INTRODUCTION

The estimated amount of Spent Nuclear Fuel (SNF) inventory of the United States is about 80,000 ton [1]. The Minor Actinides (MAs) of the SNF and the long-lived fission products (LFP) generate most of the decay heat and the long-term radiotoxicity of the SNF. Using accelerator driven subcritical (ADS) systems to eliminate the MAs and the LFP reduces the SNF radiotoxicity to less than the radiotoxicity of the natural uranium ore in several hundred years [2-3]. The MAs amount in the discharged SNF assemblies varies according to the reactor type and the irradiation history. Based on the DOE studies, the total amount of MAs fuel in the US SNF is ~131 tons. Np237, Am241, and Am243 are the three major isotopes in the MAs. Eliminating and burning the MA isotopes can eliminate the long term radiotoxicity and reduce the decay heat in the spent nuclear fuel so as to avoid the need for a large and long-term geological storage facility. The MAs isotopes have very small delayed neutron fractions, which constrain their utilization in the fission power reactors.

ADS system has been studied for safe burning of the MAs [2-5]. The ADS system is characterized as a coupled system of a subcritical fission assembly and a high-energy proton accelerator. Most of these studies selected the traditional solid fuel form for the subcritical fission blanket. In order to consume the solid MAs fuel, it has to be recycled several times. Due to the continuous change of the fuel compositions in each fuel burnup cycle, this approach will require the development of new solid fuel forms for the different fuel burnup stages, as well as the modification of the current fuel reprocessing techniques to accommodate the varied fuel forms. These developments are expensive and time consuming.

Argonne National Laboratory (ANL) performed studies of the ADS systems for transmuting the MAs using the mobile fuel concept [6-10]. In the past, the molten salt reactor utilized the mobile fuel concept by dissolving the nuclear fuel in the moving salt. However, due to the light elements in the FLIBE salt and the graphite moderator, the neutron spectrum is either thermal or epithermal, which is not desirable for burning the MAs fuel [11]. The mobile fuel concept used in in the ANL ADS concept utilizes liquid metals [12]. Lead-Bismuth Eutectic (LBE) and liquid lead have low melting temperatures, which can be used in the subcritical fission blanket. These liquid provide fast neutron spectrum for burning the MAs. Generally, the actinides have very low solubility in these liquids. Therefore, the ANL mobile fuel is a slurry fuel with micro particles of the actinide oxide mixed and suspended in the liquid metal. The mobile fuel concept eliminates the need for developing new solid fuel forms. The slurry fuel can stay in the subcritical fission blanket for long time to achieve deep burnup since no fuel-clad material is used. In addition, this concept does not require extra fuel reprocessing steps.

This paper summarizes the physics analyses performed for the ANL conceptual configurations. Specifically, physics analysis using the Monte Carlo code MCNP6 [13], MCB5 [14] and SERPENT [15] are performed to determine the detail configurations of ADS system and to evaluate the effectiveness of the concept in utilizing the MAs fuel. In addition, the important physics aspects the ADS system, which are different from tradition critical nuclear reactors, are addressed.

SPALLATION NEUTRON TARGET

In the ANL ADS conceptual configuration, the accelerator and the fission blanket are coupled by the neutrons generated from the spallation process in the target zone. The protons from the accelerator are guided to strike the liquid lead target to create the source neutrons as shown in Figure 1. The target has a beam window to separate the accelerator vacuum and the liquid lead target material. The liquid lead is circulated from the target zone to the outside heat exchanger to remove the generated heat from the spallation process.

![Fig. 1. Spallation neutron target of the ANL ADS conceptual design (Blue: liquid lead, Red: HT-9 structure, arrow →: liquid lead flow direction).](image-url)
The MCNP6 computer program was used to simulate the spallation process of the high-energy protons striking the liquid lead target. The number of the spallation neutrons per proton particle increases as the proton energy increases. However, Monte Carlo simulations showed that the number of the spallation neutrons per MW beam power start to saturate as the proton beam reach 1 GeV. In addition, the proton travelling distance in the target increase as the proton energy increases, which increases the required target length to stop the protons. High-energy protons generates more high-energy neutrons in the spallation process and it requires extra shielding thickness. Based on these considerations, the selected proton beam energy is 1 GeV. The selected beam power is 25 MW to generate ~1 GW, from fission blanket with 0.98 effective neutron multiplication.

Physics analyses using the MCNP6 computer program and the thermal hydraulic analyses using the commercial computational fluid dynamics code STAR-CCM+ were performed to define the detail design of the target assembly shown in Figure 1 and characterized in Table I [16]. The average velocity of lead inside the target zone is limited to 3 m/s. The target structure material is HT-9 to achieve a long operating life with the liquid lead. The nuclear heat deposited in the window is limited to the 24 kW/cm² [17].

Table I. Main parameters of the liquid lead target design.

<table>
<thead>
<tr>
<th>Neutron Target</th>
<th>Pb</th>
</tr>
</thead>
<tbody>
<tr>
<td>Proton Beam Energy, MeV</td>
<td>1000</td>
</tr>
<tr>
<td>Proton Beam Power, MW</td>
<td>25</td>
</tr>
<tr>
<td>Proton Beam Radius with halo, cm</td>
<td>20.07</td>
</tr>
<tr>
<td>Coolant Outlet Flow Channel Radius, cm</td>
<td>27.43</td>
</tr>
<tr>
<td>Coolant Inlet Flow Channel Radius, cm</td>
<td>33.13</td>
</tr>
<tr>
<td>Steel Outer Wall Radius, cm</td>
<td>34.27</td>
</tr>
<tr>
<td>Target Length, cm</td>
<td>75.0</td>
</tr>
<tr>
<td>Target Radius, cm</td>
<td>33.13</td>
</tr>
</tbody>
</table>

SUBCRITICAL FISSION BLANKET

The subcritical fission blanket is a cylindrical tank loaded with bundle tubes and the cylindrical tank is divided to six sectors. Figure 2 shows horizontal and vertical cross section of one of the sectors. The neutron target is placed at the center. The tubes have a regular triangular lattice with 4 cm lattice pitch, and they are connected to common manifold at the bottom and the top. The tubes have the mobile slurry fuel slowly circulated inside. A very small stream of the slurry fuel is circulated for chemistry control, removing the short-lived fission products, and adding MAs during the operation. The fission power generated in the fuel tubes is removed by the primary liquid lead coolant in the cylindrical tank. The coolant of each sector is connected to an outside heat exchanger. Figure 2 shows the Monte Carlo model of one sector.

The effective neutron multiplication factor ($k_{eff}$) of the subcritical fission blanket is set to 0.98 to provide enough safety margin from criticality. The $k_{eff}$ is also high enough to provide enough neutron multiplications in the fission blanket. The estimated neutron multiplication per source neutron is ~50. The MCNP6 computer program is used to perform the proton-neutron-photon coupled simulation, which showed that the total thermal power generated in this system is ~2.67 GW. It is close to the total fission power of a typical nuclear power plant.

The ANL conceptual study started with a PWR discharged SNF fuel as the reference fuel compositions. It has ~33 MWd/MT burnup and 25 years cooling time with all the short-lived fission products and 99.995% of uranium removed [18]. In this fuel composition, the MAs is about 15% of the total actinides. Since the majority of the MA isotopes is fertile, i.e., Am-241, Np-237, and Am243, a fraction of the plutonium has been used in the ANL conceptual design to reach the $k_{eff}$ value of 0.98.

Physics analyses with the MCNP6 computer program have been performed to define the amount of plutonium required for the ADS system [6]. In an ADS system, the MAs consumption rate is determined by the amount loaded.
in the system, the neutron spectrum, and the total fission power of the system. The total amount of MAs loaded in the liquid lead is defined based on the requirement of 0.98 neutron multiplication factor, the fission blanket geometrical configuration, the slurry fuel stability. Table II shows the slurry fuel compositions selected in the ANL conceptual design. The actinide fuel is separated into two parts, one part referred as “Pu” part which has all the plutonium isotopes and uranium leftovers, and the other part “MAs” which has all the Np, Am and Cm isotopes. The isotopic composition of each part remains the same as the reference composition. However, the fraction of the two parts in the fuel composition will be adjusted to maintain the effective neutron multiplication value of 0.98 and to have more MAs fuel loaded in the liquid lead.

Table II. Main parameters of the ANL ADS concept.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Actinide oxide volume fraction in slurry</td>
<td>9%</td>
</tr>
<tr>
<td>Pu part atomic fraction</td>
<td>47.52%</td>
</tr>
<tr>
<td>Tank outer radius, cm</td>
<td>229.0</td>
</tr>
<tr>
<td>Tank Height, cm</td>
<td>440.0</td>
</tr>
<tr>
<td>Fuel volume, m³</td>
<td>31.49</td>
</tr>
<tr>
<td>Fuel tube lattice pitch, cm</td>
<td>4.0</td>
</tr>
<tr>
<td>Fuel tube diameters, cm</td>
<td>2.3 - 2.9</td>
</tr>
<tr>
<td>Number of fuel tubes</td>
<td>11142</td>
</tr>
<tr>
<td>Fuel volume fraction, m³</td>
<td>44.96%</td>
</tr>
<tr>
<td>Actinide Fuel inventory, ton</td>
<td>27.4</td>
</tr>
</tbody>
</table>

The ADS system only needs a single SNF processing step to prepare for the initial fuel with no isotopic separation required. After separating the short-lived fission products, the uranium, and part of plutonium from the SNF, the ANL ADS concept will use the remaining plutonium, MAs, and the long-lived fission products for loading the liquid lead carrier. The plutonium is never separated alone, which enhance the proliferation resistance of the fuel cycle.

The physics analysis and the thermal hydraulic analysis were iterated to determine the detail design of the ANL ADS configurations [10]. The engineering constrains limit the average lead coolant velocity to 3 m/s and the maximum temperature of structure surface to 600°C. Fuel tubes with a small diameter are used close to the target zone as shown in Figure 2. The power density close to the target zone is very high due to the presence of the spallation neutron source. The fuel tubes with a small diameter help reduce the amount of heat generated in these tubes to satisfy the 600°C temperature constrain for the structure material.

**MONTE CARLO FUEL BURNUP ANALYSES**

Monte Carlo fuel burnup analyses were performed to calculate the amount of MAs fuel transmuted or burned in the ANL ADS concept. For simulating the fuel burnup in ADS systems, the neutron transport equation with an external source was solved to provide the actual neutron flux distribution and the fission power. The critical eigenvalue calculations at the beginning and the end of each fuel burnup step were calculated for monitoring the system subcriticality.

The current MCNP6/MCNPX computer program does not have an option for the fuel burnup analysis of subcritical system with external neutron source. Both the MCB5 and SERPENT codes were utilized to perform the fuel burnup analyses for the ANL ADS conceptual configuration [10]. The MCNP6/MCNPX code was used to simulate the physics of the 1 GeV protons striking the liquid lead target to generate the neutron source file for the MCB5 and SERPENT fuel burnup calculations.

High-energy neutrons are generated in the spallation process. Based on the MCNP6 simulations, about 15.6 neutrons are generated per 1 GeV proton with the lead target. About 16% of the generated neutrons have energy greater than 20 MeV and about 2% have energy greater than 200 MeV. The current neutron cross section libraries have cross section data for most of the isotopes up to 20 MeV and some isotopes up to 150 MeV [9]. The MCNP6 uses physics model whenever the neutron cross section library is absent. However, both MCB5 and SERPENT computer programs are lacking the physics model to handle the high-energy neutron interactions properly above 20 MeV. Numerical simulations with MCB5 and SERPENT showed that using the spallation source directly without proper considerations of the high-energy neutrons underestimates the total power generated in the core by more than 20%.

The MAs consumption rates in the ANL ADS conceptual design are governed by the neutron spectrum and the total fission power of the system. Because of the neutron multiplications inside the subcritical fission blanket, the source neutrons contribute less than 2% of the neutron population in the core. Monte Carlo simulations showed that there are no significant difference among the neutron spectra calculated by the MCNP6 proton neutron coupled simulation, MCB5, or SERPENT coupled with the external source generated from MCNP6 proton neutron simulation [9].

Typically, an ADS system may be operated with one of the two scenarios. In the first scenario, the system operates with at a fixed power with the accelerator beam power increases to compensate for the reactivity losses due to the fuel burnup. In the second scenario, the system operates at a fixed power with a fresh fuel continuously feed into the system to compensation for the reactivity losses. Increasing the accelerator beam power is not considered in the ANL ADS conceptual design since the beam power is already high. It is also more challenging and uneconomical to build a high power accelerator and it is used at low power during the operation. The ANL ADS system selects the second scenario, in which the reactivity loss within each burnup step is compensated by feeding fresh fuel particles to the blanket.
The fuel burnup simulations were performed by MCB5 and SERPENT computer programs using the generated external neutron source from the MCNP6 high-energy physics simulations. The total fission power is fixed during the fuel burnup, with the values specified to be consistent with the value calculated with the MCNP6 proton neutron photon coupled simulation [10]. The reactivity compensation is done at the end of each burnup step to bring the reactivity back to the initial of 0.98. The burnup step is 90 days. The ENDF/B-VII.0 library is used in all the simulations. In the analyses, the ANL ADS conceptual design is operated continuously for 35 full power years.

The MCB5 results show that the ANL ADS conceptual design operating for 35 full power years consumes about 24.5 tons MAs, and about 11 tons of Plutonium, as listed in Table III. At the same time, it also produces about 0.83 ton of Uranium, and about 0.97 ton of Curium. The SERPENT simulation showed very similar consumption and production values. To consume the 131 tons of MAs of the current US SNF inventory, 5.3 ANL ADS systems are needed.

Table III. Total amount of actinides transmuted in the ANL ADS conceptual design.

<table>
<thead>
<tr>
<th>Element</th>
<th>SERPENT (ton)</th>
<th>MCB5 (ton)</th>
</tr>
</thead>
<tbody>
<tr>
<td>U</td>
<td>-0.90</td>
<td>-0.83</td>
</tr>
<tr>
<td>Pu</td>
<td>10.57</td>
<td>10.98</td>
</tr>
<tr>
<td>Np</td>
<td>8.97</td>
<td>8.87</td>
</tr>
<tr>
<td>Am</td>
<td>17.05</td>
<td>16.59</td>
</tr>
<tr>
<td>Cm</td>
<td>-0.96</td>
<td>-0.97</td>
</tr>
<tr>
<td>Total MAs</td>
<td>25.05</td>
<td>24.49</td>
</tr>
<tr>
<td>Total Actinide</td>
<td>34.72</td>
<td>34.64</td>
</tr>
</tbody>
</table>

SUMMARY

This paper discussed the physics analyses of the ANL ADS system for utilizing the 131 tons of MAs of the current US SNF inventory. Physics analyses were performed to define the main parameters of an ADS system for generating about 1 GWt. An accelerator generating 25 MW beam with 1 GeV protons is used to generate the neutron source for driving the subcritical fission blanket. MCNP6 computer program was used to generate the spallation neutron source from the interaction of the proton beam with the lead target. The ADS subcritical fission blanket utilizes the mobile slurry fuel form for continuous loading of the MAs, which avoids the need for extra fuel processing steps and the development of new solid fuel forms. Physics analyses and thermal-hydraulic analyses were iterated to define the detail configuration of the subcritical fission blanket for operating with $k_{eff}$ 0.98. The Monte Carlo fuel burnup analyses using the MCNP6, MCB5, and SERPENT code have been performed and showed that the 131 tons MAs can be consumed by 5.3 unit of ANL ADS systems operating for 35 full-power years.

ACKNOWLEDGEMENT

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REFERENCES