

Burnup Analyses of An Accelerator-Driven Subcritical System Utilizing Minor Actinides

Yan Cao, Yousry Gohar and Adam R. Kraus

Nuclear Engineering Division, Argonne National Laboratory 9700 S. Cass Avenue, Argonne, IL 60439

July 31 –August 4, 2017





Accelerator-Driven Subcritical System Utilizing Minor Actinides

- The current US spent nuclear fuel (SNF) inventory contains about 115 tons of MAs, which are the main contributor of the long-term radiotoxicity and the decay heat in the SNF.
- Accelerator-Driven Subcritical (ADS) system can utilize the MAs without concern about their small delayed neutron fraction.
- Argonne National Laboratory developed conceptual ADS systems to burn the MAs in the US SNF inventory:
 - A 1-GeV 25 MW proton beam accelerator drives the subcritical fission blanket.
 - Molten lead is used as the spallation neutron target.
 - Fission blanket has keff ~0.98, generates fission power of ~ 3 GW.
 - Fission blanket uses the mobile slurry fuel which has MAs small micro particles suspended in the liquid lead.
- Targeted ADS system performance:
 - Four to five ADS systems operate for 35 years to consume the 115 tons of MAs.



ANL ADS Conceptual Configuration

- Neutron target is placed at the center. Fission blanket is a cylindrical layer wrapped outside the target, with stainless steel supporting the subcritical blanket.
- The spallation neutron target is molten lead, and is self cooled by circulating the liquid lead outside for cooling.
- The fission blanket is formed by the slurry fuel tubes and the liquid lead coolant. The blanket vessel is filled with the coolant and is segmented into six pieces, with each of them connected to an outside coolant loop.
- The HT-9 is the structure material for the target zone and is also for the fuel tubes.
- The fuel tubes are connected at the top and bottom. The slurry fuel is slowly circulated inside the tubes. A small stream of the fuel is circulated outside the blanket for fission products removal, fuel cleanup and feeding fresh fuels.







ANL Conceptual Configuration of ADS System - cont'd

- The ANL conceptual configuration is developed to ensure the satisfactory HT-9 ferritic steel performances:
 - maximum flow velocity less than 3 m/s at the steel surface.
 - maximum steel surface temperature less than or equal 600 °C.
- The reactor physics analyses and the thermal-hydraulic analyses have been coupled to determine the detail configurations of the conceptual design.

Core Configurations:	Conceptual Design
Actinide oxide volume fraction in slurry:	9%
Pu atomic fraction in the fuel:	47.52%
Target outer radius (cm)	34.27
Subcritical Assembly outer radius (cm)	229.0
Subcritical Assembly height (cm)	440.0
Subcritical Assembly fuel volume (m ³)	31.49
Fuel tube lattice pitch (cm)	4.0
Fuel tube radius (cm)	1.15 - 1.45
Number of fuel tubes	11142
Fuel volume fraction (m ³)	44.96%
Effective neutron multiplication factor k _{eff}	0.98
Generated fission power (GW)	2.67
Subcritical Assembly mobile fuel inventory (ton)	27.9
Subcritical Assembly MAs initial inventory (ton)	14.7



Monte Carlo Fuel Depletion Analysis for ADS System

- Monte Carlo fuel burnup simulations were performed to calculate the amount of MAs which can be consumed by the ANL ADS conceptual configuration, assuming that the system can be operated for 35 full power years.
- The ADS system is a coupled system. The external neutron source generated from the spallation process drives the subcritical fission blanket. Thus, the fuel burnup analyses are performed by two stages:
 - First, MCNPX/MCNP6 is utilized to simulate the 1 GeV protons striking the molten Pb target at 600 K. The fission blanket and the reflector is assumed to be vacuum.
 (a) An external volume neutron source file is produced by saving the spallation neutrons at their born sites, or (b) a surface neutron source file is produced by saving the neutron tracks while they leak out of the target zone.
 - Second, both the Monte Carlo fuel burnup code MCB5 and the SERPENT code were used to perform the fuel burnup analysis. The external neutron source files were sampled into these codes for performing Monte Caro transport calculations. The neutron target, the fission blanket and the reflector were explicitly modeled.
- The nuclear data file ENDF/B-VII.0 was used for the two simulation stages.



External Neutron Source

- The spallation process produces a significant amount of high energy neutrons. The neutron energy spectrum was calculated for the volume source file and the surface source neutrons.
- The 1 GeV protons strike the lead target. The MCNPX simulations show that about 15.6 neutrons are generated per proton at their born sites. Among them, about 16% of the neutrons have energy larger than 20 MeV, and about 2% of them have energy larger than 200 MeV.
- The high energy neutrons are transported in the target zone and lost their energy through the (n, xn) reactions. In total there are about 29.5 neutrons per proton leaking out the target zone. Only about 1.6% of the neutrons have energy larger than 20 MeV, and about 0.1% with energy larger than





Modeling High Energy Neutrons in ADS System

- For most nuclides, the ENDF/B-VII data sets have neutron cross section data below 20 MeV (U-235, Pu-239). A few isotopes have neutron cross section data extended to 150 MeV (Pb-208) or to 200 MeV (Pb-207).
- MCNP6/MCNPX has the physics model to simulate the interactions when the neutron energy is above the maximum energy of the cross section library.
- Both the MCB5 and the SERPENT code are lacking of physics models :
 - For MCB5, when the neutron energy is above the maximum energy of the corresponding cross section library, the cross section at the maximum energy is taken in the transport simulations.
 - For SERPENT code, the cross section is set to zero if the neutron energy is above the maximum energy of the cross section library.
- Without the proper treatment of the interactions with high energy neutrons, the neutron fluxes (or power) calculated in the fuel depletion calculation may not be accurate.



Modeling High Energy Neutrons in ADS System – Cont'd

- Monte Carlo simulations are performed to examine the total neutron fluxes in the subcritical fission blanket using the MCNPX, MCB5 and SERPENT code respectively.
 - The ANL ADS conceptual configuration is used in the simulation.
 - The used neutron source is a point isotropic mono-energetic source with the neutron energy varying from 20 MeV to 300 MeV.
 - The neutron fluxes are underestimated at the high energy cases both by the MCB5 and SERPENT code.

Source Energy (MeV)	MCNPX	МСВ		SERPENT	
	Flux(std)	Flux(std)	(MCNPX – MCB)/MCNPX)	Flux(std)	(MCNPX – SERPENT)/MCNP X)
10	2.92E4(0.4%)	2.96E4(0.4%)	-1.8%	2.95E4(0.5%)	-1.02%
100	1.55E5(0.4%)	1.52E5(0.4%)	2.04%	1.49E5(0.7%)	3.67%
200	2.86E5(0.4%)	2.34E5 (0.5%)	18.4%	2.83E5(0.7%)	1.08%
300	4.16E5(0.4%)	2.35E5(0.5%)	43.6%	2.86E5(0.7%)	31.2%



Power Normalization in Fuel Depletion Analysis

- If the external volume source is used for the transport calculation, the total fission power calculated by SERPENT for the ANL ADS conceptual configuration is ~2.2 GW, 17.6% less than the 2.67 GW calculated by MCNPX., the MCB5 corresponding value is 2.07 GW.
- If the the surface source is used, the total power calculated by SERPENT is 2.52 GW, ~ 6% less than the MCNPX value.
- The neutron flux spectrum in the fission blanket are mainly determined by the core configurations. The spectrum calculated by SERPENT using the volume source or surface source which are almost exactly the same as the MCNPX calculated.
- In the fuel burnup analyses, the ADS system is assumed to be operated at a constant fission power of 2.67 GW, which is calculated from the MCNP6/MCNPX simulation using the high energy physics model. The surface neutron source file or the volume neutron source file can then be used for the burnup calculations, since the fission blanket neutron spectrum can be accurately predicted by the two codes.





ADS Reactivity Compensation

- During the fuel burnup, the reactivity of the subcritical fission blanket decreases due to the consumption of the fissile materials.
- In the ANL ADS conceptual configuration, the reactivity loss is compensated by feeding fresh fuel particles back to the slurry fuel and removing the fission gasses.
- The fuel burnup time step is selected to be 90 days.
- The fresh fuel particles are fed into the blanket at the end of each burnup step. Its amount is calculated to increase the keff of the subcritical fission blanket back to the initial value of 0.98:

$$f_{j} = \frac{\left(\frac{1}{k_{1}} - \frac{1}{k_{0}}\right)\sum_{i} \chi_{i} \upsilon \sigma_{f,i} N_{i,1} \phi_{1}}{\frac{1}{k_{0}}\sum_{i} \chi_{i} \upsilon \sigma_{f,i} N_{i,0} \phi_{1} - \sum_{i} \sigma_{r,i} N_{i,0} \phi_{1}}.$$

- Where, k_0 is the k_{eff} at the beginning of the jth time step, k_1 is the k_{eff} at the end of the jth time step without the feed, $\sum_i \chi_i \upsilon \sigma_{f,i} N_{i,0} \phi_1$ and $\sum_i \chi_i \upsilon \sigma_{f,i} N_{i,1} \phi_1$ are total neutron production rates at the beginning and the end of the jth time step, $\sum_i \sigma_{r,i} N_{i,0} \phi_1$ is the capture rates using the neutron fluxes at the beginning of the step.
- In the burnup simulation, the fission gasses are removed instantly at the end of each burnup step.



Fuel Burnup Results - Reactivity Control

- Serpent calculated k_{eff} of the subcritical fission blanket at the beginning and end of each burnup step using the surface neutron source.
 - The keff of the subcritical blanket is successfully maintained at the designed value 0.98.





Fuel Burnup Results - Fuel Transmutation Rates

- Serpent calculated transmutation rates (ton/year) for each fuel element:
 - The transmutation rates are almost identical using either the volume source file or the surface source file.
 - The total amount of actinides transmuted every year is about 0.99 ton.
 - The transmutation rate of MAs fuel gradually decreases after the peak, and takes more than 20 years to reach the equilibrium rates.



Fuel Burnup Results - Fuel Compositions

- Serpent calculated fuel compositions in the subcritical fission blanket at different burnup years:
 - At the beginning of fuel burnup stage, the slurry fuel contains about 17.5% Np-237, 29.7% of Pu-239 and 31.3% Am-241.
 - The MAs concentrations (Am-241, and Np-237) in the fission blanket decrease gradually during the burnup and the Pu-238, Pu-240, and Pu-242 concentrations are increased.
 - At the end of 35 year fuel burnup, the slurry fuel contains ~8.9% Np-237, 17.2% Pu-239 and 15.2% Am-241. It also contains about 16.7% Pu-238, 19.9% Pu-240 and 8.8% Pu-242.

Fuel Burnup Results - Fuel Inventory and Feed

- Serpent calculated the fuel inventory in the ANL ADS conceptual design, and the amount of fresh fuel fed into the blanket:
 - The fuel inventory decreases in the first 10 fuel burnup years, due to the conversion of the fertile isotope Am-241 to the fissile isotope Am-242m/Am-242g.
 - The fuel inventory starts to increase gradually after the first 10 fuel burnup years, due to the buildup of fission products in the slurry fuel.
 - Extra fissile materials are required to produce neutrons to compensate the neutron losses to those fission products.
 - The total amount of fuel fed into the fission blanket is around 40 tons.

Fuel Burnup Results - Actinide Fuel Transmutation

- Serpent calculated the total amount of actinide fuels consumed by the ADS system:
 - For operation lifetime of 35 full-power-years, the ANL ADS conceptual configuration can consume about 34.7 tons of nuclear fuel, among which 25.1 tons are MAs.
 - It also consumes about 10.6 tons Pu, with only producing about 0.9 tons of extra U and 1.0 tons Cm.
 - About 5 ADS units can consume the total 115 tons of MAs.

Fuel	Initial Inventory (ton)	Total Feed (ton)	Inventory at 35 years (ton)	Burnt/Transmuted (ton)
U	0.076	0.109	1.08	-0.895
Pu	13.2	19.0	21.6	10.6
MAs	14.7	21.1	10.7	25.07
Np	4.89	7.04	2.96	8.97
Am	9.65	13.9	6.48	17.06
Cm	0.113	0.162	1.24	-0.965
Total	27.9	40.1	33.3	34.7

MCB5 and SERPENT Fuel Burnup Results Comparison

- The MCB5 is another Monte Carlo fuel depletion code which is used to analyze the fuel burnup in the ANL ADS conceptual configuration:
 - The MCB5 simulation used the same external surface source file. It also used the same strategy to compensate the reactivity losses.
 - Overall, the transmutation rates calculated by the MCB5 code agrees very well with the SERPENT code.
 - The differences shown in the first few years in the Am and Pu transmutation rates are due to the different branch ratios of Am-241 (n, γ) to Am-242/Am-242m used in the MCB5 and SERPENT code.

Conclusions

- Monte Carlo fuel burnup analyses have been performed to analyze the amount of the MAs fuel which can be consumed by the ANL ADS conceptual configuration for disposing of the SNF.
- The fuel burnup analyses were performed in two stages. In the first stage, the MCNP6/MCNPX code was utilized to simulate the 1 GeV proton beam striking the molten lead target. The spallation neutrons were saved in source files. In the second stage, the SERPENT or MCB5 code used the neutron source files and simulated the fuel depletion in the ADS system.
- The MCNP6/MCNPX simulation showed that a significant number of neutrons produced from the spallation process have higher energies than the maximum energy of the current neutron cross section libraries.
- Both the MCB5 and SERPENT code are lacking the high energy physics model to simulate the neutron interactions in the high energy range. This results in underestimating the total power generated in the ADS system when the power is normalized to the proton beam power.
- The neutron energy spectrum in the subcritical fission blanket is mainly determined by the core configurations, and the high energy source neutrons have very little impacts on the neutron spectrum in the fission blanket. Therefore, in the fuel burnup calculations, the fission power is fixed to be the initial value of 2.67 GW.

Conclusions – Cont'd

- To maintain the constant fission power within each burnup step, fresh fuels were fed into the blanket to compensate the reactivity losses. Monte Carlo fuel burnup analysis showed that the k_{eff} of the subcritical blanket can be successfully maintained at the initial value with the fuel feed estimated along the burnup simulations.
- Overall, the fuel burnup simulations showed that the ANL ADS conceptual design can consume a significant amount of MAs. Assuming that the reactor operation life to be 35 years, the total amount of actinides transmuted by the ANL ADS conceptual configuration is about 34.7 ton. It consumes about 25.1 ton of MAs, 10.6 tons of Pu. It also produces about 1 ton of Cm and about 0.9 ton of U.
- The fuel burnup analyses showed that about 5 of such ADS units can utilize the total 115 tons of MAs in the US SNF inventory.