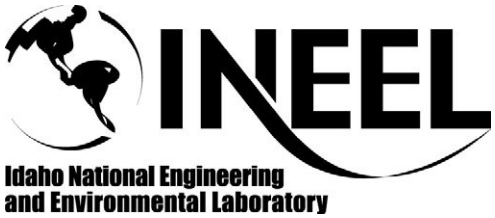

Idaho National Engineering
and Environmental Laboratory

INEEL/CON-00-01243
PREPRINT

Performance Comparison Of Metallic, Actinide Burning Fuel In Lead-Bismuth And Sodium Cooled Fast Reactors

K. D. Weaver
J. S. Herring
P. E. MacDonald

April 8, 2001

9th International Conference On Nuclear Engineering (ICONE-9)

PERFORMANCE COMPARISON OF METALLIC, ACTINIDE BURNING FUEL IN LEAD-BISMUTH AND SODIUM COOLED FAST REACTORS

K. D. WEAVER, J. S. HERRING, P. E. MACDONALD

Idaho National Engineering and Environmental Laboratory
Advanced Nuclear Energy
P.O. Box 1625
Idaho Falls, Idaho 83415-3850 USA
Phone: 208-526-0321
FAX: 208-526-2930
E-mail: weavkd@inel.gov

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

ABSTRACT

Various methods have been proposed to "incinerate" or "transmutate" the current inventory of trans-uranic waste (TRU) that exits in spent light-water-reactor (LWR) fuel, and weapons plutonium. These methods include both critical (e.g., fast reactors) and non-critical (e.g., accelerator transmutation) systems. The work discussed here is part of a larger effort at the Idaho National Engineering and Environmental Laboratory (INEEL) and at the Massachusetts Institute of Technology (MIT) to investigate the suitability of lead and lead-alloy cooled fast reactors for producing low-cost electricity as well as for actinide burning. The neutronics of non-fertile fuel loaded with 20 or 30-wt% light water reactor (LWR) plutonium plus minor actinides for use in a lead-bismuth cooled fast reactor are discussed in this paper, with an emphasis on the fuel cycle life and isotopic content. Calculations show that the average actinide burn rate is similar for both the sodium and lead-bismuth cooled cases ranging from -1.02 to -1.16 g/MWd, compared to a typical LWR actinide generation rate of 0.303 g/MWd. However, when using the same parameters, the sodium-cooled case went subcritical after 0.2 to 0.8 effective full power years, and the lead-bismuth cooled case ranged from 1.5 to 4.5 effective full power years.

BACKGROUND AND 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 to weapons material. In an effort to address the waste disposal and proliferation concerns, systems capable of transmuting (i.e., fissioning) the long-lived higher actinides have been proposed. These include both sub-critical (accelerator) and critical systems. The Los Alamos National Laboratory (LANL) has proposed a lead-bismuth cooled, accelerator-driven, sub-

critical actinide burner for burning actinides and long-life fission products from spent light water reactor 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 (Hill et al., 1995) 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 limiting neutron fluences. 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 following material characteristics: chemical inertness of lead-bismuth with air and water; high atomic number; low absorption cross section; low vapor pressure; and a small volume change upon solidification. These basic properties lead to the following advantages for a lead-bismuth coolant:

- harder neutron spectrum and, therefore, improved neutron economy, especially when burning actinides;
- better reflective properties, making it is possible to get breeding even without blankets;
- better shielding against gamma-rays and energetic neutrons;
- high boiling temperature and high heat of vaporization of lead-bismuth (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;
- simpler containment structure due to the impossibility of fires and explosions; and
- a small volume change upon solidification.

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.

A significant inventory of actinides exists in spent LWR fuel (estimated at approximately 165,000 tonnes of heavy metal, with 1500 tonnes of that being plutonium), which can be used and burned in a fast reactor. 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., (1998) at UC Berkeley has 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 can have 'modules' added for extra capacity. However, these concepts use fertile fuels while 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.

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). However, the production of Po-210 can also be regarded as a proliferation resistant feature.

Fluence limits are also a concern, where the harder spectrum found in lead-based systems will reduce the burnup capability unless they are derated. Although important, this is a materials issue, which is discussed in more detail by Hill et al. (1999).

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 (Adamov et al., 1997) and performance of different fuel types (Weaver et al., 2000). In the remainder of this paper, we will limit our discussion to the actinide burning performance of non-fertile fuel in lead-bismuth and sodium cooled fast reactors.

To effectively transmute plutonium and minor actinides from spent LWR fuel, it is desirable to minimize the waste of neutrons in order to attain a large surplus available for transmutation. 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 spectrum achievable because the fuel does not contain any moderating isotopes.

To maximize the *actinide transmutation* capability of the system, breeding of new fissile material must be minimized or eliminated, making the presence of fertile

isotopes undesirable. The choice of fuel composition for maximum actinide transmutation is then restrained to the to-be-transmuted plutonium and minor actinides (20 to 30-wt%) and to the zirconium matrix (70 to 80-wt%), constituting the structural component of the fuel rods. It should be noted that 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.

The choice of a non-fertile metallic fuel 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 amount of fertile isotopes in the non-fertile metallic fuel is very small (mostly Pu-238 and Pu-240). Moreover, the hard spectrum leads to a decrease of absorption rate in the resonance peaks. Both these factors result in a very small Doppler feedback, and measures to attain a reasonably negative Doppler coefficient must be employed for non-fertile fuel. This is especially true for lead or lead-bismuth cooled cores.

Reactivity Swing with Burnup

A major consequence of the absence of fertile isotopes in the non-fertile fuel is the lack of breeding, and therefore a faster net depletion of the fissionable

material, with the result being a marked reduction of reactivity during a cycle. To ensure the reactor will remain critical for a reasonable amount of time, the excess reactivity at beginning-of-life (BOL) must be large, making this design potentially vulnerable to accidents related to malfunction of the control rods (e.g., rapid control rod ejection).

NEUTRONIC ANALYSIS

Analysis Tools and Fuel Parameters

The current work uses the MOCUP (MCNP-ORIGEN2.1 Coupled Utility Program) code 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 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 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 fuel studied was a non-fertile metallic fuel that 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 20-30 wt%. Table 1 gives a summary of the fuel composition for fertile and non-fertile fuel.

Table 1. BOL fuel 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%**
Zirconium Matrix	**80%**

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. Also, we were not interested in a high breeding ratio typical of most fast reactor systems.

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 an upper and lower bound. The power level issue 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 adjusted or 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
Gap Material	33wt% Pb – 33wt% Sn – 33wt% Bi
Cladding Thickness	0.063 cm
Cladding OD	1.03 cm
P/D	1.6
Active Fuel Height	120 cm
Gas Plenum Height	90 cm

The metallic fuel has two interesting properties that are important to this type of fuel:

- the gas plenum above the fuel,
- axial expansion, and
- the materials used in the gap between the fuel and the cladding.

The gas plenum is used to store the fission gases as they diffuse from the metallic fuel, and the gap material enhances the thermal conductivity.

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 from a steam bubble passing through the core due to a steam tube rupture. Table 3 shows the reactivity results of voiding the center half of the fuel.

Table 3. Calculated local void reactivities at BOL.

TRU Loading (wt%)	**Coolant**	**Void Reactivity** (%Δk/k)
20	Na	0.73%
20	Pb-Bi	1.09%
30	Na	1.38%

Note that the worth of the reactivity is dependent on the delayed neutron fraction (β), which itself is dependent on the content of the fuel. MCNP calculations show that the local void reactivity worth is greater than \$4. 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., (1999a) devised a passive "streaming" assembly that allows for neutron leakage in the axial and radial directions (see Fig. 1). The design compensates for the reactivity increase, and creates a negative coefficient due to local voiding.

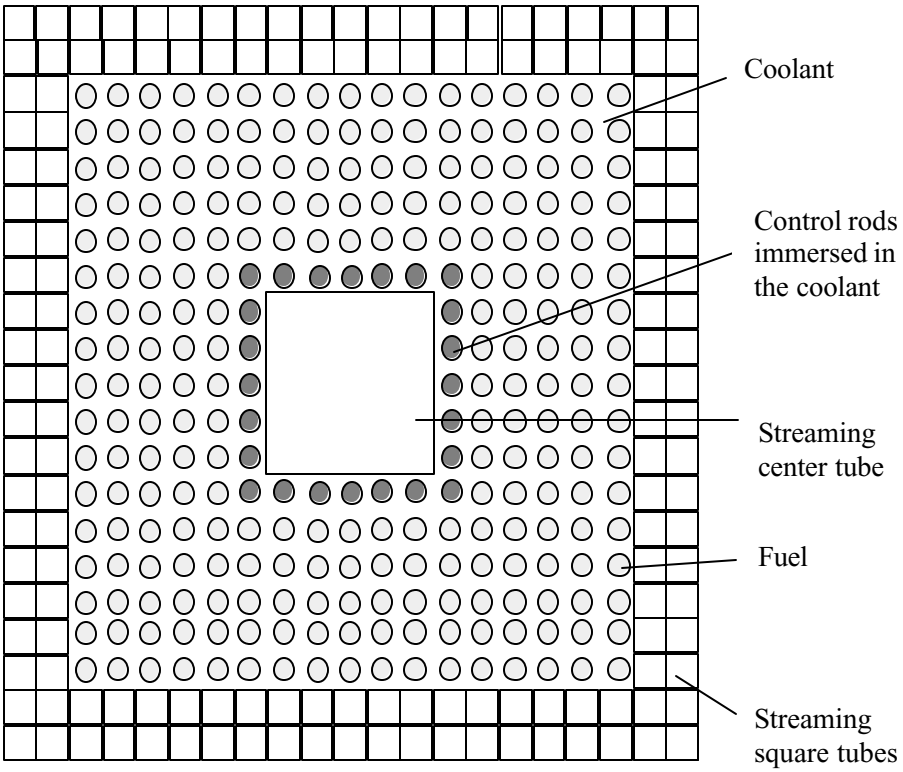

Figure 1. Schematic of Hejzlar streaming assembly.

Doppler Reactivity Coefficient

Work done at MIT (Hejzlar et al., 1999b) has shown that, although small, the Doppler coefficient is negative for the studied non-fertile fuel. 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 after 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 Fig. 2. Note the extremely short fuel life in the sodium-cooled case as compared to the lead-bismuth-cooled case.

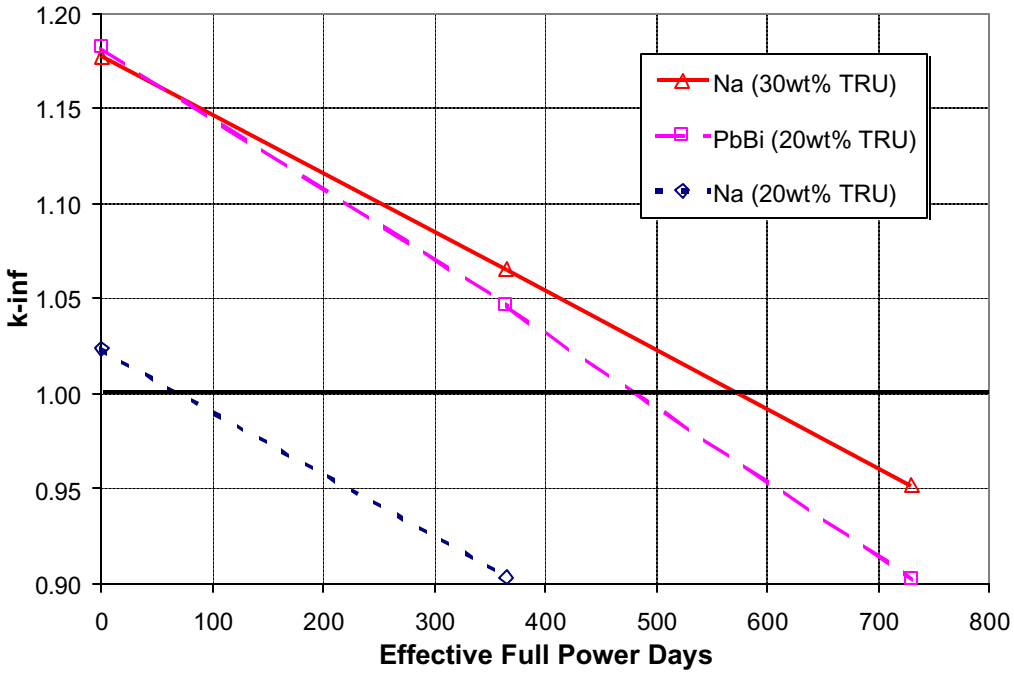

Figure 2. Reactivity comparison at 367.5 W/cm.

In order to extend the fuel life in the sodium-cooled case to make it comparable to the lead-bismuth-cooled case, an increase of approximately 10 wt% in added TRU is needed. The consequences of this will be discussed later.

Figure 3 shows the results for a comparison of a derated linear power at 86 W/cm. While increasing the overall fuel life, an increase in beginning-of-life TRU is required in the sodium cooled case to extend its life further, and thus increase the total amount of actinides burned in the cycle.

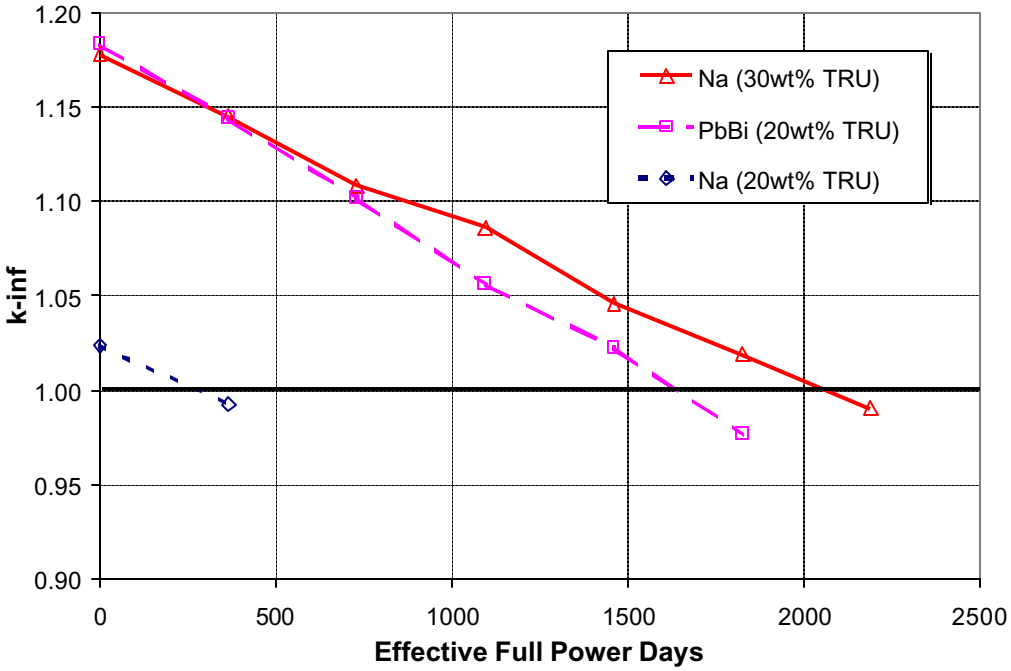

Figure 3. Reactivity comparison at 86 W/cm.

ISOTOPIC ANALYSIS

Actinide Destruction and Generation Rates

As stated previously, the actinide transmutation rate is dependent on whether fertile or non-fertile fuel is used. A higher net actinide destruction rate of the initially loaded plutonium and minor actinides is achieved with the non-fertile fuel, with the sodium and lead-bismuth cases being approximately equal, as can be seen in Fig. 4-5.

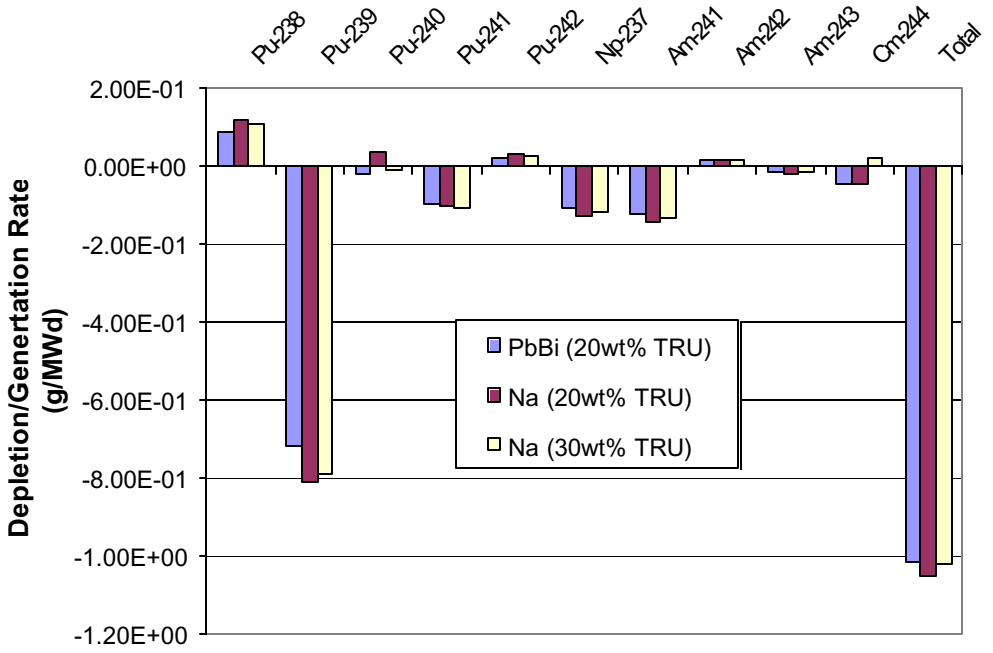

Figure 4. Average isotopic burnup rate at a linear power of 367.5 W/cm.

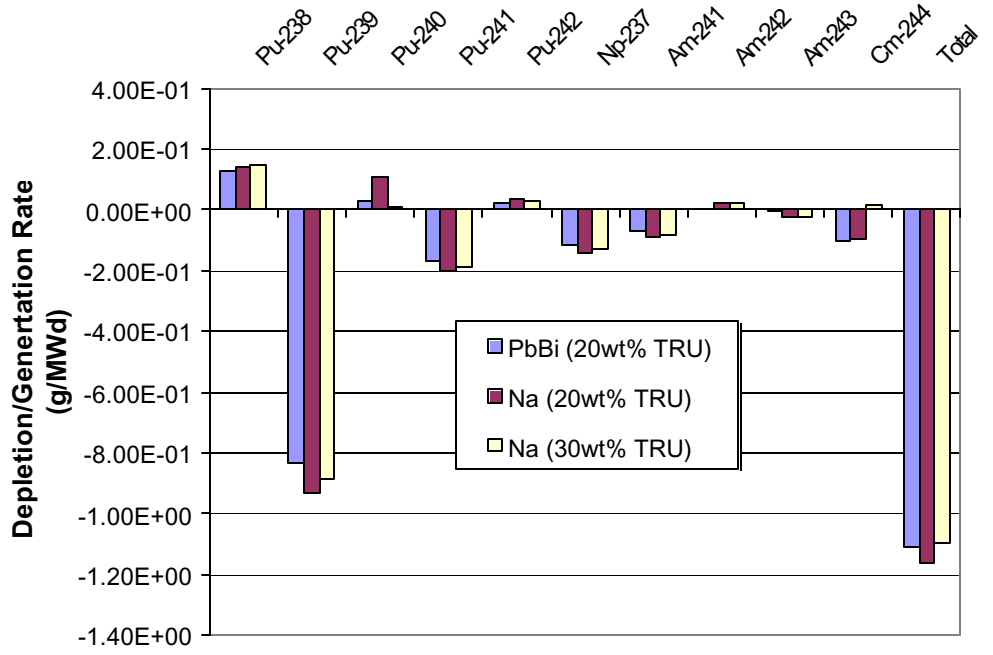

Figure 5. Average isotopic burnup rate at a linear power of 86 W/cm.

The plutonium destruction rates, particularly the Pu-239, gives a slight advantage to the 20-wt% loaded, sodium cooled case. However, as was seen previously, the shorter fuel life will not allow this particular case to “burn” enough of the plutonium to be competitive with the lead-bismuth. A normalized overall actinide

destruction rate for a 1-batch core can be seen in Fig. 6 that compares the total burnup capability based on burnup rates and total core life.

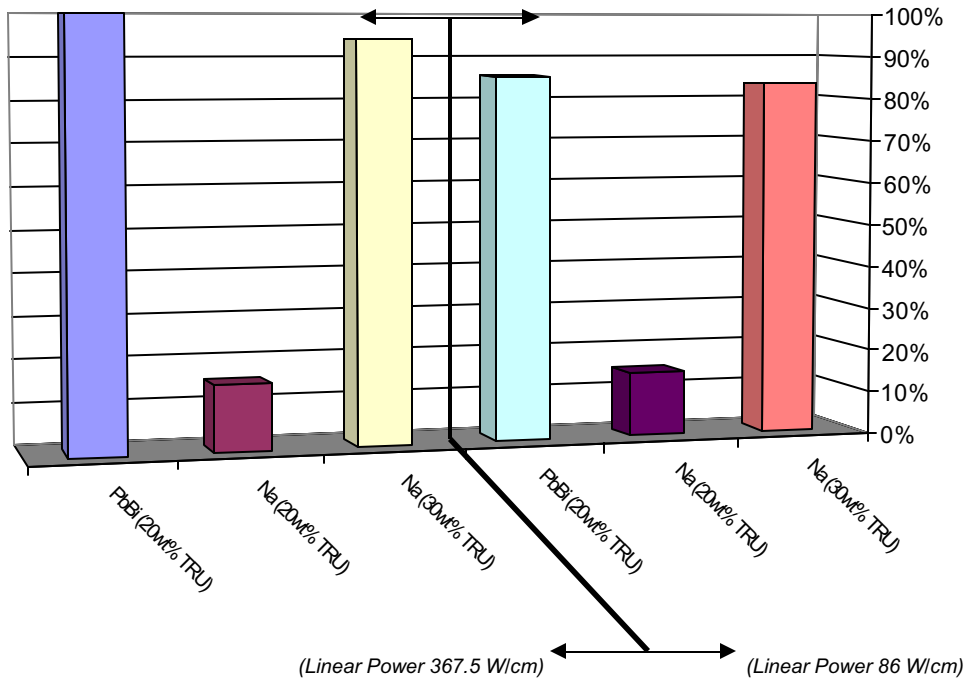

Figure 6. Normalized 1-batch core actinide burnup comparison.

The highest actinide burnup is achieved in the lead-bismuth case at a linear power of 367.5 W/cm. Note that this case was used as the final normalizing factor.

Hill et al., (1995) studied a sodium cooled fast reactor as a pure actinide burner using metallic fuel, and estimated an actinide destruction rate of 308 kg/FPY for an 840 MW(th) reactor. By scaling the power to 830 MW(th), the equivalent destruction rate would be 304 kg/FPY for a sodium-cooled reactor. The cases studied in this paper have an average actinide destruction rate that is approximately 10% greater than that reported by Hill et al. (1995). Most of this can be explained by the fact that not all capture and decay gamma energy was included in the MCNP calculations. 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 makes it more suitable for actinide burning.

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 discharge would be considerably less for the lead-bismuth cases 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%. If reprocessing is assumed, a further reduction in discharged actinides will occur. However, reprocessing in the U.S. is not option at this time and is not considered further.

Plutonium Content

Of the total discharged actinides, the plutonium content varies from 76-wt% to 82-wt%. For plutonium proliferation concerns, 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 4.

Table 4. Plutonium isotopics for once through cycle.

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%)				
PbBi	20	367.5	482	7.3%	42.5%	34.9%	8.7%	6.6%
Na	20	367.5	72	4.9%	49.4%	31.0%	9.3%	5.3%
Na	30	367.5	575	6.5%	45.5%	33.0%	9.0%	6.1%
PbBi	20	86	1639	5.9%	48.3%	32.5%	7.8%	5.5%
Na	20	86	276	2.7%	56.0%	27.5%	9.5%	4.3%
Na	30	86	2070	5.9%	49.1%	31.7%	7.7%	5.6%

Of special interest is the change in the ratio of both Pu-238 and Pu-239, where the Pu-238 ratio increases in all cases. The Pu-239 content is depleted significantly in the cases where the linear power is high, and the Pu-238 and Pu-240 ratios increase in all cases. The Pu-238 and Pu-240 isotopes are important for non-proliferation concerns because of the decay heat produced by the Pu-238, and the spontaneous neutrons produced by the Pu-240. 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.

SUMMARY AND CONCLUSIONS

The choice of fuel for a fast reactor depends on the objective of the reactor. If the goal is to burn the greatest amount of actinides possible, then a non-fertile metallic fuel is the best choice. The next choice for an actinide-burning reactor is the coolant to be used. In this paper, we chose a metallic fuel 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 by 10-wt% (to 30-wt% Pu+MA), the sodium cases had a similar BOL excess reactivity, but outperformed the lead-bismuth cases in cycle length. The need for a higher loading of TRU in the sodium cases 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 -1 g/MWd. When compared to the generation rates from LWR's (at +0.303 g/MWd), the consumption rate of one actinide burning reactor should be able to accommodate three LWR's (assuming similar power outputs). However, the longer in-core residence times for the lead-bismuth cases result in a higher total burnup at EOL, which can be up to 85% higher for similar initial TRU loadings. Higher TRU loadings for the sodium cooled cases will result in higher EOL discharges, and short in-core residence times may be a concern based on the need for more frequent refueling.

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 Pu-238 and Pu-240 percentages, and lower Pu-239 percentage). The higher weight percentage of Pu-238 and Pu-240 in the fuel would make them undesirable as a source for weapons grade material.

Based on the calculations performed, fast reactors can significantly reduce the current inventory of plutonium and minor actinides. However, the choice of coolant 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.

ACKNOWLEDGMENTS

This work was supported through the INEEL Laboratory Directed Research and Development Program under DOE Idaho Operations Office Contract DE-AC07-99ID13727.

REFERENCES

Adamov, E., Orlov, V., Filin, A., Leonov, V., Sila-Novitski, A., Smirnov, V., and Tsikunov V., 1997, "The Next Generation of Fast Reactors," Nuclear Engineering and Design, Vol. 173, pp. 143-150.

Greenspan, E., Elias, E., Kastenberg, W. E., and Stone, N., 1998, "Non-Proliferating Liquid Metal Cooled Reactors for Developing Countries," Technical Report CNTWM98-2, Univ. of California at Berkeley, Berkeley, CA.

Hejzlar, P., Driscoll, M., and Kazimi, M., 1999a, "Void Reactivity Performance in Lead-Bismuth-Cooled Reactors for Actinide Transmutation," Topical Report MIT-ANP-TR-068, Massachusetts Institute of Technology, Cambridge, MA.

Hejzlar, P., Driscoll, M., and Kazimi, M., 1999b, "Neutronic Design for a Pb-Bi-Cooled Actinide Burner Fast Reactor," Transactions of the American Nuclear Society, Vol. 81, pp. 271-273.

Hill, R. N., Wade, D. C., Liaw, J. R., and Fujita, E. K., 1995, "Physics Studies of Weapons Plutonium Disposition in the Integral Fast-Reactor Closed Fuel-Cycle," Nuclear Science and Engineering, Vol. 121, pp. 17-31.

Hill, R. N., Cahalan, J. E., Khalil, H. S., and Wade, D. C., 1999, "Development of Small, Fast Reactor Core Designs Using Lead-Based Coolant," Proceedings of GLOBAL 1999, Jackson Hole, WY.

Weaver, K. D., Herring, J. S., and MacDonald, P. E., 2000, "Performance Modeling of Metallic and Nitride Fuels in Advanced Lead-Bismuth Cooled Fast Reactors," Proceedings of ICONE-8, Baltimore, MD.