

Estimation of Residual Activity in a Molybdenum Research & Development Experiment using FLUKA, MCNP and PHITS

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Argonne National Laboratory successfully ran a Molybdenum Research Experiment (AMORE) to study production of ^{99}Mo in which accelerator-driven subcritical fission of an aqueous LEU uranyl sulfate solution was demonstrated using an electron accelerator. Depleted uranium was used as the electron-to-photon converter and the neutrons produced irradiated a 20-liter uranyl sulfate solution (145 g/liter of uranium enriched to just under 20%) which was surrounded by a water reflector tank. The geometry was constructed for MCNP and was exported to FLUKA and PHITS using FLAIR Graphical User Interface. The target and solution were contained in a lead shielded cell. The experiment produced 87.5 mCi of $^{99}\text{Mo}/\text{kWh-kg-}^{235}\text{U}$. Once the experiment was concluded, the uranyl sulfate solution was drained off, and the remaining components were kept for Decontamination and Decommissioning (DnD). The components were characterized for the residual activity, radionuclide distribution and residual dose rates. While FLUKA was used to estimate the residual dose rates, MCNP/CINDER and PHITS along with FLUKA were used to estimate the residual activity and residual nuclei distribution. To address some of the differences in the yield of radionuclides, PHITS calculations were repeated with ENDF/B-VIII coupled with CP2020, JEFF 4.0 in addition to the default JENDL5 PHITS uses. Residual activity from MCNP results were obtained from MCNP/CINDER calculations. MCNP simulations used cross section table for neutrons below 20 MeV (coinciding with the highest energy in most of the CINDER data libraries) and model physics above. In addition, model physics was used to determine residual nuclides from photonuclear reactions and from neutron-induced reactions with $E_n > 20$ MeV.

Experiments were run at 8-17 kW beam power at 40 MeV electron energy over 3 years, between 2018-2021. The exact irradiation history was used in calculations to estimate the residual nuclide yield after 5 years of cooling.

Depleted uranium had the highest activity at about 4.7×10^8 Bq, dominated by ^{147}Pm at 21%, ^{137}Cs at 16%, $^{137\text{m}}\text{Ba}$ at 15%, ^{90}Y and ^{90}Sr at about 11% each, and ^{125}Sb at 8%. These were followed by the $^{106}\text{Ru}/^{106}\text{Rh}$ and $^{144}\text{Ce}/^{144}\text{Pr}$ parent-daughter pairs, each at about 3% per nuclide. FLUKA and MCNP agreed in terms of the highest yield radionuclides while PHITS predicted ^{137}Cs as the highest yield radionuclide followed by ^{147}Pm , for all cross-section data used. A notable difference in the depleted U converter was the yield of ^{239}Pu which was ~ 7 times lower in FLUKA compared to MCNP/CINDER while PHITS reported 30% of MCNP results. The lower yield of ^{239}Pu in FLUKA was attributed to the lower GDR photoneutron yield in the 20-40 MeV energy range, because of the Bezier fits used. Within PHITS, the three data sets gave almost the same yield, not only for ^{239}Pu but also for the other radionuclides except for ^{14}C which showed 12% lower yield with ENDF data. Results from the simulations will be discussed for depleted U as well as the other components.