Transmutation of Radioactive Nuclear Waste – Present Status and Requirement for the Problem-Oriented Nuclear Database: Approach to Scheduling the Experiments (Reactor, Target, Blanket)

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Abstract. Transmutation of nuclear wastes (Minor Actinides and Long-Lived Fission Products) remains an important option to reduce the burden of high-level waste on final waste disposal in deep geological structures. Accelerator-Driven Systems (ADS) are considered as possible candidates to perform transmutation due to their subcritical operation mode that eliminates some of the serious safety penalties unavoidable in critical reactors. Specific requirements to nuclear data necessary for ADS transmutation analysis is the main subject of the ISTC Project #2578 which started in 2004 to identify the areas of research priorities in the future. The present paper gives a summary of ongoing project stressing the importance of nuclear data for blanket performance (reactivity behavior with associated safety characteristics) and uncertainties that affect characteristics of neutron producing target.

INTRODUCTION

There are two main features attributed to accelerator-driven transmuters that impose specific nuclear data requirements. The first relates to essential broadening in the nucleon energy compared to traditional critical reactors. This covers the range up to GeV region characteristic to the energy of initial protons incident on the neutron-producing target. The second one comes from fuel forms that presuppose large fraction of minor actinides (MA) to be transmuted in the blanket.

It is to be stressed that for such advanced fuels there is a lot of uncertainties even in traditional reactor energy interval (0-20 MeV). Figure 1 gives an impressive example of large discrepancy in the benchmark calculations performed on subcritical reactor core with lead-bismuth coolant and homogenized MA fuel [1]. By $k_{\text{eff}}$ they mean the homogeneous $k_{\text{eff}}$ calculations i.e. without the external (spallation) neutron source, so the discrepancy is largely determined by effect of different data for MAs in reactor energy interval incorporated in the databases coupled with neutronics codes.

The importance of high energy component is well explained by the data comprised in Table 1 that shows the difference in some of the integral parameters obtained with the use of database with upper energy 20 MeV and its extended version up to 150 MeV, correspondingly. It is seen that the effect of high-energy nucleons on the power performance (criticality, peaking) is not so prominent, but the effect on radiation damage characteristics is rather essential.

These two examples deal with basic ADS characteristics and unambiguously appeal for the need
in data improvement. In the framework of International Science and Technology Center (ISTC) activities, were have been done several studies on ADS related data. In view of complexity of the experiments it seems to be important to identify the areas of priorities and this forms the subject of a special ISTC Project #2578 started in 2004. The subsequent chapters give an outlook at the nuclear data requirements stem partly from some of the completed ISTC projects and partly from new findings in ADS analysis.

![Reactivity swings in the ADS blanket. Results of benchmark [1].](image)

**FIGURE 1.** Reactivity swings in the ADS blanket. Results of benchmark [1].

### Analysis of the Spectral Dependence of the Sensitivity to Nuclear Data of the Actinide Absorption and Fission Rate in the Blanket with Fast Spectrum

To analyze the sensitivity to nuclear data of actinide absorption and fission in the fast spectrum blanket, a calculational model of the ADS blanket was developed. This model was based on the BREST-OD-300 fast breeder core cooled by liquid lead. The proton-conducting tube of 3 cm in diameter is implanted 10 cm deep into the target. The energy of the proton beam is 660 MeV.

The subcritical blanket consists of 144 fuel assemblies, including 44 A31-type fuel assemblies, 64 A32-type fuel assemblies and 36 A33-type fuel assemblies. Each A31-type fuel assembly contains 156 fuel elements with the fuel pellets 7.9 mm in diameter. Each A32-type fuel assembly contains 160 fuel elements whose fuel pellets have the diameter of 8.3 mm. Each A33-type fuel assembly contains 48 fuel elements with the fuel pellet diameter of 8.3 mm and 112 fuel elements with the fuel pellet diameter of 9.0 mm. The section of the fuel element containing fuel is 110 mm in height.

### Review of Integral Experiments

This section gives brief summary of the ISTC supported integral experiments.

**Project #2884** “Integral experiments on the critical BFS facilities to validate the MA transmutation and analysis of the experiments”. The project envisions MA investigations on benchmark critical assemblies

### TABLE 1. 20 to 150 MeV library comparison on reference uncertainties of the main integral parameters of interest for ADS neutronics [2].

<table>
<thead>
<tr>
<th></th>
<th>$k_{eff}$</th>
<th>Power Peak</th>
<th>Max. DPA [sec$^{-1}$xcm$^{-3}$]</th>
<th>Max. He [sec$^{-1}$xcm$^{-3}$]</th>
<th>Max. H [sec$^{-1}$xcm$^{-3}$]</th>
</tr>
</thead>
<tbody>
<tr>
<td>150 MeV</td>
<td>2.81%</td>
<td>22.3%</td>
<td>37.4%</td>
<td>35.8%</td>
<td>31.2%</td>
</tr>
<tr>
<td>20 MeV</td>
<td>2.81%</td>
<td>22.1%</td>
<td>36.9%</td>
<td>32.1%</td>
<td>30.1%</td>
</tr>
<tr>
<td>Δ(%)</td>
<td>0</td>
<td>1</td>
<td>1</td>
<td>10</td>
<td>4</td>
</tr>
</tbody>
</table>

All the fuel elements of the reactor were loaded with the actinide mixture, which had the same MA composition as the spent nuclear fuel from the VVER-1000 reactor with the fuel burnup rate of 4% and after 10 years of cooling (44.5% Np, 48.6% Am-241, 6.9% Am-243). The actinides were loaded in the form of nitride with the density of 6.8 g/cm$^3$. The density was chosen based on the preliminary calculations to provide deep subcriticality. As a whole, the blanket contained 9279 kg of actinide nitride. For this fuel load $k_{eff}$ for the blanket is equal to 0.947.

The neutron absorption rate (without fissioning) and the fission reaction rate for actinide isotopes in the facility spectrum were calculated for two domains: for the internal domain of the blanket which included 8 A31-type fuel assemblies adjacent to the target and for the rest of the blanket. It was found that the neutron energy range from 0.0002 to 6 MeV is of primary importance. The weights of the group reaction rates in the above range are greater than $10^{-3}$. Besides, no significant difference has been identified between the domain adjacent to the target and the whole target.
which model lead-cooled fast reactors and molten salt reactors. The measured characteristics are:

- fission cross-section ratios for the isotopes of Np, Am, Cu (to the benchmark isotopes of $^{235}$U and $^{239}$Pu);
- central reactivity coefficients for $^{237}$Np, $^{241}$Am;
- the Doppler-effect for the isotopes of Np, Am and the benchmark isotope $^{238}$U;

**Project #1372** “Characteristics of the transmutation effects in samples” This project deals with the analysis of the experiments with the samples irradiated in fast reactor BN-350.

**Project #2680** “MATINE – the study of MA transmutation in the nitride fuel: modeling and pre-reactor tests”. The project was concerned with the validation of the serviceability of the nitride fuel containing MA inclusions and with the study of the fuel characteristics achieved through fabrication.

**Project #2661** “Calculational and experimental validation of the neutronics characteristics of the lead-cooled fast reactors”. The project envisions a complete description of the BFS facility – a model of the mixed uranium-plutonium nitride-fuelled lead-cooled fast reactor. The main task was to develop a benchmark to test the lead neutron data in the fast reactor spectrum (there are considerable discrepancies in the neutron data libraries concerning both the total cross-section and the inelastic scattering cross-section), and to test other materials used in the facility. The following investigations (apart from determining criticality and measuring operational characteristics) were performed:

- spectral indexes ($F_8$/$F_9$, $F_7$/$F_8$, $C_8$/$F_5$ et al.);
- central reactivity coefficients of the main core materials;
- spatial distribution of the fission reaction rate for the main isotopes;
- void effect.

**Project #2582** “Experimental study of MA transmutation in the critical fast facility BFS-73-1”. This facility is a conceptual model of a fast reactor fuelled by metallic uranium (zirconium alloy) and cooled by sodium. The following measurements were made in the above facility:

- ratios of $^{237}$Np, $^{241}$Am, $^{243}$Am fission cross-sections to the benchmark isotope measured by means of the ionization chambers;
- central reactivity coefficient for $^{237}$Np;
- capture cross-section for $^{237}$Np measured by activation of foils.

**Project #304** “Accelerator-based measurements of minor actinides”. Although the main objective of this project (which has already been completed) was measuring MA microscopic data, it is also concerned with MA integral experiments on the facilities modeling fast cores using MOX fuel and sodium coolant.

**NUCLEAR DATA REQUIREMENTS FOR SPALLATION TARGET PERFORMANCE**

High-energy nucleonics is stressed generally in view of radiation damage of structural material. However, designing the spallation target brings about essentially new engineering problems associated with accumulation in situ various spallation products that have not been of concern in traditional nuclear power technology. The main concern here is rather toxic alpha-emitters of Rare Earth elements found to be the most contributors to the toxicity at certain irradiation conditions [3]. In addition to this, some of the Rare Earths are known to be effective neutron absorbers and thus, might affect the overall ADS neutronics. Figure 2 gives an image of this effect giving the blanket reactivity as a function of $^{152}$Gd accumulation in the lead neutron-producing target in the center of the ADS core. As one can see even the presence of several ppm gives rather appreciable affect on the blanket criticality.

![FIGURE 2. Effect of $^{152}$Gd accumulation in the lead target on the blanket reactivity.](image-url)

The summary of identified requirements for nuclear data is summarized in Table 2.
### TABLE 2. Required accuracy in simulation of blanket neutronics behavior. Review of ISTC projects.

<table>
<thead>
<tr>
<th>Characteristic</th>
<th>Blanket</th>
<th>Accuracy Achieved in Computer Simulations</th>
<th>Required</th>
</tr>
</thead>
<tbody>
<tr>
<td>$k_{eff}$</td>
<td>0.98; 1%</td>
<td>0.98; (+0.0), (-0.005)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.95; 2-3%</td>
<td>0.95; (+0.01), (-0.01)</td>
<td></td>
</tr>
<tr>
<td>Efficiency of reactor control system</td>
<td>5-20%</td>
<td>5-20%</td>
<td></td>
</tr>
<tr>
<td>Distribution of heat deposition through fuel assembly</td>
<td>20-30%</td>
<td>10%</td>
<td></td>
</tr>
<tr>
<td>Transmutation rate of minor actinides</td>
<td>10% for fissiles; 30% for fissionable</td>
<td>5-7%</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Characteristic</th>
<th>Target</th>
<th>Accuracy Achieved in Computer Simulations</th>
<th>Required</th>
</tr>
</thead>
<tbody>
<tr>
<td>Neutron yield</td>
<td>10% - liquid metals, 30% - solid targets</td>
<td>5%</td>
<td></td>
</tr>
<tr>
<td>Neutron spectra (inside and on the target surface)</td>
<td>10%, $E_n &lt;$20 MeV; 50%, $E_n &gt;$20 MeV</td>
<td>no special requirements revealed for thermal systems; 10-20% for fast systems</td>
<td></td>
</tr>
<tr>
<td>Radiation damage in structural materials surrounding target</td>
<td>100%</td>
<td>30%</td>
<td></td>
</tr>
<tr>
<td>Heat deposition in the target and beam window</td>
<td>10% total deposition in the target; 30% specific heat deposition (beam window)</td>
<td>5-10% -total; 10%-specific (beam window)</td>
<td></td>
</tr>
<tr>
<td>Yield of residual nuclides</td>
<td>50-300% - spallation products; 100-500% -fission products; 200-1000% - clusters</td>
<td>30%</td>
<td></td>
</tr>
<tr>
<td>Activation of target and structural materials (including accelerator structure and beam duct)</td>
<td>30-100%</td>
<td>10%</td>
<td></td>
</tr>
<tr>
<td>Accumulation of neutron absorbers: $^{22}$Na, $^{58}$Co, $^{87}$Kr, $^{103}$Rh, $^{113}$Cd, $^{135}$Xe, $^{148}$Pm, $^{149}$Sm, $^{151}$Sm, $^{152}$Eu, $^{152}$Gd, $^{155}$Gd, $^{157}$Gd, $^{198}$Au</td>
<td>100-500%</td>
<td>20-50%</td>
<td></td>
</tr>
</tbody>
</table>

### ACKNOWLEDGMENTS

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### REFERENCES