The Radiological and Thermal Characteristics of Fission Waste from a Deep-Burn Fusion-Fission Hybrid (LIFE) and Implications for Repository Performance

Henry F. Shaw
Lawrence Livermore National Laboratory
7000 East Avenue
Livermore, California 94550

Contributors: Jim Blink, Joseph Farmer, Jeff Latkowski, Kevin Kramer

We are studying the use of a Laser Inertial-confinement Fusion Engine (LIFE) to drive a hybrid fusion-fission system that can generate electrical power and/or burn nuclear waste [1] The system uses the neutrons from laser-driven ICF to produce tritium and to drive nuclear reactions in a subcritical fission blanket. The fusion neutron source obviates the need for a self-sustaining chain reaction in the fission blanket. Either fissile or fertile could be used as fission fuel, thus eliminating the need for isotopic enrichment. The "driven" system potentially allows very high levels of burnup to be reached, extracting a large fraction of the available energy in the fission fuel without the need for reprocessing. In this note, we discuss the radionuclide inventory of a depleted uranium (DU) fuel burned to greater than 95% FIMA (Fissions per Initial heavy Metal Atom), the implications for thermal management of the resulting waste, and the implications of this waste for meeting the dose standards for releases from a geological repository for high-level waste [2].

The fission waste discussed here would be that produced by a LIFE hybrid with a 500-MW fusion source. The fusion neutrons are multiplied and moderated by a sequence of concentric shells of materials before encountering the fission fuel, and fission in this region is largely due to thermal neutrons. The fission blanket consists of 40 metric tons (MT) of DU, assumed to be in the form of TRISO-like UOC fuel particles embedded in 2-cm-diameter graphite pebbles. (It is recognized that TRISO-based fuel may not reach the high burnup of the fertile fuel considered here, and other fuel options are being investigated. We postulate the existence of a fuel that can reach >95% FIMA so that the waste disposal implications of high burnup can be assessed.) The engine and plant design considered here would receive one load of fission fuel and produce ~2 GWt of power (fusion + fission) over its 50- to 70-year lifetime.

Neutron and photon transport calculations were performed using MCNP5 [3]. Burnup calculations were performed using a modified version of Monteburns 2.0 [4]. The nuclear data used were from ENDF/B-VII [5]. Additional details of the burn calculations can be found in [6]. For comparison to spent fuel from light water reactors (LWRs), we use the projected initial inventory of PWR and BWR fuels (current average age of 23 years since discharge) used for the Yucca Mountain Project Final Environmental Impact Statement [7]. The decay of this initial inventory to 1 million years was calculated using ORIGEN2 [8].

The hybrid system considered here would have generated ~44 GWe-yr of energy at 99% FIMA [2, 6]. The energy generated per MT is therefore about 1100 MWe-yr/MT. In contrast, using average burnups of 41.2 GWt-day/MT and 33.6 GWt-day/MT for the PWR and BWR fuel slated for disposal at Yucca Mtn. [7], and assuming a thermal-electric conversion efficiency of ~33%, the total energy generated by the 68,000 MT "Yucca Mtn. inventory" is ~2500 GWe-yr, or ~37 MWe-yr/MT, which is ~30 times less energy per MT than the waste from the hybrid. Clearly, relative to the current once-through fuel cycle, the use of a deep-burn hybrid to generate electricity would significantly reduce the need for repository capacity.

For the first ~300 years after discharge, the specific activity (Ci/MTIHM) for deep-burn waste with > 95% FIMA is significantly higher than that of average LWR fuel (Fig. 1a). Furthermore, the specific activity of 95% FIMA waste remains above that of average LWR fuel for all times. Waste with a burnup of 99% FIMA has a specific activity similar to that of average spent LWR fuel up between ~300 years to ~100,000 years post discharge, while the 99.9% FIMA waste has a specific activity less than that of average LWR fuel from ~300 years to 100,000 years post discharge. At very long times (>300,000 years), the specific activities of the spent hybrid fuels for all three burnups are somewhat higher than that of current average spent LWR fuel. However, when normalized to the total electrical energy generated (Fig. 1b), the radioactivity per-unit-energy-generated of spent hybrid fuel is much less than that of similarly normalized spent LWR fuel, and hence the benefit-to-hazard ratio of the hybrid waste is significantly better than that of spent LWR fuel. Nevertheless, the spent fission fuel from a deep-burn hybrid engine is a hazardous material that would require isolation from the biosphere for hundreds of thousands of years.

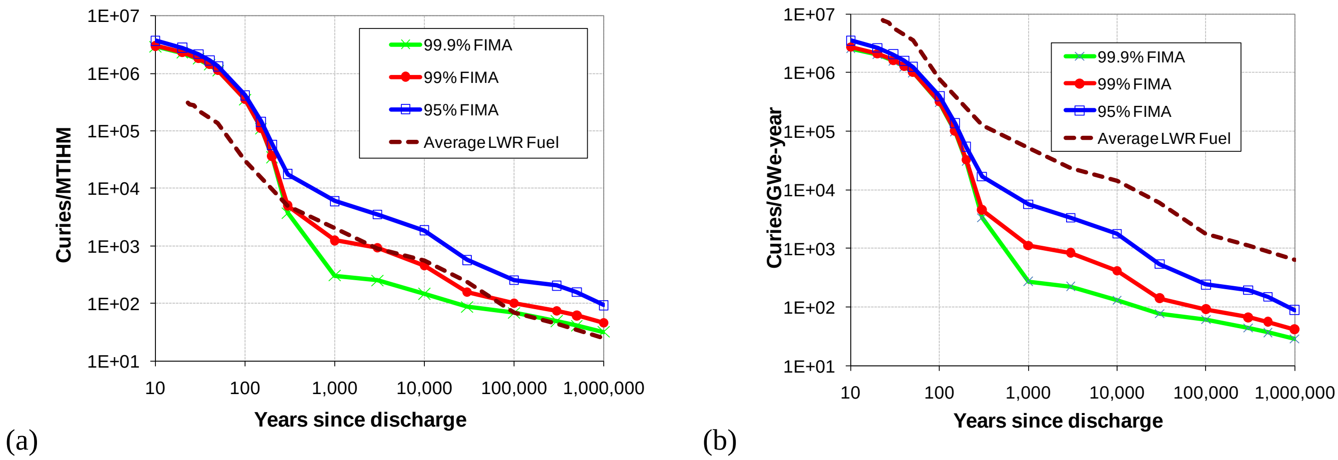

Figure 1. Activity of deep-burn fission waste compared to average spent LWR fuel

As a consequence of the higher specific activity, the hybrid waste also has a higher specific thermal power than spent LWR fuel. Management of the short-term decay heat of the high-burnup waste will likely require cooling using a thermal transfer medium. Calculations of the heat-transfer requirements indicate that by using appropriate designs, the heat load can be managed during interim storage of the fuel [9]. We have also conducted calculations for the thermal evolution of a Yucca-Mtn.-like repository being loaded with very young (5-years post discharge), high-burnup hybrid fuel, and shown that by spreading the thermal load over several emplacement drifts, the Yucca Mountain thermal limits on waste, waste package, drift wall, and mid-pillar temperatures can be met [9].

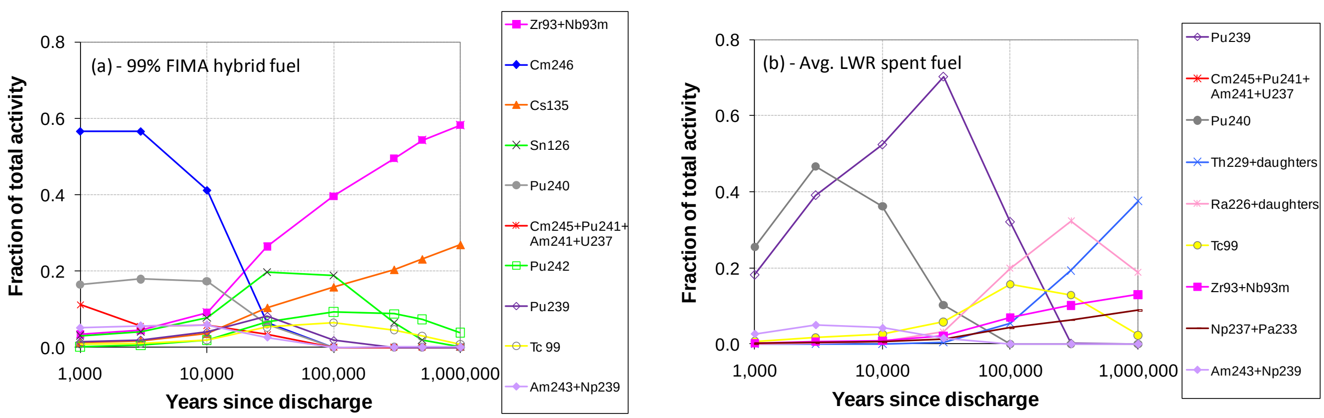

Figure 2. Fractional contribution of individual nuclides and decay chains to the total activity of (a) 99% FIMA hybrid fuel and (b) average LWR spent fuel for the period 1000 to 1,000,000 years post discharge. Figures include all nuclides or chains contributing > 5% to the total activity at any time during these periods.

For decay times of less than ~300 years, the activity of spent hybrid fuel is dominated by short-lived fission ^{137}Cs + ^{137m}Ba, and ^{90}Sr + ^{90}Y, which are the same nuclides responsible for most of the activity of spent LWR fuel during this time. Between 300 and a few tens of thousands of years ^{246}Cm, ^{245}Cm+daughters, and ^{240}Pu are the dominant sources of radioactivity in the spent hybrid fuel (Fig. 2a). At times greater than ~20,000 years, fission products (^{135}Cs, ^{126}Sn, ^{93}Zr + ^{93m}Nb and ^{99}Tc) again become the dominant activities. ^{242}Pu is the only actinide that contributes more than 5% of the total activity during the post-100,000-year time period. This differs substantially from average spent LWR fuel, in which the long-term activity is dominated by the decay of ^{99}Tc, ^{241}Am, ^{239}Pu, ^{240}Pu, ^{242}Pu, ^{226}Ra+daughters, and ^{229}Th+ daughters (Fig 2b).

Table 1. Uranium and transuranic (TRU) element content of hybrid fuel as a function of burnup. Data are also provided for average spent LWR fuel.

	kg per MT initial U at discharge at indicated burnup						kg per GWe-year at discharge at indicated burnup					
% FIMA	60%	80%	95%	99%	99.9%	avg. LWR	60%	80%	95%	99%	99.9%	avg. LWR
U	243	106	24	4.5	0.21	955	366	121	23	4.1	0.18	24476
Np	0.91	0.47	0.16	0.039	0.007	0.68	1.4	0.53	0.15	0.035	0.006	17
Pu	135	69	12	0.7	0.02	10	203	79	11	0.65	0.018	254
Am	10	7.3	1.8	0.17	0.004	1.2	15.3	8.3	1.7	0.16	0.004	30
Cm	13	16	11	4.4	1.0	0.03	19	18	11	4.0	0.93	0.69
Bk	0.008	0.021	0.026	0.016	0.001	--	0.012	0.023	0.025	0.014	0.001	--
Cf	0.026	0.054	0.072	0.100	0.010	--	0.039	0.061	0.068	0.090	0.009	--
Total TRU	158	93	25	5.4	1.1	12	239	106	24	4.9	0.97	302

The actinide concentration in the hybrid fuel as a function of burnup is given in Table 1. From 20% to ~60% FIMA, the fuel contains roughly 10 times the concentration of transuranic elements (TRU) than average LWR fuel at the time of discharge. With increasing FIMA, these elements begin to "burn out", and by 95% FIMA, their concentration is approximately twice that in average spent LWR fuel. By 99% FIMA, they are at half the concentration in LWR fuel, and at 99.9% FIMA, they are about a tenth the LWR concentration. Comparing the production of TRU as a function of the energy produced by the hybrid *vs.* a conventional LWR with no reprocessing, the hybrid produces less TRU per unit energy generated for all burnups greater than ~50%. Per unit energy generated, a hybrid operating to a burnup of 99% FIMA would produce ~60 times less TRU waste than an average LWR.

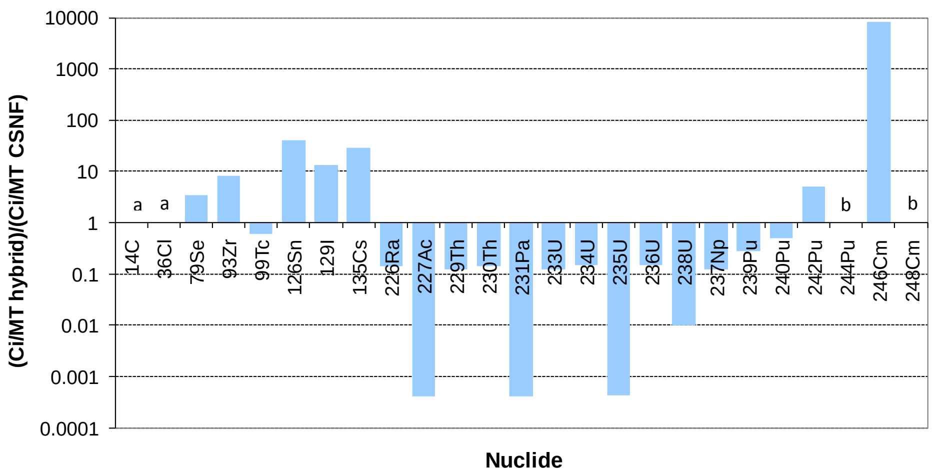

Figure 3. Ratios of the specific activities of radionuclides important for long-term repository performance in spent hybrid fuel (99% FIMA) to their values for average spent LWR fuel. Notes: (a) data on ^{14}C and ^{36}Cl are not available for hybrid waste due to limitations in the nuclear cross section data used; (b) ^{242}Pu and ^{248}Cm are not present in significant quantities in spent LWR fuel.

Total system performance assessment (TSPA) results for the Yucca Mtn. license application [12] indicate that ^{14}C, ^{99}Tc, ^{129}I, and ^{239}Pu would dominate the dose to the maximally exposed individual for the first 10,000 years after repository closure. ^{239}Pu, ^{129}I, and ^{226}Ra dominate the dose for the next 90,000 years, and ^{226}Ra, ^{237}Np, and ^{242}Pu, dominate the dose from 100,000 to 1,000,000 years. Figure 3 shows the ratios of the specific activities of these radionuclides in hybrid waste (99% FIMA) to their values for average LWR fuel. When this ratio varies over time, the maximum value is shown. With few exceptions, the specific activity of the actinides and daughter products are much lower in the hybrid waste than in average LWR fuel; hybrid waste has higher activities of ^{242}Pu, ^{244}Pu, ^{246}Cm, and ^{248}Cm. As would be expected from the higher burnups, all the fission products, except ^{99}Tc have higher specific activities in the hybrid waste. When normalized to the energy produced, however, only ^{126}Sn and ^{246}Cm have higher abundances in the hybrid waste.

By making the simplifying assumption that the dose from a repository to the maximally exposed individual for a given radionuclide is proportional to its inventory in the repository, one can make a first-order assessment of the impact of replacing the spent LWR fuel in a repository with deep-burn hybrid waste. This calculation entails a number of additional, generally conservative, assumptions that are discussed in [2]. The results indicate that for the post-25,000 year period, the doses from a "hybrid repository" containing the same mass of initial heavy metal (at 99% FIMA) as proposed for Yucca Mtn. would be ~5 times higher than the YMP TSPA-LA results for this time period, though the waste in the repository would have produced ~28 times the energy. The doses do not come close to approaching the proposed NRC limit of 350 mrem/yr [10, 11]. Most of the dose from the hydrid waste would come from ^{129}I, ^{126}Sn, ^{135}Cs, and ^{242}Pu. Note that this calculation provides insight into which radionuclides in hybrid waste would be likely to contribute significantly to the dose from a repository similar to the proposed repository at Yucca Mtn.; other geologic settings or repository designs could have significantly different results. Note also that this assessment only pertains to the post-25,000-year performance period. A similar assessment for times < 25,000 years cannot be done yet because we lack information on the ^{14}C content (a major contributor to does during this period) of hybrid waste due to limitations in the cross-section data used for the burn calculations and poor constraints on the ^{14}N content (the primary precursor of ^{14}C) of the hybrid fuel.

CONCLUSIONS

Hybrid fusion-fission systems could allow very high burnups to be achieved in a subcritical fission blanket consisting of non-enriched fuel. Spent, deep-burn fuel will have higher specific activity and thermal power than spent LWR fuel. Plausible designs for interim storage containers and cooling configuration can remove the heat without exceeding fuel temperature limits. Calculations also indicate that a repository for such fuel can be designed that would perform within the thermal limits established for the proposed Yucca Mtn. repository. Because of the more efficient use of the uranium fuel, the amount of heavy metal waste generated per unit energy from a deep-burn hybrid would be 25-30 times less than for the current once-through fuel cycle, and would not require multiple cycles of reprocessing. The dose from a properly designed and sited repository containing such waste should be well below current regulatory standards, and would be dominated by ^{129}I, ^{126}Sn, ^{135}Cs, and ^{242}Pu activities.

This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344. LLNL-CONF-416698.

References

1. Moses, E.I., de la Rubia, T.D., Storm, E., Latkowski, J.F., Farmer, J.C., Abbott, R.P., Kramer, K.J., Peterson, P.F., Shaw, H.F., Lehman, R.F., "A Sustainable Nuclear Fuel Cycle Based on Laser Inertial Fusion Energy", *Fusion Sci. Tech.*, 56, 547-565, (2009).
2. Shaw, H.F., and Blink, J.A., LIFE Materials: Fuel Cycle and Repository, Volume 11. LLNL-TR-409725. Lawrence Livermore National Laboratory, Livermore, CA. 100pp (2008).
3. Los Alamos National Laboratory "MCNP - A General Monte Carlo N-Particle Transport Code," LA-UR-03-1987, Los Alamos National Laboratory, (2003).
4. D. Poston and H. Trellue, "Users Manual, Version 2.0 for MONTEBURNS Version 1.0," Los Alamos National Laboratory, (1999).
5. M.B. Chadwick, P. Oblozinsky, M. Herman *at al.,* "ENDF/B-VII.0: Next Generation Evaluated Nuclear Data Library for Nuclear Science and Technology", *Nuclear Data Sheets*, vol. 107, pp. 2931-3060, (2006).
6. K.J. Kramer, J.F. Latkowski, R.P. Abbott, D. E. Cullen, J.K. Boyd, J.J. Powers, and J.E. Seifried, Neutron Transport and Nuclear Burnup Analysis for the Laser Inertial Confinement Fusion-Fission (LIFE) Engine. *Fusion Sci. Tech*, **56**, 625-631, (2009).
7. USDOE, Final Environmental Impact Statement for a Geologic Repository for the Disposal of Spent Nuclear Fuel and High-Level Radioactive Waste at Yucca Mountain, Nye County, Nevada. Appendix A, Table A-11, (2002).
8. A.G. Croff, "ORIGEN2: A Versatile Computer Code For Calculating the Nuclide Compositions and Characteristics of Nuclear Materials," *Nuclear Technology*, **62**, 335-352, (1983).
9. Blink, J.A., Chipman, V., Farmer, J., Shaw, H.F., and Zhao, P., Thermal performance of deep-burn fusion-fission hybrid waste in a repository. *Mat. Res. Soc. Symp. Proc*. **1125**, 87-94 (2009).
10. USNRC, 10 CFR 63. Energy: Disposal of High-Level Radioactive Wastes in a Geologic Repository at Yucca Mountain, Nevada, (2007).
11. USNRC, 70 FR 53313. Implementation of a Dose Standard After 10,000 Years.
12. Sandia National Laboratories, Total System Performance Assessment Model/Analysis for the License Application. Sandia National Laboratories, Las Vegas, NV MDL-WIS-PA-000005 REV00, (2008).