Calculations vs. measurements of remnant dose rates for SNS spent structures

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Abstract. Residual dose rate measurements were conducted on target vessel #13 and proton beam window #5 after extraction from their service locations. These measurements were used to verify calculation methods of radionuclide inventory assessment that are typically performed for nuclear waste characterization and transportation of these structures. Neutronics analyses for predicting residual dose rates were carried out using the transport code MCNPX and the transmutation code CINDER90. For transport analyses complex and rigorous geometry model of the structures and their surrounding are applied. The neutronics analyses were carried out using Bertini and CEM high energy physics models for simulating particles interaction. Obtained preliminary calculational results were analysed and compared to the measured dose rates and overall are showing good agreement with in 40% in average.

1. Introduction
The Spallation Neutron Source (SNS) in Oak Ridge, Tennessee, is an accelerator driven neutron scattering facility for materials research. Presently SNS is capable and approved to operate at 1.4 MW proton beam power incident on a mercury target with a proton beam energy of 1 GeV and 60 Hz repetition rate.

SNS target system components are periodically replaced because they reach their end-of-life due to radiation induced material damage. The target vessel, which houses the mercury target, is exchanged about two-three times per year and the proton beam window (PBW) is exchanged ones per two – three years. Both target vessel and PBW are exposed to severe radiation environment, and are replaced to avoid events of spills due to structural failure caused by excessive material degradation and fatigue.

All spent components must be safely removed, placed in a container for temporary on-site storage, and ultimately transported off-site to a nuclear waste disposal site.

In order to characterize and classify spent components, accurate estimates of radionuclide inventory are performed. A bounding case considering the maximum possible radionuclide inventory, based on a scenario with maximum possible irradiation exposed during the life-time, is established for the respective component. Using these data, the spent component is characterized and classified, and an appropriate container for temporary storage on-site and subsequent transport off-site is suggested.

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Once the container has been selected, transport calculations for the Department of Transportation (DOT) package are performed to ensure that the transport package is compliant with transportation and waste management regulations. Supporting documentation with radionuclide inventory and dose rate prediction for the time of the transportation is prepared for each shipment. Neutronics analyses are performed, assuming realistic irradiation history and decay case to ensure that the container/package, housing the structure, is compliant with the waste management regulations. Analyses are complicated due to geometry, multi-code usage and following data treatment. To validate analyses, dose rates from the spent target vessel #13 and PBW module #5 were measured. Neutronics analyses were performed to calculate residual dose rates from both structures for the time of measurements. This work presents comparison of preliminary analyses vs. measurements of the residual dose rates.

2. Methods for neutronics analyses

Complete analyses for the spent component radionuclide inventory and dose rates are performed for each off-site shipment, containing target vessel or PBW module, with realistic irradiation history and for the time of the shipment. Because these analyses are occurring on regular basis, to facilitate the process, the automated script system, which runs the analyses and generates the report, for both target vessel and PBW was developed.

Full three-dimensional radiation transport calculations with the state-of-the-art Monte-Carlo code MCNPX Version 2.7.0 [1] and the latest as-built target station model including the PBW module (Figure 1) are performed to simulate the radiation environment. Specifically, isotope production rates due to spallation reactions and the below-20-MeV neutron fluxes in the 63 group CINDER90 group structure were calculated for target facility components of interest. The calculations were performed twice with the two different high energy physics models Bertini and CEM.

![Figure 1. MCNPX model of target station, including PBW module, vertical cross section through the beam center line.](image)

The obtained isotope reaction rates and neutron fluxes were extracted from the transport calculation output and were fed into the CINDER90 transmutation code [2] [5] using the standardized ACTIVATION_SCRIPT [3] to calculate the radionuclide inventory of the components. In order to obtain local distributions of radionuclide inventory and subsequent decay source terms, the components were subdivided into small pieces. The target vessel model was represented by 140 cells and the PBW model by 116 cells. The target vessel radionuclide inventory includes the target vessel,
which is made from stainless steel 316L, and, as we assume, 200 g of activated mercury dispersed in the target together with 10% of the mercury radionuclide inventory (other than mercury, gold and noble gas isotopes) deposited on the mercury exposed steel piping and target structure of the mercury loop. The PBW module is comprised from an Inconel-718 window and is encased in a 316-stainless steel holder, and a water-cooled collimator assembly also of 316-stainless steel.

Realistic irradiation history and decay time, reconstructed from archived measured accumulated energy of SNS operations, are used in the analyses. Decay gamma sources for a defined history of build-up and decay were extracted from each cell and compiled to a source term in MCNPX language by running the GAMMA_SOURCE_SCRIPT [4]. The decay gamma source was prepared for the time, when measurements of the dose rates occurred. Then, the decay gamma source terms were utilized in photon transport calculations for both target vessel and PBW module. To simulate measurements, box-type volumes of 2x2x2cm³ at the detector positions were defined around the components at the locations of the measurements. For the residual dose rate scoring in the detector volume, a track-length flux tally, was applied. Dose rates were obtained by folding fluxes with SNS specific flux to dose conversion coefficients [6].

3. Measurements
The specialized high-range radiation detection instrumentation Ludlum Model 9-7 with 9-7-BH Detector (Figure 4) was used for the measurements of residual dose rates. According to specification, the detector range is 0.01 – 19.99 kR/hr, with the resolution of 0.01 kR/h and the linearity of 10%. The active detector volume (ion chamber) is 7cm³, with 2.54cm diameter and 1.64 cm thickness. Detector calibration was performed at ORNL with a Cs-137 source. Certificate of calibration was issued.

4. Results
The analyses for residual dose rates were performed for the time of measurements, which is 106 days for the target #13 and 80 days for the PBW module #5 after beam termination on target. The standard deviation for most of the calculated dose rate values was less than 1%, beside a few locations, where the standard deviation was slightly higher but lower that 4%.

4.1. Target
Service lifetime [8] for target vessel #13 is approximately 0.50 years in which it accumulated slightly more than 2588.55 MWh proton beam energy.

For the dose rate analyses the fluxes under 20 MeV and the high-energy radionuclide production rates were obtained using:

- MCNPX version 2.5 with Bertini model for high-energy particles interactions;
- MCNPX version 2.7 with Bertini model for high-energy particles interactions;
- MCNPX version 2.7 with CEM model for high-energy particles interactions.

Mercury, which is left over after the drainage from the target vessel, was modeled in two way – dispersed inside the target vessel, and as a paddle at of the target nose. Results are presented in Table 1. C/M refers to calculation over measurements ratio.

Overall the calculated dose rates are in a good agreement with the measured dose rates – within about 40%. Calculated dose rates at most locations have higher values compared to measured data. Using CEM model vs. Bertini model for high-energy particles physics does not show significant impact on calculations. Using reaction rates and fluxes generated by MCNPX version 2.7.0 compared by MCNPX version 2.5.0 improves the C/M comparison. Modeling 200g of mercury as a puddle vs. distributed mercury at lower density does not impact the dose rates significantly.

Table 1. Calculated dose rates vs measured dose rates for target vessel #13, mrem/h.

| Detector# | Measurements | MCNPX 2.5.0  | MCNPX 2.7.0 | MCNPX 2.7.0, CEM | Mercury paddle |
|-----------|--------------|--------------|-------------|------------------|----------------|
|           |              | 2.04E+6      | 3.35E+6     | 1.64             | 1.44            | 2.94E+6        | 1.47     |
| 1         |              | 2.42E+6      | 3.80E+6     | 1.57             | 1.36            | 3.36E+6        | 1.39     |
| 2         |              | 1.96E+6      | 3.34E+6     | 1.70             | 1.51            | 3.02E+6        | 1.54     |
| 3         |              | 1.40E+6      | 1.92E+6     | 1.37             | 1.22            | 1.65E+6        | 1.18     |
| 4         |              | 1.60E+6      | 2.31E+6     | 1.44             | 1.28            | 1.99E+6        | 1.24     |
| 5         |              | 1.45E+6      | 2.25E+6     | 1.55             | 1.38            | 1.93E+6        | 1.33     |
| 6         |              | 1.09E+6      | 1.85E+6     | 1.70             | 1.50            | 1.58E+6        | 1.45     |
| 7         |              | 6.60E+5      | 1.08E+6     | 1.64             | 1.50            | 9.69E+5        | 1.47     |
| 8         |              | 3.70E+5      | 5.50E+5     | 1.49             | 1.38            | 4.92E+5        | 1.33     |
| 9         |              | 2.20E+5      | 2.45E+5     | 1.12             | 1.22            | 2.41E+5        | 1.21     |
| 10        |              | 1.60E+5      | 1.45E+5     | 0.91             | 0.98            | 1.34E+5        | 1.00     |
| 11        |              |              |             |                  |                |                |          |

4.2. PBW

For the residual dose rate analyses, the fluxes under 20 MeV and the reaction rates were obtained using MCNPX version 2.7 with the Bertini model for high-energy particles interactions. Results are presented at Table 2.

Table 2. Calculated dose rates vs measured dose rates for PBW #5, kRem/h.

| Detector # | Measurements | Calculations | C/M |
|------------|--------------|--------------|-----|
| 1          | 0.16         | 0.22         | 1.38|
| 2          | 0.22         | 0.24         | 1.09|
| 3          | 0.17         | 0.2          | 1.18|
| 4          | 0.38         | 0.4          | 1.05|
| 5          | 1.15         | 1.03         | 0.90|
| 6          | 0.52         | 0.38         | 0.73|
| 7          | 0.22         | 0.23         | 1.05|
| 8          | 0.26         | 0.31         | 1.19|
| 9          | 0.21         | 0.24         | 1.14|

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Most of the calculated results are within 20% of the measured data, and are generally in the higher side. There are two locations where the dose rates differ by 40%. This difference could be caused by uncertainty in the detector position during the measurements.

5. Conclusions
In order to validate neutronics analyses, measurements of dose rates from a spent target vessel and a PBW module were performed. Preliminary neutronics analyses were conducted to calculate residual dose rates from the target vessel and PBW for the time of the measurement. Overall calculated dose rates are in agreement with measured dose rate to within 40%, beside for a few locations that lie within 60%. We are pleased with the outcome of this validation effort considering the complexity of the geometry and involvement of different calculational codes.

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References
[1] D. Pellowitz, ed., “MCNPX User’s Manual, Version 2.7.0,” LA-CP-11-00438, Los Alamos National Laboratory, Los Alamos, New Mexico (April 2011).
[2] D. Pellowitz, ed., “MCNPX User’s Manual, Version 2.5.0,” LA-CP-05-0369, Los Alamos National Laboratory, Los Alamos, New Mexico (April 2005).
[3] W. B. Wilson, S.T. Cowell, T. R. England, A.C.Hayes, P. Möller, A Manual for Cinder’90 Version07.4, LA-UR-07-8412, Los Alamos National Laboratory, Los Alamos, (2007).
[4] F. X. Gallmeier and M. Wohlmuther, Activation Script Version 1.0 User Guide, ORNL-TM-2008/031, Oak Ridge National Laboratory, August 2008
[5] M. Wohlmuther and F.X. Gallmeier, User Guide for the Gamma Source Perl Script 1.0, PSI-TM-85-08-02, Paul Scherrer Institute, July 2008.
[6] P. D. Ferguson, CINDER’90 for SNS Activation Studies, SNS-106100200-TR0142-R00, Oak Ridge National Laboratory, March 2006.
[7] I. Popova, Flux to Dose Conversion Factors, SNS-NFDD-NSD-TR-0001-R00, Oak Ridge National Laboratory (October 2009)
[8] 8. I.I. Popova, “Activation and Waste Handling of Target Vessel #13, SNS106100200-DA0074-R01, Oak Ridge National Laboratory (October 2009), August 2016.