VIII.4 R&D for Safety, Codes and Standards: Hydrogen Behavior

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Project Start Date: October 1, 2003
Project End Date: Project continuation and direction determined annually by DOE

Overall Objectives

- Develop a science and engineering basis for the release, ignition, and combustion behavior of hydrogen across its range of use (including high pressure and cryogenic).
- Create models and engineering tools that enable the assessment of the safety (risk) of hydrogen systems, the revision of regulations, codes, and standards, and the permitting hydrogen fueling stations.

Fiscal Year (FY) 2017 Objectives

- Measure cryogenic hydrogen dispersion (hydrogen at <50 K) in lab-scale experiments with precise control of boundary conditions using high-fidelity imaging diagnostics. Use this data to validate the COLDPlume model for cryogenic hydrogen dispersion that will be used to provide scientific basis for safety distances for liquefied hydrogen systems.
- Design large-scale experiments to enable validated modeling of pooling and vaporization of liquid hydrogen.

Technical Barriers

This project addresses the following technical barriers from the Hydrogen Safety, Codes and Standards section of the Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan.

(A) Safety Data and Information: Limited Access and Availability

(G) Insufficient Technical Data to Revise Standards

Contribution to Achievement of DOE Safety, Codes & Standards Milestones

This project will contribute to achievement of the following DOE milestones from the Hydrogen Safety Codes and Standards section of the Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan.

- Milestone 2.13: Develop and validate simplified predictive engineering models of hydrogen dispersion and ignition. (4Q, 2015)
- Milestone 2.19: Validate inherently safe design for hydrogen fueling infrastructure. (4Q, 2019)
- Milestone 4.8: Revision of NFPA 2 to incorporate advanced fueling and storage systems and specific requirements for infrastructure elements such as garages and vehicle maintenance facilities. (3Q, 2016)

FY 2017 Accomplishments

- Developed a novel diagnostic to simultaneously measure cryogenic hydrogen concentration and mixture temperature in two dimensions, providing high-quality data for model validation.
- Measured the dispersion and warming characteristics for a range of hydrogen releases, including some with a source temperature below 50 K, extending the hydrogen dispersion validation data set to cryogenic sources.
- Initiated the validation process by comparing the concentration and temperature data to an existing model of cold hydrogen dispersion that will be used for risk assessment and to provide the scientific basis for risk-informed safety distances, with initial results showing reasonable agreement between the experimental results and the model simulations.

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INTRODUCTION

Fire codes govern the required distances between hydrogen sources (e.g., a liquid hydrogen tank at a fueling station) and hazards (e.g., ignition sources). Revisions to the fire code distances require justification, which is facilitated by models. These models must be validated with carefully controlled experiments, under relevant conditions, which can include high pressures (10,000 psi) or cryogenic temperatures (20 K). Over the course of this project, experiments have been designed and run to provide validation data for models. Models have been developed and exercised to inform the fire codes. This work has enabled quantitative risk assessments of hydrogen systems, and subsequent reduction of setback distances from high pressure hydrogen sources. Currently, we are focusing on developing a scientific basis for modeling dispersion and flames from cryogenic (liquid) hydrogen sources. Validated models from this project will be exercised to provide a technical basis for the revision of fire codes related to liquid hydrogen.

APPROACH

The goals of this work are to develop and validate scientific models to accurately predict hazards and harm from unintentional hydrogen releases. In this project, we previously developed one-dimensional and engineering models of hydrogen dispersion and flames that can run quickly on a personal computer. While these models are one-dimensional, they include enough physics (e.g., the effect of buoyancy) to be accurate under a wide range of scenarios. These models are able to characterize the hazards from hydrogen releases and flames and are fast enough that they can be run multiple times and incorporated into a quantitative risk assessment framework. While the models we used to inform fire codes have been validated, carefully controlled experiments are required to validate and develop new models for cryogenic hydrogen to have an impact on liquid hydrogen separation distances. Advanced optical and laser diagnostics are used, along with more conventional diagnostics (e.g., thermocouples) to characterize the dispersion and flame properties of releases, at a lab scale. The temperature, pressure, and orifice of the unignited releases and flames are controlled while characteristics are measured (e.g., concentration, flame temperature, radiative heat flux).

RESULTS

The cryogenic hydrogen release laboratory was constructed and commissioned last fiscal year. This fiscal year, the laboratory was used to measure the concentration and temperature fields of a range of cryogenic hydrogen releases. We have performed a comparison of this data to a previously developed model for the dispersion of cryogenic hydrogen. We are planning to gather more data through the end of the fiscal year. By the end of the fiscal year, we will conclude the required development and validation of this model.

One accomplishment this fiscal year was developing and implementing the diagnostic that could be used to measure the concentration and temperature in two dimensions. Previous work in the laboratory with room temperature releases of hydrogen used planar laser Rayleigh scattering to measure the concentration of hydrogen. However, cryogenic hydrogen entrains humid air and the water vapor from the air condenses in the ultra-cold flow. The scattered light from the water vapor scatters light much more intensely than the Rayleigh scatter from the gas molecules, saturating the cameras and precluding this method as a diagnostic. After unsuccessful attempts to implement filtered Rayleigh scattering to reduce the light scattered off of the water vapor, we were successful at imaging planar laser Raman scattered light with the experimental setup shown in Figure 1. This technique takes advantage of the known large Raman wavelength shift of light scattered off of different gas molecules. In the laboratory setup, we measure Raman scattering off of both hydrogen and nitrogen. The 532-nm wavelength laser light is shifted to 607 nm when the light scatters off of nitrogen molecules, and 683 nm when it scatters off of hydrogen molecules. We use two scientific cameras, each with several bandpass filters and a notch filter at 532 nm to suppress the light by a factor of 18 at the laser wavelength (532 nm), and a factor of at least 12 for all wavelengths except for a 10-nm wide passband near the wavelength of interest (the passband for nitrogen is centered at 610 nm and the passband for hydrogen is centered at 685 nm). By simultaneously measuring light scattered off of nitrogen and hydrogen, we are able to determine the instantaneous mole fraction of hydrogen, mole fraction of nitrogen, and temperature in the two-dimensional plane of the laser sheet. Much like previous experiments in this laboratory, we keep the laser and cameras fixed and move the release point to gather statistics on how cryogenic hydrogen disperses and warms at different distances from the release point.

A model, known as ColdPLUME, that was developed at Sandia [1,2], has shown reasonable agreement with some limited centerline concentration measurements of hydrogen cooled to liquid nitrogen temperatures (80 K). ColdPLUME includes equations to describe accelerating flow through the leak, the expansion of an under-expanded jet, initial

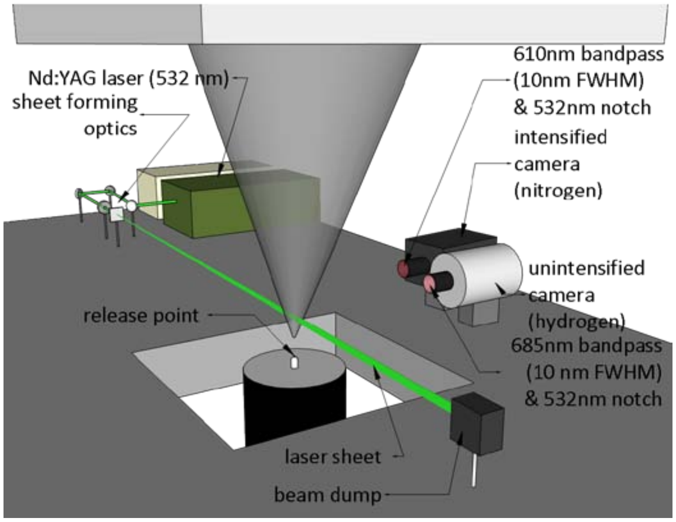

FIGURE 1. Diagnostic setup of experiment. A frequency doubled neodymium-doped yttrium aluminum garnet laser is formed into a sheet, and Raman signals at two specific wavelengths are collected simultaneously by two cameras.

entrainment and heating, establishment into a turbulent plume, followed by a one-dimensional set of ordinary differential equations that conserve mass, momentum and energy along the streamline of that turbulent plume. Although the model is one-dimensional along the streamline, buoyancy is taken into account, and the plume can rise or fall, depending on the density of the plume along the streamline relative to the surrounding air. The model relies on the assumption that within the established flow, the mean velocity and density profiles (looking radially) are Gaussian shaped, as has been shown previously for turbulent plumes. That these profiles are Gaussian for cryogenic hydrogen has been confirmed by the experiments performed this fiscal year. This can be seen in Figure 2, where the normalized distribution of mass fraction and temperature both align well with a Gaussian curve at all distances downstream. In addition to the experimental data points, Figure 2 shows two Gaussian distributions: a curve fit to the data by the thick red line, and the previously measured distribution of mass fraction of room temperature hydrogen and other gases by the thin black line [3,4]. The fit of the mass fraction data for this particular release is quite close to the literature fit for room temperature gases, although the fit to the data for all nine of the releases performed thus far of cryogenic hydrogen is a little further from this particular fit. This may be due to experimental noise, or could suggest that some of the empirical parameters in the model, such as the entrainment rate of air, may need to be modified for these cryogenic hydrogen releases as compared to room temperature hydrogen releases. The data also shows that the normalized temperature distribution is Gaussian, for which there is no literature data, due to the fact that previous measurements were all on room temperature releases and there was no variation in temperature across the plume. The temperature data is noisier than the mass fraction data, but the data suggests that the distribution is wider for temperature than for mass fraction.

Figure 3 shows the mean mole fraction and temperature fields observed experimentally by the thick dashed lines and shading, and those predicted by ColdPLUME by the thin solid lines for one of the release experiments. The model is doing a good job of predicting the mole fraction contours for this data set. In terms of temperature, the model seems to be a bit further off, predicting cold temperatures along the centerline that penetrate further downstream than the data would suggest, although the data is noisier in this case. This figure demonstrates the amount of data generated by the planar laser Raman imaging—two-dimensional measurements of concentration and temperature—is ideal for validating a model that is generating two-dimensional predictions of concentration and temperature. This data is superior to sparse centerline measurements of concentration or temperature only. We extracted the centerline inverse mass fraction from our experiments and found that the decay rate scaled linearly with the normalized (by the effective diameter) distance from the release, and was quite close to the inverse mass fraction decay rate of room temperature hydrogen and other gases.

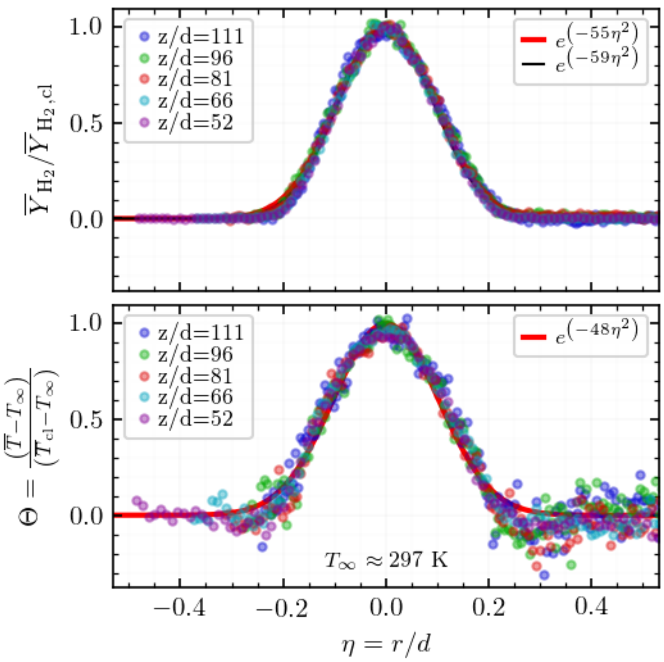

FIGURE 2. Radial mass fractions at selected distances, normalized by the centerline mass fraction (top) and radial temperatures at selected distances, normalized by the temperature excursion from the atmosphere (bottom). Data is shown by the points; the red line shows the fit to the data, and the black line shows the expected distribution for atmospheric temperature gases [3,4].

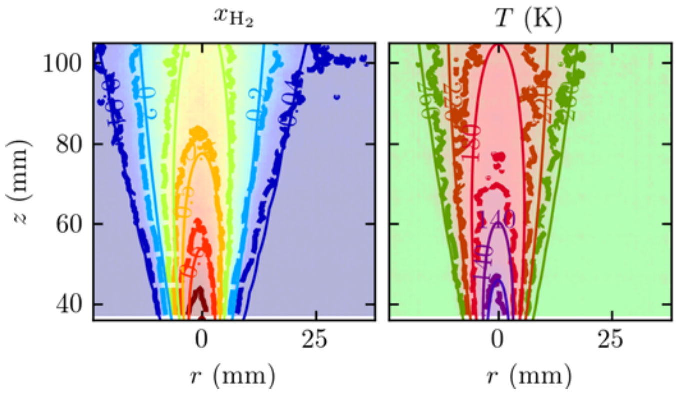

FIGURE 3. Comparison of the model predictions shown by the solid, thin lines, to the experimental data, shown by the thick, dashed lines and shading for mole fraction (left) and temperature (right).

CONCLUSIONS AND UPCOMING ACTIVITIES

We have successfully measured the concentration and temperature fields for a range of cryogenic hydrogen releases. This data is being compared to a previously developed model, which will be subsequently validated, or modified to be valid to predict the flow field of cryogenic hydrogen. Through the end of this fiscal year, we will be performing more experiments to increase the range of known model validity and conclude model development and validation activities.

Other fundamental aspects of liquid hydrogen releases, such as the pooling and evaporation from liquid hydrogen pools are poorly understood and modeling capabilities are limited or nonexistent. This fiscal year, we are also planning additional research and development looking at these aspects of liquid hydrogen. Developing a diagnostic that can be used on a larger release that would actually pool and evaporate is one thrust of the work this fiscal year. Developing the experimental platform on which to use the diagnostic will occur next fiscal year.

FY 2017 PUBLICATIONS/PRESENTATIONS

1. C. San Marchi, E.S. Hecht, I.W. Ekoto, K.M. Groth, C. LaFleur, B.P. Somerday, R. Mukundan, T. Rockward, J. Keller & C.W. James, "Overview of the DOE hydrogen safety, codes and standards program, part 3: Advances in Research and Development to Enhance the Scientific Basis for Hydrogen regulations, Codes and Standards," *International Journal of Hydrogen Energy* 42 (2017): 7263–7274.

2. K.M. Groth & E.S. Hecht, "HyRAM: A methodology and toolkit for Quantitative Risk Assessment of hydrogen systems," *International Journal of Hydrogen Energy* 42 (2017): 7485–7493.

3. P.P. Panda & E.S. Hecht, "Ignition and flame characteristics of cryogenic hydrogen releases," *International Journal of Hydrogen Energy* 62 (2017): 775–785.

4. E.S. Hecht & P. Panda. "Liquid Hydrogen Behavior Studies," Presented to the LH2 Separation Distance Technical Panel, June 2016, (SAND2016-6149 PE).

5. E.S. Hecht. "Cryogenic Hydrogen Plume Behavior," Prepared for the HySAFE Research Priorites Workshop, Petten, The Netherlands, September 2016, (SAND2016-9482 C).

6. E.S. Hecht. "Accidental Hydrogen Ignition," Prepared for the HySAFE Research Priorites Workshop, Petten, The Netherlands, September 2016, (SAND2016-9481 C).

7. E.S. Hecht. "Non-Premixed Hydrogen Combustion," Prepared for the HySAFE Research Priorites Workshop, Petten, The Netherlands, September 2016, (SAND2016-9480 C).

8. E.S. Hecht, P. Panda. "Validation data for cryogenic hydrogen releases and flames." Presented to the Hydrogen Codes and Standards Tech Team, October 2016, (SAND2016-10307 PE).

9. E.S. Hecht, P. Panda. "Validation data for cryogenic hydrogen releases and flames," Presented to the LH2 Separation Distance Stakeholders, November 2016, (SAND2016-11548 PE).

10. E.S. Hecht. "Validation data for cryogenic hydrogen releases and flames," Presented to the NFPA 2 task group, February 2017, SAND2017-2308 PE.

11. E.S. Hecht, P. Panda "Mixing and Warming of Cryogenic Hydrogen Releases," Accepted for the International Conference on Hydrogen Safety, Hamburg, Germany, September 2017. (SAND2017-7481 C).

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3. C.D. Richards, W.M. Pitts, "Global density effects on the self-preservation behavior of turbulent free jets," *Journal of Fluid Mechanics* 254 (1993): 417–435.

4. A.J. Ruggles, "Statistically advanced, self-similar, radial probability density functions of atmospheric and under-expanded hydrogen jets," *Experiments in Fluids* 56 (2015): 202.