TRANSMUTATION STUDIES FOR TUNGSTEN AND MERCURY TARGETS WITH THE CINDER'90 AND ORIHET'95 CODE

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SUMMARY
The total activity, decay heat, and DOE-STD-1027 hazard classification category 3 threshold quantity ratio were calculated for simple tungsten and iron-clad mercury spallation targets using MCNPX and the two radionuclide inventory codes CINDER'90 and the Activation Analysis System, based on ORIHET. In addition, the effect of an updated nuclear structure library, PHTLIB, was studied for the tungsten target.

Total activity predictions agreed well for the two code systems. Decay heat predictions showed considerable differences between the CINDER'90 and ORIHET codes. These differences were mostly accounted for by using an updated decay library with the ORIHET calculations. The hazard classification calculations tended to follow the activity calculations and agreed well at most time steps. Larger differences were observed for specific nuclides for the mercury target material and the iron cladding. Remaining differences deserve further studies and will probably lead to improvements in both code systems.

I. INTRODUCTION

The radionuclide inventory of an accelerator driven system is a critical component of the system safety documentation. For typical tungsten and mercury target systems many important quantities, such as decay heat and gamma-ray emission rates, are integral properties obtained by simply summing the contributions of all radionuclides. However, some quantities, such as the facility hazard classification as defined by DOE-STD-1027,[1,2] may be dominated by a few specific radionuclides that may or may not be key contributors to the total activity. As a result, methods are constantly being sought to improve upon the accuracy of radionuclide inventory predictions and several computer codes have been developed to calculate the inventory.

To study the effect of using different programs to predict radionuclide inventories, calculations were performed using MCNPX[3] in conjunction with two transmutation codes: CINDER'90[4] and the Activation Analysis System- AAS.[5] AAS is a collection of codes and procedures built around the ORIHET[6] code.

Two simple spallation target models were considered: 1) a bare tungsten cylinder and 2) a mercury cylinder clad in iron and surrounded by a beryllium reflector. Tungsten was selected because it is a typical solid target material and a bare cylinder was used to emphasize spallation reactions over activation products. In contrast, the reflected, iron-clad mercury target is typical of a liquid target selection and the beryllium reflector enhances the low-energy neutron flux, therefore adding a neutron activation component to the inventory.

The predictions of the buildup and decay of the total activity, decay heat, and hazard classification, obtained using the two transmutation codes are compared and discussed for each of the targets. In addition, the effect of an updated nuclear structure library for MCNPX, PHTLIB/SPEC1,[8] was studied for the tungsten target.

II. CALCULATIONAL METHODOLOGY

MCNPX calculations for these comparisons were completed using a parallel version of MCNPX[7] on a linux beowulf cluster. Using the family naming feature to work around the 2 GB file size limit in

linux, a series of history files totaling more than 7 GB were written to insure the relative statistical errors on many spallation product production rates were below 0.10. Details of the calculations for each target are described below.

A. Bare Tungsten Target

MCNPX calculations for a bare tungsten target, one centimeter in diameter by one centimeter long, were completed. A 1-GeV, 1-mA pencil beam of protons was incident on the center of the cylinder. The neutron flux (E<20 MeV) within the cell was tallied and a history file was written and post-processed for the nuclide production and destruction rates. Identical spallation nuclide production data was passed to CINDER'90 and ORIHET. The low-energy neutron flux was input to CINDER'90, while the AAS processed cross-sections and folded them with the flux to produce low-energy reaction rates for ORIHET.

The MCNPX calculations were repeated using a newer version of the nuclear structure library, PHTLIB.[8] The new PHTLIB (SPEC1) was developed to eliminate inconsistencies between the radionuclide production in MCNPX and the CINDER'90 inventory code. Although inconsistencies still exist, the number has been greatly reduced.

B. Reflected Mercury Target

The mercury target consisted of a two-centimeter diameter by two-centimeter long cylinder of mercury in an iron container with two-millimeter thick walls. A beryllium reflector, twenty-one centimeters long by twenty centimeters in diameter surrounded the target. The proton beam flightpath removed from the reflector was matched to the target, cylindrical in cross section with a diameter of 2.4 cm, and extended through the reflector.

As with the tungsten target, a 1-GeV, 1-mA pencil beam of protons was incident on the center of the target. The same nuclide production data was passed to CINDER'90 and ORIHET, in this case for both the mercury and the iron cladding. Only the new PHTLIB library was used for the mercury target calculations.

III. RESULTS

A. Bare Tungsten Target

MCNPX calculations for the bare tungsten target were completed with both the old and new versions of PHTLIB. With the new PHTLIB, 198 additional metastable states and 11 additional ground states were identified for input to the inventory calculations. Of the 11 additional ground states, 5 of the nuclides were attributed to poor statistics with only a single event out of 5E+7 incident protons. As expected, the calculation using the old version of PHTLIB also showed 2 nuclides that did not appear with the new version of PHTLIB, both with poor statistics. In addition, two nuclides were generated in the MCNPX analysis that were outside the footprint on the chart of the nuclides covered by the radionuclide inventory codes. Each event was a single event out of a large run, but they stand as good reminders of both the lack of complete coverage of the chart of nuclides by the inventory codes and the fact that the physics models in MCNPX can predict a nuclide that does not physically exist.

The activity, decay heat, and hazard classification category 3 threshold quantity (Cat-3 TQ) ratio for the bare tungsten target are plotted as a function of time in Figs. 1-3 for a 208 day (~5,000 hours, the equivalent of one operation year at the SNS) irradiation followed by a 90 day decay. In each plot, points exist for CINDER'90 using the old and new PHTLIB files, the original ORIHET calculations, and a modified ORIHET calculation using the old and new PHTLIB files.

The modified ORIHET calculations use the same ORIHET code, but a modified nuclide decay library. The modified decay library contains the same nuclides as the original ORIHET library, which contains fewer nuclides than the CINDER`90 library. However, the reaction Q values and nuclide half-lives were replaced with those taken from the ORIGEN2S decay library, which was in turn converted from an older version of the CINDER'90 data library. The ORIGEN2S code was obtained from Brookhaven National Laboratory.[9]

Figure 1 shows the total activity for each of the calculation methods. At each time step, all of the calculations agree reasonably well. Using the CINDER'90 calculation with the new PHTLIB as the basis, the largest variation in the calculations occurs at short times, where the original ORIHET calculation is ~15% low. All of the other

calculations are within ~5% of each other. At long times, all of the calculations agree to within 5%. From the two CINDER'90 calculations, the new PHTLIB appears to have little effect on the activity with the two calculations differing by less than 0.1% at all times. The two modified ORIHET calculations give similar results, with an average difference of 1.5%, although some individual time steps differ by as much as 3.7%. So, the new PHTLIB appears to have little effect on the bulk activity of the target.

Figure 2 displays the decay heat calculations as a function of time for each of the calculation methods. The original ORIHET calculation is lower than the other calculations, initially by a factor of two and diminishing to ~20% at the end of the irradiation. After one day of decay, all of the calculations agree within ~10%. Experiments have demonstrated the ability of CINDER'90 to calculate the decay heat of a simple tungsten target to within 15%,[10,11] so the focus was turned to the ORIHET calculation. Comparing the individual nuclide contributions to decay heat from the various calculations, it was discovered that several of the metastable tungsten nuclides were not accounted for in the original ORIHET calculation. This fact is also shown, but not nearly as dramatic, in the activity comparison (Fig. 1). So, the difference in decay heat calculations and the time variation of the difference are understood.

Figure 3 shows the hazard classification Cat-3 TQ ratio as a function of time for each calculation method. For each time step, all of the calculations agree within 9%. Although some variations in the level of agreement exist as a function of time, the fluctuations appear to be fairly small. The CINDER'90 calculations do appear to consistently be higher than the ORIHET calculations, but at the 10% level, the calculations are the same.

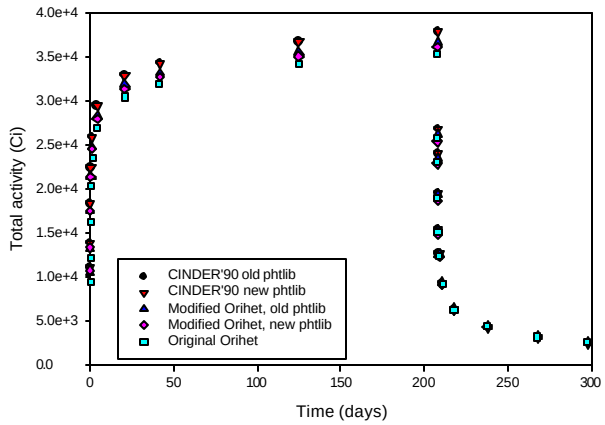

Figure 1. Total activity for the bare tungsten target.

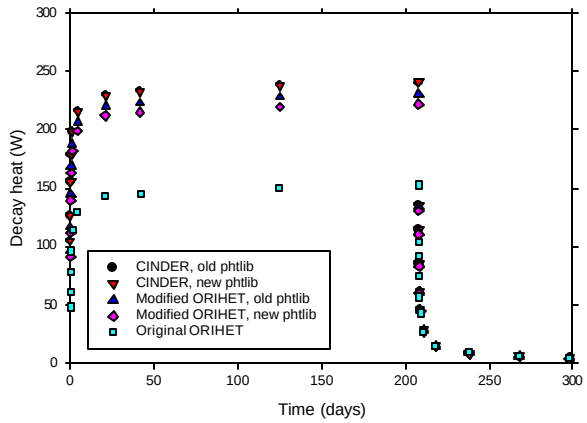

Figure 2. Decay heat for the bare tungsten target.

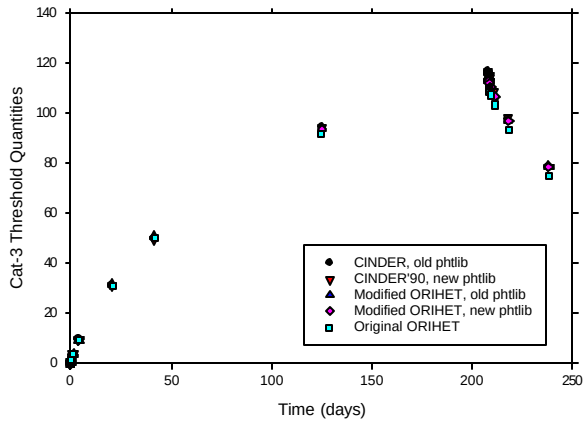

Figure 3. Hazard classification for the bare tungsten target.

B. Reflected Mercury Target

All calculations for the reflected mercury target were completed using the new PHTLIB. Figures 4-6

show the total activity, decay heat, and hazard classification for the mercury in the mercury target. Figures 7-9 show the same set of plots for the iron cladding on the mercury target.

As with the tungsten target, calculations for the total activity for the mercury (Fig. 4) agree well, within 8% at all time steps. However, at every time step in the analysis the CINDER'90 calculations are greater than the ORIHET calculations. The average difference is approximately 4% for the modified ORIHET calculations and 6% for the original ORIHET calculations. While the differences are small, the behavior is different from the tungsten target analysis.

Figure 5 displays the decay heat calculations as a function of time for the mercury. Again the original ORIHET calculations are lower than the other calculations. However, for the mercury, the CINDER'90 and modified ORIHET calculations are also different at approximately the 10-15% level during the irradiation. A comparison of the top twenty contributors to the decay heat for each calculation method indicates that the majority of the top contributors to the decay heat agree well. One notable exception is Au-187, where CINDER'90 predicts more than twice as much decay heat than the modified ORIHET calculation. These results deserve some additional study of the individual codes.

Figure 6 shows the hazard classification Cat-3 TQ ratios for the three calculation methods. As with the tungsten, the CINDER'90 calculations are higher than the two ORIHET calculations. Although the modified ORIHET calculations agree well with the CINDER'90 calculations with an average of 4% difference, at short times the original ORIHET calculations disagree. The differences approach 30% at short times where metastable state contributions are more important. At longer times, the differences become much smaller.

For the iron cladding around the mercury target, the total activity is plotted in Fig. 7. While the calculations agree well at long times, at very short times both ORIHET calculations are up to 50% greater than the CINDER'90 calculations. By one day into the irradiation, the difference is less than 20%. At all other times, including the majority of times of interest to operating facilities, the calculations agree to within 10%.

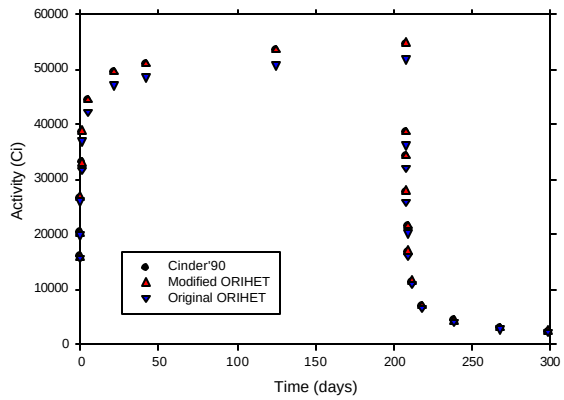

Figure 4. Total activity for the mercury in the reflected mercury target.

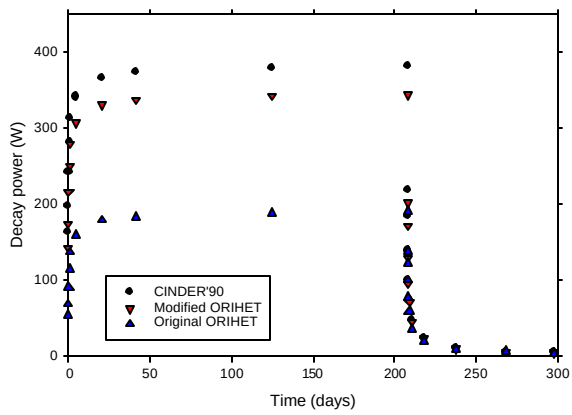

Figure 5. Decay heat for the mercury in the reflected mercury target.

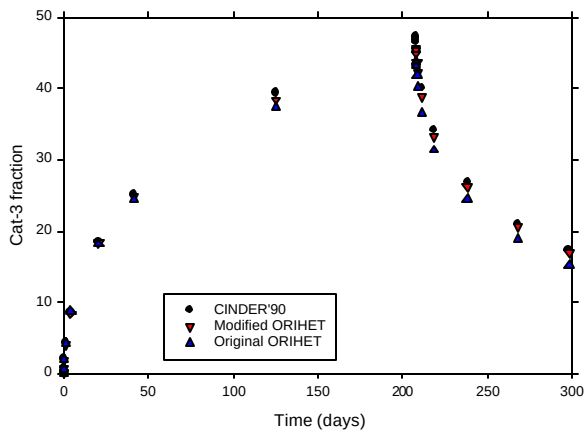

Figure 6. Hazard classification for the mercury in the reflected mercury target.

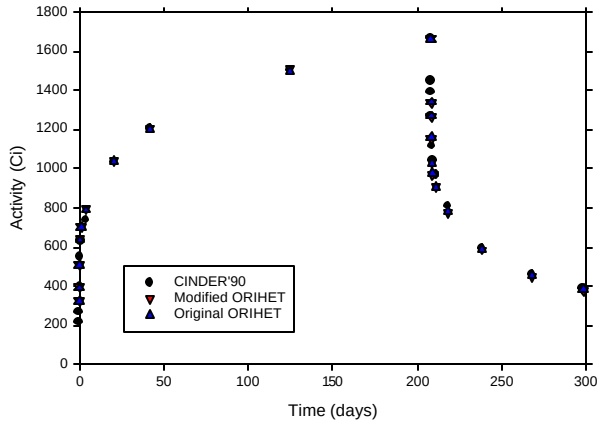

Figure 7. Total activity for the iron in the reflected mercury target.

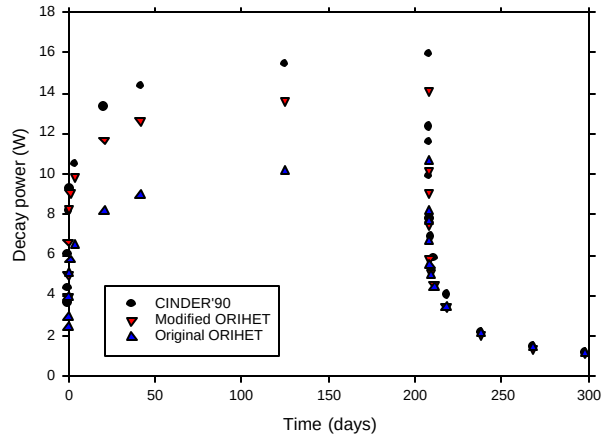

Figure 8. Decay heat for the iron in the reflected mercury target.

Figure 8 shows the decay heat calculations as a function of time for the iron cladding. As with the other materials, the original ORIHET calculations underpredict the decay heat. Similar to the mercury calculations, the CINDER'90 calculations are higher than the modified ORIHET calculations by slightly more than 10%. Comparisons of the top twenty nuclide contributions to decay heat show that the two calculations have 16 of the top 20 in common, although in a slightly different order. Two of the four nuclides from the CINDER'90 calculation are not included in the ORIHET library and are therefore not in the ORIHET calculation. Similar to Au-187 in the mercury calculation, the CINDER'90 calculation predicts almost four times as much Na-24 as the ORIHET calculation. This difference should be studied in more detail.

The hazard classification analysis (Fig. 9) follows trends similar to the mercury calculations. CINDER'90 predicts higher values than the ORIHET calculations, although the majority of the time step comparisons are close (within 8%). The notable exception would be near the one-day mark, when the difference between the CINDER'90 and ORIHET calculations are on the order of 15%. One of the driving nuclides behind this difference is Na-24, as identified in the decay heat comparison. Reconciling the Na-24 activity would resolve the differences in the decay heat and hazard classification.

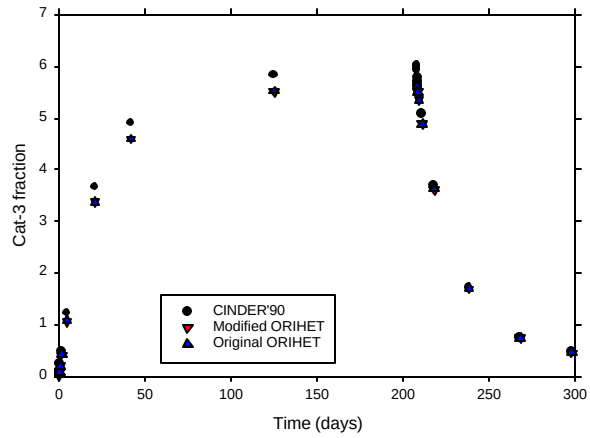

Figure 9. Hazard classification for the iron in the reflected mercury target.

IV. CONCLUSIONS

Two different radionuclide inventory codes were used in conjunction with MCNPX to calculate activity, decay heat, and hazard classification for two typical spallation source targets. For the majority of the time steps, the total activity agreed well for all calculation methods. The decay heat calculations showed the largest differences, near a factor of two for some cases. Changes made to the ORIHET decay library and Q values resulted in better agreement. However, there are still individual radionuclides that can be identified where the differences are a factor of two or more. The study of these nuclides would make a good starting point for future work.

The hazard classification comparisons agreed better than the decay heat comparisons. Because of the relatively large weighting factors for some nuclides and the differences discovered in the

decay heat calculations, the agreement is somewhat surprising. For target materials other than those studied for this work, the agreement of the calculated hazard classification Cat-3 TQ ratio may not be as good.

Further investigations of the observed differences will likely lead to improvements in both CINDER'90 and AAS codes.

ACKNOWLEDGMENTS
SNS is managed by UT-Battelle, LLC, under contract DE-AC05-00OR22725 for the U.S. Department of Energy.

The authors would like to thank Michael Todosow and his colleagues at Brookhaven National Laboratory for providing the ORIGEN2S code and libraries.

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