Hydrogen release rate from the NPDGamma liquid hydrogen target into the BL-13 shielding structure in a failure of target vacuum and hydrogen boundaries

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Object: Object of this report is to define an upper limit to the hydrogen release rate into the experimental shielding structure of the BL-13 in a case of a failure of both the isolation vacuum and the hydrogen boundaries of the NPDGamma hydrogen target. The report summarizes briefly some of experimental results, found from literature and results with the NPDGamma hydrogen target, describes results from FLUENT simulations and then estimates the heat transfer rate to the cryogenic hydrogen in the NPDGamma target when the cryostat isolation vacuum is filled by air as a consequence of a vacuum failure. The heat transfer rate is the variable that defines the liquid hydrogen boil off rate in the target and thus the hydrogen release rate into the BL-13 enclosure if the hydrogen boundaries are failed during the event.

The NPDGamma experiment in the beam line (BL-13) requires a 16 liter liquid hydrogen (LH₂) target for a measurement of the directional correlation $A_\gamma$ between the momentum of the 2.2 MeV $\gamma$-ray and the neutron spin in the reaction of $\bar{n} + p \rightarrow d + \gamma$ where polarized cold neutrons are captured by hydrogen. In normal mode the LH₂ target is operated at temperatures below 20 K cooled by two mechanical cryocoolers [1]. The liquid hydrogen is contained in a cylindrical aluminum vessel which is connected to the relief system located outside the BL-13 cave by a vent line which conducts the H₂ gas safely outside of the Target Building when the target is emptied. The heat conductivity from outside of the cryostat into the LH₂ has been minimized by applying normal cryogenic technique such as vacuum (cryostat vacuum or isolation vacuum), heat shield (radiation shielding), and layers of superinsulation, see figure 1 which shows a schematic of a cross section of the NPDGamma hydrogen target cryostat.

In the case of a catastrophic event where the isolation vacuum fails and consequently the vacuum is filled by air, a large heat flow (energy flow) to the LH₂ causes a rapid boil off of the cryogenic hydrogen. If in addition the vent line is broken inside the BL-13 enclosure, the boiling hydrogen gas will flow into the enclosure (cave). The release rate of hydrogen into the cave depends upon the heat transfer rate into the LH₂ from the outer walls of the cryostat carried by air in the vacuum space. In this report we estimate the upper limit for the heat transfer rate and thus the release rate limit of hydrogen gas into the cave.
Table 1. Values for volumes and surface areas in the NPDGamma cryostat.

<table>
<thead>
<tr>
<th>Component</th>
<th>Value</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cryostat vacuum - volume</td>
<td>1.5×10^2 (0.15)</td>
<td>liter (m^3)</td>
</tr>
<tr>
<td>Cryostat vacuum – surface area</td>
<td>1.8</td>
<td>m^2</td>
</tr>
<tr>
<td>Cryostat vacuum - material</td>
<td>Al</td>
<td></td>
</tr>
<tr>
<td>LH2 target - volume</td>
<td>16 (0.016)</td>
<td>liter (m^3)</td>
</tr>
<tr>
<td>LH2 target – surface area</td>
<td>0.4</td>
<td>m^2</td>
</tr>
<tr>
<td>LH2 target - material</td>
<td>Al</td>
<td></td>
</tr>
</tbody>
</table>

Figure 1. Cross sectional sketch of the cryostat and the LH2 vessel. Around the LH2 vessel are several layers of superinsulation, also around the copper radiation shielding is an about 1-2 cm thick superinsulation layer. The function of the superinsulation is to prevent radiative and convection heat, and reduce conductive heat transfer to the liquid hydrogen, LH2. The continuously cooled Cu-radiation shielding establishes an intermittent temperature boundary at below100 K.

The function of the isolation vacuum is to provide a thermal isolation for the inner components of the cryostat such as the cryocoolers, LH2 target, and Cu-radiation shielding which is kept at below 100 K to shield the LH2 target from radiative heating.
The vacuum prevents heat transfer through conduction and convection but heat transported by radiation is still possible and therefore several layers of superinsulation have been wrapped on the Cu-radiation shielding and on the LH$_2$ target vessel to reduce radiative heating of the liquid hydrogen. On the target vessel are about 10 layers and around the Cu-shielding the thickness of the layer is about 1-2 cm.

When air spoils the cryostat vacuum of a cold cryostat, the air will first condense on the cold surfaces like the Cu-heat shielding, superinsulation, and the LH$_2$ vessel and effectively transporting heat to these surfaces through condensation which forms a solid layer of iced air, which, at a steady-state conditions, acts as a thermal resistance on the surface. The main heat transfer mechanism when superinsulation has been used, is conduction and to some extend convection. The high heat load to the LH$_2$ will produce a boiling film on the inner surface of the LH$_2$ vessel which will then transport heat to the liquid body in parallel with conduction. Also on the outside of the LH$_2$ vessel film boiling is possible.

The third possible reduction of the heat transfer through the aluminum wall into the liquid hydrogen is caused by the so-called boundary resistance which is an acoustic phonon mismatch between solid and liquid. Energy transfer between solid and liquid at low temperatures can occur only via phonon transmission and when temperature decreases (T<4 K) phonon spectra in solid and liquid become more and more different and thus reducing phonon scattering from one material to another. At 20 K, the thermal boundary resistance between the aluminum wall and liquid hydrogen does not play a significant role in the energy transportation and does not need to be considered.

The heat transfer mechanisms from the warm outer walls of the cryostat into the LH$_2$ are complicated and are difficult to calculate reliably [2]. However, in the literature several reports can be found where heat transfer rates are measured between two surfaces that are at different temperatures and when between the surfaces is heat transporting gas and/or isolating material. Most of the measurements have been performed using liquid helium as a cryogenic fluid, hydrogen measurements are sparse, however, the difference between liquid helium and liquid hydrogen results is not significant in our uncertainties.

At LANL in the design of the NPDGamma LH$_2$ target and its safety, the heat transfer into the LH$_2$ target in different accident scenarios was first estimated by calculating and extrapolating from available data [1,2]. When permission to operate the LH$_2$ target at LANSCE was obtained, these heat transfer estimates were verified by performing measurements where the isolation vacuum was partially filled with a He and Ar gas mixture. The results indicated that the originally selected heat transfer rate was an overestimate and thus provided to the target an extra safety.
1. **Estimated heat transfer based on some experimental results available in literature**

When the cryostat vacuum fails and air gets into vacuum, there are two main processes transferring heat into the LH$_2$. Condensation and freezing of air onto the outer surface of the LH$_2$ vessel will load the LH$_2$ with the enthalpy of condensation and fusion. The other heat transfer process is convection which is the main mechanism in transferring heat between the warm cryostat walls and the cold LH$_2$ vessel when the isolation vacuum is lost. If the vacuum is spoiled by H$_2$ or He gas, then the condensation does not take place (required temperatures are < 20 K or < 4 K, respectively). In both cases, the poor thermal conductivity of the superinsulation is the largest thermal resistance for the heat transport into the LH$_2$.

Table 2 lists some of reported results from the heat transfer rate measurements in failure of the isolation vacuum at different experimental configurations. Mainly these measurements have been performed when the cryogenic fluid is liquid helium, $T_2$=4 K, where the film boiling of LHe is slightly different than the film boiling of LH$_2$.

<table>
<thead>
<tr>
<th>No.</th>
<th>Condition; thickness of superinsulation and gas filling the isolation vacuum</th>
<th>$\dot{q}_{\text{eff}}$ (W/m$^2$)</th>
<th>Temperature of warm ($T_1$) and cold ($T_2$) surfaces [K]</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>No superinsulation, 1 atm air</td>
<td>2.7×10$^4$</td>
<td>$T_1$=300 $T_2$=78</td>
<td>[3]</td>
</tr>
<tr>
<td>2</td>
<td>No superinsulation, 1 atm H$_2$</td>
<td>2×10$^5$</td>
<td>$T_1$=300 $T_2$=20</td>
<td>[3]</td>
</tr>
<tr>
<td>3</td>
<td>No superinsulation, vacuum</td>
<td>600</td>
<td>$T_1$=300 $T_2$=20</td>
<td>[3]</td>
</tr>
<tr>
<td>4</td>
<td>No superinsulation, 1 atm air</td>
<td>(1-6)×10$^4$</td>
<td>$T_1$=300 $T_2$=4</td>
<td>[4]</td>
</tr>
<tr>
<td>5</td>
<td>1&quot; thick superinsulation, 1 atm air</td>
<td>1.3×10$^3$</td>
<td>$T_1$=300 $T_2$=4</td>
<td>[4]</td>
</tr>
<tr>
<td>6</td>
<td>3 mm superinsulation, 1 atm air</td>
<td>4.4×10$^3$</td>
<td>$T_1$=300 $T_2$=4</td>
<td>[5]</td>
</tr>
<tr>
<td>7</td>
<td>Thick layer of superinsulation, 1 atm Ar$^+$</td>
<td>4×10$^3$</td>
<td>$T_1$=300 $T_2$=78</td>
<td>[1]</td>
</tr>
<tr>
<td>8</td>
<td>Thick layer of superinsulation &lt;1 atm Ar/He mixture</td>
<td>600</td>
<td>$T_1$=300 $T_2$=20</td>
<td>[6]</td>
</tr>
</tbody>
</table>

(*) See section 3 below.

2. **Estimation of the heat transfer rate based on FLUENT simulations**

One of the goals of the NPDGamma liquid hydrogen target project has been to create a capability to perform verified simulations of heat transfer problems and other hydrogen target related thermodynamic problems such as dimensioning of the relief paths and devices and hydrogen combustion at different circumstances. We have adapted FLUENT a computational fluid dynamic (CFD) software package [7] to perform these simulations. The complexity of the problem makes it very hard to construct an enough representative
model of these processes in a rational time. However, a model that includes the cryostat, the LH$_2$ target, and the FNPB cave has been made and the following results were obtained from simulations.

For hydrogen boil-off three cases were simulated; a) the target vessel completely superinsulated with 25 layers, b) only 80% of the LH$_2$ vessel covered by the superinsulation, and c) the vessel completely without superinsulation and in all the cases the isolation vacuum is filled with either air or hydrogen.

Table 3. Heat transfer rates to the LH$_2$ per area unit when air or hydrogen filling the isolation vacuum.

<table>
<thead>
<tr>
<th>Condition</th>
<th>$\dot{q}_{\text{eff}}$ (W/m$^2$) with air</th>
<th>$\dot{q}_{\text{eff}}$ (W/m$^2$) with hydrogen</th>
</tr>
</thead>
<tbody>
<tr>
<td>No superinsulation</td>
<td>$1.6 \times 10^4$</td>
<td>$2.1 \times 10^4$</td>
</tr>
<tr>
<td>80% vessel covered*</td>
<td>$4.1 \times 10^3$</td>
<td>$5.1 \times 10^3$</td>
</tr>
<tr>
<td>25 layers of superinsulation</td>
<td>860</td>
<td>860</td>
</tr>
</tbody>
</table>

*) In calculation heat transfer rates from rows 1 and 3 have been used.

Table 4 lists the release mass flow rates and vaporization times for the 1.2 kg LH$_2$ target when the vessel is fully superinsulated with 25 layers, 20% of the vessel not covered, and with no superinsulation. In these cases the heat transfer to the LH$_2$ is a product of conduction, convection, and film boiling depending on the fraction of the vessel surface covered by superinsulation.

Table 4. Mass flow rates and emptying times for the 1.2 kg LH$_2$ target.

<table>
<thead>
<tr>
<th>Superinsulation</th>
<th>Air</th>
<th>Hydrogen</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$m$ (g/s)</td>
<td>$t$ (s)</td>
</tr>
<tr>
<td>25 layers of superinsulation</td>
<td>0.80</td>
<td>1503</td>
</tr>
<tr>
<td>80% of vessel covered</td>
<td>3.7</td>
<td>328</td>
</tr>
<tr>
<td>No superinsulation</td>
<td>15.3</td>
<td>79</td>
</tr>
</tbody>
</table>

In the FLUENT simulations the film boiling and convections on the walls, inside and outside, of the target vessel are included. Heat transfer rate via film boiling or convection is described by

$$\dot{q} = h(T_s - T_f)A,$$

where $h$ is the heat transfer coefficient for film boiling, $h_{fb}$ and for convection, $h_{cv}$, $T_s$ is the wall temperature and $T_f$ is temperature of the fluid, and $A$ is the surface area. Table 5 lists the heat transfer coefficients for film boiling and for convection when the fluid is either air or hydrogen.
Table 5. Heat transfer coefficients for film boiling and convection when the fluid is air or hydrogen.

<table>
<thead>
<tr>
<th>Heat transfer coefficient</th>
<th>( h ) (W/Km(^2)) with air</th>
<th>( h ) (W/Km(^2)) with hydrogen</th>
</tr>
</thead>
<tbody>
<tr>
<td>Film boiling - ( h_{fb} )</td>
<td>101.3</td>
<td>99.7</td>
</tr>
<tr>
<td>Convection - ( h_{cv} )</td>
<td>142.8</td>
<td>322.8</td>
</tr>
</tbody>
</table>

3. Estimation of the heat transfer based on field data with the NPDGamma hydrogen target

3.1 Field data

For the NPDGamma hydrogen target three different measured heat transfer results are available. In these measurements the isolation vacuum was filled with argon or with a 50/50 Ar-He gas mixture.

3.1.1 Boil off of LN\(_2\) when Ar gas was introduced into the isolation vacuum:
The LH\(_2\) vessel was filled with LN\(_2\) and then the isolation vacuum was filled with Ar gas and kept at pressure of 1 atm. This test is described in Ref. [1]. The effective heat transfer rate of \( \dot{q}_{\text{eff}} \approx 4200 \text{ W/m}^2 \) was obtained. The LN\(_2\) temperature is significantly higher than the LH\(_2\) temperature and therefore this heat transfer rate cannot be directly applied to consideration of the heat transfer into the LH\(_2\).

3.1.2 Intentional boil off of LH\(_2\) by filling isolation vacuum with 50/50 gas mixture of Ar and He:
After the NPDGamma LH\(_2\) target operation was approved at LANSCE, a permission was requested and granted to perform a test where the full LH\(_2\) target would be boiled off by filling the isolation vacuum with a 50/50 mixture of Ar and He gas. The goal of the measurement was to define the boil off time of the full target for emergencies where the target has to be emptied. The target emptying time was measured to be about 45 min when a STP volume of the Ar-He mixture was about the volume of the isolation vacuum. Ar-He pressure in the vacuum during the boil off was less then 1 atm caused by condensation of the Ar and reduced pressure of He by low temperatures. The release rate of H\(_2\) corresponds to the effective heat flow of \( \dot{q}_{\text{eff}} \approx 500 \text{ W/m}^2 \).

3.1.3 Inadvertent boil off of LH\(_2\) after accidental filling of the isolation vacuum with the Ar-He mixture:
One of the safety features of the NPDGamma target at LANSCE was that in a certain hydrogen target interlock event, the LH\(_2\) target was to be able to be emptied by dumping Ar-He gas mixture into the isolation vacuum, see section 3.1.2. Once during the three month long run at LANSCE, the interlock system was activated and the valve that was holding the Ar-He mixture was opened, the gas filled the isolation vacuum and boiled off
the liquid hydrogen target in about 40 min. This gives the effective heat transfer of

\[ \dot{q}_{\text{eff}} \approx 530 \text{ W/m}^2. \]

### 3.2 An estimate for heat transfer rate in the NPDGamma target after the vacuum and H\(_2\) boundaries fail

The experimental heat transfer results shown in Table 2 do not give directly the value of the heat transfer rate to the LH\(_2\) in the case of the NPDGamma target. The reported measurements have not been performed particularly in the configuration of the NPDGamma target. Also they do not describe enough details of the geometry and arrangement of the superinsulation and radiation shielding. According to Table 2 the most representative measurements with air that meet the conditions of the NPDGamma target are number 5) and 6). In the both cases the isolation vacuum is filled with air to 1 atm but in the both cases the cryogenic fluid was LHe. In measurement 6) the thickness of the superinsulation is only 3 mm which is very thin compared to the many mm thick superinsulation layer of the NPDGamma target. An extrapolation with some assumptions from these two measurements gives the range for the thermal conductivity in superinsulation of \( \kappa = 0.05 - 0.11 \text{ W/m} \cdot \text{K} \). On the other hand, the thermal conductivity of 30 layers of superinsulation immersed in gas is measured to be about \( \kappa = 0.07 \text{ W/m} \cdot \text{K} \) [8]. The average value of these thermal conductivities gives the effective heat transfer rate of \( \dot{q}_{\text{eff}} = 2000 \text{ W/m}^2 \) for the NPDGamma target where the thickness of the superinsulation is assumed to be 1 cm consisting of about 30 layers. The selected heat transfer rate of 2000 W/m\(^2\) can be considered as an upper limit since, for instance, in the corresponding safety calculations at Jefferson Lab the thermal conductivity of \( \kappa = 0.02 \text{ W/m} \cdot \text{K} \) for their superinsulation is used [9]. The FLUENT simulation result in Table 3 for air gives to the thermal conductivity in superinsulation of \( \kappa = 0.03 \text{ W/m} \cdot \text{K} \).

### 4. Hydrogen release rate into the BL-13 enclosure

In the previous section, the heat transfer rate to LH\(_2\) during the assumed multiple boundary failure accident is estimated to be \( \dot{q}_{\text{eff}} = 2000 \text{ W/m}^2 \).

Enthalpy of vaporization for hydrogen is \( h_v = 445.6 \text{ kJ/kg} \), density of LH\(_2\) at normal boiling point is \( \rho = 70.78 \text{ kg/m}^3 \), and mass of 16 liter of LH\(_2\) is then 1.14 kg \( \approx 1.2 \text{ kg} \). Amount of energy required to boil off the 16 liter of LH\(_2\) is

\[ Q = h_v m = 445.6 \frac{\text{kJ}}{\text{kg}} \times 1.2 \text{ kg} = 535 \text{ kJ}. \]

For the boil off time we get when the vessel surface area is 0.4 m\(^2\),

\[ \Delta t = \frac{Q}{\dot{q}_{\text{eff}} A} = \frac{\left(535 \times 10^3 \text{ J}\right)}{2000 \frac{\text{J}}{\text{s} \cdot \text{m}^2} \times 0.4 \text{ m}^2} = 668 \text{ s}. \]

For the mass flow rate out from the cryostat into the BL-13 cave we obtain
\[ \dot{m} = \frac{1.2 \text{ kg}}{668 \text{ s}} \approx 0.0018 \frac{\text{kg}}{\text{s}}. \]

This corresponds to a volumetric flow rate at STP of

\[ \dot{V} = \frac{14.3 \text{ m}^3}{668 \text{ s}} = 0.021 \frac{\text{m}^3}{\text{s}} \approx 21 \frac{\text{liters}}{\text{s}}. \]

REFERENCES


