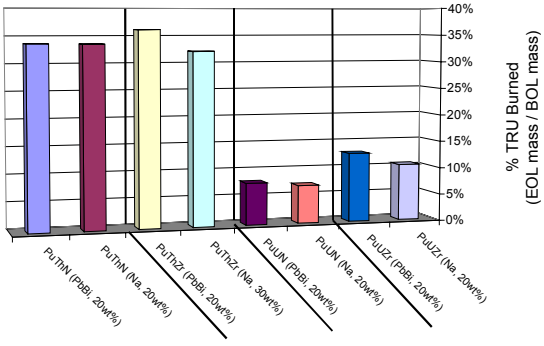

Figure 4b : 1-batch core actinide burnup comparison at a linear power of 86 W/cm.

The highest actinide burnup is achieved with the ThN based fuel in the lead-bismuth case at a linear power of 367.5 W/cm. In all cases, the actinide burnup using the lead-bismuth coolant is as good or better than the sodium cooled cases.

The hard spectrum in both lead-bismuth and sodium cooled reactors result in a high fission to capture ratio, making them attractive as an actinide burners. However, the longer core life for comparable initial actinide loadings in the lead-bismuth-cooled reactor appears to make it more suitable for actinide burning and long life.

Actinide Discharge. The end-of-life (EOL) actinide discharge for each case depends highly on the total actinides burned. When scaled on a per year basis, the sodium and lead-bismuth cooled cases are almost identical. But if no reprocessing or fuel shuffling is assumed, the actinide discharge would be considerably smaller in most cases for the lead-bismuth coolant when using comparable initial TRU loadings. Further reduction of the actinide discharge for a once through (no reprocessing) cycle can be accomplished by using a 3-batch cycle, which would increase the lifetime and, therefore, the total destruction by 50% [12]. However, the fluence limits would preclude the use of a multi-batch refueling scheme in these reactors.

Plutonium Content. Of the total discharged actinides, the total plutonium content decreases in all cases, with the fraction (or weight percent) of each plutonium isotope also changing. However, the isotopic fractions are dependent on the coolant and the effective-full-power-days, as can be seen in Table 3.

Table 3 : plutonium isotopics for once through cycle.

Fuel	Coolant	Initial TRU Loading (wt%)	Linear Power (W/cm)	EFPD	BOL Plutonium Fractions, All Cases (wt%) Pu-238	Pu-239	Pu-240	Pu-241	Pu-242
					2.0%	58.0%	26.0%	10.0%	4.0%
					EOL Plutonium Fractions (wt%)				
PuThN	PbBi	20	367.5	4120	13.4%	21.6%	45.3%	8.0%	11.6%
	Na	20	367.5	2499	12.2%	30.6%	40.2%	8.4%	8.7%
	PbBi	20	86	7300	10.0%	40.4%	37.3%	4.9%	7.3%
	Na	20	86	7300	10.7%	38.4%	38.2%	5.4%	7.3%
PuThZr	PbBi	20	367.5	2021	10.2%	34.5%	39.3%	7.4%	8.5%
	Na	*30*	*367.5*	*2313*	10.2%	35.6%	38.5%	7.5%	8.1%
	PbBi	20	86	6325	9.2%	41.3%	37.1%	5.0%	7.4%
	Na	*30*	*86*	*7300*	8.6%	43.4%	36.3%	4.8%	6.9%
PuUN	PbBi	20	367.5	6361	4.9%	55.1%	30.5%	4.4%	5.1%
	Na	20	367.5	4957	6.4%	52.6%	30.8%	5.0%	5.3%
	PbBi	20	86	7300	6.4%	57.8%	27.4%	3.5%	4.9%
	Na	20	86	7300	6.8%	56.2%	28.3%	3.8%	4.9%
PuUZr	PbBi	20	367.5	6015	5.5%	56.7%	28.5%	3.7%	5.6%
	Na	20	367.5	4757	6.5%	55.2%	28.8%	4.1%	5.4%
	PbBi	20	86	7300	5.9%	57.8%	27.8%	3.5%	5.1%
	Na	20	86	7300	6.0%	57.6%	27.9%	3.5%	5.0%

Of special interest is the change in the ratio of both ^{238}Pu and ^{239}Pu, where the ^{238}Pu ratio increases in all cases. The ^{239}Pu content is depleted significantly in the thorium base fuels, and the ^{238}Pu and ^{240}Pu contents increase in all cases. The ^{238}Pu and ^{240}Pu isotopes are important for non-proliferation concerns because of the decay heat produced by the ^{238}Pu, and the spontaneous neutrons produced by the ^{240}Pu. If significant percentages of these two isotopes are present in the plutonium, it will considerably reduce the yield of a weapon, and make handling of the plutonium difficult.

5 - CONCLUSION

In this paper, we chose metallic and nitride fuel using uranium or thorium, and compared the reactivity swings and actinide burning capabilities for two different coolants; sodium and lead-bismuth. The comparison was based on a constant P/D ratio, similar physical and material fuel parameters, similar fuel constituents, and one variation in the amount of loaded TRU.

The reactivity swing of each coolant case varied, depending on the initially loaded plutonium and minor actinides. Using equal TRU loadings, the lead-bismuth-cooled cases had the highest excess reactivity at BOL and the longest fuel cycles. By increasing the TRU loading from 20wt% to 30wt% in the ThZr based fuel, the sodium case had a higher BOL excess reactivity, and outperformed the lead-bismuth cases in cycle length. The

need for a higher loading of TRU in this sodium case can be attributed to the somewhat softer spectrum and, therefore, higher capture to fission ratio than with the lead-bismuth coolant. Also important are the superior reflective properties of lead-bismuth compared to sodium, thus requiring less enrichment than sodium.

In the case of actinide consumption rates, the coolants show similar effects; where the actinides are consumed at a rate of approximately 0.5-0.9 g/MWd. The longer in-core residence times for the lead-bismuth cases result in a higher total burnup at EOL, which can be up to 40% higher than the sodium cases for similar initial TRU loadings. Higher TRU loadings for the sodium cooled cases will result in higher EOL discharges.

The decrease in total TRU destruction at EOL for the low power option, as compared to the higher power option, is due to the increase in some minor actinides from generation and decay. It thus appears that a higher power will transmute more of the initial actinide inventory. Based on the linear reactivity model [12], a 3-batch refueling scheme would give 50% more burnup, which would further enhance the TRU transmutation. However, the cost of more frequent refueling may offset the gain in TRU destruction, and the fluence limits will restrict the actual lifetime.

The plutonium content of the fuel is depleted in all cases, and the plutonium isotopics of the fuel are superior by way of proliferation resistance compared to the other fuel forms (based on the higher ^{238}Pu and ^{240}Pu percentages, and lower ^{239}Pu percentage). The higher weight percentage of ^{238}Pu and ^{240}Pu in the fuel would make the fuel undesirable as a source for weapons grade material.

Based on the calculations performed, fast reactors can reduce the current inventory of plutonium and minor actinides found in spent LWR fuel, while attaining long core life. However, the choice of coolant and power density can affect the total amount of TRU consumed. Our calculations have shown that when comparing reactivity coefficients (e.g., using "streaming" assemblies to make the void coefficient negative), reactivity swing, and the amount of TRU consumed, lead-bismuth-cooled fast reactors can outperform their sodium-cooled counterparts.

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