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Chemistry and Physics Challenges in

Spallation Neutron Source Safety Analyses

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Introduction

This paper describes several unique issues that have been addressed in the accident and source term analyses performed in support of the Preliminary Safety Analysis Report (PSAR) for the Spallation Neutron Source (SNS) Target Building. The inventory of radioactive material associated with the SNS is in the Target Building, which is a nuclear facility. Areas of interest for safety analyses include determining the major contributors to source terms from the spallation process within the mercury target, the effect of water and mercury interaction following a postulated target failure, and the combined toxicological and radiological impacts of events with airborne mercury release.

Background

The SNS is a Department of Energy (DOE) research facility under construction near Oak Ridge, Tennessee. The SNS includes a 300-m long, 1 GeV, 2 MW, linear accelerator that produces neutrons by collisions of high-energy protons with mercury target nuclei. The mercury target atoms are in a circulating mercury loop that is water-cooled. The mercury loop operates at a nominal average temperature of 75° C (60° C nominal cold leg temperature and 90° C nominal hot leg temperature). The overall target system also includes circulating fluid systems for supercritical cryogenic hydrogen (to moderate product neutrons to low energy), heavy water (for cooling of shielding), and several light water systems (for shielding cooling, proton beam window and neutron beam window cooling, and to moderate neutrons to energies higher than those from the cryogenic hydrogen moderator).

Figure 1 provides an overview of the entire SNS facility. Figure 2 shows a cutaway view of the Target Building. Figure 3 shows the equipment arrangement for the circulating mercury loop. Figure 4 shows the area within the Core Vessel where the proton beam strikes the mercury target. Additional general descriptive information about the SNS is available online at: *www.sns.gov*.

Material at Risk

After sufficient "beam on" operations, the primary radioactive material at risk will include radioactive spallation product atoms, radioactive mercury atoms (from neutron capture in mercury), and tritium (from capture in water cooling systems and from decay of spallation products). The spallation process generates a broad range of radioactive nuclides with atomic numbers below 80 (mercury is atomic number 80). Operation of the target system will slowly build in spallation products (which act like impurity materials in the mercury). The spallation products can be viewed as being dissolved in or alloyed with the liquid mercury. Accident analyses are based on the calculated radionuclide inventory after 40 years of operation.

Except for volatile elements and compounds, radioisotopes will be contained within the circulating mercury loop during normal operation. If the mercury is spilled as the result of an accident, the volatiles will be released from the loop and spallation products dissolved in the mercury (or alloyed with the mercury) will accompany the spilled mercury.

Approximately 50% of the potential hazard from radionuclides is from radioactive mercury, while the remaining approximately 50% is from spallation products. Specific radioactive spallation product nuclides that are of special concern in consequence assessments are Gd-148 (an alpha emitter), radioisotopes of iodine (which have the potential to be highly volatile at system operating temperatures), radioisotopes of xenon (which is a gas), and radioisotopes of rhenium (Re) and osmium (Os)(which have the potential to be converted into gaseous or volatile oxides in some accident scenarios).

Toxic chemical hazards of mercury are also considered in the safety analysis. In some postulated accident scenarios, the resulting mercury exposure could exceed toxicological criteria without exceeding established radiological guidelines. The toxic nature of the mercury means that certain safety-related controls will be imposed on facility operations even before the mercury has become sufficiently radioactive as to require controls.

Accident Scenarios

Several postulated accident scenarios lead to spills of radioactive mercury (containing radioactive spallation products) within the SNS Target Building. The spilled mercury is initially at the mercury loop operating temperature (or even higher in some scenarios that involve either facility fires or reduced heat removal from the mercury prior to the spill).

Seismic events have the potential to cause spills both of mercury and of other coolants (such as water and heavy water). These events therefore lead to scenarios with the potential for mercury and liquid coolants to come into contact. Because of the possibility of such contact, the accident analyses have included reviews of: (1) the chemistry of mercury and of spallation products in the mercury in the presence of water; (2) dispersive mechanisms involving water, including energetics and spallation product dissolution; and (3) transport of radioactive spallation products in both mercury and water.

Airborne Release Mechanisms

The potential for generation of airborne mercury aerosols, mercury vapor, and other gaseous and volatile radioactive materials from spilled material is a major focus of the accident and source term analyses. Mercury aerosols would include radioactive spallation products "dissolved" in the liquid mercury. Mercury vapor would contain radioactive mercury isotopes, but spallation products would generally be left behind during the evaporation process. Gaseous and volatile materials among the spallation products include tritium, iodine, and xenon isotopes.

The evaluations performed for aerosols include both mercury aerosols and aqueous aerosols. The aerosolization processes that have been evaluated include: atomization of mercury, mechanical dispersion of mercury, boiling of water (including boiling due to contact with hot mercury), and dispersion and evaporation of aqueous solutions. In addition, a determination was made that water and mercury temperatures would not support significant energetics (vigorous boiling or steam explosions).

The analyses have assessed several possible interactions between mercury and water that could exacerbate accident consequences – including the possibility that spallation products could (1) be transferred from mercury to water during the accident scenario and (2) then become airborne from the water due, for example, to vigorous water boiling. The analyses have also assessed the possibility that airborne releases from mercury could be reduced by the presence of water, such as by spilled water forming a vapor barrier over the mercury and by spilled water cooling the hot mercury.

Early in the SNS target design process (when this work was initiated), the nominal mercury loop hot leg temperature was expected to be over 100°C during operation, which introduced the possibility that contact between mercury and water during accident scenarios could cause boiling of the water. Assessments were performed which lead to the conclusion that this mercury temperature is too low to trigger steam explosions if hot mercury came into contact with water during accident scenarios.

Subsequent design changes have resulted in even lower values for the expected mercury temperatures; the nominal hot leg temperature is 90°C in the current design. Even allowing for the most adverse combination of instrument uncertainty and operating condition variations, the maximum mercury temperature during normal operation is now expected to be, at most, only a few degrees above the boiling temperature of water.

Higher temperatures may be associated with certain accidents, such as fires, and this potential is explicitly included in the analyses for those events.

For completeness, the analysis has identified and evaluated several other potential mechanisms and phenomena that could cause radioactive material to become airborne during postulated SNS accident scenarios (or that could increase the amount of radioactive that becomes airborne). Items that have been evaluated include: "flaking" or air entrainment of solid oxides of spallation products that could be floating on the free surface of any spilled mercury, the heat added by exothermic oxidation of spallation products, and the possibility of the formation of volatile spallation product oxides (two such oxides have been identified: OsO_4 and Re_2O_7, however, these can only be formed at temperatures much higher than those present during normal operation, so they could be formed only during certain accident scenarios). These potential release mechanisms and phenomena have been incorporated into the analyses, where appropriate.

Mercury and Spallation Product Chemistry

In order to fully evaluate the various potential release mechanisms, supporting analyses have included assessments of the behavior of: (1) alloys (amalgams) of the mercury and metallic spallation products; (2) oxides of the spallation products; and (3) other compounds that might reasonably be expected, such as mercurous or mercuric iodide. This subsection briefly summarizes some of the results of the assessments.

Mercury is chemically inert toward water so the interaction of mercury and water is treated on purely physical grounds in the accident analyses. Important physical characteristics of mercury include its high density of 13,600 kg/m^3, substantial surface tension, and its slow evaporation at the temperatures of interest in accident scenarios. The density difference between water and mercury and the mercury surface tension restrict its contact with water under accident conditions; usually, such contact would involve only a limited amount of surface between pools of mercury and overlying water.

Liquid mercury is a fairly aggressive solvent toward many other metals, forming alloys called amalgams. In sufficient amounts these amalgams may crystallize out of solution. Most are lighter than mercury, so they will float on a mercury pool.

Amalgams are typically very chemically reactive. Depending on the elements involved, they may react spontaneously with air, water, or even organic materials. The resulting oxides, or other non-metallic products of such reactions, are typically very insoluble in liquid mercury. As these oxides are generated, they will form a skin or dross on the surface of the mercury pool, but some may collect on the surface of the mercury vessels and piping.

A few of the spallation product elements, such as tantalum, do not form alloys or react with mercury(1). There are expected to precipitate from the liquid during operation. Most such precipitates are less dense than mercury, so they will float to the surface. However, tantalum and tungsten are more dense than mercury and will sink, unless they oxidize; these two elements may collect as sediments at low points in the circulating loop. The spallation product elements of primary radiological concern may be divided into chemical categories. These are identified and briefly discussed in the following paragraphs.

Isotopes of the rare earths Gd, Lu, Tb, Ho, Dy, Er, and Tm -- The most important of these with respect to radiological consequences is Gd-148. The chemical behavior of all these materials is very similar. In the absence of air they remain dissolved in the liquid mercury. If contacted by air or water, they form oxides that precipitate from the liquid[(2)]. The rare earth oxides resulting from such reactions are only very slightly soluble in water.

Hafnium isotopes -- The most important of these with respect to radiological consequences is Hf-172. Hafnium is similar in its chemistry to zirconium. One authoritative source states that zirconium was not observed to alloy with mercury.[(3)] Another source, however, reports zirconium as being very soluble in mercury.[(4)] Whether soluble or not, hafnium in solution or fine division is expected to oxidize rapidly if exposed to air. The resulting oxide is rather inert and has a very low solubility in water.

Gold isotopes -- The most important of these with respect to radiological consequences are Au-188 and Au-195. These are expected to dissolve homogeneously in the liquid mercury. They are inert to water and air and will remain associated with the mercury.

Tungsten isotopes -- The most important of these with respect to radiological consequences are W-172, W-173, and W-174. Tungsten is chemically inert toward mercury and toward water. It is likely to precipitate from the liquid mercury, and may oxidize if exposed to air. The resulting oxides are chemically stable and inert, and do not dissolve in water near pH 7.

Tantalum isotopes -- The most important of these with respect to radiological consequences are Ta-170 and Ta-171. Tantalum is chemically inert toward mercury and quite insoluble in water. It oxidizes in air when heated, and will probably oxidize at room temperature if finely divided. The resulting oxide is also insoluble in water. Tantalum will sink to the bottom of a mercury pool, and so it may not all be exposed to air even if the mercury spills during an accident.

Osmium isotopes -- The most important of these with respect to radiological consequences are Os-179 and Os-183m. Osmium is one of the platinum group of metals. As such it is normally inert toward attack by water. Information is lacking on whether it forms an amalgam with liquid mercury, but other metals in this group do so. Osmium is reported to oxidize slowly when exposed to air, and amalgamated or finely divided osmium from the liquid mercury loop should oxidize rapidly on contact with air[(6)] The resulting osmium dioxide disproportionates when heated, forming volatile osmium tetroxide. Consequently, under fire conditions, a significant fraction of the osmium may be converted to and dispersed as OsO_4 gas (because of this, Os is treated as a gas in source term calculations for events where the mercury can reach elevated temperatures and be exposed to air). This tetroxide is a powerful oxidant, and reacts rapidly with many organic compounds. It can also dissolve in water[5]. Osmium dioxide may also dissolve to some extent in water.

Iodine isotopes -- The most important of these with respect to radiological consequences are I-124, I-125, and I-126. These will probably be in the form of mercurous or mercuric iodide. These both have a slight solubility in water and so might partly or completely dissolve in water that contacts them.

Radioactive isotopes of mercury --The most important of these with respect to radiological consequences are Hg-189, Hg-193, Hg-194, Hg-195, Hg-197, and Hg-203. The radioactive mercury will be homogeneously mixed with the stable mercury.

Elements of lesser radiological concern include Tl, Pt, Ir, and Re. Tl is a reactive metal, and its amalgam will react with air to form oxides that are not volatile at ordinary temperatures. Pt and Ir are likely to remain in mercury solution. The behavior of Re is uncertain; it may also oxidize, and one of its oxides is somewhat volatile (because of this, Re is treated as a gas in source term calculations for events where the mercury can reach elevated temperatures and be exposed to air).

The total amounts of material formed by spallation are small. Typical amounts for each category of element described above are between 100 and 1000 grams. This includes both radioactive isotopes of these elements and stable isotopes for each of these elements, which will also be present after 40 years of operation.

Scoping calculations have been performed to evaluate the possibility that, under accident conditions, spallation products or their oxides could (1) transferred from spilled mercury into spilled water and then (2) become airborne from the water. The results of these calculations indicate that, primarily because of low solubilities, large quantities of water (several cubic meters) would be required to be in contact with the mercury for substantial dissolution of spallation products. Even if this amount of water were present, the transfer would be a very slow process (probably requiring many days). Both stable and radioactive atoms would be transferred to the water, which contributes to the slowness of this process. Processes that tend to transfer the radioactive isotopes of a given spallation product element to water will also tend to transfer the non-radioactive isotopes.

The large amount of water and the relatively long time period required for significant spallation product transfer means that after such a transfer, the mercury would no longer be at high temperature. Therefore vigorous water boiling (due to contact with hot mercury) would not be possible. Boiling from contact with hot mercury during postulated SNS accident scenarios is the primary mechanism that has been identified with the potential to liberate dissolved spallation products from water.

Any water spilled in conjunction with a mercury spill during an accident would float on the mercury. This floating water would act as a vapor barrier, which would retard radioactive mercury vapor from becoming airborne. Scoping calculations indicate the ability of a water spill to reduce mercury evaporation rates more than offsets the radiation dose consequences from any spallation products released due to transfer of spallation products to water.

Conclusions

The analyses have concluded that the presence of water, which will "float" on the mercury, has the net effect of reducing overall consequences from events where both water and mercury are released from their normal piping and come into contact with each other.

Due to low solubility, only small quantities of spallation products could be transferred to water during accident scenarios. In addition, due to the relatively low mercury temperature (near 100° C), there is no mechanism for releasing a substantial fraction of the small amount of spallation product that could be transferred to the water or for the water to provide energetics that could increase the mercury release. Therefore, the consequences of any potential releases of these soluble spallation products are more than offset by reduced mercury vapor formation due to water serving as a mercury vapor barrier.

References

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Figure 1

SNS Facility Arrangement

Figure 2

SNS Target Building Cutaway Drawing

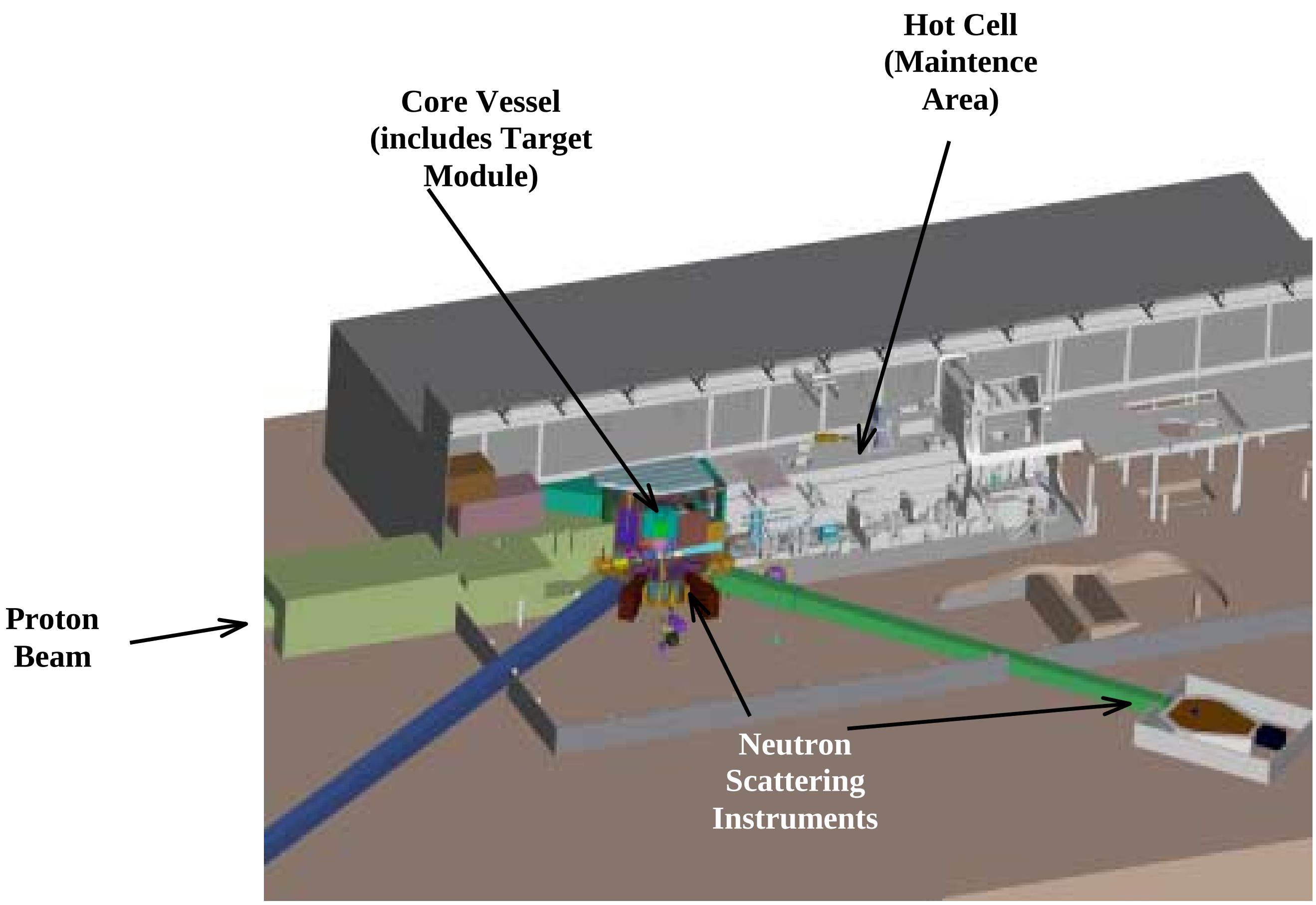

Figure 3

Mercury Circulating System Arragnement

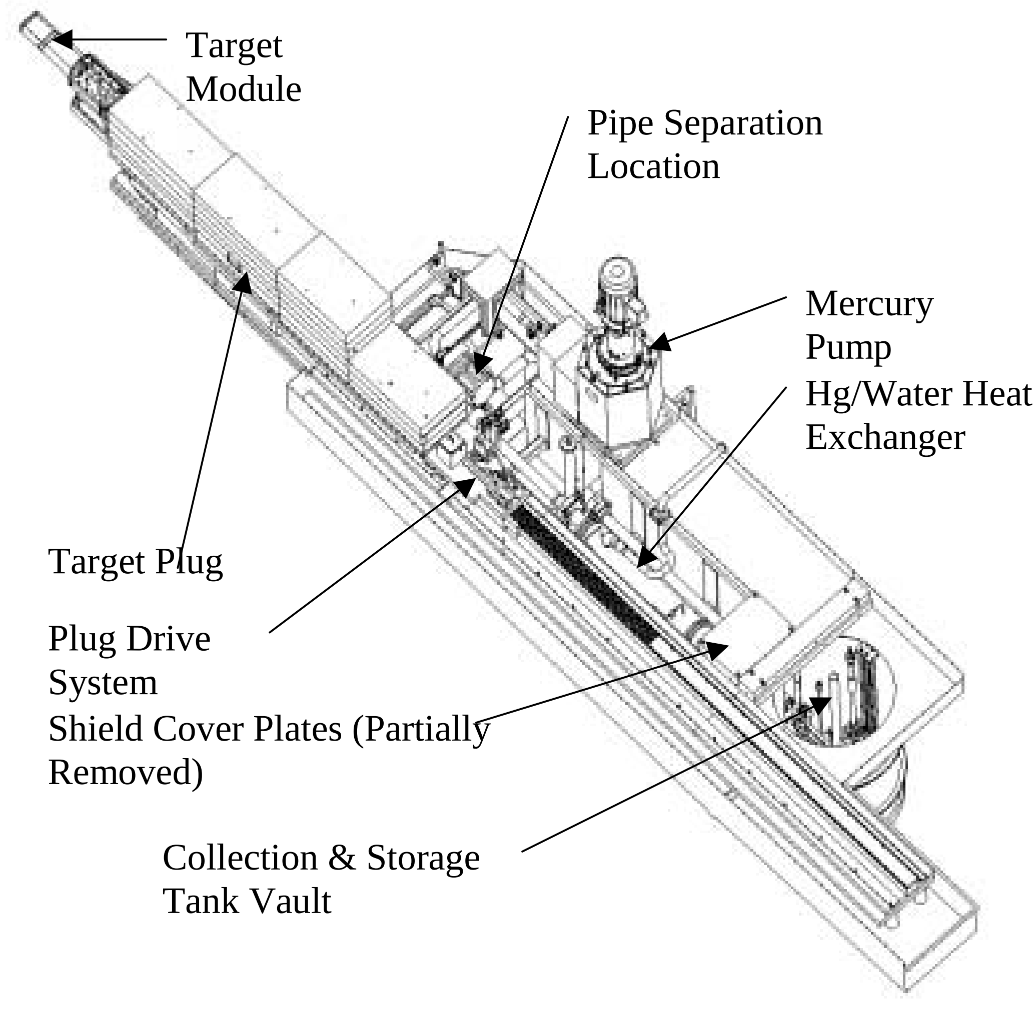

Figure 4

Core Vessel Interior Arrangement