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INITIAL INVESTIGATIONS OF SNS TARGET FACILITY ACCIDENT SOURCE TERMS*

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ABSTRACT

The Spallation Neutron Source (SNS) is a Department of Energy, accelerator-based neutron source proposed for construction at the Oak Ridge National Laboratory. The project is currently nearing the end of the conceptual design stage. The objective of the target facility is to provide beams of pulsed thermal and sub-thermal neutrons for research purposes. The neutrons are created by the action of highly energetic protons (~1 GeV) on a mercury target. The proton beam power will be 1 MW with planned upgrades to 2 MW and, eventually, to 4 MW. Over the course of facility life, significant inventories of spallation and activation products will build up in the target mercury. Accordingly, the facility is being designed to prevent or minimize potential environmental source terms.

The results of calculations of the SNS target mercury radionuclide inventories and the characteristics of the dominant radionuclides are presented. The effect of the activation/spallation product chemical and physical characteristics on dispersability is discussed. Energy sources that could drive potential releases, credible initiating events and facility preventive and mitigative features are described. The source term for the limiting extremely unlikely mercury spill accident scenario is presented. These results support the conclusion that the facility has a low hazard profile with regard to the accidental release of radioactive material.

I. INTRODUCTION

The SNS accelerator produces pulses of 1 GeV protons to bombard the mercury target, causing spallation of the mercury nuclei and consequent pulses of neutrons that are moderated and channeled along beam tubes and guide tubes, to research instrument stations in the experiment hall. The mercury target holds about 25-liters of mercury; however, considering mercury system components such as the pump, heat exchanger, and piping, the total volume of mercury in the system approaches 1 m^3. Transmutation of the target mercury is sufficiently low that it is not necessary to change the mercury during the life of the facility. Spallation and activation products are distributed throughout the entire (<1 m^3) volume of mercury in the target system.

During operation of the proton beam, the mercury is pumped through a heat exchanger to dissipate the heat absorbed from the proton beam. Normal mercury target vessel outlet temperature is 110 °C and inlet temperature is 80 °C. The heat exchanger is double walled with monitoring for interspace leakage to eliminate the possibility of mercury contaminating the cooling water, or vice versa.

The mercury system is closed except for a very low rate of helium purge. This helium flow transports gaseous spallation and activation products from the mercury system to the off-gas treatment system. Multiple stages of treatment ensure that normal environmental releases of the radioactive nuclides are extremely low. These stages include cooling of the helium off-gas to the temperature of liquid nitrogen to condense any mercury vapor present in the exhausted helium, compression and storage of non-condensible gases in a decay tank or their adsorption on a cryogenic activated charcoal bed to allow decay of their radioactivity before release, and chemical removal of tritium in a hydride bed.

Design features have been selected to ensure that potential accident source terms are very low. First, the normal temperature of 80 - 110 °C is consistent with a low mercury vapor pressure, e.g . $5.9(10)^{-4}$atm at 110 °C. Mercury evaporates very slowly at this temperature. Redundant, diverse and independent proton initial spallation product transport studies have concentrated on iodine, mercury, and gadolinium transport.

Table 1 Dominant target radionuclides after 40-y of operation at 1 MW

Nuclides present with hazard index > .01			Dispersability assessment
Nuclide inventories[a] (Ci)	Half life	Hazard index (no units)	
Gaseous			Can be released if air enters the hydride bed in the hot off-gas system.
H-3: $4(10)^3$	12.3 y	.013	
Volatile			Iodine combines chemically with Hg to form Hg_2I_2, but the accident source terms assume 100% release to ensure conservatism.
I-125: $7.5(10)^1$	60.1 days	0.03	
Semi-volatile			
Hg-194: $1.1(10)^3$	262 y	0.017	
Hg-197: $1.2(10)^5$	2.67 days	0.025	Hg can evaporate if exposed to air.
Hg-203: $8.3(10)^4$	46.6 days	0.132	
Non-volatile			No transport mechanism has been identified: these elements have essentially zero vapor pressure at normal and accident temperatures. They are either dissolved in the Hg or have plated out on an interior Hg system surface or been filtered out of the Hg.
Gd-148: $7.9(10)^2$	74.5 y	0.57	
Hf-172: $1.6(10)^4$	1.87 y	0.035	

III. SPALLATION PRODUCT TRANSPORT

The discussion in the section is based on the very preliminary look that has been given to spallation product chemistry and physics. Additional work is planned, as needed to obtain a complete understanding of the transport processes.

Tritium on the hydride bed can be released if a fault occurs in the hydride bed boundary or associated piping that allows oxygen to enter the bed. The reaction results in oxidation of the hydrided tritium, releasing the tritium in the form of tritiated water vapor. This water vapor can be assumed to escape readily and completely from the target hot cell.

It is not likely that iodine in a Hg spallation neutron source would be in the form of unreacted elemental iodine. In pure mercury it would react to form mercurous iodide Hg_2I_2. However, iodine forms compounds with spallation products such as cesium, barium, and the rare earths that are much more stable than Hg_2I_2. The vapor pressure of I_2 over Hg_2I_2 is very low. The value calculated at 110°C for the reaction: $Hg_2I_2 = 2\ Hg + I_2\ (gas)$ was only $\sim 10^{-16}$ atmospheres. However, mercurous iodide Hg_2I_2 can dissociate into mercury and mercuric iodide, HgI_2:

$$Hg_2I_2 = Hg + HgI_2\ (gas).$$

At 110°C the partial pressure of the HgI_2 (gas) was calculated as $\sim 7 \times 10^{-6}$ atmospheres. This is still not very high, but some iodine could be lost over long periods of time. It should be noted, however, that air would react with the iodides and convert them to oxides while releasing iodine as elemental iodine. This is of concern in an accident situation because the dynamics of the oxidation reaction have not yet been studied; therefore, accident studies, to date, have conservatively assumed 100% release of any iodine present in spilled Hg that is exposed to air. Studies are planned to determine the extent to which Hg_2I_2 exposed to air will oxidize and release iodine. In addition, studies are also planned to determine the extent to which iodine may form stable iodides with some spallation product nuclides (e.g. lanthanum).

Mercury is subject to release by evaporation if a spill is assumed to occur in conjunction with the features that are designed to ensure that spilled mercury drains to the collection tank. Mercury can, of course, be made to boil -- given enough energy input -- but the decay heat (initially ~0.25 W/kg of Hg and decreasing thereafter) is too low to bring mercury to the boiling point (357 °C at 1 atm) and it is not credible to postulate that the 1 MW proton beam continues indefinitely while mercury temperature exceeds the normal range, which is more than 200 °C below the boiling point.

For an accident involving a mercury spill, the source term for release of Hg from the target hot cell can, depending on accident specifics, be limited by one of two factors: the rate at which evaporation can occur from the surface of the mercury, or the rate at which mercury-saturated air can transport the mercury out of the enclosure or hot cell.

The flow of air inside the mercury enclosure is very slow but a turbulent mass transfer correlation was used to provide a conservative estimate of the evaporative tendency:

$$\text{Flux} = k_g C_g$$

where,

Flux = evaporative mass flux of Hg (mass per unit time per unit area)

k_g = mass transfer coefficient

C_g = mercury vapor concentration at the mercury/air interface, calculated from the saturated vapor pressure by the ideal gas equation.

The mass transfer coefficient is determined from a correlation for turbulent plane flow:

$$k_g = D/L\,(0.036\,Re^{.8}\,Sc^{.33}) = 2.74(10)^{-4}\ ms^{-1}$$

Substituting values applicable to a widespread spill of Hg with an assumed temperature of 110 °C, the mercury flux is found to be 3.6 g/ m^2h from a mercury surface exposed to a turbulent flow air. The evaporation rate can at other temperatures can be estimated similarly. For example, at 50 °C it would be reduced to 0.12 g/ m^2h. This approach does not account for very thin oxide films that can coat mercury surfaces exposed to oxygen and retard evaporation[2].

If the Hg vapor source term for a mercury spill is not evaporation-limited then it will be limited by the saturation of the ventilation air with mercury vapor. As an example, consider that the mercury enclosure's 11 m^3/min flow of air is exhausted at 50 °C, a feasible post-spill mercury enclosure exhaust temperature. The corresponding transport rate for mercury would be 81 g Hg/h from the enclosure. Evaporation, whether limited by the conditions at the air-mercury interface or by the gross ventilation system air flow rate, provides a mechanism for the release of small amounts of Hg in accidents where failure of the mercury to drain as designed is postulated. The amounts and rates of release will depend upon the details of the accident assumptions.

Hafnium and gadolinium are very reactive with oxygen. This is true of the other rare earths as well. This means that any oxygen in the He purge gas would be scavenged to form an oxide. Thus, depending on the purity of the He, hafnium and gadolinium could be in the mercury as metals or as the oxides HfO_2 or Gd_2O_3. The solubility of Gd in Hg at 100°C has been reported as 5×10^{-2} atom%[3] and this concentration is not exceeded after 40 years of operation. Several rare earth-mercury compounds are known. As noted, these compounds would most likely contain a variety of rare earth elements. There are no data available for the solubility of hafnium in mercury, but by comparison with zirconium, it is very low. A hafnium-mercury compound Hf_2Hg could form. In summary, gadolinium could be in the form of an oxide, it could be dissolved in liquid mercury, or it could form an intermetallic compound that may or may not contain Hg. It is extremely difficult to conceive of any mechanism where gadolinium could be airborne at 110°C. The vapor pressure of Gd dissolved in Hg would be less than the vapor pressure of elemental Gd at this temperature which is negligibly small. Consequently, the accident releases of Gd and Hf from spilled Hg are thought to be negligibly small. Studies of spallation product chemistry will continue, as needed to understand the interactions of mercury and its spallation products.

IV. SOURCE TERM ESTIMATES

A full range of target system and target off-gas system accidents has been considered, and source terms estimated in support of the SNS Environmental Impact Statement. All events capable of releasing radioactivity were considered, and placed in frequency categories from anticipated ($>2(10)^{-2}$/y), to unlikely (10^{-4}/y<frequency<$2(10)^{-2}$/y), to extremely unlikley (10^{-6}/y<frequency<10^{-4}/y), to beyond design basis (frequency <10^{-6}/y). Engineering judgement was used to place events in the correct bin by considering the number and type of faults defining the event. For example, an operator error or a pump coastdown would be in the anticipated category, whereas a major structural failure would be considered unlikely or extremely unlikely.

This section concentrates on the limiting extremely unlikley (EU) mercury spill event identified for the SNS target facility. EIS source terms tend to be extremely conservative since there is the need to bound possible consequences for the consideration of potential health effects (not addressed in this paper). Simple analytical metehods -- essentially hand calculations -- were used for the source term estimates in order for the source terms to be demonstrably conservative. A more realistic estimate of the same source term is presented for comparison purposes.

The EU limiting mercury spill event is defined by the following postulated failures: (1) simultaneous loss of mercury system integrity and loss of the mercury heat exchanger cooling water, (2) failure of the anticipatory proton beam cut-off based on cooling water pump status, allowing the mercury to heat up by ~20 C before the beam is cut off on high mercury temperature, and (3) failure of the mercury enclosure engineered splash shield and drainage features, allowing the mercury leakage to be spread over the interior of the whole hot cell. The leak is catastrophic enough for all the mercury to have leaked out within the first 10 minutes (at which time the operator would stop the pump and drain any unspilled mercury to the drain tank).

Conservatisms applied to both the bounding EIS analysis and the more realistic case include: (1) hot cell ventilation flow continues at the normal 136 m^3/min rate throughout (source term would be much lower without continuing forced cell air flow), (2) cell air inlet temperature is 30 C (air conditioning failure during the hottest month of the year). The different bounding assmptions employed for each analysis are illustrated as follows:

Analysis assumption	Bounding EIS analysis	Realistic conservative analysis
Air exit temperature for first 10 minutes of accident	86 C ... based on assumed thermal equilibrium between leaked mercury and cell air	40 C ... based on assessment that the surface area and conditions would not provide significant heating of cell air
Air exit saturation with mercury during first 10 minutes	100% ... assumed efficient and complete contact of air with leaked mercury	50% ... only air flowing along floor would achieve significant mercury saturation.
Uncertainty factor applied to estimated evaporation rate between 10 minutes and 10 days	10	2
Surface area exposed to air for evaporation	100% of the cell floor area	20% of the cell floor area

Utilizing the above stated assumptions, the source term for mercury loss was calculated for each part of the accident -- the initial phase during the first 10 minutes during which time the mercury is leaking out, and the long term phase, between 10 minutes and 10 days. After 10 days it is assumed that accident recovery and clean up operations would be successful in reducing the mercury evaporation to a very low rate. The source term estimates for this event are as follows:

- First 10 minutes: 1500 g Hg for the EIS analysis vs. 41 g Hg for the realistic analysis
- 10 minutes to 10 days: 1900 g Hg/day for the EIS analysis vs. 76 g Hg/day for the realistic analysis.

As previously discussed, the above source term for release of radioactive mercury from the hot cell must be assumed to be transported by the ventilation system to the target facility stack for release to the environment. For the EIS analysis it was assumed that all the iodine would be released in conjunction with the mercury release due to the assumed oxidation of Hg_2I_2. A more realistic estimate of the iodine release was not attempted because the Hg_2I_2 oxidation reaction has not been sufficiently studied at this time. No gaseous spallation products are released with the evaporated mercury because of the action of the helium purge flow which transfers them continuously to the off-gas system. None of the solid spallation products are released because these are typically dissolved in the mercury, with negligibly small vapor pressures (as discussed in the previous section for gadolinium and hafnium) at the temperature range possible in this accident.

V. CONCLUSIONS

Accident characteristics of the SNS target system have been investigated and initial surveys of the chemistry and physics of spallation product trasnport completed. The source term for the limiting extremely unlikely mercury spill event has been estimated using bounding assumptions and compared to the source term estimated using realistic but still conservative analysis assumptions. The results show that the EIS source term is conservative by more than an order of magnitude.

Although the scope of this paper includes source terms and not health effects, it is evident that the radiological hazard asociated with the limiting mercury spill accident is small considering the very low frequency of the event. Referring to Table 1 in Section II, and the explanation that a hazard index of one means that an accident involving the corresponding amount of each nuclide (and assumed release fractions of 1% for Hg and 50% for I) would cause a 1 rem dose at 300-m for a ground level release, we can estimate that, if the EIS source term were a ground level release, the integrated 10-day dose at 300-m would be on the order of 0.1 rem. This is below the annual background exposure. Furthermore, since the limiting extremely unlikley release would be from the stack, and thus elevated for more dispersion, the actual exposure would be expected to be significantly lower than 0.1 rem. It is concluded that the radiological hazard associated with SNS target facility accidents is low with regard to members of the public or for workers at other facilities.

VI. REFERENCES

1. DOE Standard DOE-STD-1027-92, "Hazard Categorization and Accident Analysis Techniques for Compliance with DOE Order 5480.23, Nuclear Safety Analysis Reports," U. S. Department of Energy, December 1992.

2. D. J. Banjamin, "The Effect of Gases and Vapours on Mercury Evaporation," Mat. Res. Bull., Vol. 19, pp. 443-450, 1984. Pergammon Press Ltd.

3. A. F. Messing and O. C. Dean, *Solubilities of Selected Metals in Mercury: Hermex Process*, ORNL-2871, June 1960.