INTERNATIONAL JOURNAL OF HYDROGEN ENERGY 34 (2009) 5174-5182



# Limits for hydrogen leaks that can support stable flames

# M.S. Butler<sup>a</sup>, C.W. Moran<sup>b</sup>, P.B. Sunderland<sup>b,\*</sup>, R.L. Axelbaum<sup>a</sup>

<sup>a</sup>Department of Energy, Environmental and Chemical Engineering, Washington University, St. Louis, MO 63130, USA <sup>b</sup>Department of Fire Protection Engineering, University of Maryland, College Park, MD 20742, USA

#### ARTICLE INFO

Article history: Received 31 January 2009 Received in revised form 4 April 2009 Accepted 5 April 2009 Available online 9 May 2009

Keywords: Compression fittings Fire safety Laminar flames Leakage Quenching limits Microcombustion Microflames

# ABSTRACT

Quenching and blowoff limits of hydrogen diffusion flames on small burners were observed. Four burner types, with diameters as small as 8  $\mu$ m, were considered: pinhole burners, curved-wall burners, tube burners, and leaky fittings. In terms of mass flow rate, hydrogen had a lower quenching limit and a higher blowoff limit than either methane or propane. Hydrogen flames at their quenching limits were the weakest flames recorded to date, with mass flow rates and heat release rates as low as 3.9  $\mu$ g/s and 0.46 W. The quenching limit for a hydrogen flame at a 6 mm leaky compression fitting was found to be 28  $\mu$ g/s. This limit was independent of supply pressure (up to 131 bar) and about an order of magnitude lower than the corresponding limits for methane and propane.

© 2009 International Association for Hydrogen Energy. Published by Elsevier Ltd. All rights reserved.

# 1. Introduction

Hydrogen is attractive as an energy carrier for highway vehicles. It can power fuel cells or engines with only water vapor as exhaust. With oil supplying 33% of the world's primary energy, hydrogen could help mitigate concerns over fossil fuel consumption if it is produced from renewable energy sources or nuclear energy [1,2]. Hydrogen can be stored as a gas, liquid, or a solid (in metal hydrides), and can be transported using pipelines, tankers or rail trucks [3].

Hydrogen is an unusual fuel. It has a high leak propensity and wide flammability limits, 4–75% by volume [4]. Among all fuels, hydrogen has the lowest molecular weight, the lowest quenching distance (0.51 mm), the smallest ignition energy in air (28  $\mu$ J), the lowest auto-ignition temperature by a heated air jet (640 °C), the highest laminar burning velocity in air (2.91 m/s), and the highest heat of combustion (119.9 kJ/g) [4]. Hydrogen flames are the dimmest of any fuel. Hydrogen embrittles and attacks metals more than any other fuel.

Hydrogen may ultimately prove to be no more hazardous from a fire safety standpoint than gasoline or diesel. However, gasoline and diesel have undergone over a century of widespread vehicle use, and this has resulted in codes and standards that have yielded an acceptable fire safety record. Further research is necessary if hydrogen is to be rapidly introduced with a similar safety record. It is expected that existing codes and standards will require updating to ensure safe use of hydrogen in highway vehicles [5].

The goal of this study is to investigate small hydrogen flames-flames that can be characterized as "leak flames," i.e., small flames that could exist as a result of hydrogen leaking

<sup>\*</sup> Corresponding author. Tel.: +1 301 405 3095; fax: +1 301 405 9383. E-mail address: pbs@umd.edu (P.B. Sunderland).

<sup>0360-3199/\$ –</sup> see front matter © 2009 International Association for Hydrogen Energy. Published by Elsevier Ltd. All rights reserved. doi:10.1016/j.ijhydene.2009.04.012

from a containment vessel. The scenario of concern is that a small leak in a hydrogen system could ignite, burn undetected for a long time, and potentially degrade surrounding materials or ignite secondary fires.

Several studies have been conducted to evaluate the characteristics of hydrogen leaks without combustion. Lee et al. [6] conducted hydrogen and helium leak rate experiments on micromachined orifices of different sizes and shapes. They examined the differences in flow rates among circular, square, and elliptical slit orifices as a function of pressure, and in most cases the flow was choked. Schefer et al. [7] investigated leaks where the flow was due to pressure-driven convection and permeation through metals. They obtained analytical relationships for flow rates of choked flows, subsonic laminar flows, and turbulent flows. Hydrogen leakage in stainless steel threaded pipe fittings was considered by Ge and Sutton [8]. They found that a larger tightening torque is less important in leak prevention than choice and proper application of thread sealant. The tests were run at up to 70 bar and typical hydrogen leak rates through these fittings were found to be  $1 \mu g/s$ .

Studies have also evaluated the risks of hydrogen leaks with respect to accumulation and explosion of hydrogen [9–11]. Swain and Swain [12] examined risks associated with leaks of hydrogen, methane, and propane from leaky fittings into an enclosure. Their measured leak rates showed that for a given supply pressure (except at the lowest supply pressures), hydrogen had a significantly higher volumetric flow rate than methane or propane. This behavior is expected for choked flows, owing to the high speed of sound of hydrogen gas.

Leak flames resemble the micro diffusion flames that have been observed in other laboratories and, while only one previous study specifically evaluated leak flames [13], there have been many studies of micro diffusion flames [14–19]. Micro diffusion flames are typically associated with an application, e.g., a microcombustor for power generation. Nonetheless, it is possible that they could arise unexpectedly. For example, if a fuel leak from a crack or hole in a fitting, tube or storage vessel of a plumbing system were ignited, this could be characterized as a micro diffusion flame.

Ban et al. [14] investigated micro diffusion flames that were 2-3 mm long on round burners with inner diameters of 0.15-0.4 mm. Three fuels were considered: ethane, ethylene and acetylene. The experiments agreed with predictions of flame shapes in the absence of buoyancy. The flames were nearly spherical and their shapes were unaltered when burner orientation was changed with respect to gravity. Cheng et al. [17] performed detailed measurements and computations on small hydrogen flames burning on similar burners. Buoyancy was found to be insignificant for these flames. Nakamura et al. [19] simulated methane micro diffusion flames supported on circular burners with diameters less than 1 mm. They, too, found nearly spherical flames, as a result of the weak buoyancy forces. They also considered quenching limits of the methane flames. Baker et al. [15] studied micro-slot burners (with port widths of 0.1–0.76 mm) and developed a flame height expression for purely diffusion-controlled propane/air nonpremixed flames.

Quenching and blowoff limits refer to flames with the smallest and highest fuel flow rates for sustained burning. Matta et al. [16] measured quenching limits for propane on small round burners. Similar experiments were performed by Cheng et al. [18] for methane. Both studies found that quenching mass flow rate was largely independent of burner diameter. Prior to the present work, quenching flow rates had not been measured for hydrogen. Blowoff limits for diffusion flames on small burners have been measured for a variety of fuels [16,20].

Thus motivated, this study includes experiments and analysis to identify which hydrogen leaks can support flames. A scaling analysis is presented to estimate the fuel mass flow rate at the quenching limit. Measured mass flow rates, both at the quenching and blowoff limits, are presented for hydrogen, methane and propane on round-hole burners. Flame quenching limits for leaky compression fittings are also presented. Further details are available in Butler [21] and Moran [22].

# 2. Scaling for flame quenching limits

A scaling analysis is presented here for flame quenching limits. This is similar to past work of Matta et al. [16]. The stoichiometric length  $L_f$  of laminar gas jet diffusion flames on round-hole burners is given by:

$$L_{\rm f}/d = a \ {\rm Re} = 4m_{\rm fuel}a/(\pi \ \mu d) \tag{1}$$

where *d* is the burner inside diameter, *a* is a fuel-specific coefficient, Re is fuel port Reynolds number,  $m_{\text{fuel}}$  is the fuel mass flow rate, and  $\mu$  is fuel dynamic viscosity. The scaling of Eq. (1) arises from many theoretical and experimental studies, including Roper [23], Sunderland et al. [24], and references cited therein.

The base of an attached jet diffusion flame is quenched by the burner and is premixed. The flame's standoff distance can be approximated as the standoff distance (i.e., 50% of the quenching distance) of the corresponding stoichiometric premixed flame. The 50% modification arises here because premixed flame quenching distances typically are reported as the minimum tube diameter or plate separation distance,  $L_q$ , through which a premixed flame can pass [4]. It is assumed here that a jet flame is above its quenching limit if its stoichiometric length exceeds its standoff distance:

$$L_{f} > L_{a}/2.$$
<sup>(2)</sup>

Combining Eqs. (1) and (2) yields the fuel mass flow rate at the quenching limit:

$$m_{\rm fuel} = \pi L_{\rm q} \mu / (8a) \tag{3}$$

Eq. (3) predicts that the fuel mass flow rate at the quenching limit is a fuel property that is independent of burner diameter, which was similarly predicted by Matta et al. [16]. When measured values of  $L_q$ ,  $\mu$ , and *a* (see Table 1) are inserted into Eq. (3), predicted fuel mass flow rates at the quenching limit are obtained, as listed in Table 1.

#### 3. Experimental

#### 3.1. Round-hole burners

Three types of round-hole burners were considered: pinhole burners, curved-wall burners, and tube burners, as illustrated in Fig. 1. Each type included various hole diameters, as

			erties of hy quenching		
-		-			

	Fuel	а	L <sub>q</sub> (mm)	S <sub>L</sub> (cm/s)	μ (g/m-s)	m <sub>fuel</sub> (μg/s)
	H <sub>2</sub>	0.236	0.51	291	8.76E-3	8
	$CH_4$	0.136	2.3	37.3	1.09E-2	85
	$C_3H_8$	0.108	1.78	42.9	7.95E-3	63
_						

Values for *a* are from [24],  $L_q$  and  $S_L$  (laminar burning velocity) are from [4], and  $\mu$  is from [25]. The quantity  $m_{\text{fuel}}$  is predicted from Eq. (3).

tabulated in Table 2. The pinhole burners (such as the one in Fig. 2) were stainless steel nozzles that were manufactured for solid-stream spray generation. The top surface of each burner (except the two smallest ones, which were planar) is a curved surface (radius of curvature of 9.6 mm) with a hole passing through its axis, as depicted in Fig. 1. The pinhole burners represent pinhole leaks in pressure vessels where the radius of curvature of the wall is large or infinite.

The curved-wall burners were constructed from stainless steel tubes with two outside diameters: 1.6 and 6.4 mm. A radial hole was drilled in the wall of each tube. These burners represent pinhole leaks that might occur in fuel supply tubing.

The tube burners were made from stainless steel hypodermic tubes. Although they do not represent practical leak flames, they are a fundamental configuration that leads to a flame from a wall where the radius of curvature approaches zero. Their observation should lead to an improved understanding of microinjectors in small-scale microelectromechanical power generators [16].

Fuel was delivered via a pressure regulator and a needle valve, and all tests were performed at normal lab pressure and temperature. Measuring the small flow rates at the quenching limits required special procedures. A glass soap-bubble meter was installed upstream of the burners. Quenching flow rates were measured by establishing a small flame near the quenching limit and gradually decreasing the flow rate until the flame extinguished. The fuel flow rate was then measured using the soap-bubble meter.

Determining the existence of hydrogen flames near their quenching limits was complicated by their small size and low luminosity. The flames were detected with a K-type thermocouple positioned 10 mm above the burner, yielding detection that is believed to be more sensitive than available camera technology could provide.

Burners were allowed to warm slightly above room temperature to prevent water condensation. This was necessary because condensation was found to disturb the flows from the small burners, sometimes extinguishing the flames.

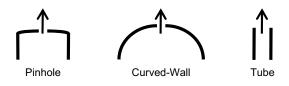


Fig. 1 – Schematics of the round-hole burners. Arrows show the fuel flow direction. Burner dimensions are shown in Table 2.

Table 2 – F burners.	Iole diameters (mm	) for the round-hol	е			
Pinhole	Curved wall <sup>a</sup>	Curved wall <sup>b</sup>	Tube			
0.008	0.41	0.41	0.051			
0.13	0.53	1.75	0.152			
0.36	0.74	2.46	0.406			
0.53	0.86	3.12	0.838			
0.71	1.02		1.194			
0.84			2.21			
1.01						
1.40						
1.78						
2.39						
3.18						
a Tube diameter of 1.6 mm. b Tube diameter of 6.4 mm.						

Using similar burners, Takahashi et al. [13] also reported complications of water condensation. Tests performed here at different burner temperatures, up to about 200 °C, found quenching flow rate to be largely independent of burner temperature provided condensation was avoided. Tests were also conducted with varying ambient humidity, and quenching limits were found to be generally independent of the relative humidity of the air in the range of 46–90%.

Hydrogen flow rate at blowoff was measured with a soap bubble meter. A stable flame was established and then the flow rate was increased until the flame first lifted and then extinguished. For blowoff tests the flames were detected visually. Hearing protection was required for the blowoff tests for the larger burners.

Additional tests were performed to consider buoyancy effects. To this end quenching flow rates were found for pinhole and tube burners in the vertical, horizontal, and inverted orientations.

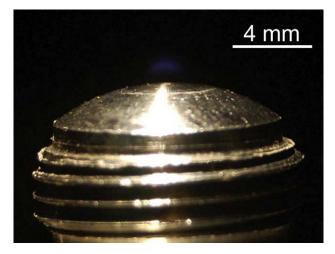


Fig. 2 – Image of a hydrogen flame slightly above its quenching limit of 7.5  $\mu$ g/s (original in color). The burner is a 0.36 mm pinhole burner, with a surface radius of curvature of 9.6 mm, and is oriented vertically. The image was recorded in a dim room at//4.2 with an exposure time of 30 s.

# 3.2. Leaky fittings

Quenching limits were also measured for leaky compression fittings. The tests involved leaks between Swagelok<sup>®</sup> stainless steel tube union compression fittings and stainless steel tubes. Tube diameters were 3.2, 6.4, and 12.7 mm. The end of each union opposite the tube was sealed. New tubes and fittings were assembled according to manufacturer instructions and were rejected if bubbles appeared in an applied soap water solution when pressurized with hydrogen to 8 bar absolute. Leaks were then introduced in one of three ways: by reducing the torque on the threaded nut, by over tightening the nut, or by scratching the front ferrule sealing surface. These types of leaks are occasionally encountered in plumbing systems. The three leak types were found to yield the same flame quenching limits, so only results for reduced torque fittings are presented here. Reduced torque fittings allow quenching limits to be measured for different upstream pressures by adjusting the torque, i.e., an increased torque requires an increased upstream pressure at the quenching limit. Most tests were performed with the fitting in the vertical orientation, with the leaky fitting at the top end of the tube, but several horizontal and inverted orientations also were considered.

Quenching limits were found by igniting the fuel (with an external flame) and then reducing fuel flow rate until extinction. Fuel flow rate was controlled with upstream pressure, primarily, and with torque on the threaded nut. Upstream pressure was set with a pressure regulator. When a quenching limit was established, a plastic tube was installed over the fitting such that the leak flow was measured with a downstream soap bubble meter at laboratory pressure. The quenching limits were obtained with fittings at nearly laboratory temperature.

The hydrogen flames generally were not visible even in dark conditions. As with the round-hole burners, the presence of hydrogen flames was determined with a thermocouple positioned 10 mm above the flame position. Quenching limits for methane and propane were identified visually because thermocouples confirmed these flames to be visible in all cases.

Uncertainties for the round-hole and leaky fitting tests are estimated at  $\pm$ 5% for hole diameter, fuel flow rate, and upstream pressure.

# 4. Results

# 4.1. Round-hole burners

Fig. 2 shows an image of a hydrogen flame burning slightly above its quenching limit on a 0.36 mm pinhole burner. The flame appearance is similar to that of all the quenching limit hydrogen flames on round-hole burners. The hydrogen quenching distance of Table 1 suggests steady hydrogen diffusion flames should be anchored about 0.26 mm from the burner surface. Fig. 2 indicates that this is reasonable for the hydrogen flames near their quenching limits. Because the maximum flame dimension is comparable to its standoff distance, this flame closely resembles a flat premixed flame [16].

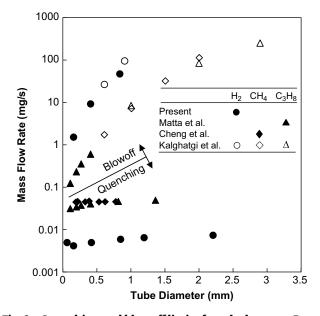


Fig. 3 – Quenching and blowoff limits for tube burners. Past measurements shown are from Refs. [16,18,20].

Measured quenching and blowoff limits for hydrogen, methane, and propane on tube burners are shown in Fig. 3. Measurements shown for methane and propane come from past work, and were confirmed in our laboratory for representative conditions. Consistent with the prediction of Eq. (3), the mass flow rate at quenching is not strongly dependent on burner diameter. The mean quenching limit flow rates are in reasonable agreement with the predictions of Table 1. The mean quenching limits for hydrogen are about an order of magnitude lower than those of the other fuels. Table 1 and Eq. (3) indicate this arises from hydrogen's short quenching distance (primarily) and from its large flame length relative to its mass flow rate (i.e., large coefficient *a*).

The mass flow rate at blowoff, see Fig. 3, increases with tube diameter because for these flames blowoff occurs when local velocities exceed local flame speeds. The blowoff limits for hydrogen are about an order of magnitude larger than those of the other fuels, largely owing to the high burning velocity of hydrogen. Regardless of tube diameter, the limits of stable combustion of hydrogen are much wider than those for propane or methane. The limits narrow with decreasing tube diameter, as was shown for propane in Matta et al. [16], but they never meet.

Various existing codes and standards require leak rates below 20 scc/h (0.46  $\mu$ g/s) for hydrogen and below 200 scc/h (36  $\mu$ g/s) for natural gas [13]. For hydrogen this leak rate is well below the measured flame quenching limits, whereas for methane this leak rate is only slightly below the quenching limits. Referring to the hydrogen measurements in Fig. 3, SAE J2579 [26] now limits localized hydrogen leaks in highway vehicles to a maximum of 5  $\mu$ g/s.

Fig. 4 shows the hydrogen quenching limits for the three types of round burners. Fig. 4 indicates that quenching limits are affected by burner type and diameter for the smaller burner diameters. Where the limits are affected by burner type, the differences can largely be attributed to varying

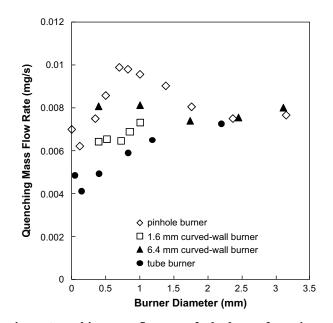


Fig. 4 – Quenching mass flow rates for hydrogen for various burner types and diameters. See Fig. 1 for burner schematics.

amounts of heat loss, as discussed next. An increase in heat loss requires a higher quenching flow rate to sustain the flame.

For burner diameters below 1.5 mm, pinhole burners have the highest quenching flow rates, while tube burners have the lowest. Pinhole burners have greater heat losses owing to their shape and mass. Heat conducted to the pinhole burners is mostly transferred to the ambient and constitutes a loss of enthalpy from the system. On the other hand, much of the heat conducted to the tube burners is transferred to the fuel and oxidizer and thus is not intrinsically lost from the system. For tube burners the primary mechanism of heat loss is expected to be radiation from the tube.

The effects of burner curvature on heat loss were investigated by observing quenching limits on curved-wall burners. An increase in curvature is expected to reduce wall heat loss. Indeed the 1.6 mm curved-wall burners yield quenching limits approaching those of the tube burners, whereas the 6.4 mm burners have quenching limits closer to those of the pinhole burners.

The dependence of quenching limits on burner diameter is complicated for small hole diameters, as shown in Fig. 4. For tube burners the quenching flow rate generally increases with increasing burner diameter. This also was observed for propane by Matta et al. [16], as shown in Fig. 3. This is attributed to increasing heat loss with increasing burner diameter. In contrast pinhole burners do not exhibit this trend because their heat loss is nearly independent of burner hole size. The reason for the local maximum in Fig. 4 for pinhole burners is unknown. For burner diameters above 1.5 mm, the quenching flow rates for both pinhole and tube burners agree reasonably well with the 8  $\mu$ g/s prediction of Eq. (3), shown in Table 1.

Quenching mass flow rate increases slightly when hole diameters are decreased below 0.1 mm for both pinhole and tube burners (see Fig. 4). It is likely that the flow field is affecting the quenching limits at these very small sizes. In particular, the measured quenching flow rates for these two burners indicate sonic velocities and Reynolds numbers of 14–130. For all other hole sizes, velocities are well below sonic and Reynolds numbers are small (with a range of 0.4–7).

While the results above are for vertical burner orientation (upward flowing fuel), hydrogen quenching limits also were measured for inverted and horizontal orientations. The results, shown in Fig. 5, reveal that quenching limits for the pinhole and tube burners are not dependent on orientation, indicating that these limit flames are not affected significantly by buoyancy. This can be elucidated by considering their Froude numbers, given by

$$Fr = u^2/(qd) \tag{4}$$

where u is the fuel velocity at the burner port and g is the acceleration of gravity. For the present quenching limit flames on pinhole and tube burners, the Fr range is 0.17–39. These are generally in the nonbuoyant regime for micro-flames of Fr > 1 [14]. Thus, flame structure at the quenching limits should not vary with orientation. Furthermore, as mentioned in the experimental section, small changes in burner temperature do not affect the quenching limits, so any effects of orientation on burner heating should not affect these limits.

The above results demonstrate that flame quenching limits depend on fuel type, with burner type, diameter, and orientation having far less significant effects. For a given fuel the mass flow rate from the leak is a function of the leak geometry and the upstream pressure. This relationship depends on whether the leak regime is diffusional, subsonic laminar, subsonic turbulent, isentropic choked, supersonic, etc., as considered by Schefer et al. [7]. While the leaks considered here would not be considered isentropic, insight into the quenching limits can be obtained by considering isentropic

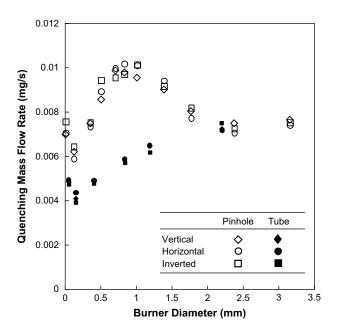


Fig. 5 – Quenching mass flow rates for hydrogen for pinhole and tube burners in horizontal, inverted, and vertical orientations.

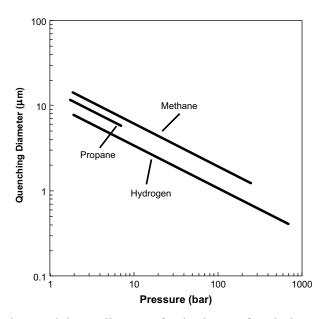


Fig. 6 – Minimum diameter of a circular port for a leak flame as a function of upstream absolute pressure assuming isentropic choked flow.

choked flows through round holes at the quenching limits. This could, for example, be relevant to a pinhole leak in a thin wall tube. Under these conditions the fuel flow rate is linear with respect to leak area A and upstream absolute pressure  $p_0$  as follows [7]:

$$m_{\rm fuel} = Ap_{\rm o} \left(\frac{kMW}{T_0 R_{\rm u}}\right)^{1/2} \left(\frac{2}{k+1}\right)^{k+1/2(k-1)}$$
(5)

where k is fuel specific heat ratio, MW is fuel molecular weight,  $T_0$  is upstream stagnation pressure, and  $R_u$  is the universal gas constant.

For a given fuel and upstream pressure, Eq. (5) can predict the hole diameter associated with the quenching limit. The results of these predictions are shown in Fig. 6 for hydrogen, methane, and propane. For this plot the quenching limit flow rates were taken as the average values for tube burners in Fig. 3. Each line in Fig. 6 starts at the minimum upstream pressure for choked flow and ends at the maximum pressure anticipated in alternative fuel vehicles. This plot predicts that for a given storage pressure, hydrogen is susceptible to leak flames for hole diameters that are smaller than those for methane or propane. Furthermore, at hydrogen's maximum anticipated storage pressure of 690 bar (10,000 psi), a hole diameter of just 0.4  $\mu$ m is predicted to support a flame.

The limit flames in this study are believed to be among the weakest steady flames ever recorded. The weakest of these had a hydrogen flow rate of  $3.9 \,\mu$ g/s (see Fig. 5), which corresponds to a heat release rate of 0.46 W based on hydrogen's lower heating value of 119.9 kJ/g. Previously, Ronney et al. [27] documented hydrogen flame balls produced under microgravity conditions with a power output of about 1 W. In subsequent tests they achieved flames as weak as 0.5 W. It is believed these were the weakest flames recorded until the present work.

Flame balls were first predicted by Zeldovich [28], who proposed that a solution exists to the steady heat and mass conservation equations corresponding to a stationary, spherical premixed flame. The phenomenon was discovered 40 years after Zeldovich's work by Ronney et al. [29] in drop tower experiments with lean hydrogen-air mixtures. The microgravity environment was necessary to obtain the spherical symmetry and to avoid extinction brought on by buoyancy. Flame balls have only been achieved in microgravity conditions with very lean hydrogen mixtures. With no mechanism other than radiative heat loss, and this being low for hydrogen flames, it was reasonable to assume that these flames were the weakest possible flames. The present results indicate that, despite the possibility of heat loss to the tube, the present diffusion flame geometry is actually slightly more resistant to extinction than a flame ball. As noted earlier, heat transfer to the hypodermic tube does not intrinsically imply a significant loss of enthalpy as much of this heat preheats the reactants.

# 4.2. Leaky fittings

Fig. 7 shows images of hydrogen, methane, and propane flames on 6.3 mm leaky compression fittings in the vertical orientation. These images were recorded slightly above the quenching limits. The hydrogen flame is significantly smaller and dimmer than the others. Near their quenching limits the flames do not burn along the entire fitting annulus. For all three flames the shortest distance between the flame and the metal material is approximately 50% of the quenching distances given in Table 1. No yellow soot luminosity was visible in any of the flames near their quenching limits. The leaks associated with the images of Fig. 7 produced readily visible bubbles when soap water solution was applied.

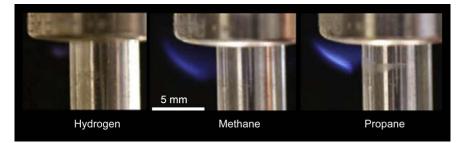


Fig. 7 – Flames near extinction on leaky compression fittings for hydrogen, methane, and propane (original in color). Also visible is the 6.3 mm tube and the threaded nut. The fitting was vertically oriented and the upstream absolute pressure was 4 bar.

An audible pop occurred upon hydrogen ignition, but not upon ignition of methane or propane. When the external flame was removed, ignition was quickly followed by extinction for fuel flow rates below the quenching limit. For fuel flow rates above the quenching limit, ignition was followed by a stable flame.

Fig. 8 shows measured quenching limits for hydrogen, propane, and methane for 6.3 mm fittings in the vertical orientation. The data at the higher line pressures were obtained by increasing the torque on the fitting, thus reducing the leak size. The upper limit on line pressure for propane is lower than that of the others because the vapor pressure of propane is only 9.1 bar absolute (142 psia) at 25 °C.

Within experimental uncertainties, the quenching limits of Fig. 8 are independent of line pressure for each fuel. This suggests that, as anticipated from the round-hole burner results of Fig. 4, the key parameter controlling quenching limits is the fuel mass flow rate. For these low flow rates and small leak sizes the line pressure does not significantly affect the velocity field in the flame region.

The mean hydrogen flow rate at the quenching limit is about an order of magnitude lower than for the other fuels, which is consistent with the results of Fig. 3. For leaky fittings, the quenching flow rates for hydrogen, methane, and propane are about an order of magnitude higher than the corresponding flows for tube burners (see Fig. 3). This is attributed to additional heat loss in the leaky fittings, where the flames burn near thick concave metal surfaces.

The effects of fitting orientation are shown in Fig. 9, and reveal that orientation has little or no effect on the quenching limits of the hydrogen flames. This is anticipated as these flames are small at their limits and the results are consistent with the tube and pinhole burner hydrogen measurements of Fig. 5. Nonetheless, fitting orientation did affect the quenching

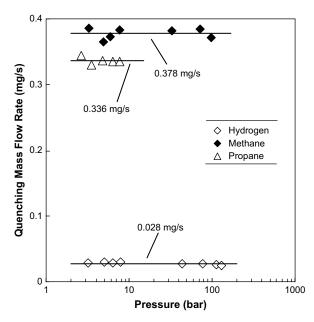


Fig. 8 – Quenching limit (minimum flaming flow rate) versus upstream absolute pressure for a 6.3 mm leaky compression fitting in the vertical orientation. The line fits represent mean values of the measured quenching limits.

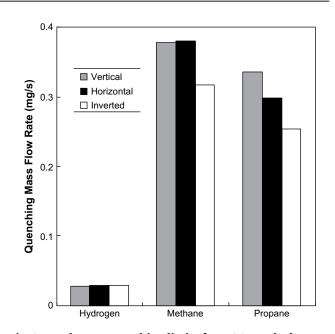


Fig. 9 – Hydrogen quenching limits for a 6.3 mm leaky compression fitting in vertical, horizontal and inverted orientations. The upstream absolute pressure was 4 bar.

limits of propane and methane flames, with the inverted orientation yielding limits about 20% lower than the vertical orientation. These flames are larger than the corresponding hydrogen flames such that buoyancy begins to have a significant effect on the flow field. The inverted orientation has the fitting below the tube, and this minimizes the surface area of the flame impingement, leading to a reduced heat loss and thus lower quenching limits.

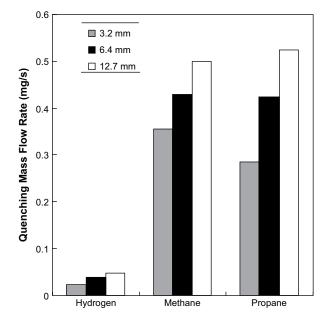


Fig. 10 – Hydrogen quenching limits for leaky compression fittings for tube diameters of 3.1, 6.3, and 12.6 mm. The fittings were vertically oriented and the upstream absolute pressure was 4 bar.

The effects of fitting diameter on quenching limits are examined in Fig. 10. Diameter had a significant impact on the quenching limits in all cases; on average as tube diameter doubles, the fuel mass flow rate at quenching increases 30%. This is attributed to increased heat losses associated with larger fittings.

This study has identified new questions that warrant further study. For example, what corrosive effects do leak flames have on containment materials? Can permeation leaks support flames? Can surface coatings, e.g., intumescent paints, be applied that will increase the quenching fuel flow rate? An improved understanding of hydrogen leaks is necessary to ensure safe use of hydrogen in the public sector.

# 5. Conclusions

Flame quenching and blowoff limits were measured for diffusion flames resulting from fuel issuing from small burners. Hydrogen fuel was emphasized, and was compared with methane and propane for a variety of burner types that characterized a range of heat losses. The key findings are as follows.

- 1. Flame quenching limits for round-hole burners were measured for hydrogen, methane, and propane. For hydrogen these are in the range of  $4-10 \ \mu g/s$  depending on burner diameter, shape, and orientation. The limits for methane and propane are about 10 times as high. The measurements indicate that many existing safety codes and standards should revisit their allowable fuel leak rates in consideration of these quenching limits.
- Flame blowoff limits increase with burner diameter and for hydrogen are about 10 times as high as the corresponding limits for methane and propane. Thus the limits of stable combustion are much wider for hydrogen than for the other fuels considered.
- 3. For an isentropic choked leak, hydrogen is susceptible to leak flames for hole diameters that are smaller than those for methane or propane. This suggests that for hydrogen stored at 690 bar, a pinhole leak as small as  $0.4 \,\mu\text{m}$  could support a stable flame.
- 4. The weakest hydrogen flame found in this study, which was generated with a hypodermic needle with an inner diameter of 0.152 mm, is believed to be the weakest flame ever observed, with a fuel flow rate of  $3.9 \,\mu$ g/s and a heat release rate of 0.46 W.
- 5. The minimum flow rate necessary for sustaining a hydrogen flame on a leaky 6.3 mm tube compression fitting is 28  $\mu$ g/s. This is about an order of magnitude lower than for propane or methane. The minimum mass flow rate for all fuels is independent of upstream (line) pressure, and is sufficient to produce bubbles when a soap-water solution is applied.
- 6. Fitting orientation had about a 20% effect on the quenching limits of the leaky compression fittings considered here, while a doubling of tube size on average results in a 30% increase in quenching flow rate.

This work was undertaken because hydrogen is such an unusual fuel and its quenching limits had not previously been measured. Hydrogen's low quenching limits, combined with its high leak propensity, could present unusual risks in a hydrogen economy. These risks should be explored in further research and should be incorporated into the many relevant safety codes and standards.

# Acknowledgments

This work was supported by NIST under the technical direction of J. Yang. The assistance of B.H. Chao and N.R. Morton is appreciated.

# REFERENCES

- Marban G, Valdes-Solis T. Towards the hydrogen economy? Int J Hydrogen Energy 2007;32:1625–37.
- [2] Yamawaki M, Nishihara T, Inagaki Y, Minato K, Oigawa H, Onuki K, et al. Application of nuclear energy for environmentally friendly hydrogen generation. Int J Hydrogen Energy 2007;32:2719–25.
- [3] Sherif SA, Barbir F, Veziroglu TN. Wind energy and the hydrogen economy—review of the technology. Solar Energy 2005;78:647–60.
- [4] Kanury AM. Introduction to combustion phenomena. New York: Gordon and Breach; 1975. p. 131.
- [5] MacIntyre I, Tchouvelev AV, Hay DR, Wong J, Grant J, Benard P. Canadian hydrogen safety program. Int J Hydrogen Energy 2007;32:2134–43.
- [6] Lee ID, Smith OI, Karagozian AR. Hydrogen and helium leak rates from micromachined orifices. AIAA J 2003;41(3):457–63.
- [7] Schefer RW, Houf WG, San Marchi C, Chernicoff WP, Englom L. Characterization of leaks from compressed hydrogen dispensing systems and related components. Int J Hydrogen Energy 2006;31:1247–60.
- [8] Ge X, Sutton WH. Analysis and test of compressed hydrogen interface leakage by commercial stainless steel (NPT) fittings. SAE Int 2006:35–47.
- [9] Dorofeev SB. Evaluation of safety distances related to unconfined hydrogen explosions. Int J Hydrogen Energy 2007; 32:2118–24.
- [10] Takeno K, Okabayashi K, Kouchi A, Nonaka T, Hashiguchi K, Chitose K. Dispersion and explosion field tests for 40 MPa pressurized hydrogen. Int J Hydrogen Energy 2007;32(13): 2144–53.
- [11] Zbikowski M, Makarov D, Molkov V. LES model of large scale hydrogen-air planar detonations: verification by the ZND theory. Int J Hydrogen Energy 2008;33:4884–92.
- [12] Swain MR, Swain MN. A comparison of  $H_2$ ,  $CH_4$ , and  $C_3H_8$ fuel leakage in residential settings. Int J Hydrogen Energy 1992;17: 807–15.
- [13] Takahashi M, Tamura Y, Suzuki J, Watanabe S. Investigation of the allowable flow rate of hydrogen leakage on receptacle. Detroit: SAE International; April 2008. Paper 2008-01-724.
- [14] Ban H, Venkatesh S, Saito K. Convection-diffusion controlled laminar micro flames. Trans ASME 1994;116:954–9.
- [15] Baker J, Calvert ME, Murphy DW. Structure and dynamics of laminar jet micro-slot diffusion flames. J Heat Transfer 2002; 124:783–90.
- [16] Matta LM, Neumeier Y, Lemon B, Zinn BT. Characteristics of microscale diffusion flames. Proc Combust Inst 2002;29:933–8.
- [17] Cheng TS, Chao Y-C, Wu C-Y, Li Y-H, Nakamura Y, Lee K-Y, et al. Experimental and numerical investigation of

microscale hydrogen diffusion flames. Proc Combust Inst 2005;30:2489–97.

- [18] Cheng TS, Chen CP, Chen CS, Li YH, Wu CY, Chao YC. Characteristics of microjet methane diffusion flames. Combust Theory Modeling 2006;10:861–81.
- [19] Nakamura Y, Yamashita H, Saito K. A numerical study on extinction behaviour of laminar micro-diffusion flames. Combust Theory Modeling 2006;10(6):927–38.
- [20] Kalghatgi GT. Blow-out stability of gaseous jet diffusion flames. Part I: In still air. Combust Sci Technol 1981;26(5): 233–9.
- [21] Butler MS. Flame quenching and materials degradation of hydrogen leaks. M.S. Thesis. St. Louis: Washington University; May 2008. 50 pp.
- [22] Moran CW. Flame quenching limits of hydrogen leaks. M.S. Thesis. University of Maryland; May 2008. 48 pp.
- [23] Roper FG. The prediction of laminar jet diffusion flame sizes: Part 1. Theoretical model. Combust. Flame 1977;29:219–26.

- [24] Sunderland PB, Mendelson BJ, Yuan ZG, Urban DL. Shapes of buoyant and nonbuoyant laminar jet diffusion flames. Combust Flame 1999;116:376–86.
- [25] Weast RC, Astle MJ. CRC Handbook of chemistry and physics. 59th ed. West Palm Beach: CRC Press Inc; 1979. p. F-58.
- [26] SAEJ2579. Recommended practice for general fuel cell vehicle safety, a surface vehicle recommended practice. Detroit, MI: SAE International; January, 2009.
- [27] Ronney PD, Wu MS, Pearlman HG. Structure of flame balls at low Lewis-number (SOFBALL): preliminary results from the STS-83 and STS-94 space flight experiments. Paper AIAA-1998-463. Reno, Nevada: Aerospace Sciences Meeting; January 1998.
- [28] Zeldovich Y. Theory of combustion and detonation of gases. Moscow, Russia: Academy of Science; 1994.
- [29] Ronney PD, Wu MS, Pearlman HG. Experimental study of flame balls in space: preliminary results from STS-83. AIAA J 1998;36(8):1361–8.