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Technical Notes

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Hydrogen–Helium Leak Detection at Elevated Pressures and Low Temperatures

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Nomenclature

A_{orifice}	=	effective cross-sectional area of the leak source
DC	=	discharge coefficient
Kn	=	Knudsen number, λ/L
L	=	characteristic length
$\dot{m}$	=	mass flow rate
p	=	static pressure
p_t	=	stagnation pressure
R	=	specific gas constant
T_t	=	stagnation temperature
γ	=	ratio of specific heats, c_p/c_v
λ	=	mean free path
ρ	=	gas density

I. Introduction

HYDROGEN is widely used as a primary fuel source for rocket engines due to its beneficial combustion properties. Compared with hydrocarbon reactions, hydrogen–oxygen combustion produces high engine specific impulse. Hydrogen is also readily storable in the condensed phase, yet its low flammability limit and low ignition energy make hydrogen potentially explosive when leaked into an oxidizing environment. Because of this potential hazard, the development of accurate hydrogen leak-detection techniques is critical for rocket engine testing and operation, in addition to the development of hydrogen fuel cell and other hydrogen-based energy systems.

The helium signature test (HST) was developed by NASA as a systematic leak-detection method for cryogenic hydrogen fuel tanks to ascertain their safety [1]. The HST procedure described here is based on the discussion by Hass et al. [2]. The test involves pressurizing the system in question with an inert gas (helium) at relatively low pressures and room temperature to obtain mass flow rates that arise due to leakage. With the measured helium leak-rate data and the assumptions of ideal-gas behavior and choked orifice flow, hydrogen leak rates from the system at high pressures and low temperatures can be extrapolated. This procedure has been used in NASA X-33 propulsion hazard mitigation tests, for example [3]. Exploration of the HST is the focus of the present study.

II. Background on the Helium Signature Test

The HST experimental procedure has two steps to obtain helium leak rates and involves a pressurized helium tank and propellant feed system contained within a chamber into which nitrogen can be injected. First, known flow rates of helium and nitrogen are inserted into the purged system, and calibration curves relating helium concentrations and the response of the mass spectrometer are determined. Second, a backpressure of helium from the tank is applied to the feed system and the mass spectrometer measures the altered helium concentration due to leakage in the chamber environment. Using the helium concentration data and the calibration curves, the helium leak rate can be calculated for a range of backpressures, which are much lower than the typical operating pressures for hydrogen storage tanks. Using data gathered in these two steps, a response curve is generated for the helium mass flow rate due to leakage for given operational pressures.

From the measured helium leak rate, the cross-sectional area of the leak orifice, A_{orifice}, can be calculated from an isentropic flow relation that assumes a calorically perfect gas and choked flow at the leak source:

$$A_{\text{orifice}} = \dot{m}\sqrt{T_t} \bigg/ \left[\mathrm{DC}\left(\frac{\gamma}{R}\right)^{\frac{1}{2}}\left(\frac{2}{\gamma+1}\right)^{\frac{\gamma+1}{2(\gamma-1)}} p_t\right] \tag{1}$$

where $\dot{m}$ represents the mass flow rate of gas exiting from the pressurized tank through the leak orifice, γ is the ratio of specific heats, and T_t and p_t represent the stagnation temperature and pressure, respectively, within the tank upstream of the leak orifice. Consistent with the assumption of isentropic flow, the discharge coefficient DC is assumed to be unity in the application of the standard HST. Once the orifice area has been estimated for the helium leak rate using the properties of helium in Eq. (1), the hydrogen leak rate in an equivalent system can be predicted. Equation (1) is used once more, this time with hydrogen gas properties, to calculate the predicted hydrogen mass flow rate associated with the same orifice area A_{orifice}, but at a much higher equivalent tank pressure p_t. Although the fundamental theory behind the HST technique is straightforward, the physics of the flow through the leak sources may not adhere to the assumptions. A discharge coefficient less than unity is required to incorporate dissipative effects, and different orifice shapes will typically have different effective DC values.

Previous studies by our group [4,5] on helium and hydrogen leak detection sought to answer questions pertaining to the flow regime, the choked-flow condition, the discharge coefficients, and the validity of the HST by performing direct mass-flow-rate comparisons. Micro orifices of known shapes and sizes were fabricated out of silicon to serve as leak sources for gaseous helium and hydrogen from high-pressure storage tanks. Tank pressures of up to 13.65 atm were applied to the orifices at room temperature. For

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these conditions, it was determined that the Knudsen number associated with orifice flow was, at most, on the order of 0.006, so that the flow through the micro orifices remained in the continuum regime.

To determine the actual discharge coefficient from Eq. (1) in these previous studies, it had to be verified that the flow through the micro orifice was choked. There is a critical value of the ratio of the orifice (or downstream) pressure p to the tank (or upstream) pressure p_t, below which the mass flow rate approaches a choked condition, so that the DC asymptotes to a constant value. By systematically increasing the tank pressure for either helium or hydrogen and a given orifice, the pressure ratio could be decreased below the critical values, so that plots of DC vs p/p_t were produced. Then using the HST procedure, comparisons were made in our previous studies [4,5] between actual measured hydrogen leak rates at 13.65 atm and those predicted from lower-pressure (6.85 atm) helium leak-rate measurements. In general, the HST was seen to underpredict hydrogen leak rates by as much as 27%. When the HST is modified to use a higher helium stagnation pressure, however (13.65 atm, the same as the pressure of the desired hydrogen leak rate), this error was reduced to a maximum 6% underprediction.

Because the actual HST procedure in [1] is usually performed with a much lower helium stagnation pressure than the pressure for which the hydrogen leak rate is sought, it is possible that errors in the standard HST could be much higher. Our previous studies [4,5] were all performed at room temperature and at a highest upstream pressure of 13.65 atm, but practical systems store hydrogen fuel at very high pressures, on the order of hundreds of atmospheres, near cryogenic temperatures. The aim of the present study is to extend the investigation of the HST to higher upstream pressures ($\sim$100 atm) and lower temperatures ($\sim$195 K) and to estimate its validity at these somewhat-more-realistic conditions. Because the previous studies discovered that the prediction of hydrogen leak rates could be improved by using helium leak-rate data gathered at higher pressures, it is also of interest to determine how high the helium stagnation pressure should be raised in the HST procedure, at either room temperature or a lower temperature, to obtain a relatively accurate estimation of the hydrogen leak rate. Using higher upstream pressure conditions, this question can be answered using measured helium leak-rate data. The findings will determine at what pressure the HST should be performed to obtain an accurate hydrogen leak-rate prediction from helium leak-rate data.

III. Experimental Configuration

Details on the system configuration for the present experiments are given by Lee [4], Lee et al. [5], and Mak [6]. In these experiments, high-pressure gas (either helium or hydrogen) was stored in tanks and regulated up to 1600 psig (109 atm). The tanks were connected to a holder in which a silicon wafer, with a hole or orifice of known shape and cross-sectional area, was located. Helium or hydrogen leaked through the orifice at rates that depended on its geometry and the difference between the tank pressure p_t and the line flow pressure downstream of the orifice (p). The downstream line pressure was established by a nitrogen carrier gas and was set at 30 psig (2.04 atm) using a manual shutoff valve as well as a pressure relief valve. The carrier gas was stored in a separate tank and its pressure was regulated to about 100 psig (6.80 atm) to maintain a steady flow in the downstream line. Helium or hydrogen leaking through the orifice was mixed with the nitrogen carrier gas downstream of the silicon wafer. This mixture or sample gas flowed to a dual thermal conductivity detector (TCD) (SRI model 100), in which it was compared with a stream of pure nitrogen as the reference gas. By comparing thermal conductivities of the sample gas and the reference gas, the TCD measured the concentration of either helium or hydrogen in the sample gas mixture. As discussed in [4–6], a linear fit for the TCD reading with respect to hydrogen and helium concentrations was used, with a bias error below 5%. The leak rates of either helium or hydrogen through the orifice were calculated from the measured concentration data.

The TCD had an uncertainty of 100 ppm. For the TCD to accurately compare the sample gas and reference gas, two flow controllers were installed in the system to maintain the flow rates of these two streams. The flow rate of the nitrogen carrier gas was measured by one of two flow meters: a Tylan FC260 flow meter/controller, which was capable of measuring flow rates up to 7.5 slpm, and a Sierra Instruments 826 flow meter, with a factory-calibrated range of up to 100 slpm. Errors in the volumetric flow rates were 0.1% of full scale. A specially designed flange was used to hold the micromachined silicon orifices at the measuring position. Ideally, leakage of helium and hydrogen should only occur through the prescribed orifice, and so the purpose of the flange was to eliminate leakage both around the wafer and to the atmosphere. Pressure was measured upstream of the micro orifice using a gauge connected to the helium or hydrogen gas regulator and with another gauge downstream of the micro orifice in which the leakage mixed with the carrier gas. These pressure measurements were used to analyze the leakage concentration data measured by the TCD. Bias errors in the pressure measurements were a maximum of 3% for the range of conditions explored.

It was of particular interest to examine hydrogen leak rates at temperatures much lower than room temperature. To lower the temperature of the sample gas and the leak orifice, a bath of dry ice and acetone in an insulated container was used to submerge the flange and its connected pipes. To ensure that an equilibrium temperature was reached, the apparatus remained immersed for at least 30 min before the experimental procedure was initiated.

The leak orifices of prescribed shapes and cross-sectional areas were manufactured from silicon wafers using a slight modification of the standard lithography method, followed by the deep reactive ion etching (DRIE) technique. The goal of the microfabrication process was to produce the smallest possible bore dimension on the thickest possible silicon wafer to maintain structural integrity when exposed to normal and shear stresses. An orifice with these attributes will have a high aspect ratio, which is defined as the etched hole depth to the width or other characteristic length scale. To produce these orifices, very-high-aspect-ratio etching of the silicon wafers was necessary. This phase of the microfabrication process presented some difficulties, and slight modifications to the standard procedures (noted subsequently) were required to achieve the state-of-the-art limits for DRIE. Details on the complete photolithography and DRIE processes used here may be found in [4,5]. A range of orifice shapes (circular and elliptical or slit) and sizes could be created via this approach.

In most cases, the leak sources were created by etching completely through a 250 μm wafer. When fabricating the smallest micro orifices, for orifices with a diameter or smallest dimension of 15 μm or lower, aspect-ratio nonuniformities were possible because the DRIE etching rate decreases with the depth of the etch, and the etch aspect ratio was higher than the equipment could achieve. This problem was solved by performing double-sided etching of the smaller orifices, in which the photolithography and DRIE process were applied to both sides of the silicon wafer, as described in [4,5]. On one side of the wafer, for example, a 10-μm-diam circular orifice was etched for a depth of 70 μm, whereas on the other side of the wafer, a 500-μm-diam circular orifice was etched for a depth of 180 μm. This created a micro orifice with a diameter of 10 μm and an effective orifice thickness of 70 μm. Sample photographs of an elliptical or slit orifice fabricated using the double-sided etching technique are shown in Figs. 1a and 1b, taken using an optical microscope with a 40$\times$ magnification. As a result of the double-sided etching technique, relatively uniform bore shapes were observed in both top and bottom views of the orifices.

IV. Results and Discussion

Using the experimental procedure described previously for helium and hydrogen leak detection, mass flow rates were measured by the TCD for the higher pressures and lower temperatures examined in this study. The TCD measurements allowed for the calculation of the mass flow rates $\dot{m}_{\mathrm{He,actual}}$ and $\dot{m}_{\mathrm{H_2,actual}}$ for a range of tank pressures,

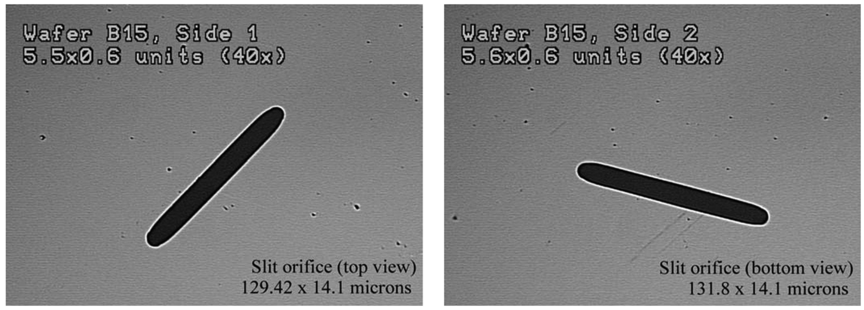


Fig. 1 Optical microscope images of a sample micro orifice etched using the double-sided etching technique. Shown are both sides of a slit or elliptical orifice with an effective hydraulic diameter of 31.8 μm (with major and minor axes as indicated).

which are equivalent to the leak rates of helium and hydrogen through the micro orifices. From these mass flow rates, Eq. (1) can be used to calculate the discharge coefficient DC associated with the various flow conditions. Calculated bias errors in mass flow rates were determined to be a maximum of 4% for the Sierra Instruments flow meter and a maximum of 3% for the Tylan flow meter.

In these experiments, the upstream stagnation pressure p_t was read from the regulator pressure gauge and the area of the orifices A_{orifice} was found by measuring them under a microscope. For the low-temperature tests, the stagnation temperature T_t was equal to $-109°$F (195 K), determined from the temperature of the dry ice and acetone bath within an accuracy of 1%. From the flow conditions investigated in the present study, it was determined that the Knudsen number was, at most, 0.0022. This value again verified that the compressible flow through the micro orifices remained in the continuum regime.

The choked-flow condition in the micro orifices is an assumption of the HST and must be verified so that the DC can be calculated using Eq. (1). Several orifices of different sizes and shapes were used to verify the choked condition for both room temperature and for the dry ice and acetone bath temperature and for pressures up to 1450 psi (98 atm). By systematically increasing the tank pressure for either helium or hydrogen, the pressure ratio p/p_t was decreased below the critical values (0.487 for helium and 0.528 for hydrogen), and the DC could be plotted as a function of pressure ratio.

An example of the behavior of the discharge coefficient as a function of pressure ratio for a circular orifice labeled $1F$ (with a diameter of 57 μm) is shown in Fig. 2. For most conditions, the DC gradually increased and asymptoted as p/p_t was reduced, but in the case of hydrogen at room temperature, the DC began to decrease slightly with decreasing pressure ratio. This slight drop in DC at very high upstream pressures was also seen for certain orifices at low values of p/p_t in our previous studies [4,5]. This observation is consistent with Jackson's findings [7] indicating that flows through knife-edged or very thin orifices may not reach choked conditions under the theoretical critical pressure conditions.

The HST method was then tested. Measured hydrogen leak rates at high pressure ($p_{t,\text{high}} \sim 100$ atm) and either low temperature ($T_t \sim 195$ K) or room temperature ($T_t \sim 295$ K) could be compared with predicted hydrogen leaks rates derived from room temperature helium at lower pressures. Figure 3a, for example, shows the ratio of the actual hydrogen leak rate at 98 atm and at two different temperatures to the predicted hydrogen leak rate for the slit orifice $3D$ (with a hydraulic diameter of 15.8 μm). The predicted hydrogen leak rate is derived from helium leakage at room temperature and at a range of pressures, plotted on the abscissa. Figure 3b shows the same plot but for the prediction of hydrogen leak rates at room temperature for the circular orifice $1F$ (with a diameter of 57 μm). If the HST were to work perfectly, the value of this ratio would be unity for the range of upstream helium pressures considered. Figure 3a shows that the actual hydrogen leak rate at 195 K and 98 atm exceeded the predicted value by more than 40% when very low helium stagnation pressures at room temperature were used. The prediction became more accurate as the helium pressure was increased, however, and the error was reduced below 10% when the upstream helium pressure was raised above 20 atm. For the prediction of hydrogen leakage at high pressures and at room temperature, the experimental data in Figs. 3a and 3b also indicate that the HST was a reasonable prediction of the actual hydrogen leak rate when the helium tests were conducted above 20 atm; in fact, for circular orifice $1F$, a helium stagnation pressure of 12 atm was sufficient to be able to predict hydrogen leak rates at 98 atm with reasonable accuracy. Data for other conditions [6] similarly indicated that helium tests at 20 atm were sufficient to predict hydrogen leak rates at higher pressures, at least as high as 100 atm.

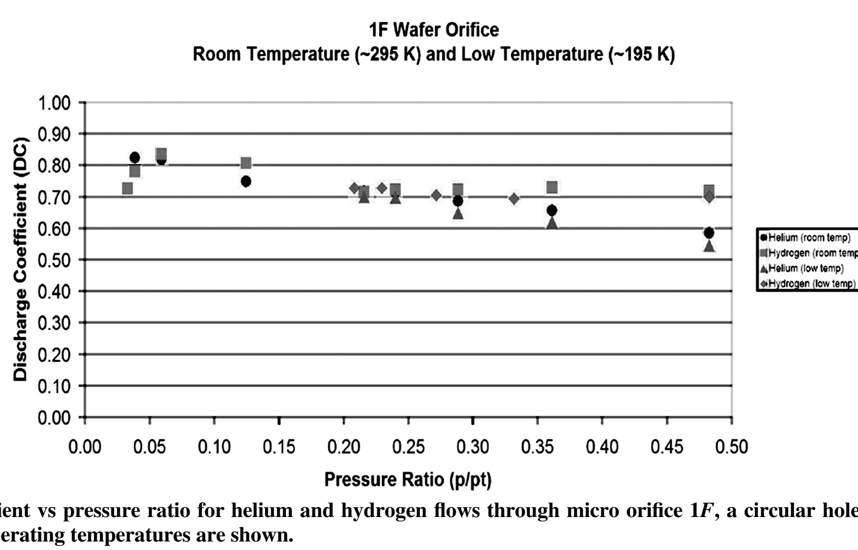
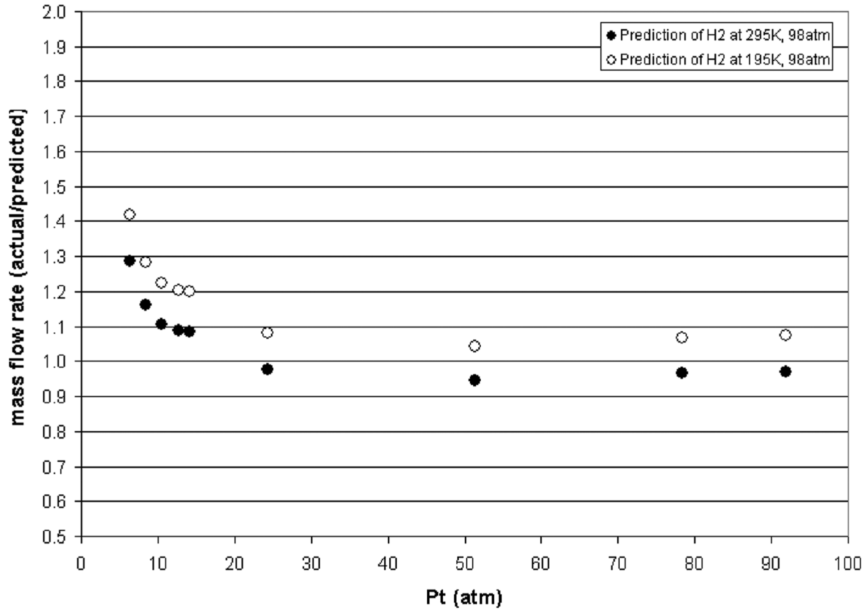
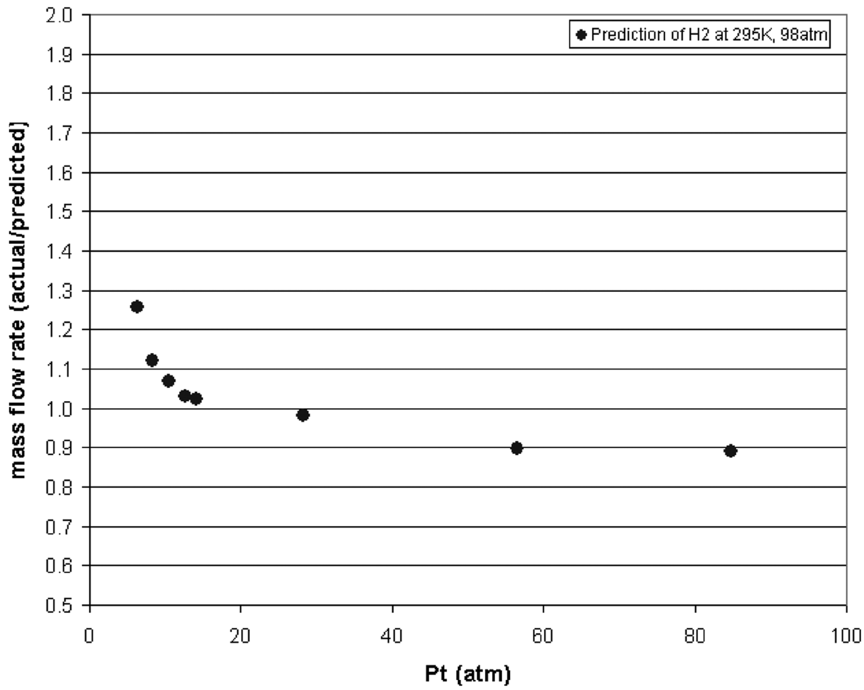


Fig. 2 Discharge coefficient vs pressure ratio for helium and hydrogen flows through micro orifice $1F$, a circular hole with a diameter of 57 μm. Conditions at different operating temperatures are shown.



a)



b)

Fig. 3 Actual hydrogen leak rates at 98 atm and at two different temperatures, divided by those predicted from helium leak rates at different pressures p_t and at room temperature for a) slit orifice 3*D* (hydraulic diameter of 15.8 μm) and b) circular orifice 1*F* (diameter of 57 μm).

V. Conclusions

The relationship between helium and hydrogen leak rates has been explored, using micromachined orifices of prescribed shapes and sizes, via a thermal conductivity detector to measure the mass flow rates of gas through the orifices. Comparisons were made between actual measured hydrogen leak rates at high pressures (∼100 atm) and at both room temperature (295 K) and low temperature (∼195 K) with those predicted from lower-pressure helium leak-rate measurements using the standard helium signature test (HST) procedure. In general, the HST was shown to underpredict the low-temperature hydrogen leak rate, possibly by as much as 40% in some cases. Yet when the HST was conducted using helium at room temperature and at higher upstream pressures, exceeding about 20 atm, this error was significantly reduced. Thus, based on the present tests, a helium stagnation pressure of about 20 atm and even at room temperature should be sufficient to accurately predict hydrogen leak rates at very high pressures (up to 100 atm and perhaps more) and low temperatures. These findings on the sufficient pressure and temperature conditions in the HST have important implications for the development of hydrogen storage and safety systems for a range of aerospace and ground vehicle applications.

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