

Flame Quenching Limits of Hydrogen Leaks

M. S. Butler and R. L. Axelbaum

Washington Univ., St. Louis

C. W. Moran and P. B. Sunderland

Univ. of Maryland

ABSTRACT

This study examines the types of hydrogen leaks that can support combustion. Hydrogen, methane, and propane diffusion flames on round burners and leaky compression fittings were observed. Measurements included limits of quenching and blowoff for round burners with diameters of 0.006 - 3.18 mm. The measured mass flow rates at the quenching limits were found to be generally independent of burner diameter at relatively large burner diameters. In terms of mass flow rate, hydrogen had the lowest quenching limit and the highest blowoff limit of the fuels considered, which means that there are high and low flow rates where hydrogen is able to support a flame while methane and propane are not able to. The quenching limits for hydrogen diffusion flames on round burners with thick walls were found to be higher than for thin walls. The limits were also found to be independent of burner orientation; leaks with low flow rates are able to support flames independent of their orientation. The minimum mass flow rate of hydrogen that can support combustion from a leaking compression fitting was found to be 0.028 mg/s. This flow was independent of pressure (up to 131 bar) and about an order of magnitude lower than the corresponding methane and propane flow rates. The implications for fire safety are discussed.

INTRODUCTION

Concerns about the emissions of greenhouse gases have led to extensive consideration of hydrogen as an energy carrier. Currently, oil constitutes about 33% of the world's primary energy, and it is primarily produced in countries that are replete with political instability as viewed from a western point of view [1]. With crude oil approaching \$100/barrel, supply is not keeping up with demand and political instability makes the price even more tenuous. In addition, our society's dependence on fossil fuels possibly results in global warming, acid rain, etc. It is also predicted that crude oil supplies will run dry by 2043 [2]. Hydrogen as an energy carrier can help mitigate these issues [3]. Some benefits of hydrogen include that it can be converted to electricity with a high efficiency, its raw materials and the majority of its products are water, it can be stored as liquid, gas or

solid, and it can be transported using pipelines, tankers or rail trucks over long distances [4]. It is also projected that hydrogen production can be produced efficiently using renewable methods such as wind [5].

There are, however, several hazards associated with hydrogen. Applications for the gas will likely shift to include commercial applications; these hold more risk than the current, tightly-regulated industrial applications, thus codes and standards must be updated [6]. Experimental and numerical work has shown that there are some obvious safety risks in hydrogen explosions and high pressure gas releases [7,8]. Also, hydrogen itself presents several unusual fire hazards, including high leak propensity, high blowoff limits, ease of ignition, and invisible flames. These characteristics have been investigated in the context of micro diffusion flames.

There are three classifications for laminar jet flames: diffusion controlled flames such as the Burke-Schumann flame [9], diffusion and momentum controlled flames such as the Roper flame [10] and micro diffusion flames controlled by momentum and diffusion [11]. Extensive research has been done on the first two types, but only a limited amount has been done on micro diffusion flames. Baker et al. [12] devised a flame height expression for purely diffusion controlled flames capable of accurately predicting micro-slot diffusion flame heights. Useful parameters for characterizing dominant flame mechanisms were defined: a diffusion-buoyancy and a diffusion-momentum parameter. This work extended the investigations of Roper et al. [13] to smaller slot sizes. Ban et al. [11] investigated flames established on circular burners with inner diameters of 0.15, 0.25, and 0.40 mm. The experiments worked to verify the predicted flame shape of laminar flames ($Re = 1 - 10$) for three fuels: ethane, ethylene and acetylene. The work found that buoyancy effects are negligible for small flames; nearly spherical flames were established that were unaltered by rotating the burner. An in depth analysis of a micro diffusion hydrogen flame was performed by Cheng et al. [14]; their numerical solution for species was compared with experimental data. It was found that buoyancy effects were insignificant.

Nakamura et al. [15] numerically studied methane micro diffusion flames sitting on a circular burner of a diameter less than 1 mm. They found that small flames have the same, nearly spherical structure as those in microgravity, citing weak buoyancy forces. Also, the existence of a minimum flame size necessary for combustion was predicted.

Quenching and blowoff limits bound the leak flow rates that can support combustion. Matta et al. [16] found that a flame is not able to exist when its predicted length is less than the measured standoff distance. Experiments verified this analytical method of finding the quenching flow rate by establishing a propane flame over a hypodermic, stainless steel tube and decreasing the fuel flow rate until extinction occurred. They also measured blowoff. Cheng et al. [17] performed similar quenching experiments and included a dominant mechanism similar to that of Baker et al. [12], except using methane as fuel. Kalghatgi et al. [18] measured blowoff limits for a variety of fuels including propane, methane and hydrogen on circular burners. These studies show that hydrogen blowoff limits are higher than those for methane and propane. For a given leak size, there is a range of mass flow rates where hydrogen is able to support a stable flame but methane and propane would be blown off.

In order to understand how much gas is flowing through a given leak, research has been done in evaluating leak flow rates of hydrogen, methane, and propane. Swain and Swain [19] modeled and measured leak rates for diffusion, laminar, and turbulent flow regimes. They found that combustible mixtures in an enclosed space resulted more quickly for propane and hydrogen leaks than for methane leaks. It was also found that hydrogen had significantly higher volumetric flow rates through leaks than did methane and propane. Their supply pressures were the same for all fuels.

Another risk associated with hydrogen is that it is often stored at high pressures in spite of its high propensity to leak. It is predicted that hydrogen will need to be stored at up to 40 MPa to gain the efficiency desired [22]. Hydrogen's safety hazards resulted in a Department of Energy report [23] finding that hydrogen containment was the chief safety concern associated with using hydrogen as a transportation fuel and documented several catastrophic hydrogen fires. One such accident that displays hydrogen's difficulty in storage is a pressurized hydrogen tank rupture in Hanau, Frankfurt in 1991. Heavy damage was done to the rest of the plant [20].

Other characteristics of hydrogen that make it more prone to risk are that it is also easier to ignite than most other common fuels, and, once ignited, it is difficult to detect. The minimum ignition energy for hydrogen is an order of magnitude lower than those for methane and propane [21]. This means that a small spark might be able to ignite a hydrogen leak, whereas the same spark might not be able to ignite a methane or propane leak. It

has also been noted that hydrogen flames have weak luminosity [14], hence a hydrogen leak sustaining a flame would be difficult to detect by the human eye.

Considering these properties of hydrogen, the scenario of interest in this work is that a small, easily ignitable leak in a hydrogen system might ignite, support a flame that is difficult to detect, and degrade containment materials to the point of a catastrophic failure. This study includes experiments and analysis to identify which hydrogen leaks can support flames. Thus motivated, the objectives of this work are to measure limits of sustained combustion, both at quenching and blowoff, for hydrogen on round burners and lower flaming limits for hydrogen, methane and propane on leaky compression fittings.

FLAME QUENCH SCALING

A scaling analysis was developed to interpret measured flame quenching limits. These limits are the minimum flow rates required to support a diffusion flame.

The stoichiometric length L_f of laminar gas jet diffusion flames on round burners is:

$$L_f / d = a Re = a \rho u_0 d / \mu, \tag{1}$$

where d is the burner inside diameter, a is a dimensionless fuel-specific empirical constant, Re is Reynolds number, u_0 is the average fuel velocity in the burner, ρ is fuel density, and μ is fuel dynamic viscosity. The scaling of Eq. 1 arises from many theoretical and experimental studies, including Roper [10], Sunderland et al. [24], and references cited therein. Constant a here is assigned values measured by Sunderland et al. [24], as listed in Table 1.

The base of an attached jet diffusion flame is quenched by the burner. Its standoff distance can be approximated as one half of the quenching distance of a stoichiometric premixed flame. Such quenching distances typically are reported as the minimum tube diameter, L_q , through which a premixed flame can pass. It is assumed here that a jet flame can be supported only if its stoichiometric length is greater than half this quenching distance:

$$L_f \geq L_q / 2. \tag{2}$$

Measurements of L_q , shown in Table 1, are taken from

Table 1: Selected fuel properties of hydrogen, methane, and propane. Values for a are from Sunderland et al. [7], L_q and S_L (laminar burning velocity) are from Kanury [8], and μ is from Weast and Astle [9].

Fuel	a	L_q [mm]	S_L [cm/s]	μ [g/m-s]	m_{fuel} [mg/s] predicted
H ₂	0.236	0.51	291	8.76e-3	0.008
CH ₄	0.136	2.3	37.3	1.09e-2	0.085
C ₃ H ₈	0.108	1.78	42.9	1.96e-3	0.063

Kanury [25]. When combined, Eqs. (1) and (2) predict the following fuel flow rate, m_{fuel} , at the quenching limit:

$$m_{fuel} = \pi \rho u_0 d^2 / 4 = \pi L_q \mu / (8 a). \quad (3)$$

Eq. (3) indicates that the fuel mass flow rate at the quenching limit is a fuel property that is independent of burner diameter. When values of L_q , μ , and a from Table 1 are inserted into Eq. (3), the predicted fuel mass flow rates at quenching are obtained.

EXPERIMENTAL

Two different burner configurations are considered here: round burners and leaky compression fittings.

There is generally a limited range of flow rates for which a flame can be established on the present burners. Below this range, the flow is said to be below the quenching limit. Quenching occurs when there is too much heat loss for combustion to be sustained. Above this range, the flow is said to be above its blowoff limit. This limit occurs when the flow velocity in the flammable region of the flame becomes greater than the burning velocity of the mixture.

Round Burners

Three different types of round burners were considered for this work: tube burners, pinhole burners, and curved-wall pinhole burners, as displayed in Fig. 1. Each had a range of flow passage diameters. All tests were performed at ambient lab pressure, and flow was delivered via a pressure regulator and a flow controlling valve.

The tube burners were made from stainless steel hypodermic tubes, oriented vertically. Tube inside diameters were 0.006, 0.016, 0.033, 0.047, and 0.087 mm. These burners resemble microinjectors that may be used in future, small scale microelectro-mechanical power generators [16]. The pinhole burners were stainless steel nozzles that are manufactured for solid-stream spray generation. The top of each burner (except the two smallest ones) is a curved surface with a hole passing through its axis, as shown in Fig. 2. The two smallest burners had holes in planar, not curved, surfaces. It is expected that the variation of quenching flow rate as a result of this slight curvature is on the order of the error associated with measurements. The hole diameters were 0.008, 0.13, 0.36, 0.53, 0.71, 0.84, 1.01, 1.40, 1.78, 2.39, and 3.18 mm. The curved-wall pinhole burners were constructed of stainless steel tubes with two outside diameters: 1.59 and 6.35 mm. A

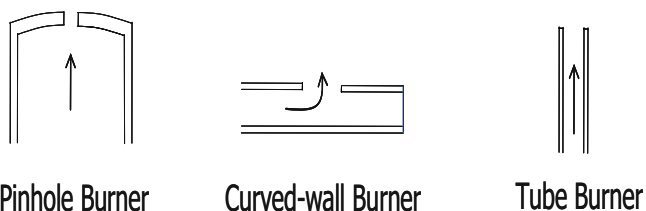


Figure 1: Burners used to find quenching limits.

radial hole was drilled in each tube. Hole sizes were 0.41, 0.53, 0.74, 0.86, and 1.02 mm (small tubes) and 0.41, 1.75, 2.46 and 3.12 mm (large tubes).

Certain complications are unique to these experiments. The present small flow rates could not be accurately measured using conventional flow meters. Instead, for tests with round burners a glass soap-bubble meter was installed upstream of the burners. Tests performed with varying air moisture content found quenching flow rate to be generally independent of air moisture content in the range of 46% to 90% relative humidity. Hydrogen flame detection was complicated by their dimness, especially at low flow rates, but this was resolved by using thermocouples to detect flames. K-type thermocouples were chosen to avoid catalysis, and were placed several flame lengths above the flames to avoid disturbances.

Quenching flow rates were measured by first establishing a small flame, decreasing the flow rate until flame extinction and then introducing a soap bubble for flow rate measurement. Each burner was allowed to warm to just above room temperature to prevent water condensation on the burner surface. This is because water condensation was found to disturb flow from the small burners, sometimes extinguishing the flames. Tests performed with different burner temperatures found quenching flow rate to be largely independent of burner temperature.

Burner temperature was not an issue during blowoff measurements because the burners remained relatively cool. The fuel flow rate at blowoff was measured in much the same way as quenching limits were. A stable, large flame was first established; the flow rate was then increased by increasing the upstream pressure via a pressure regulator until the flame first lifted and then extinguished. For these tests the flames were detected visually. Hearing protection was used for blowoff tests on larger burners.

To complete the investigation on round burners, experiments were performed to consider buoyancy effects. Quenching flow rates were found for both pinhole and tube burners in the horizontal (rotated 90°) and inverted orientations (rotated 180°). The limits were measured in the same manner.

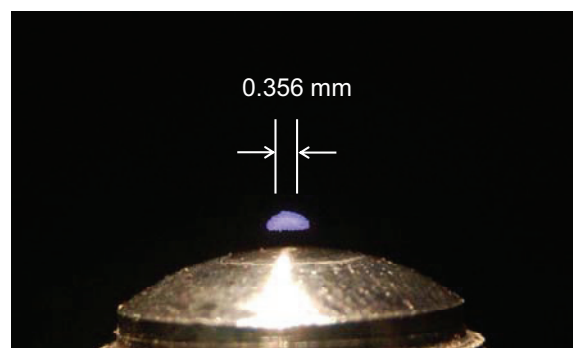


Figure 2: Contrast-enhanced image of hydrogen flame over a 0.356 mm pinhole burner (original in color).