ABSTRACT

This study examines the types of hydrogen leaks that can support combustion and the effects on stainless steel of long term hydrogen flame exposure. Experimental and analytical work is presented. Hydrogen diffusion flames on round burners were observed. Measurements included limits of quenching, blowoff, and piloted ignition for burners with diameters of 0.36 - 1.78 mm. Results are compared to measurements for methane and propane. A dimensionless crack parameter was identified to correlate the quenching limit measurements. Flow rates were 0.019 - 40 mg/s for hydrogen, 0.12 - 64 mg/s for methane, and 0.03 - 220 mg/s for propane. Hydrogen flames were found to be corrosive to 316 stainless steel tubing.

INTRODUCTION

Concerns about the emissions of greenhouse gases have led to extensive consideration of hydrogen as an energy carrier. Hydrogen presents several unusual fire hazards, including high leak propensity, ease of ignition, and invisible flames. This research concerns experiments and analysis to identify which hydrogen leaks can support flames. A small leak in a hydrogen system could ignite easily, support a flame that is difficult to detect, and degrade containment materials to the point of a catastrophic failure. Hydrogen leaks can develop in pressure vessels, piping, seals, valves, pressure regulators, and pressure relief devices.

A Department of Energy report (Cadwallader and Herring, 1999) found that hydrogen containment was the chief safety concern associated with using hydrogen as a transportation fuel. This report documents several catastrophic hydrogen fires.

Research in hydrogen combustion has increased recently, but no study to date has characterized the types of hydrogen leaks that can support a flame. Absent such information, it may be difficult for the designers of a hydrogen system to perform a cost-benefit analysis of protection against leaks.

Research has been done in quantifying leak flow rates, comparing hydrogen to methane and propane. Swain and Swain (1992) 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. However their supply pressures were the same for all fuels and thus did not reflect plans for hydrogen systems in vehicles with pressures of up to 700 bar.

The present research seeks to determine the relative fire hazards of small hydrogen leaks compared to those of methane and propane. The modeling and experimentation focus primarily on small burners and flames near the quenching limit. Experiments also consider the corrosive effects associated with the exposure of 316 stainless steel to hydrogen and methane flames.

Thus motivated, the objectives of this work are to (1) measure limits of flaming (at ignition, quenching and blowoff) for hydrogen, methane, and propane issuing from circular burners of various sizes, and (2) examine material degradation arising from hydrogen and methane diffusion flames.

FLAME QUENCHING THEORY

A theoretical model was developed to predict flame quenching limits. These limits are the minimum flow rates sufficient to support a diffusion flame. This theory also yields a dimensionless crack parameter that indicates how close a given leak is to the quenching limit.

The stoichiometric length \( L_f \) of a laminar gas jet diffusion flames on a round burner is:
\[ L_f / d = a \ Re = a \ \rho \ u_0 \ d / \mu , \]  

(1)

where \( d \) is 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, \( \rho \) is fuel density, and \( \mu \) is fuel dynamic viscosity. The scaling of Eq. (1) arises from many theoretical and experimental studies, including Roper (1977), Sunderland et al. (1996), and references cited therein. Constant \( a \) here is assigned values measured by Sunderland et al. (1996), as listed in Table 1.

Table 1: Selected fuel properties of hydrogen, methane, and propane. Values for \( a \) are from Sunderland et al. (1996), \( L_q \) and \( S_L \) are from Kanury (1975), and \( \mu \) is from Weast and Astle (1979).

<table>
<thead>
<tr>
<th>Fuel</th>
<th>( a )</th>
<th>( L_q ) [mm]</th>
<th>( S_L ) [cm/s]</th>
<th>( \mu ) [g/m-s]</th>
</tr>
</thead>
<tbody>
<tr>
<td>H(_2)</td>
<td>0.236</td>
<td>0.51</td>
<td>291</td>
<td>8.76e-3</td>
</tr>
<tr>
<td>CH(_4)</td>
<td>0.136</td>
<td>2.3</td>
<td>37.3</td>
<td>1.09e-2</td>
</tr>
<tr>
<td>C(_3)H(_8)</td>
<td>0.108</td>
<td>1.78</td>
<td>42.9</td>
<td>7.95e-3</td>
</tr>
</tbody>
</table>

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:

\[ L_f \geq L_q / 2 \]  

(2)

Measurements of \( L_q \), shown in Table 1, are taken from Kanury (1975). When combined, Eqs. (1) and (2) predict the following fuel flowrate, \( m_{\text{fuel}} \), at the quenching limit:

\[ m_{\text{fuel}} = \pi \ \rho \ u_0 \ d^2 / 4 = \pi \ L_q \ \mu / (8 \ a) . \]  

(3)

Eq. (3) indicates that the fuel mass flow rate at the quenching limit is a fuel property that is independent of burner diameter.

A crack parameter can now be derived. Assuming fully-developed laminar flow in the burner,

\[ u_0 = d^2 \ \Delta p / (32 \ \mu \ L_b) , \]  

(4)

where \( \Delta p \) is the pressure drop across the burner and \( L_b \) is the burner flow passage length (Munson et al., 2002).

Combining Eqs. (1), (2), and (4) yields

\[ CP = a \ \rho \ d^4 \ \Delta p / (16 \ \mu^2 \ L_b \ L_q) \geq 1 \]  

(5)

where \( CP \) is the dimensionless crack parameter.

EXPERIMENTAL

Two types of measurements were made. Quenching and blowoff limits of small-scale hydrogen, methane, and propane flames were measured. These tests involved five hemispherical burners of different diameters. Materials degradation tests were also performed, using tube burners. Flowrates were measured with calibrated rotameters.

The quenching and blowoff limit burners are hemispherical stainless steel nozzles that are manufactured for spray generation. At the apex of the hemisphere is a drilled hole of the specified diameter.

Fuel flow was commenced and ignited, creating a flame approximately 5 mm in size. The flow was then reduced until the flame extinguished. This was done several times for each burner and each fuel. The flames were small enough, and the experiment was done quickly enough, that there was no noticeable increase in the temperature of the stainless steel burners.

Inverted burns also were performed, in which the jet direction was downward. Hydrogen performed essentially the same; the quench limit was largely independent of burner orientation. Methane required less fuel to sustain a flame in the inverted position, and propane required a significantly larger flowrate to sustain an inverted flame.

Also measured were the blowoff flows of each fuel for each burner. Blowoff is achieved when the flammable regions flow faster than the laminar flame speed, which is shown in Table 1. Blowoff limits were measured by igniting a flow of fuel and then increasing the flow rate until the flame lifted off and extinguished.

Tests were also conducted to determine the corrosive effects of these flames on 316 stainless steel. For these tests the flames considered were those of Fig. 1.

Figure 1: Color images of a representative hydrogen flame (A) and methane flame (B).
RESULTS

Typical hydrocarbon flames burn much brighter than do hydrogen flames, as shown in Fig. 1.

The results of the quenching and blowoff studies are presented in Figs. 2 and 3. These figures also include the predictions of quenching limits from Eq. (3). Figure 2 shows the fuel mass flow rate versus diameter, while Figure 3 shows fuel velocity versus diameter for the same tests.

Figure 2: Measured fuel mass flow rate at the quenching and blowoff limits versus burner diameter. The lower set of three lines are the quenching limit theory of Eq. (3) for the fuels as shown. The upper set of three lines are the best fits of the blowoff measurements for the fuels as shown.

Figure 3: Measured fuel velocity at the quenching and blowoff limits versus burner diameter. The curves shown are the quenching limit theory of Eq. (3) for the fuels as shown. The lines are the best fits of the blowoff measurements for the fuels as shown.

different published values of quenching distances of premixed flames.

Fig. 2 demonstrates the important finding that for each fuel there is a critical mass flowrate below which combustion is impossible. Using these results, given the flowrate of a leak, it is readily known whether the leak is flammable.

The blowoff data show that methane will reach blowoff at the lowest mass flowrate, propane next, and hydrogen will require the most mass flow to achieve blowoff. These observations are qualitatively supported by the laminar flame speeds shown in Table 1.

Figure 3 shows the same measurements of Fig. 2 where the ordinate is changed to fuel velocity. This figure suggests a regime may exist at the smallest burner diameters where the blowoff limit is lower than the quenching limit. Burners smaller than those considered here will need to be tested to further evaluate this.
MATERIAL DEGRADATION

Two stainless steel burners were fabricated from 316 stainless steel. One supported a hydrogen flame and the other a methane flame, as shown in Fig. 1. Figure 4 shows the burners in their pre-test conditions.

The two flames were burned for 355 hours. The flames burned continuously except for brief periods when images were recorded. Figure 5 shows the burners after the test.

At the end of the test, there was noticeably more corrosion on the hydrogen burner, see Fig. 5. The additional corrosion on the hydrogen burner is believed to arise because hydrogen flames have relatively short standoff distances and thus result in increased material temperatures.

CONCLUSIONS

The quenching and blowoff limits for hydrogen, methane, and propane have been modeled and measured for small round burners. The fuel mass flow rate at the quenching limits was found to be independent of burner diameter. The fuel mass flow rate at blowoff was found to be considerably higher for hydrogen than for methane or propane.

Hydrogen flames were found to cause more corrosion of 316 stainless steel than methane for similar exposure conditions.

ACKNOWLEDGMENTS

This work was supported by NIST grant 60NANB5D1209 under the technical management of J. Yang. The assistance of K.B. Lim, V. Lecoustre, and C. Moran is appreciated.

REFERENCES


CONTACT

The corresponding author for this work is Peter B. Sunderland, pbs@umd.edu, www.enfp.umd.edu, (301) 405-3095.