Title: Estimate of Residual Tritium Inventory of WR-1			Date: 2019/03/13
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TABLE

1. INTRODUCTION

This document provides an evaluation of the remaining tritium inventory in the Whiteshell Reactor 1 (WR-1) moderator system. It explains the tritium production process, characterization work on the system to date, and the uncertainty associated with the inventory. The estimation methods used are described along with their assumptions and the results given. Lastly, the inventory to be carried forward in all future modelling is identified with justification.

2. BACKGROUND

WR-1 was a heavy water moderated reactor. This made it particularly susceptible to tritium production as the deuterium of the heavy water would become activated creating tritiated heavy water. The tritiated heavy water produced was contained to the moderator system which, after shutdown of the reactor, was drained and dried. However, tritium, just like stable hydrogen, is slowly absorbed into metals and hence the piping and tanks of the moderator system built up an unknown amount of tritium. Since the moderator system was drained and dried, tritium has been desorbing from the metals to the air inside of the system. The system has been connected to active ventilation and degassing tritium at a relatively stable rate.

As part of an ongoing Environmental Assesement (EA) additional characterization work was undertaken to improve confidence in the source term values for radionuclide inventory, used in performance assessment modelling.

In 2015, holes were cut into the moderator system at locations of expected higher contamination using powered saws [1]. This verified that the system had truly been drained and dried, and allowed swipe and air monitoring tests to be performed. No analysis of the embedded tritium levels was conducted, but the swipes and air monitoring testing provided some information about contaminants within the system.

Holes were cut into the system using powered saws again in 2017 as part of a wider WR-1 reactor systems characterization plan [2]. The plan was developed by Oak Ridge Associated Universities and used random selection points to verify or determine the radionuclide inventory of the remaining reactor systems. The work was executed by Whiteshell Laboratories (WL) staff and the samples cut from the moderator system were analyzed for absorbed tritium.

Compared to the preliminary tritium estimate, the analysis showed significantly less tritium remaining within the system [3] (see Table 1). The belief at the time was that the mechanical means of sample extraction generated sufficient heat and vibrations to release the absorbed tritium as during both campaigns a significant increase in tritium emissions to the ventilation stack was observed.

3. ESTIMATION ASSUMPTIONS AND METHODS

3.1 General Assumptions

Four methods were used to provide an estimation of the possible remaining tritium in the system:

- Method 1 - Amount of H-3 Released During Sampling
- Method 2 - Rate of H-3 Release During Sampling
- Method 3 - Assumed Volume of Absorbed Tritiated Heavy Water
- Method 4 - Solubility of H^+ in Stainless Steel

Estimations for Methods 1 -3 below rely on the assumption that the samples taken were liberated of their tritium content during the sample collection process. These estimates further assume that all of the tritium release to the ventilation stack was from the sampled areas only. If the first assumption is correct, this second assumption is conservative as it neglects any tritium emission from the metal immediately surrounding the sample locations which would have also been heated and agitated.

It is also possible that the samples were not liberated of their tritium content [4] and that not all of the tritium on or in the samples was accessible for analysis. An additional estimate (Method 4) that calculates the maximum amount of tritium that could be present in the metal components of the moderator system was made to account for this possibility.

3.2 Method 1 – Assumed Volume of Absorbed Tritiated Heavy Water

This method assumes that 1% of the total volume of tritiated heavy water from the system was absorbed into the metals. 1% was assumed to be conservative as it represents a significant amount (≈172 kg) of heavy water (D_2O) [5] and, because heavy water accounting was very strict, it is expected that only insignificant fractions could be lost due to absorption each year and not be tracked. The concentration of tritium was assumed to be equal to the concentration measured in the heavy water drained from the reactor at the time of shutdown, decay corrected to present day.

$$Decay\ Corrected\ {}^{3}H\ Concentration\ \left(\frac{Bq}{kg}\right) \times Mass\ of\ D_2O\ System\ Capacity\ (kg) \times 1\%$$

3.3 Method 2 – Amount of Tritium Released During Sampling

An estimation of tritium activity per surface area (SA) was calculated using the increase in total activity of tritium released that appears to be due to sampling activities and the internal surface area of the samples collected. The value was then multiplied by the internal surface area of the moderator system. This approach was applied to two data sets: the samples collected in 2015 and the ones collected in 2017.

$$\frac{Total\ {}^{3}H\ Released\ (Bq)}{Internal\ SA\ of\ Samples\ (cm^2)} \times Internal\ SA\ of\ Moderator\ System\ (cm^2)$$

3.4 Method 3 – Rate of Tritium Release During Sampling

The third method uses the peak release rate of tritium during the sampling (Bq/week) and multiplies it by the sampling period (weeks) to get an estimate of the total activity of tritium released during sampling. This is then multipled by the internal surface area ratio of the moderator system over the samples. Unlike method 2, this estimation does not factor in the routine emissions of tritium expected during the sampling period which adds to its conservatism, however, it also doesn’t account for the higher than usual emissions before and after the physical sampling. Again, a calculation was completed for both data sets.

$$Peak\ {}^{3}H\ Release\ Rate\ \left(\frac{Bq}{wk}\right) \times Sampling\ Time\ (wk) \times \frac{Internal\ SA\ of\ Moderator\ System\ (cm^2)}{Internal\ SA\ of\ Samples\ (cm^2)}$$

3.5 Method 4 – Solubility of Hydrogen in Stainless Steel

This last approach assumes that the samples were not liberated of their tritium content and that the increase in tritium emission during sampling was due to the holes in the system increasing airflow. It does not rely on emission data and is performed as a check on the theoretical maximum amount of tritium that could be absorbed by stainless steel (SS) over the operational lifetime of the reactor. The solubility limit of tritium in stainless steel is taken from a review of several experiments [6]. The total volume of metal in the system was estimated while conservatively assuming aluminum components to be stainless steel as stainless steel has a greater tritium solubility. This method does not account for tritium surface contamination, only absorbed tritium, and conservatively ignores radioactive decay and gaseous emissions from the system since shutdown. The concentration of tritium in the absorbed hydrogen isotopes was assumed to be the same as that of tritium in the heavy water at shutdown.

$$Solubility\ of\ H\ in\ SS\ \left(\frac{mol}{m^3 atm^{1/2}}\right) \times Volume\ of\ SS\ (m^3) \times Operational\ Pressure\ (atm) \times Molar\ Mass\ \left(\frac{g}{mol}\right) \times {}^{3}H\ Concentration\ \left(\frac{Bq}{kg}\right)$$

4. RESULTS

The results of the estimation methods and the absorbed tritium analysis are presented in Table 1 below. The actual calculations were carried out in a separate spreadsheet [7].

Table 1: Estimates of Remaining Tritium Activity

Method	Activity (Bq)
Absorbed Tritium Analysis	1.11×10^{9}
1 - 1% Remaining Estimate	1.27×10^{14}
2a - Amount of ^{3}H Released (2015)	2.47×10^{15}
2b - Amount of ^{3}H Released (2017)	3.80×10^{14}
3a - Rate of ^{3}H Release (2015)	2.26×10^{15}
3b - Rate of ^{3}H Release (2017)	3.14×10^{14}
4 - H Solubility Limit in SS	5.31×10^{12}

For both of the methods that used two data sets, the estimations produced by the 2015 data are higher. This may be the result of two key factors:

- The 2015 sampling work was preceded by the opening of several valves that had previously been closed and caused an increase in tritium emissions prior to the start of sampling. It is unclear what contribution of the total activity is from previously isolated pockets of tritium vapours and what is from the sampling activities. The result is likely an over estimate of the activity released per area of the samples and thus an over estimate of the activity.
- As previously stated, the 2015 work focussed on areas with the expected highest levels of tritium contamination and did not acquire data from the rest of the system. The selection of the most contaminated points skews the average contamination for the system to a higher value by disregarding areas with low or no contamination.

The 2017 results are not skewed by biased sampling, but did require the opening of valves to previously unvented portions of the system.

5. HYPOTHESIS TESTING

It was proposed that the heating and vibrations experienced by the samples may not have been sufficient to release substantial amounts of tritium from the samples and that the increase in tritium emissions was simply a result of increased airflow through the system. In such case the tritium emissions produced during the 2015 and 2017 campaign would not be related to the area of the coupons, but to the length of time of increased airflow. This would also mean that the results of the absorbed tritium analysis are more indicative than assumed.

To test this theory, a series of simple experiments were performed [4]. First, one of the sampling holes was opened up to increase airflow. Second, a sample-sized portion of the system was heated up to 500°C. For both experiments tritium emissions were monitored, but neither produced comparable emissions to the sampling campaigns.

6. CONCLUSION

Based on the hypothesis testing, it is concluded that neither increased airflow nor heating caused the increased tritium emissions during sample collection and as such no inferences will be drawn to which tritium activity best represents the actual remaining inventory. To maintain a consistent conservative approach to selection of model parameters, the source term values, used for modelling tritium in the EA [8][9], will be modified to use the most conservative estimate (2.47×10^{15} Bq) and the results will be updated as necessary.

7. REFERENCES

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