

CNG, H₂, CNG-H₂ Blends – Critical Fuel Properties and Behavior



Jay Keller,
Sandia National Laboratories

Keynote Lecture presented at:
Workshop on
Compressed Natural Gas and Hydrogen Fuels:
Lessons Learned for the Safe Deployment of Vehicles

December 10-11, 2009



Hydrogen Behavior – Myth Busting



Jay Keller,
Sandia National Laboratories

Topical Lecture
Progress in Hydrogen Safety: International Short
Course Series

June 15-19, 2009



Hydrogen Myths



- ⇒ Hydrogen Molecular Diffusivity is 3.8 times that of CH_4
 - Therefore it diffuses rapidly and mitigates any hazard
- ⇒ Hydrogen is 14.4 times lighter than air
 - Therefore it rapidly moves upward and out of the way
- ⇒ We do not know the flammability limits for H_2
- ⇒ We just do not understand hydrogen combustion behavior
 - Hydrogen release is different than other fuels
 - Radiation is different than other fuels



Hydrogen Myths



- ⇒ Hydrogen hazards can be compared favorably to experiences with other hydrocarbon fuels
 - Less dangerous than gasoline, methane ...
- ⇒ Simply adding hydrogen to natural gas improves engine efficiency and lowers emissions.
- ⇒ ICE's are 33% less efficient than are Fuel Cells (@50% DOE / FreedomCar current goal)
- ⇒ Hydrogen always ignites
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Momentum-Dominated Jets are within the Ignition Region



Unignited Jet Separation Distance Length Scales

Pressure = ~20 MPa (~3000 psig)

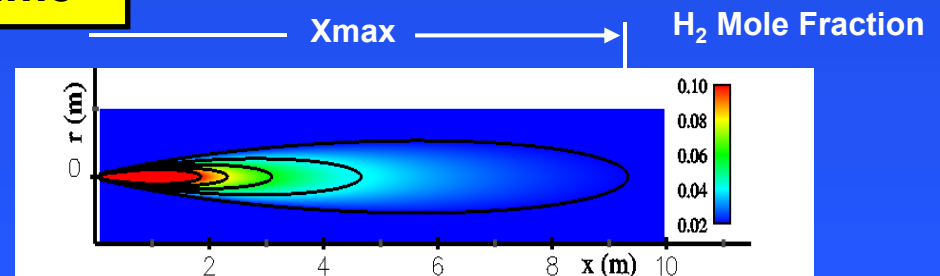
Hole Diameter	Flowrate	Xmax - Distance to 4% mole fraction	Start of Intermediate Region (Buoyancy)
1.5875 mm (1/16 inch)	(2,463 ft ³ /min)* 2.430x10 ⁻² Kg/sec (615.9 ft ³ /min)*	7.40 m (24.28 ft)	14.6 m (48.0 ft)
0.794 mm (1/32 inch)	6.075x10 ⁻³ Kg/sec (154.1 ft ³ /min)*	3.70 m (12.14 ft)	10.3 m (33.9 ft)

*@NTP = 21° C (70° F), 101 kPa (14.7 psia)

Flow between exit and 4% mole fraction is in the momentum dominated regime

- Start Intermediate Region
 $x/D = 0.5 F^{1/2} (\rho_{\text{exit}}/\rho_{\text{amb}})^{1/4}$

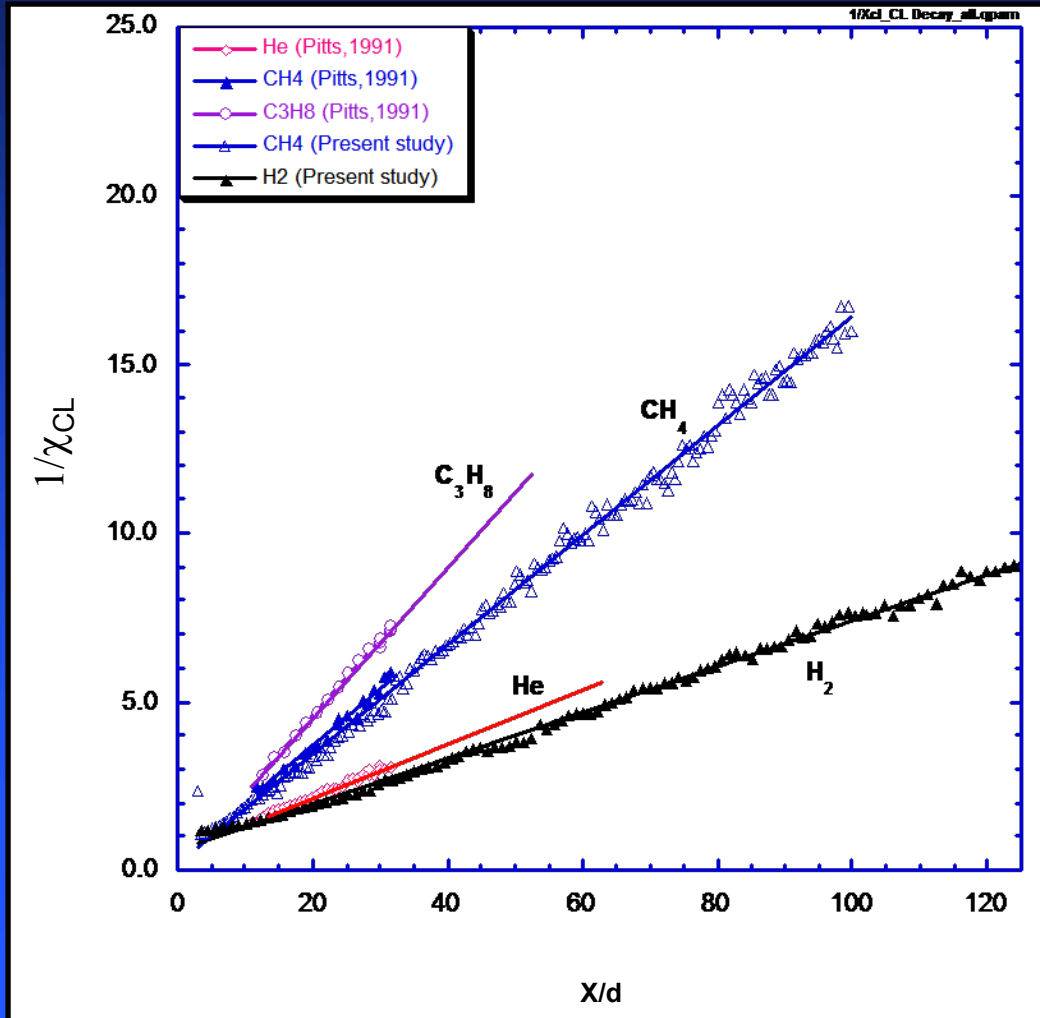
$$F = \text{Exit Froude No.} = U_{\text{exit}}^2 \rho_{\text{exit}} / (gD(\rho_{\text{amb}} - \rho_{\text{exit}}))$$



Small Unignited Releases: Momentum-Dominated Regime



Data for round turbulent jets



⇒ In momentum-dominated regime, the centerline decay rate follows a $1/\chi_{CL}$ dependence for all gases.

⇒ The mole fraction centerline decay rate increases with increasing molecular weight.

The decay rate for H₂ is significantly slower than for methane and propane.



Hydrogen Myths



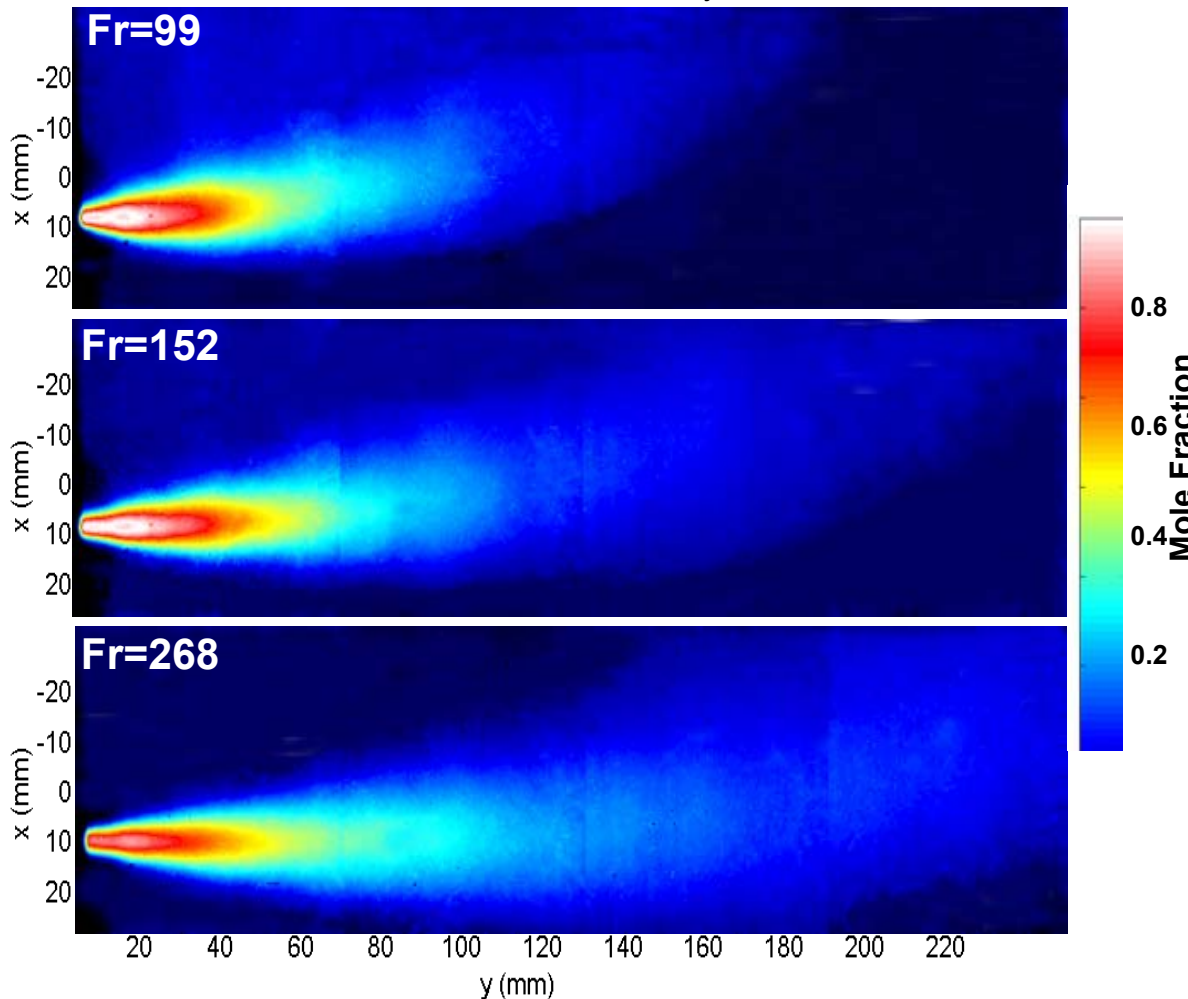
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Buoyancy effects are characterized by Froude number



Horizontal H₂ Jet (d_j=1.9 mm)



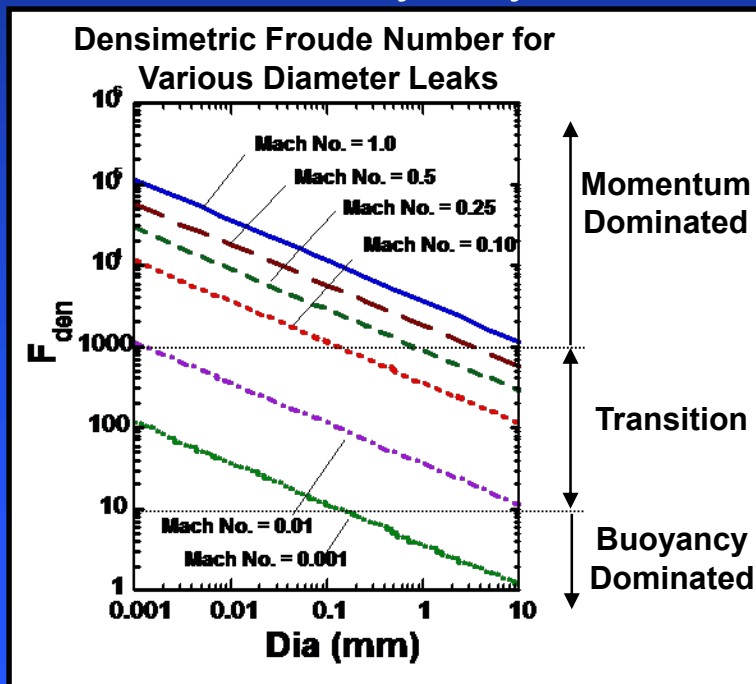
- ⇒ Time-averaged H₂ mole fraction distributions.
- ⇒ Froude number is a measure of strength of momentum force relative to the buoyant force
- ⇒ Increased upward jet curvature is due to increased importance of buoyancy at lower Froude numbers.



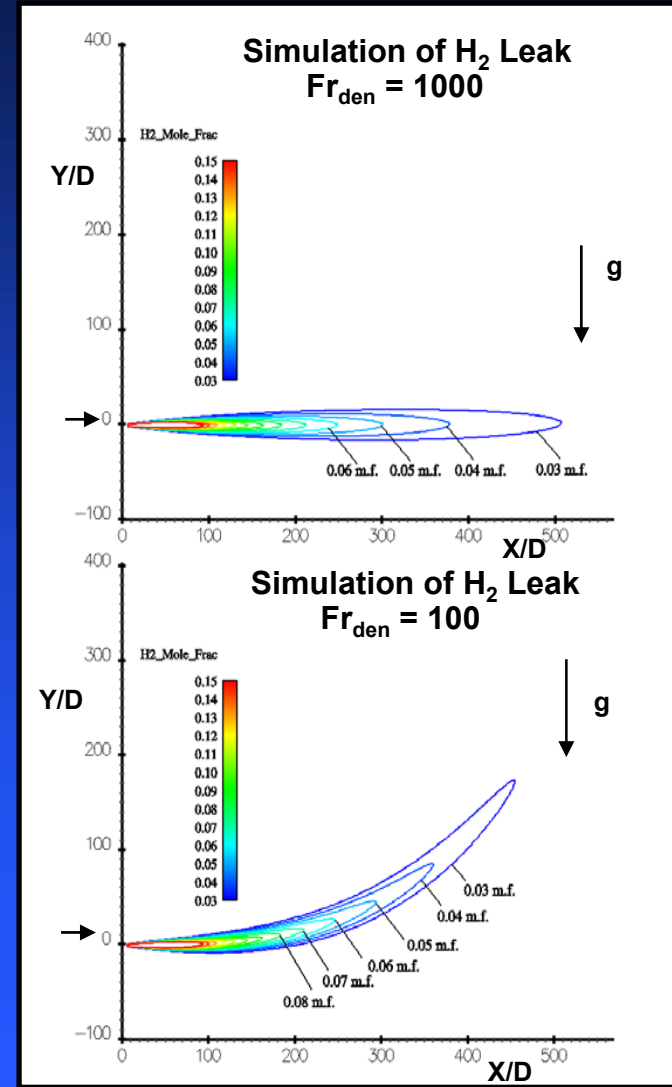
Influence of buoyancy is quantified by the Froude number



- Jets from choked flows (Mach 1.0) are typically momentum-dominated ($P_{\text{upstream}}/P_{\text{downstream}} > \sim 2$).
- Lower source pressures or very large pressure losses through cracks lead to subsonic, buoyancy-dominated plumes.



$$Fr_{\text{den}} = U_{\text{exit}} / (gD(\rho_{\text{amb}} - \rho_{\text{exit}}) / \rho_{\text{exit}})^{1/2}$$



Ricou and Spalding entrainment law (J. Fluid Mechanics, 11, 1961)

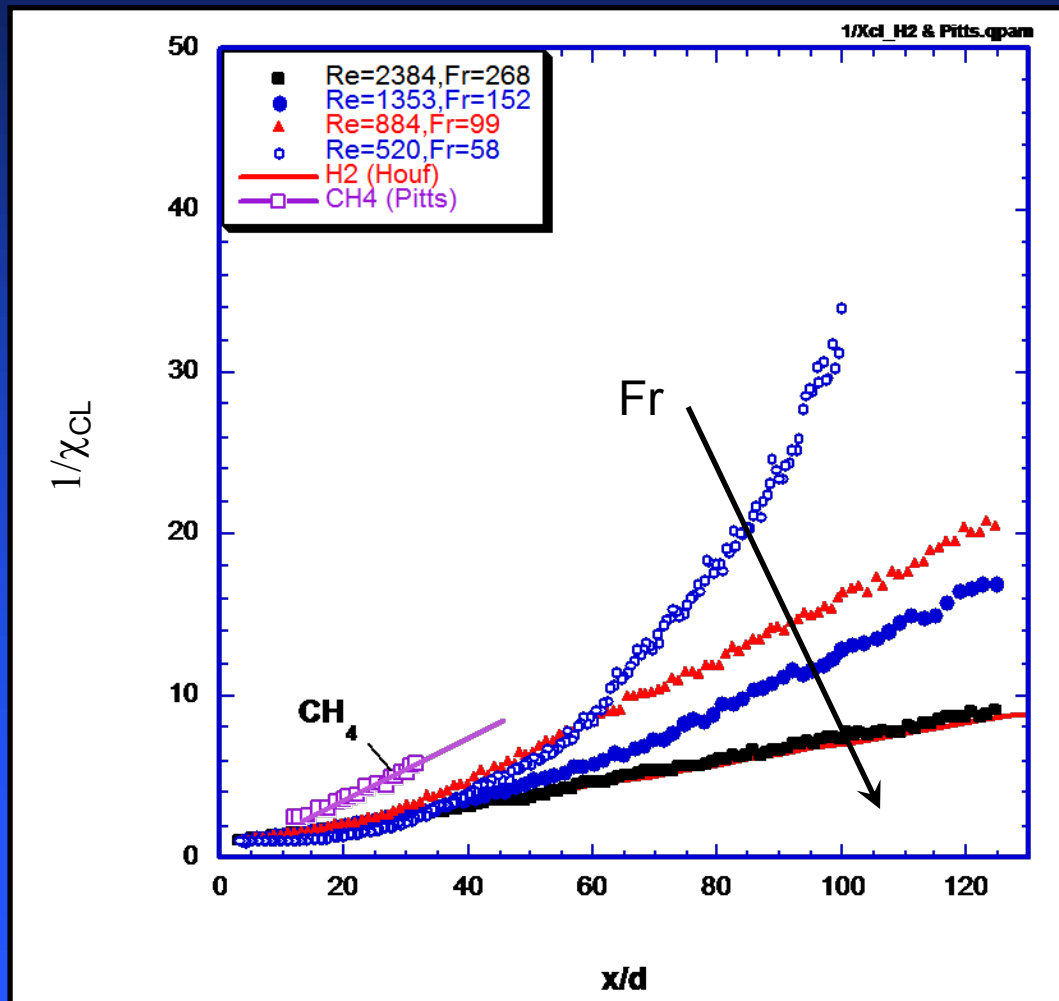
Sandia National Laboratories



Small Unignited Releases: Buoyancy Effects



→ Data for round H₂ Jets (d_j=1.91 mm)



- At the highest Fr, $1 / \chi_{CL}$ increases linearly with axial distance, indicating momentum dominates.
- As Fr increases buoyancy forces become less important and the centerline decay rate decreases.
- The transition to buoyancy-dominated regime moves downstream with increasing Fr.



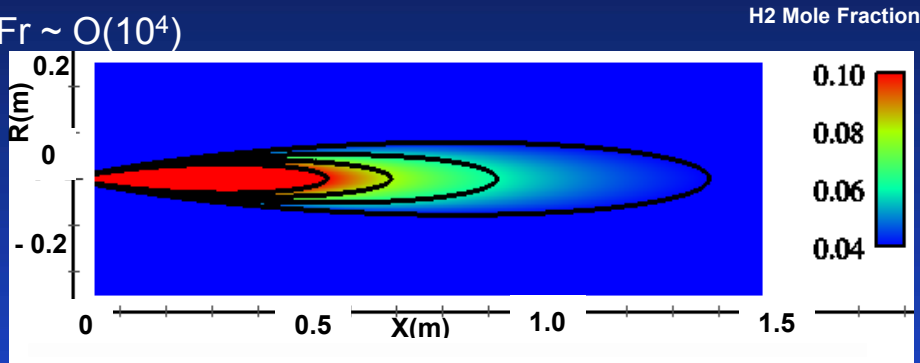
Choked & Unchoked Flows at 20 SCFM



Tank Pressure = 3000 psig, Hole Dia. = 0.297 mm

Exit Mach Number = 1.0 (Choked Flow)

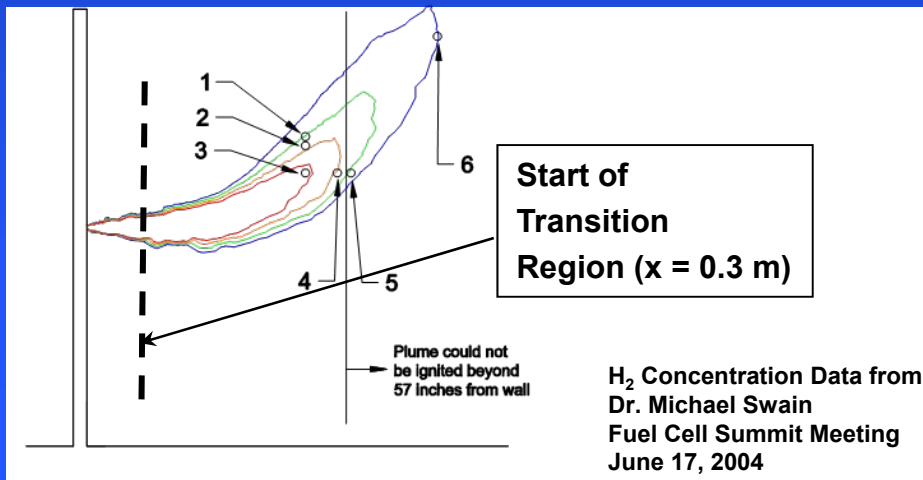
$Fr \sim O(10^4)$



Flowrate = 20 scfm, Hole Dia. = 9.44 mm

Exit Mach Number = 0.1 (Unchoked Flow)

$Fr \sim O(100)$



- ⇒ Correlations based on experimental data
- ⇒ Start Intermediate Region
 - $x/D = 0.5 F^{1/2} (\rho_{\text{exit}}/\rho_{\text{amb}})^{1/4}$
- ⇒ End Intermediate Region
 - $x/D = 5.0 F^{1/2} (\rho_{\text{exit}}/\rho_{\text{amb}})^{1/4}$
- ⇒ F = Exit Froude No.
 - = $U_{\text{exit}}^2 \rho_{\text{exit}} / (gD(\rho_{\text{amb}} - \rho_{\text{exit}}))$

Start Transition Region -> x = 6.3 m

- ⇒ Assuming gases at 1 Atm, 294K (NTP)
 - Red – 10.4%
 - Orange – 8.5%
 - Green – 5.1%
 - Blue – 2.6%

*(Chen and Rodi, 1980)



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Flammability Limits for H₂



Upward Flame Propagation

Tube Dimensions, cm		Firing end	Limits, percent		Water Vapor Content	Reference
Diameter	Length		Lower	Higher		
7.5	150	Closed	4.15	75.0	Half-saturated	356
5.3	150					
5.3	150					
5.3	150					
5.0	150					
5.0	150					
4.8	150					
4.5	80					
4.5	80					

Horizontal Flame Propagation

Tube Dimensions, cm		Firing end	Limits, percent		Water Vapor Content	Reference		
Diameter	Length		Lower	Higher				
7.5	150	Closed	6.5	-----	Half-saturated	356		
5.0	150		N	6.7			N	356
2.5	150		N	6.7			N	356

Downward Flame Propagation

Tube Dimensions, cm		Firing end	Limits
Diameter	Length		Lower
21.0	31	Open	9.3
8.0	37	Closed	8.9
7.5	150	N	8.8
7.0	150	N	-----
6.2	33	Open	8.5
6.0	120	N	9.45

Propagation in a Spherical Vessel

Capacity, cc	Firing end	Limits, percent		Water Vapor Content	Reference
		Lower	Higher		
Not stated	Closed	9.2	----	Saturated	271
Not stated	N	8.5	67.5	N	82
1,000	N	8.7	75.5	N	95
810	N	5.0	73.5	N	349
350	N	4.6	70.3	N	368
35	N	9.4	64.8	N	297



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Tube Dimensions, cm		Firing end	Limits, percent		Water Vapor Content	Reference
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- ⇒ 78 investigations of hydrogen flammability limits were identified between 1920 and 1950.
- ⇒ Hydrogen flammability limits are well established.

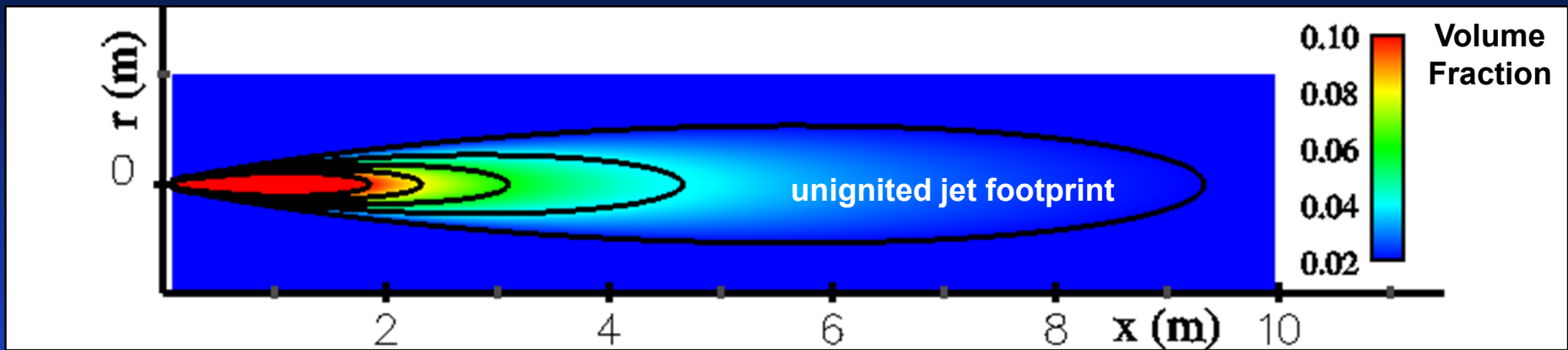
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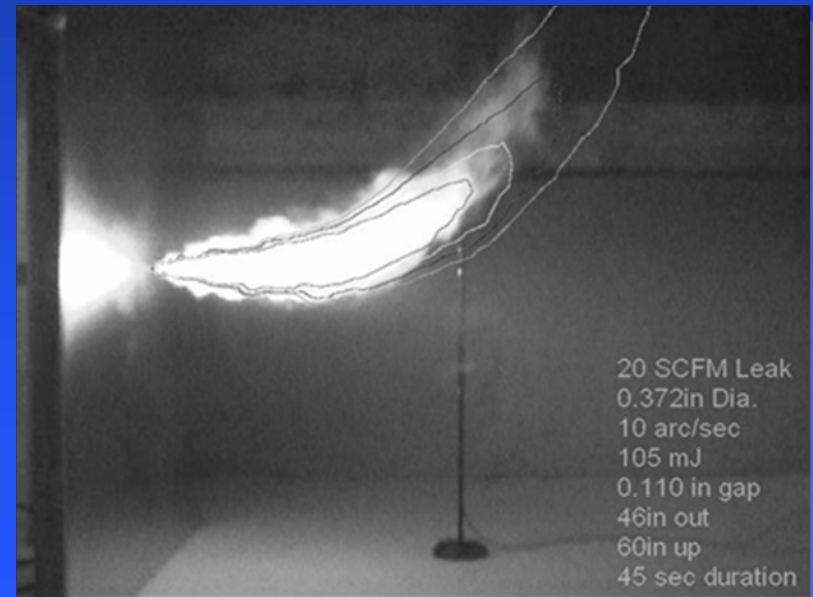
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What is a Reasonable Flame Stabilization Limit?



- ⇒ Which volume fraction contour is relevant:
 - lean flammability limit? ... 4% or 8%
 - detonation limit? ... 18%
 - a fraction of the lowest lean flammability limit? ... 1%
- ⇒ **Ignition of hydrogen in turbulent jets occurs around 8% as measured by Swain.**
 - This is consistent with the downward propagating limit of 8%



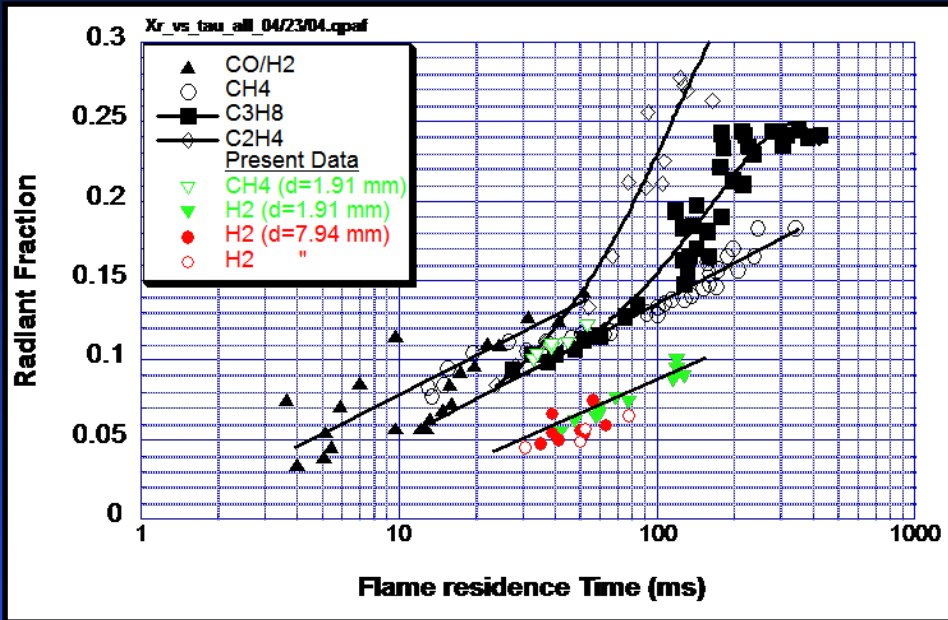
Hydrogen Myths



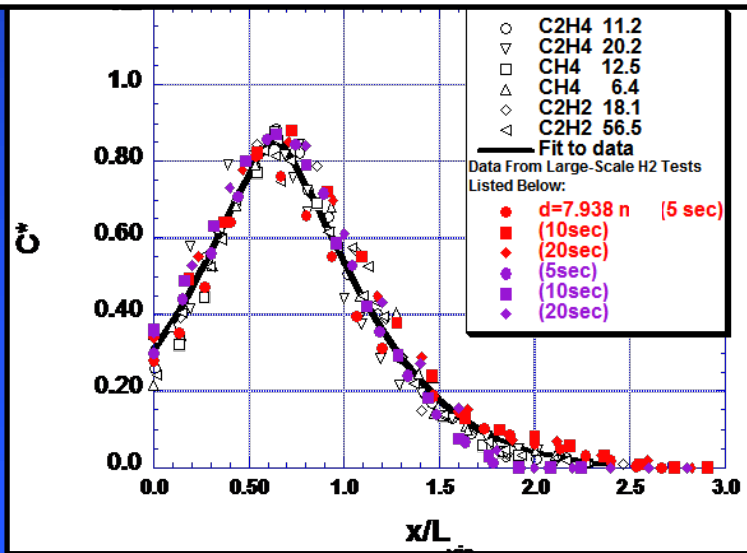
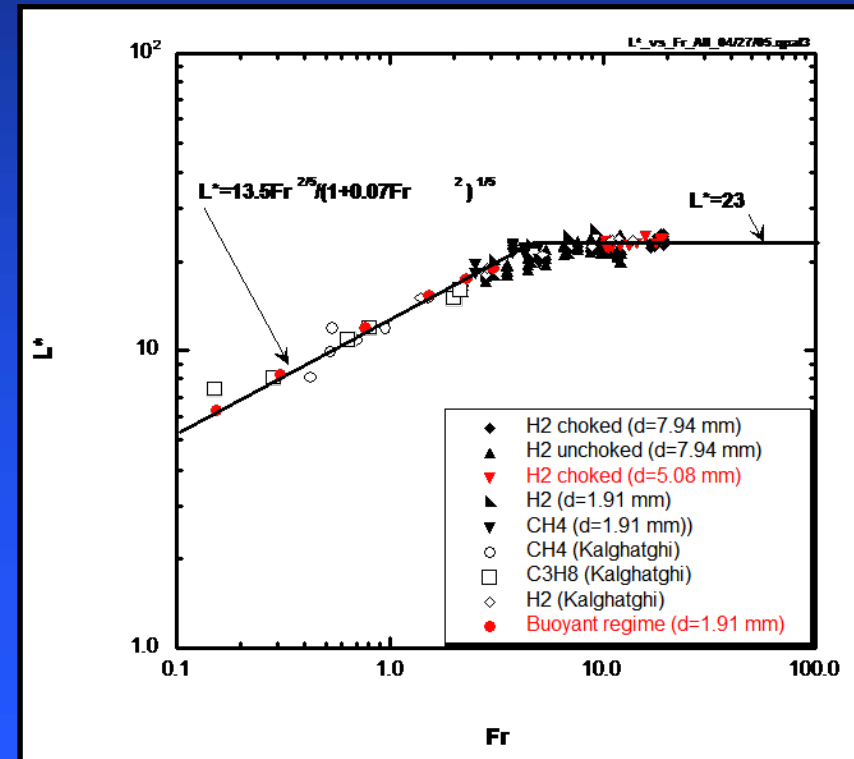
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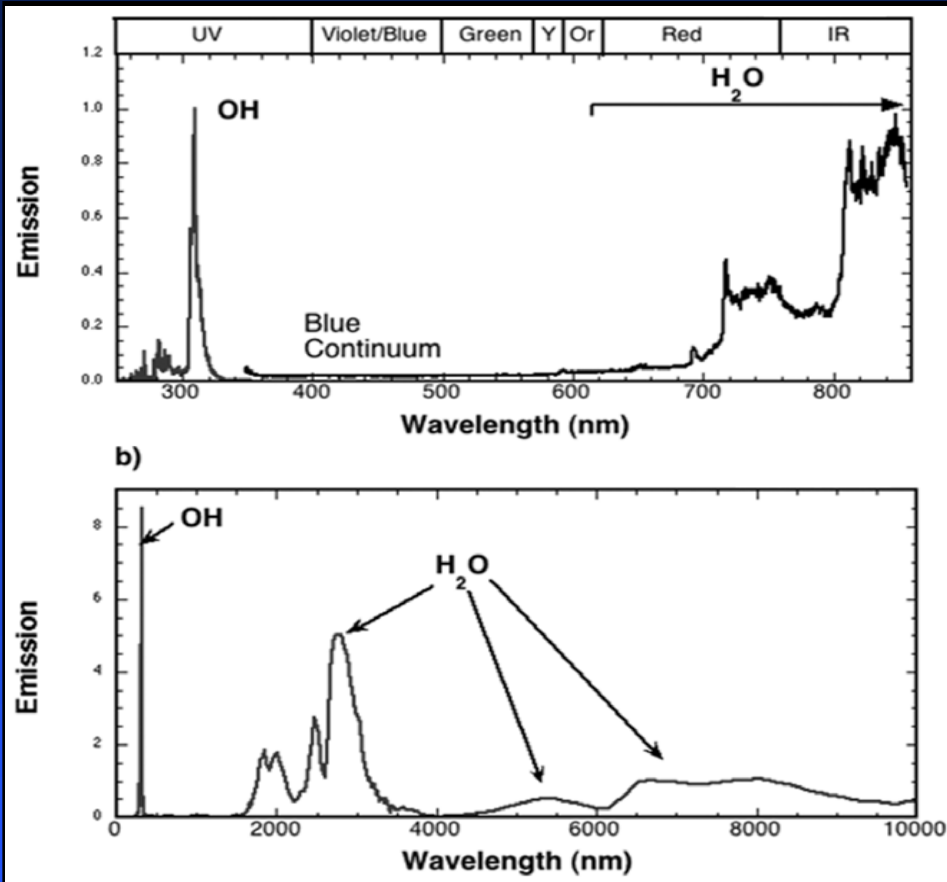
Hydrogen jets and flames are similar to other flammable gases



- ⇒ Fraction of chemical energy converted to thermal radiation
- ⇒ Radiation heat flux distribution
- ⇒ Jet length



H₂ Flame Radiation

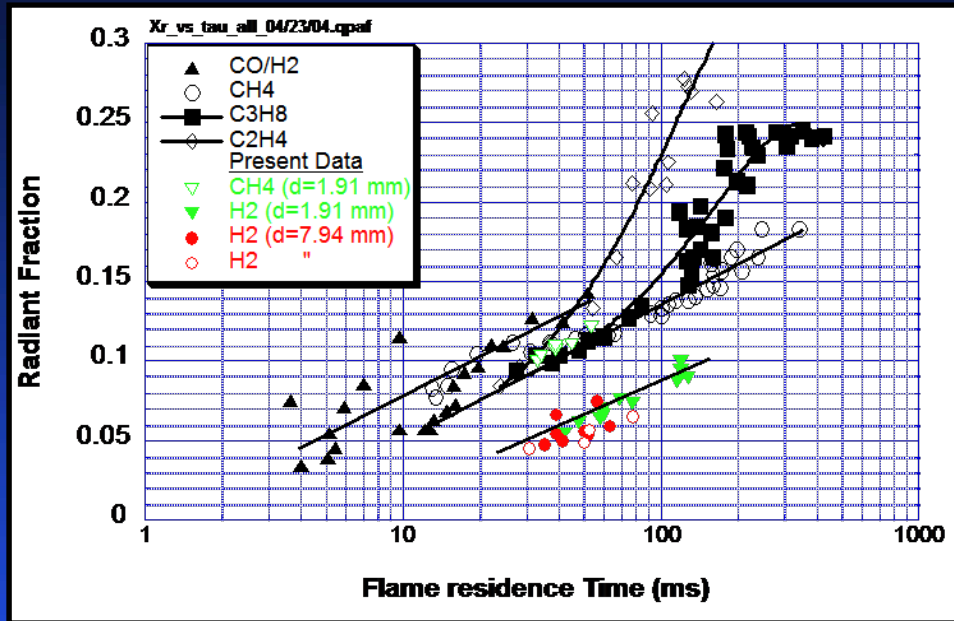


- ⇒ Orange emission due to excited H₂O vapor
- ⇒ Blue continuum due to emission from OH + H ⇒ H₂O + hν
- ⇒ UV emission due to OH*
- ⇒ IR emission due to H₂O vibration-rotation bands

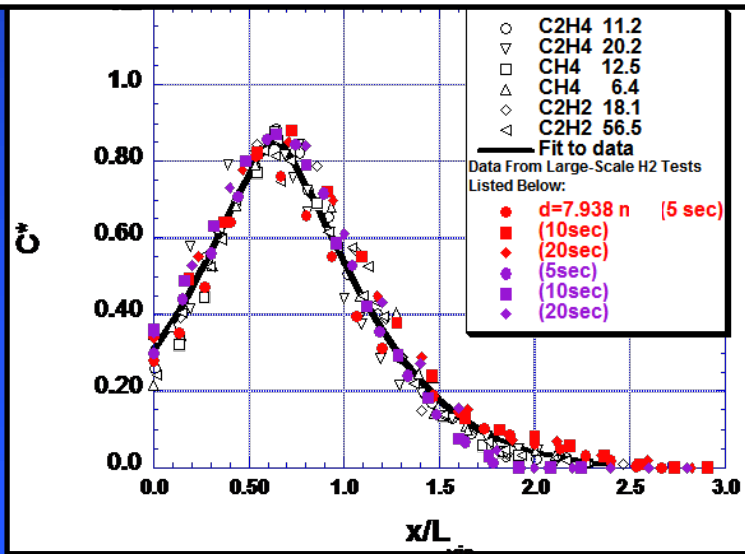
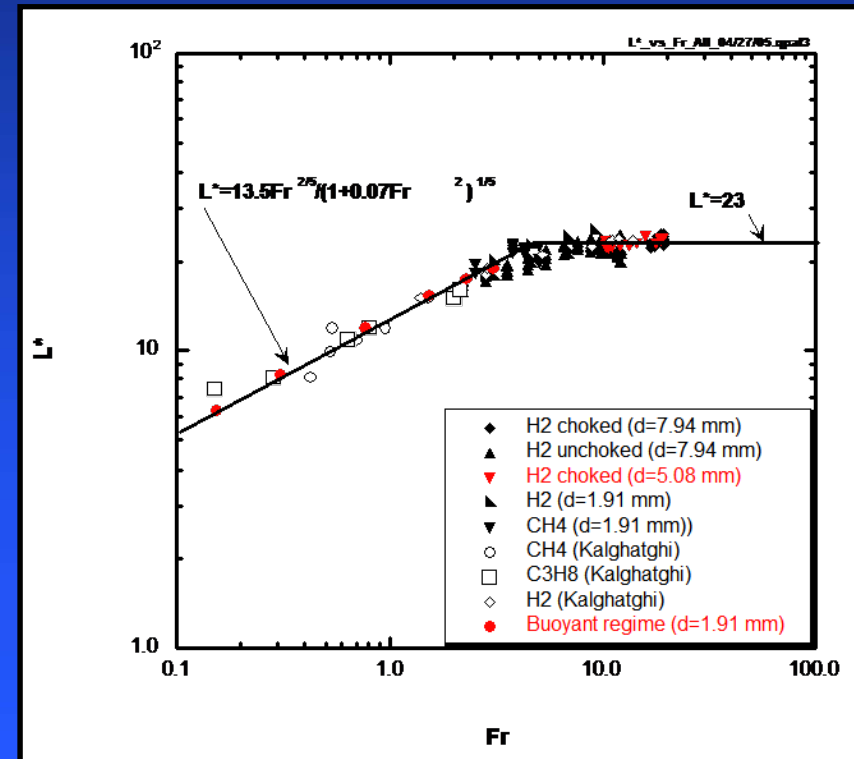
H₂O emission in IR accounts for 99.6% of flame radiation



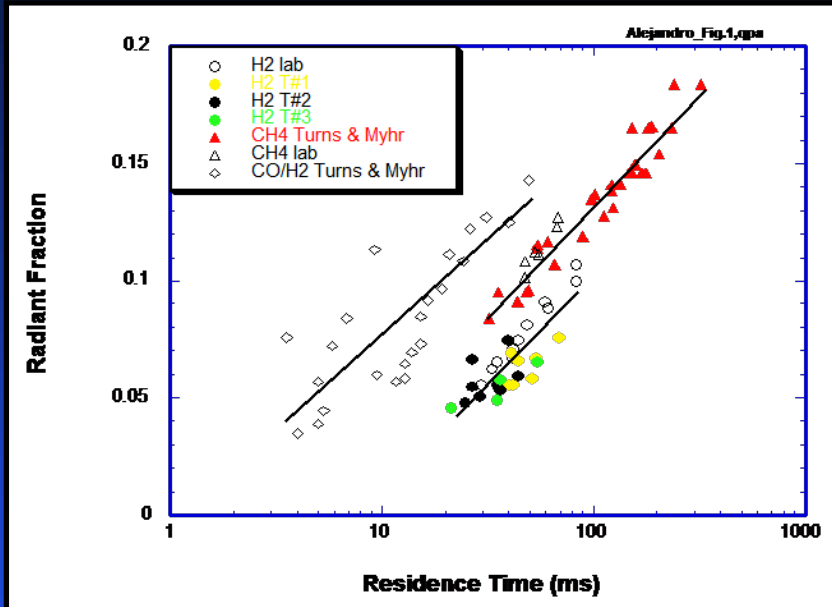
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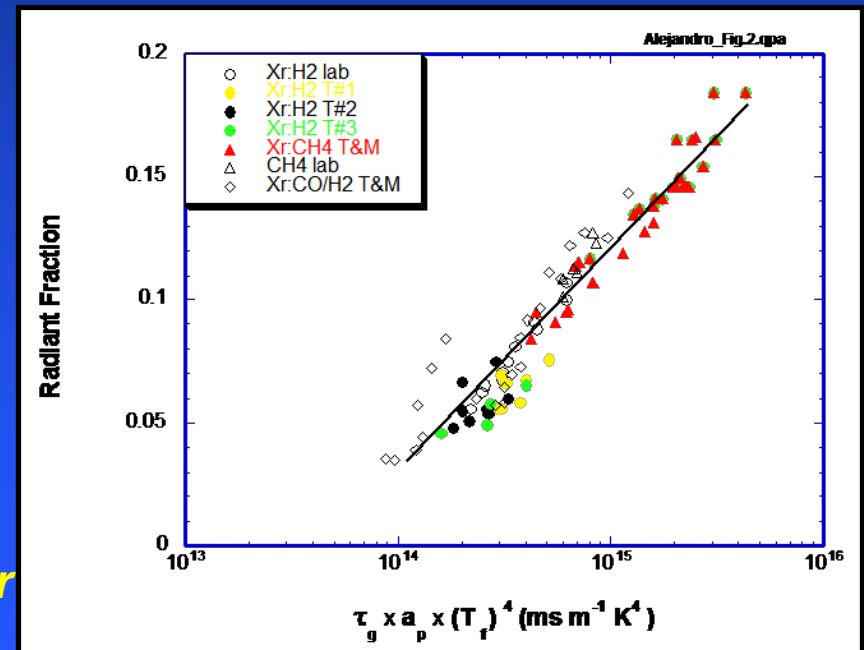


Thermal Radiation from Hydrogen Flames



- ⇒ Radiation heat flux data collapses on single line when plotted against product $\tau_G \times a_p \times T_f^4$.
- ⇒ a_p (absorption coefficient) is factor with most significant impact on data normalization
- ⇒ **Plank mean absorption coefficient for different gases must be considered**

- ⇒ Previous radiation data for *nonsooting* CO/H₂ and CH₄ flames correlate well with flame residence time.
- ⇒ Sandia's H₂ flame data is a factor of two lower than the hydrocarbon flame data.



Hydrogen Myths



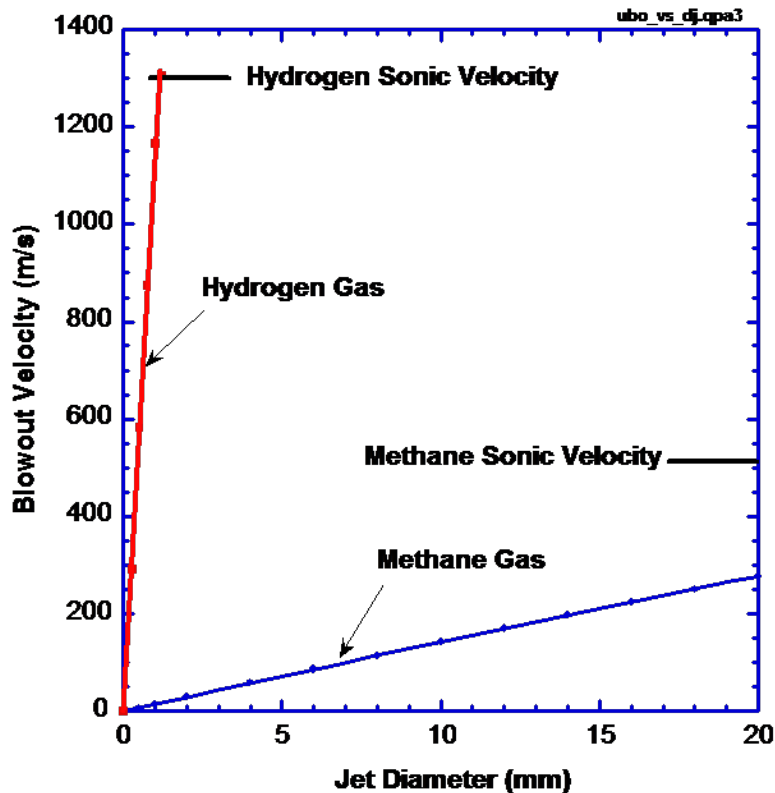
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- ⇒ Simply adding hydrogen to natural gas improves engine efficiency and lowers emissions.
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Comparisons of NG and H₂ Behaviors



Comparison of Blow-Off Velocities for Hydrogen and Natural Gas



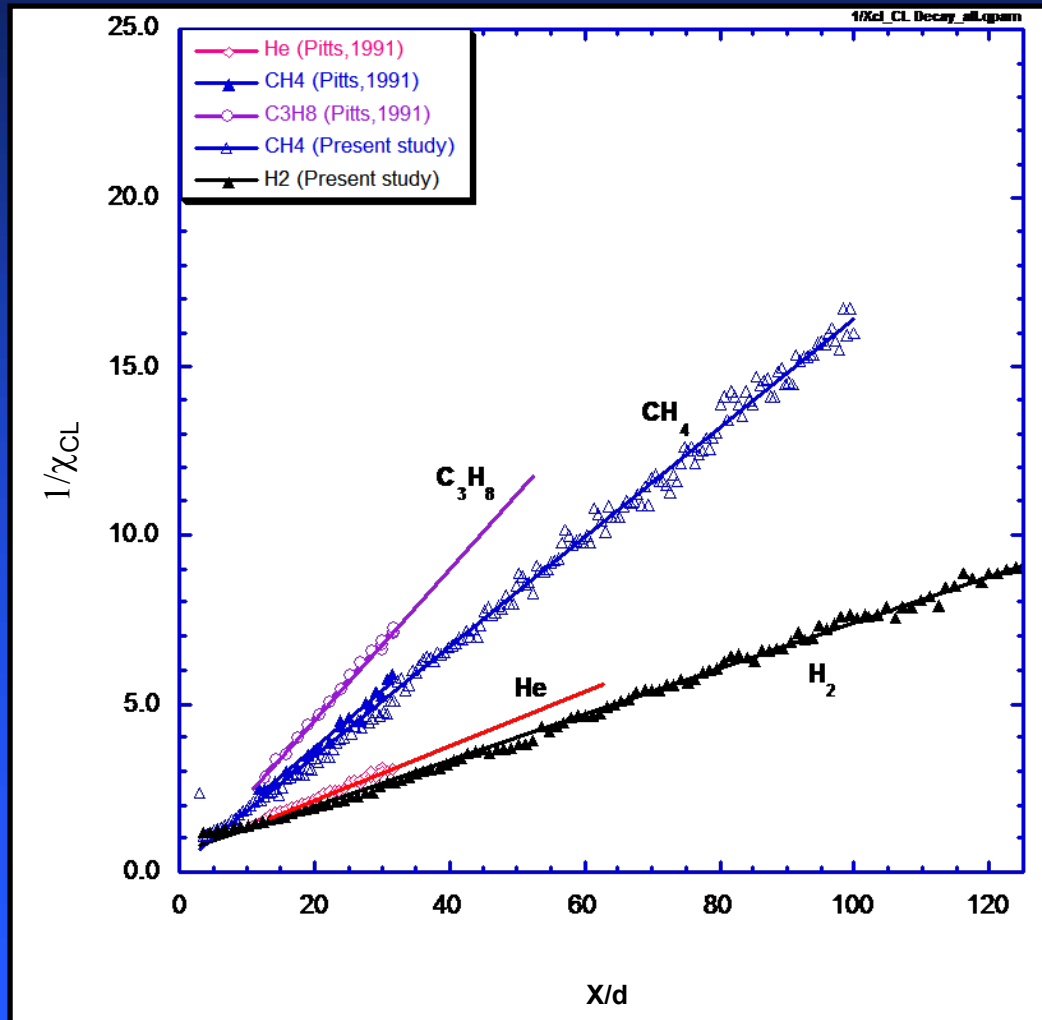
- ⇒ Assume 3.175 mm (1/8 inch) dia. hole
- ⇒ Unignited jet lower flammability limits
 - LFL H₂ - 4% mole fraction
 - LFL NG - 5% mole fraction
- ⇒ Flame blow-off velocities for H₂ are much greater than NG
- ⇒ Flow through 1/8" diameter hole is choked
 - $V_{\text{sonic}} = 450$ m/sec for NG (300K)
 - $V_{\text{sonic}} = 1320$ m/sec for H₂ (300K)
- ⇒ Hole exit (sonic) velocity for NG is **greater** than NG blow-off velocity
 - No NG jet flame for 1/8" hole
- ⇒ Hole exit (sonic) velocity for H₂ is **much less** than blow-off velocity for H₂
 - H₂ jet flame present for 1/8" hole



Small Unignited Releases: Momentum-Dominated Regime



Data for round turbulent jets



- ⇒ In momentum-dominated regime, the centerline decay rate follows a $1/\chi_{CL}$ dependence for all gases.
- ⇒ The mole fraction centerline decay rate increases with increasing molecular weight.

The decay rate for H₂ is significantly slower than for methane and propane.



Unignited jet concentration decay distances for NG and H₂.



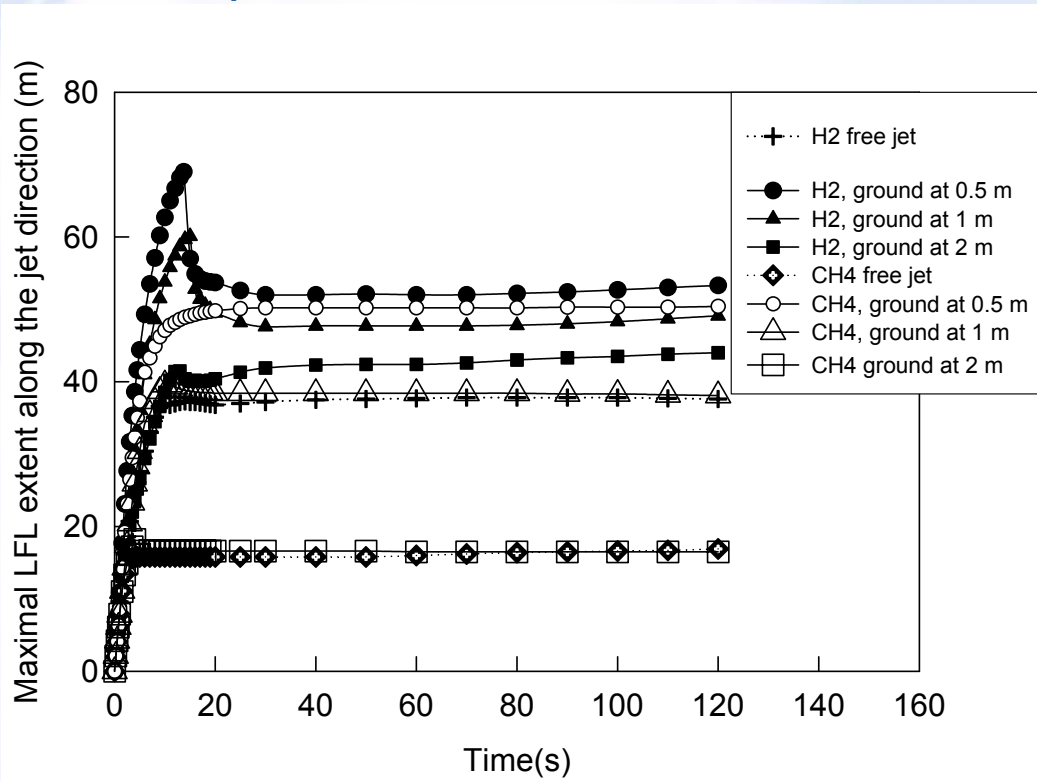
Distance on Jet Centerline to Lower Flammability Limit for Natural Gas and Hydrogen

Tank Pressure	Hole Diameter	Distance to 5% Mole Fraction Natural Gas	Distance to 4% Mole Fraction. Hydrogen
18.25 bar (250 psig)	3.175 mm (1/8 inch)	1.19 m (3.90 ft)	4.24 m (13.91 ft)
	1.587 mm (1/16 inch)	0.59 m (1.93 ft)	2.12 m (6.95 ft)
207.8 bar (3000 psig)	3.175 mm (1/8 inch)	3.92 m (12.86 ft)	13.54 m (44.42 ft)
	1.587 mm (1/16 inch)	1.96 m (6.43 ft)	6.77 m (22.21 ft)

Distance to the lower flammability limit for hydrogen is about 3 times longer than for natural gas



Maximum LFL Extents vs Time – Horizontal H₂ and CH₄ Jets along Horizontal Surface



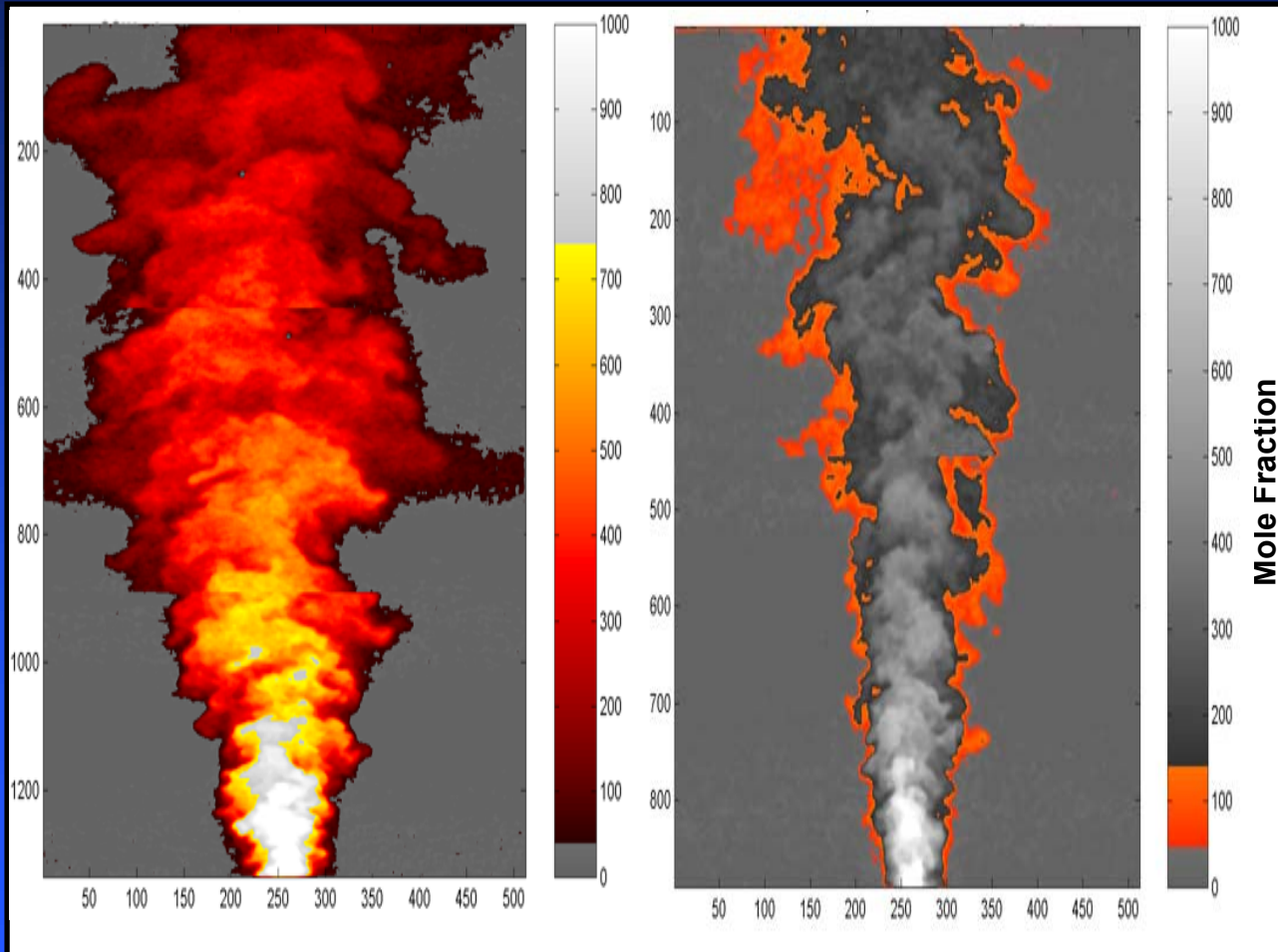
- ❑ Features: strong transient overextent of the hydrogen cloud (larger than at steady state), not observed in case of methane
- ❑ Much stronger effect of surface on methane jets vs hydrogen jets:
 - ✓ While methane free jet max LFL extent is almost 3 times shorter than that of hydrogen, at 0.5 m above ground the max LFL extents for both gases become almost equal

Small Unignited Releases: Ignitable Gas Envelope



H₂ Jet at Re=2,384; Fr = 268

CH₄ Jet at Re=6,813; Fr = 478



→ H₂ flammability limits: LFL 4.0%; RFR 75%

→ CH₄ flammability limits: LFL 5.2%; RFR 15%

Radial profiles in H₂ jet, d = 1.91 mm, Re = 2384



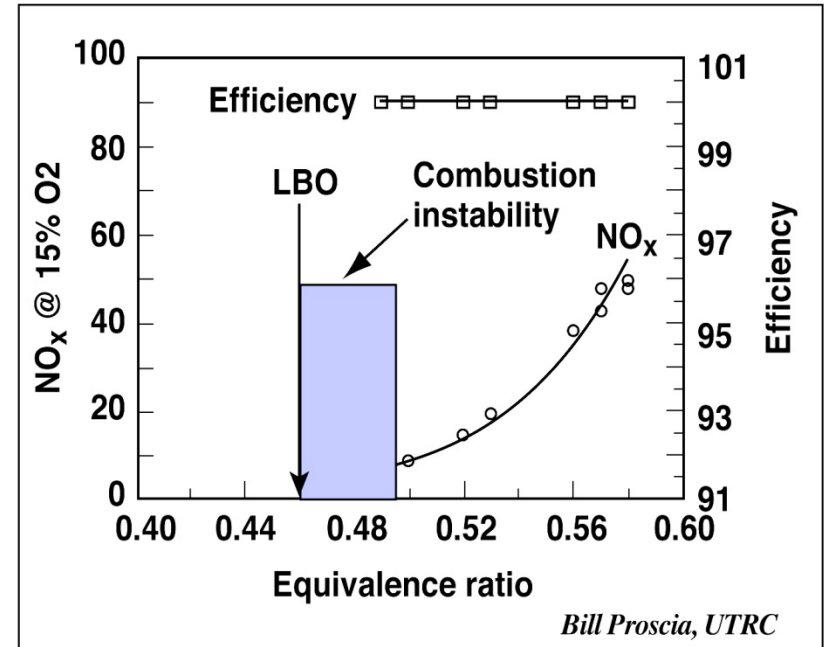
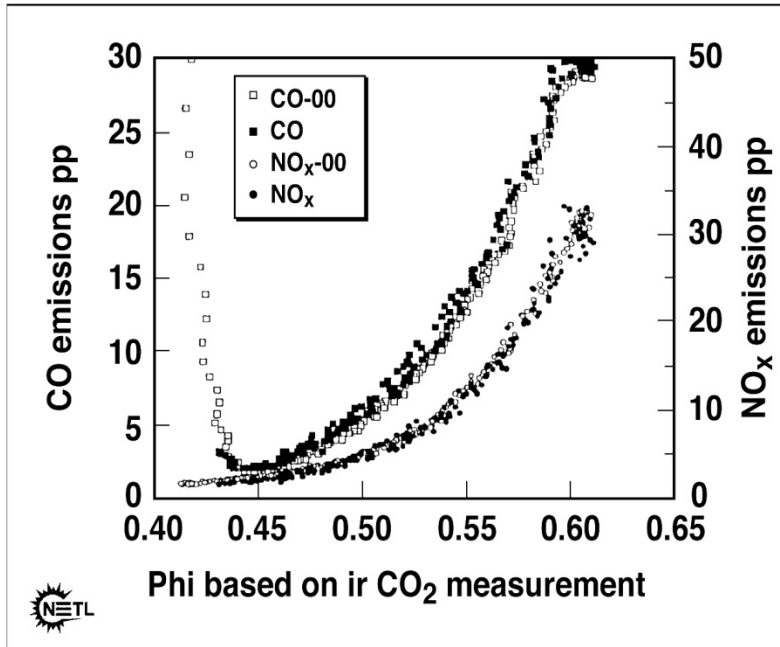
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Lean Premixed Combustion for NO_x Control



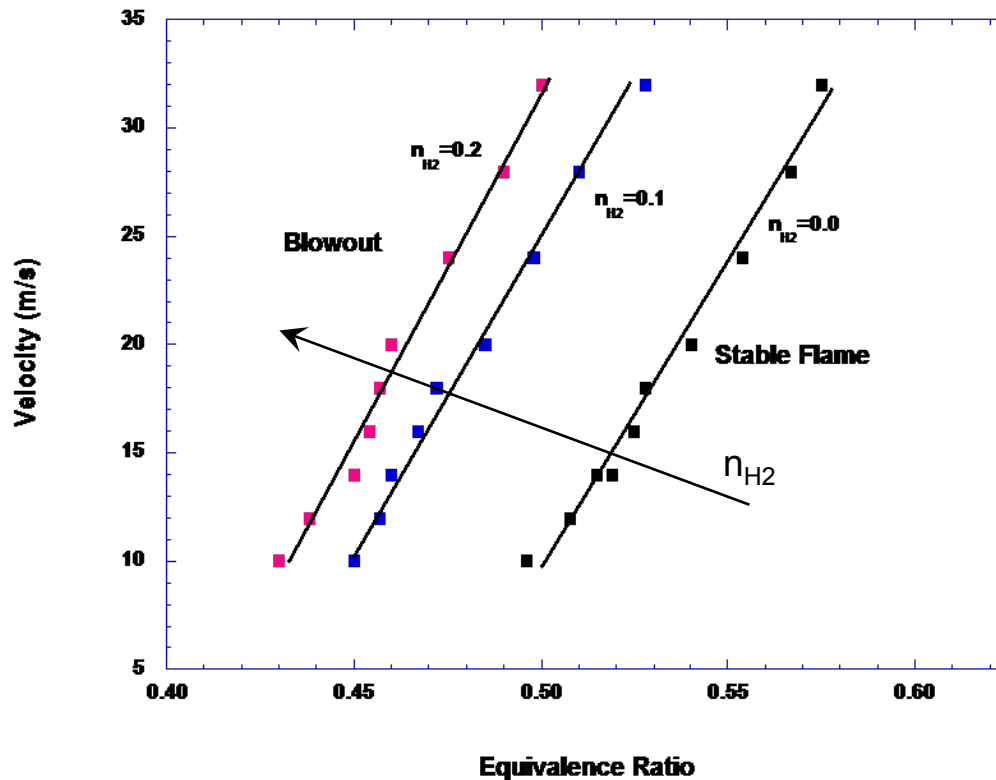
At ultra lean conditions a tradeoff exists between NO_x and CO emissions

Ultimately, lean operation is limited by the onset of flame instability and blowout

H₂ addition extends lean flammability limit and reduces CO emissions



Effect of Hydrogen -Enrichment on Flame Stability



- ⇒ Data points indicate minimum equivalence ratio at which a stable flame can be maintained
- ⇒ Hydrogen addition significantly extends lean flame stability
- ⇒ Expected NO_x levels less than 3 ppm can be achieved with 20 % hydrogen addition



Present Day H₂ICEs: Emissions

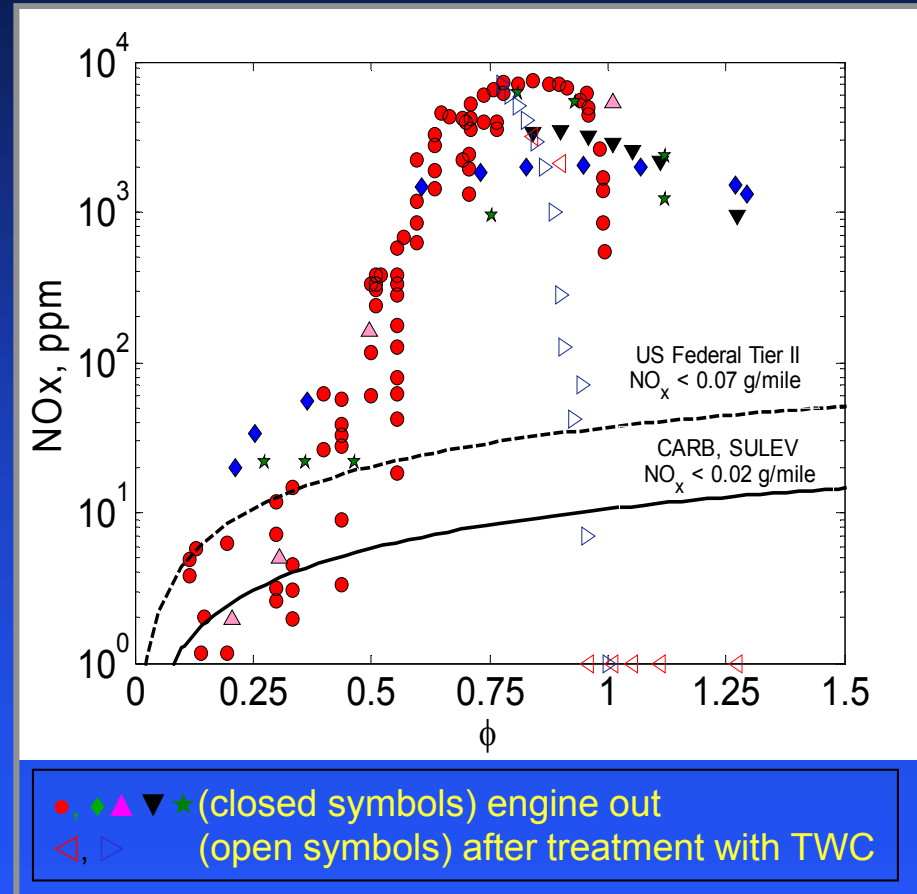


⇒ NO_x is the only non-trivial engine-out emission pollutant

- Engine out Dial-a-NO_x value ~ 5-6 ppm
- With after-treatment NO_x values can be near zero ***
 - Measured tailpipe NO_x emissions equal to ambient levels of about 50 ppb

⇒ HC, CO all near zero engine-out emissions **

- Trace amounts from lubricating oil
 - CO – O(1) ppm, HC – O(5) ppm for a reduction of a factor of 1000, 250 respectively compared to gasoline



*BMW presentation @ 2006 National Hydrogen Association Meeting March, 2006

**SAE Papers #'s 2002-01-0240 thru 0243 and 2003-01-0631; Ford Research

***James Heffel, University of California, Riverside, College of Engineering – Center for Environmental Research and Technology (CE-CERT); Personal Communication; Under the technical guidance and contract to Sandia National Laboratories, funding from the Hydrogen Program Office; OPT



Hydrogen Myths

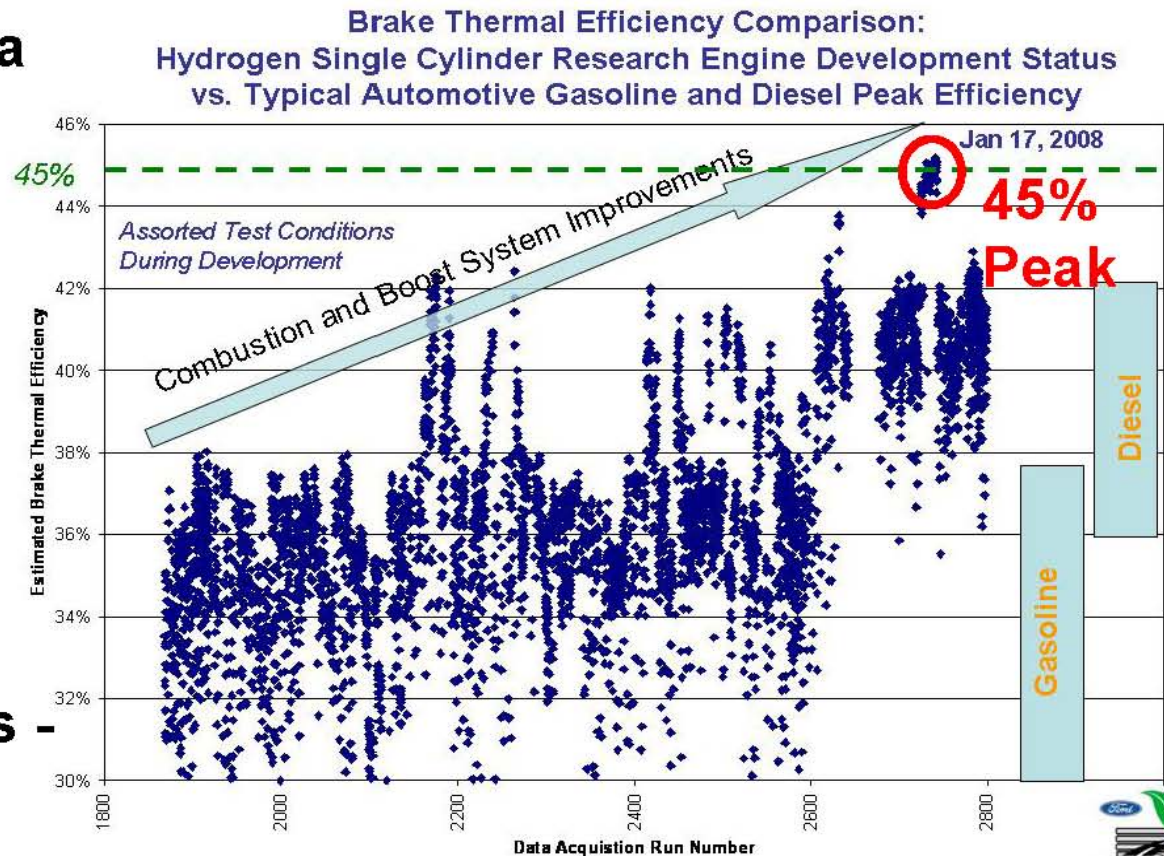


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Brake Thermal Efficiency Trend – Projected to Multi-cylinder Engine

- 3+ years of data
- Different injection rates
- Mostly 1500-4500 RPM
- Naturally aspirated & boosted.
- Discrete clouds - different boost condition.



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Proposed Mechanisms for Spontaneous Ignition



⇒ 81 ignitions of H₂ releases have been reported (MHIDAS database). In 11 cases the ignition source was identified (flame, electric, hot surface). *In the remaining 70 no ignition source could be identified.*

⇒ Proposed causes include the following:

- Joule-Thomson
- *Static charge buildup in the flow*
- *Shock heating that leads to ignition of H₂/air mixtures*
- *Catalytic reaction with materials present in the flow (iron oxide)*
- *Friction heating of particulates / hot surface ignition*

Area of current research



Proposed Mechanisms for Spontaneous Ignition



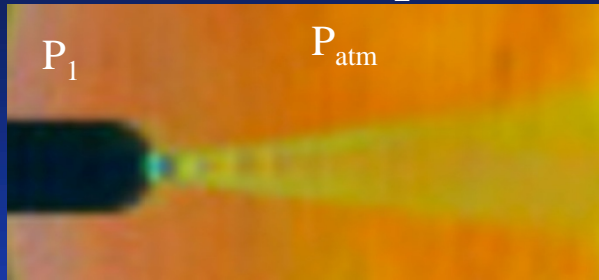
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Joule-Thomson Effect

High-pressure H₂ Jet



⇒ For initial compressed gas pressure of 14 MPa, the estimated temperature rise is approximately ~6 K.

⇒ At pressures up to 250 MPa, the maximum estimated coefficient is 0.53 K/MPa. Thus, at future H₂ storage pressures of 100 MPa, the maximum temperature rise would be 53 K.¹ If the initial temperature was 300K (room temperature) the exit gas temp would be 353K

¹Astbury and Hawksworth, "Spontaneous ignition of hydrogen leaks: postulated mechanisms", International Conference on Hydrogen Safety, Pisa, Italy (2005).

⇒ The direction and magnitude of temperature change is determined by the Joule-Thomson coefficient which is a function of upstream pressure (P₁).

$$\mu_{JT} = (\delta T / \delta P)_H = (\Delta T / \Delta P)_H$$

⇒ Above the inversion temperature, the expanding gas temperature increases.

⇒ The inversion temperature of H₂ is between 28 and 200 K (depending on pressure); at ambient temperature the expanding H₂ increases in temperature.

Given the H₂ auto-ignition temperature of 858 K, Joule-Thomson heating is insufficient to cause ignition





Acknowledgements

The author wishes to recognize the DOE Hydrogen Codes and Standards program element and Antonio Ruiz, the Technical Program Manager, for supporting most of the work reported on here.

The author also wishes to recognize the following people for their contribution to the science discussed in this presentation who are not otherwise recognized in the reference list.

Benard, Pierre, Universite du Quebec a Trios-Rivieres
Evens, Greg; Sandia National Laboratories
Groethe, Mark; SRI International
Houf, Bill; Sandia National Laboratories
Merilo, Eric; SRI International
Moen, Chris; Sandia National Laboratories
Schefer, Bob; Sandia National Laboratories
Tchouvelev, Andrei, Tchouvelev & Associates
White, Chris, Sandia National Laboratories



Publication list



11.3 m

Nighttime photograph of ~40 MPa large-scale H₂ jet-flame test ($d_j = 5.08\text{mm}$, $L_{\text{vis}} = 10.6\text{ m}$) from Sandia/SRI tests.

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Some people just do not get it!

⇒ H_2

➤ is not toxic,

➤ it is environmentally benign

➤ we just borrow it -- ($2H_2O + E \rightarrow 2H_2 + O_2$; then $2H_2 + O_2 \rightarrow 2H_2O + E$)

⇒ H_2 is a fuel and as such has stored chemical energy

➤ It has hazards associated with it

• It is no more dangerous than the other fuels that store chemical energy

• IT IS JUST different; -- ***WE UNDERSTAND THE SCIENCE***

Following NFPA 52 / 2 - Hydrogen Installations are no more dangerous than current refueling stations.

