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Design of the Cryogenic Hydrogen Release Laboratory

Ethan S. Hecht, Mark Zimmerman, A. Christine LaFleur, and Michael Ciotti

Prepared by
Sandia National Laboratories
Albuquerque, New Mexico 87185 and Livermore, California 94550

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Ethan S. Hecht and Mark Zimmerman
Combustion Research Facility
Sandia National Laboratories
P.O. Box 969
Livermore, CA 94551-9052
ehecht@sandia.gov, mdzimme@sandia.gov

A. Christine LaFleur
Sandia National Laboratories
Risk & Reliability Analysis
P.O. Box 5800
Albuquerque, NM 87185-0748
aclafle@sandia.gov

Michael Ciotti
H₂ Fueling Engineering
Linde LLC
575 Mountain Avenue
Murray Hill NJ 07974
michael.ciotti@linde.com

Abstract

A cooperative research and development agreement was made between Linde, LLC and Sandia to develop a plan for modifying the Turbulent Combustion Laboratory (TCL) with the necessary infrastructure to produce a cold (near liquid temperature) hydrogen jet. A three-stage heat exchanger will be used to cool gaseous hydrogen using liquid nitrogen, gaseous helium, and liquid helium. A cryogenic line from the heat exchanger into the lab will allow high-fidelity diagnostics already in place in the lab to be applied to cold hydrogen jets. Data from these experiments will be used to develop and validate models that inform codes and standards which specify protection criteria for unintended releases from liquid hydrogen storage, transport, and delivery infrastructure.

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1 Introduction

The Turbulent Combustion Laboratory (TCL) at Sandia National Laboratories in Livermore, CA, has been used to develop and validate models for high-pressure gaseous releases of hydrogen, utilizing high-fidelity diagnostics with well-controlled laboratory experiments [5, 8–10]. These physical models are used in quantitative risk assessment (QRA) models, and can be used to generate risk-informed separation distances [6]. Separation distances are specified in safety codes and standards, such as the *Hydrogen Technologies Code* (NFPA 2) from the National Fire Protection Agency (NFPA), that govern the allowable distances between hydrogen sources and other objects (e.g., flammable liquids, building openings or air intakes) [7]. Authorities having jurisdiction use these separation distances to qualify and approve designs that have hydrogen on site, such as a fueling station for hydrogen fuel cell vehicles. While the current requirements for bulk gaseous storage (862 bar, 100 kg) require at most 24 feet of separation (between the bulk storage and lot lines and building openings/air intakes), 75 ft of separation is required between a bulk liquid storage container (3,500-15,000 gallons) and a building opening or public assembly area [4, 7]. Discussions with members of the NFPA code committee have elucidated that the separation distances in NFPA 2 for liquid hydrogen are based on expert opinion rather than risk-informed, and may be overly conservative [2]. In this work, we describe our plans to modify the TCL to generate data, analogous to the data used to validate models for high-pressure gaseous releases of hydrogen, to validate (and develop, as necessary), models that can be used in QRA to revise the separation distances in the safety codes and standards that govern liquid hydrogen.

The cryogenic hydrogen release experiment in the TCL has been designed by staff at Sandia working with staff at Linde, through a cooperative research and development agreement (CRADA). This CRADA has allowed Linde, experts in working with cryogenic hydrogen, to assist in developing a TCL modification plan to ensure that these cryogenic hydrogen release experiments are performed safely and at relevant conditions. A weekly teleconference and several visits from Linde to Sandia facilitated discussions on design plans. This CRADA has led to a design plan that will lead to safe experiments that can generate important validation data for releases from liquid hydrogen systems.

2 Experimental Design

Liquid hydrogen tanks store hydrogen at fairly low pressure (< 10 bar), at the saturated liquid temperature (32 K at 10 bar), therefore the targeted experiments are designed for a maximum pressure of 10 bar and as close to the saturation temperature as possible. Control of the flows/pressure is an important consideration for experimental design. The design of the experiment in the TCL is very similar to an experiment that was run at the Karlsruhe Institute of Technology (KIT) [3], where room temperature hydrogen was controlled/monitored, followed by cooling in a series of heat exchangers. This allows the pressure and temperatures to be varied independently, rather than using liquid hydrogen directly. Unlike the KIT experiment, where the gaseous hydrogen was cooled by liquid hydrogen in the final stage, the experiment at Sandia will cool the atmospheric temperature hydrogen using liquid helium. The experiment we are installing also uses the cold gaseous effluent from the final stage to additionally cool the hydrogen. The KIT design only had two stages of cooling. A sketch of the experimental design is shown in Fig. 1.

As shown in Fig. 1, compressed hydrogen flows into the laboratory where flow control occurs. Flow control is achieved using a Tescom 44-3200 series pressure regulator, controlled electronically from the feedback of two pressure transducers flowing hydrogen across a critical flow orifice. The hydrogen then flows back outside the lab and into a three-stage heat exchanger. A long coil bathed in liquid nitrogen will cool the hydrogen to around 80 K. The cold hydrogen then flows through a counter-flow tube-in-tube heat exchanger where the hydrogen is further cooled to approximately 40 K using the cold helium gas. Finally, the cold hydrogen flows through a liquid helium bath where it is cooled to the saturation temperature (which is dependent on the experimental pressure, 32 K at 10 bar). This saturated hydrogen flows through a vacuum jacketed line into the laboratory. A silicon diode temperature sensor and a pressure tap near the exit will be used to monitor the exit gas conditions, and the hydrogen will be released in the lab through small orifices, around 1 mm diameter.

Schematically, the flow system is shown in Fig. 2. In stage 1, the temperature of the coolant (liquid nitrogen) can be considered a constant, and heat is extracted from the heat of vaporization of the fluid. Mathematically,

$$Q_1 = - \int_{300}^{T_{H_2,1}} \dot{m}_{H_2} c_{p,H_2} dT_{H_2} = h_{fg,N_2} \dot{m}_{N_2},$$

where Q is the heat flow, the hydrogen is assumed to be at 300 K initially, T is the temperature, \dot{m} is the mass flow rate, c_p is the heat capacity, and h_{fg} is the heat of vaporization. For stage 2, the heat balance can be written as

$$Q_2 = - \int_{T_{H_2,1}}^{T_{H_2,2}} \dot{m}_{H_2} c_{p,H_2} dT_{H_2} = \int_{T_{He,sat}}^{T_{He,exit}} \dot{m}_{He} c_{p,He} dT_{He}.$$

Stage 3 is similar to stage 1, and the heat balance can be written as

$$Q_3 = - \int_{T_{H_2,2}}^{T_{H_2,exit}} \dot{m}_{H_2} c_{p,H_2} dT_{H_2} = h_{fg,He} \dot{m}_{He}.$$

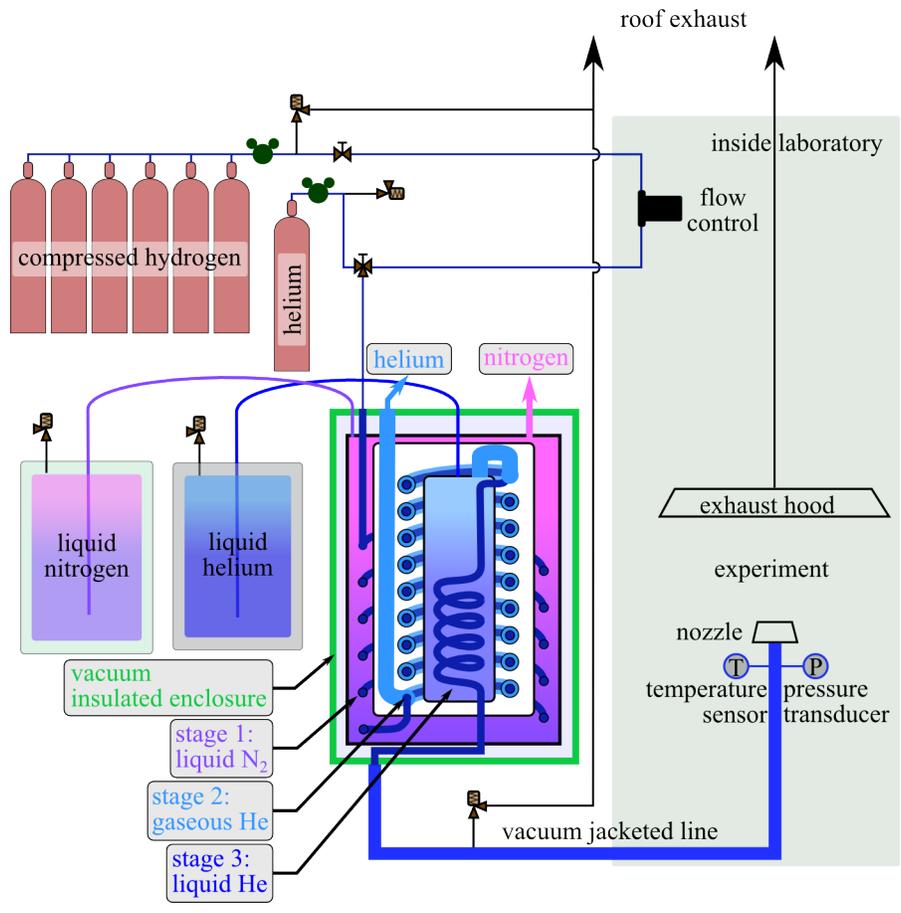


Figure 1. Sketch of the laboratory setup for the cryogenic hydrogen release experiments. Gaseous hydrogen is cooled in three stages—first using liquid nitrogen followed by gaseous helium, and finally by liquid helium.

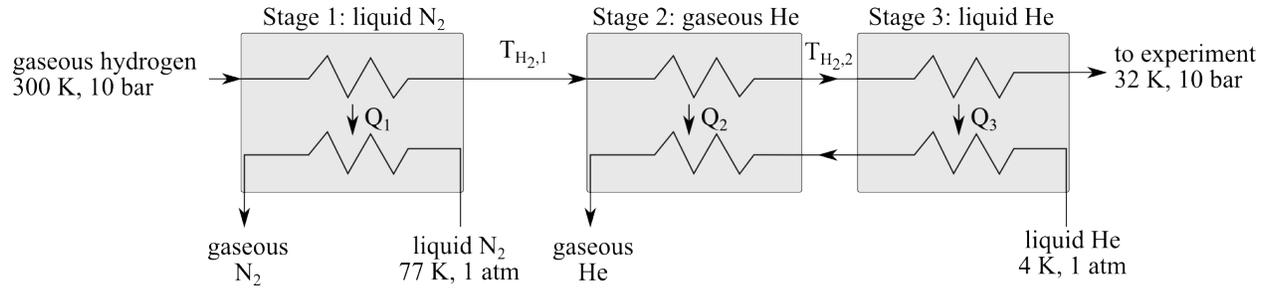


Figure 2. Schematic diagram of flows for three-stage heat exchanger.

Table 1. Normal (at atmospheric pressure) boiling and freezing points for select substances.

gas	boiling point (K)	freezing point (K)
oxygen	90.2	55
nitrogen	77.4	63
hydrogen	20.3	14
helium	4.2	N/A

Assuming a volumetric flow rate of 1000 SLPM hydrogen (or a mass flow rate of 1.5 g/s), using CoolProp [1] to calculate the heat capacity (which is a slight function of temperature), assuming that $T_{H_2,1}$ is 82.3 K (5 K above the liquid nitrogen saturation temperature) and $T_{H_2,exit}$ is 31.4 K (the saturation temperature), the first stage will exchange 4.3 kW of heat, and stages 2 and 3 will transfer 1.0 kW of heat. The heat of vaporization for liquid nitrogen is 199.1 kJ/kg, therefore, the utilization rate of liquid nitrogen will be 21.6 g/s, or 96 liquid liters/hr. The heat of vaporization for liquid helium is 20.6 kJ/kg. Assuming that the coolant helium gas is heated to the temperature of the liquid nitrogen by the end of stage 2 (77.3 K, or in other words, within 5 K of the hydrogen inlet temperature to stage 2), the utilization rate of helium will be 2.5 g/s, or 71 liquid liters/hr.

Although this heat exchanger is designed for liquid helium coolant, liquid hydrogen could also be used. In this case, the heat transferred will be equivalent in stages 2 and 3, but the utilization rate of liquid hydrogen would be slightly less than for helium, requiring only 1 g/s, or 48 liquid liters/hr. The availability and reduced safety concerns associated with liquid helium over liquid hydrogen caused us to design for helium as the coolant. However, should the costs for liquid helium increase significantly, or the availability be reduced, we can utilize liquid hydrogen in this heat exchanger.

As compared to the KIT design, the additional gas-gas stage 2 heat exchanger reduces the utilization rate of the liquid coolant significantly. For a given flow rate of gaseous hydrogen, helium utilization would be nearly 20 times greater without stage 2. If hydrogen were used as the coolant for stages 2 and 3, the utilization rate would still increase, but only by 2.4 times the utilization rate with stage 2. In addition, venting either helium or hydrogen at the low, saturation temperature that will be the exhaust of stage 3 (and the coolant exhaust, should stage 2 not exist) could lead to air freezing, since this exhaust would be below the freezing point of oxygen and nitrogen (see Table 1). Even with the current design, air condensation to liquid is possible, which poses a safety concern (albeit less than solidified air), since the boiling point of oxygen is higher than nitrogen, which causes an oxygen enriched fluid to form. For this reason, the heat exchanger will be located outside on inert concrete rather than asphalt, which can combust in an oxygen enriched environment. The 3-stage heat exchanger is also designed within a single vacuum insulated enclosure, with stage 3 located within stage 2, which is within stage 1. This minimizes the heat leakage and coolant waste heat flow to the environment.

Figure 1 also includes a three-way solenoid valve that can switch the supply gas from hydrogen to helium. This valve is included for safety reasons. Should hydrogen be detected by one of several

flammable gas sensors in the laboratory, this solenoid valve will switch, flowing helium rather than hydrogen through the heat exchanger, vacuum jacketed line, and nozzle. This will prevent the back-flow and freezing of air in the cold lines under an alarm condition.

Within the laboratory, similar experiments as have been performed on high-pressure gaseous hydrogen releases [5, 8–10] will be performed on these releases of cold hydrogen plumes. These include schlieren imaging to observe density gradients, planar laser Rayleigh scattering imaging to quantify hydrogen concentrations, and laser spark ignition to quantify the flammable boundaries of these cold jets. The diagnostics are already in place in the laboratory, although there will be additional challenges for quantifying the Rayleigh scattering images, associated with the combined temperature and concentration gradients of these cold jets.

3 Conclusion

The Office of Energy Efficiency and Renewable Energy, Fossil Energy, and the Office of Science are working to lower the cost and energy use of the hydrogen delivery infrastructure. Results from the cryogenic hydrogen experiments, made possible by the laboratory modifications described in this report, will inform revisions to codes and standards associated with siting cryogenic hydrogen at fueling stations. This should enable cryogenic hydrogen to be located at more hydrogen refueling sites, leading to lowered costs due to the economic benefits of cryogenic storage over compressed gas storage. These codes and standards revisions will also help companies, such as Linde, to build more refueling stations that store hydrogen as a liquid, their preferred and a more economical approach. The science-based codes and standards revisions will allow consumers to have a safe and positive experience refueling their hydrogen fuel cell vehicles.

References

- [1] I. H. Bell, J. Wronski, S. Quoilin, and V. Lemort. “Pure and pseudo-pure fluid thermophysical property evaluation and the open-source thermophysical property library coolprop”. *Ind. Eng. Chem. Res.* 53 (2014), 2498–2508.
- [2] I. W. Ekoto, E. S. Hecht, C. San Marchi, K. M. Groth, A. C. Lafleur, N. Natesan, M. Ciotti, and A. Harris. *Liquid Hydrogen Release and Behavior Modeling : State-of-the-Art Knowledge Gaps and Research Needs for Refueling Infrastructure Safety*. Tech. rep. SAND2014-18776. Sandia National Laboratories, 2014.
- [3] A. Friedrich, W. Breitung, G. Stern, A. Vesper, M. Kuznetsov, G. Fast, B. Oechsler, N. Kotchourko, T. Jordan, J. Travis, J. Xiao, M. Schwall, and M. Rottenecker. “Ignition and heat radiation of cryogenic hydrogen jets”. *Int. J. Hydrogen Energy* 37 (2012), 17589–17598.
- [4] A. P. Harris, D. E. Dedrick, C. Lafleur, and C. S. Marchi. *Safety , Codes and Standards for Hydrogen Installations : Hydrogen Fueling System Footprint Metric Development*. Tech. rep. SAND2014-3416. Sandia National Laboratories, 2014.
- [5] W. G. Houf and R. W. Schefer. “Analytical and experimental investigation of small-scale unintended releases of hydrogen”. *Int. J. Hydrogen Energy* 33 (2008), 1435–1444.
- [6] J. LaChance. “Risk-informed separation distances for hydrogen refueling stations”. *Int. J. Hydrogen Energy* 34 (2009), 5838–5845.
- [7] *NFPA 2: Hydrogen technologies code*. Tech. rep. NFPA 2. National Fire Protection Agency, 2011.
- [8] A. J. Ruggles and I. W. Ekoto. “Experimental investigation of nozzle aspect ratio effects on underexpanded hydrogen jet release characteristics”. *Int. J. Hydrogen Energy* 39 (2014), 20331–20338.
- [9] A. J. Ruggles and I. W. Ekoto. “Ignitability and mixing of underexpanded hydrogen jets”. *Int. J. Hydrogen Energy* 37 (2012), 17549–17560.
- [10] R. Schefer, W. Houf, and T. Williams. “Investigation of small-scale unintended releases of hydrogen: momentum-dominated regime”. *Int. J. Hydrogen Energy* 33 (2008), 6373–6384.

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1 Michael Ciotti
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