Project ID # SCS010

R&D for Safety Codes and Standards: Hydrogen Behavior

Sandia National Laboratories

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DOE EERE FCT Annual Merit Review
May 15, 2015

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Overview

Timeline

- Project start date: Oct. 2003
- Project end date: Sept. 2015
- Percent complete: *80%*

Barriers (2012 MYRD&D)

- F. Enabling national and international markets requires consistent RCS
- G. Insufficient technical data to revise standards
- L. Usage and Access Restrictions – parking structures, tunnels and other usage areas

Budget

- Total project funding (to date)
  - DOE share: $15.4M ($13.6M)*
- FY11 Funding: $1.3M ($1.2M*)
- Planned Funding in FY12: $0.8M ($0.7M*)

*(*R&D core, no IEA contracts)*

Partners

Industry: Air Products, Lincoln Composites, HIPOC, FCHEA, CTFCA, SRI

Govt: NREL, CSTT, NIST, PNNL

SDO/CDO: NFPA, ICC, ISO, CSA, SAE, ASME

International: IEA, Longitude 122 West, HYPER, HySAFE, IPHE, I2CNER

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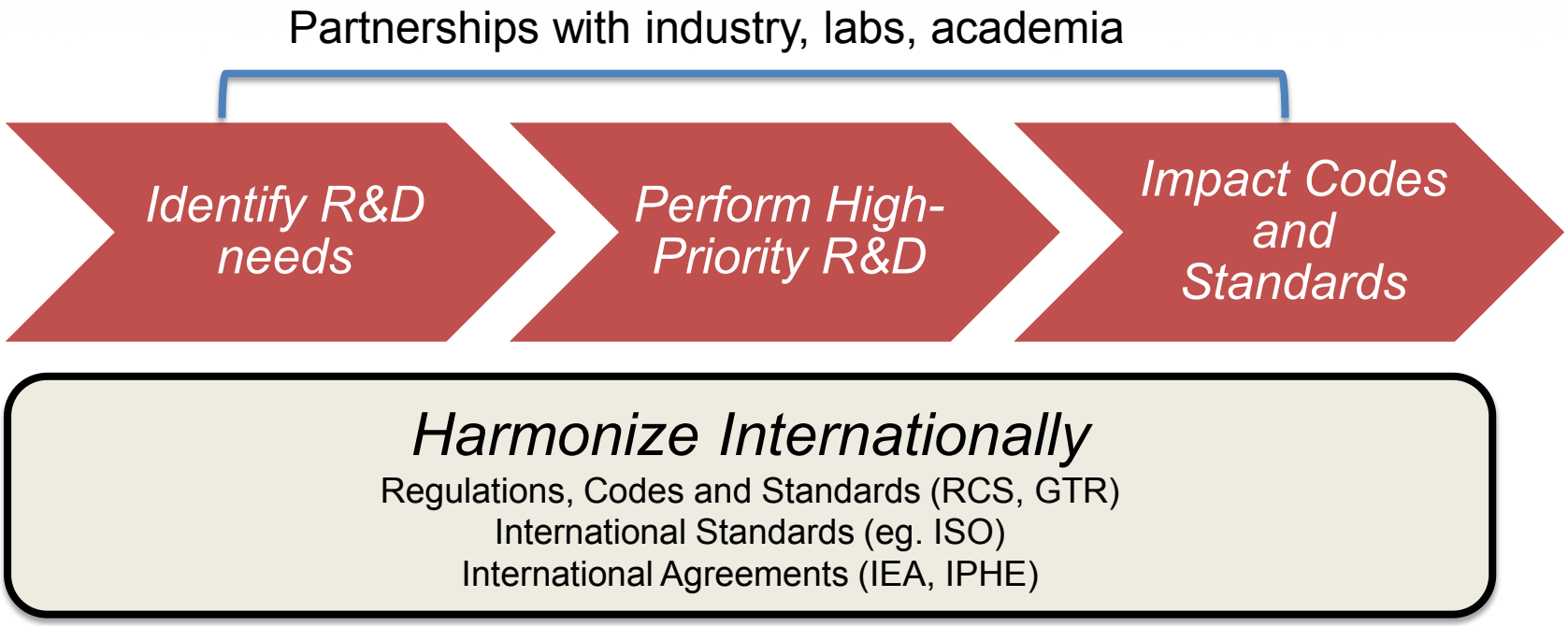
Coordinate critical stakeholders and research to remove technology deployment barriers



- Metrics for Success
  - Number of codes, standards, regulations impacted
  - Degree of harmonization

Enhanced Risk Evaluation

Risk conventionally defined as: Frequency × Consequences

A more detailed definition relates **harm probability** to **accident occurrence** as:

$$Risk \propto \sum_{i,j,k} P(\text{Release}_i) P(\text{Ignition}_j | \text{Release}_i) P(\text{Hazard}_k | \text{Ignition}_j \cap \text{Release}_i) P(\text{Harm} | \text{Hazard}_k)^{\dagger}$$

- Large potential permutations that strongly depend on release scenarios
- Assignment solely from incident statistics is prohibitive

Terms currently approximated through the use of lookup tables, e.g.,

Hydrogen Ignition Probabilities

Tchouvelev			HYSAFE	
Hydrogen Release Rate [kg/s]	Immediate Ignition Probability	Delayed Ignition Probability	Hydrogen Release Rate [kg/s]	Immediate Ignition Probability
<0.125	0.008	0.004	0.01-0.1	0.001
0.125-6.25	0.053	0.027	0.01-0.1	0.001+0.001 when P>100 bar
>6.25	0.23	0.12	1-10	0.01+0.01 when P>100 bar
			>10	0.1+0.01 or 0.02

However:

- Other factors control ignition probability (e.g., flow velocity, ignition mechanisms, mixing)
- Ignition does not have uniform hazard criteria
- No insight into risk reduction strategies possible

Deterministic modeling enhances probability quantification & provides physical insight for focused risk reduction strategies.

† $P(A|B \cap C)$ is the conditional probability that event A occurs for given events B and C.

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Objectives:

Project Milestones

SNL R&D for H_2 Safety, Codes & Standards (SCS)

Develop Science Basis for H_2 SCS

- Risk-Informed R&D
- Hydrogen Effects in Structural Materials
- **Hydrogen Behavior**
  - **Fast fill experiments/modeling**
    - Validation experiments for SAEJ2601 fill protocols
  - **Release & ignition experiments/modeling**
    - Choked flow dispersion model development
    - Qualitative high-speed flame ignition imaging
    - Flame light-up engineering model development
  - **Validated consequence modeling**
    - Large-scale jet flame radiation analysis

Harmonize H_2 SCS Development

- **Consequence & Risk:**
  - Participation in: HIPOC, ISO, NFPA, ICC
- **International Engagement:**
  - Participation in: ISO, IEA, SAE, IPHE, GTR, H2CAN

Hydrogen Behavior Project milestones

Standards advocacy ensures transfer of science-based H_2 SCS knowledge to code development committees.

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Fast Fill Modeling

Terry Johnson(PI)
SNL
Experiments

Jianjun Ye
Zhejiang University, PRC
CFD Modeling

Partners and Collaborators:
Lincoln Composites and Zhejiang University, PRC

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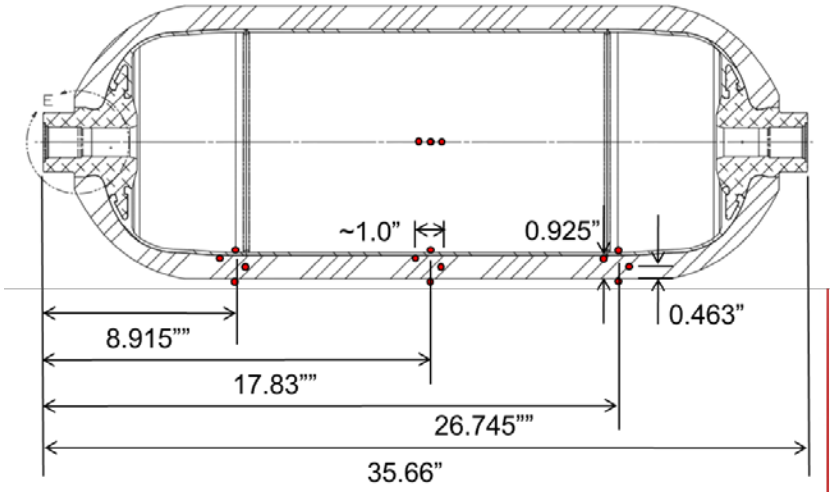
Experimental Approach

We seek to develop validation databases through H_2 fast-fill experiments at specified and relevant pressure ramp rates with measurements of:

- Transient gas tank pressure and mass-averaged tank temperature
- Total hydrogen gas enthalpy and mass flow rate entering the tank
- Final uniform tank temperature/pressure after the fill

- Tank and gas temperatures measured at discrete locations
  - 4 tank wall measurements, each at 4 depths
  - 5 gas temperature measurements
- Inlet & closed end pressure measurement
- Infrared tank exterior temperature imaging



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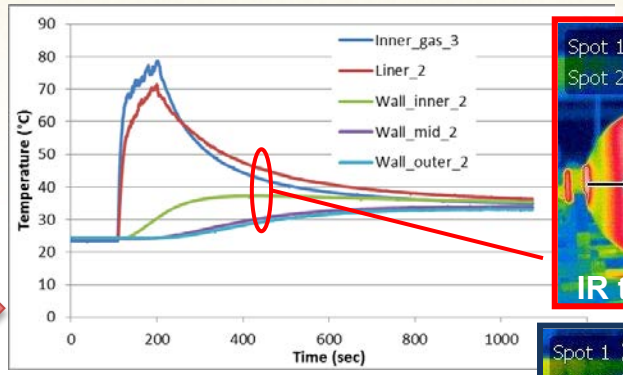
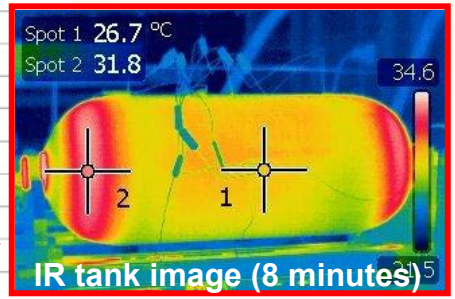
Accomplishments: Experimental Results

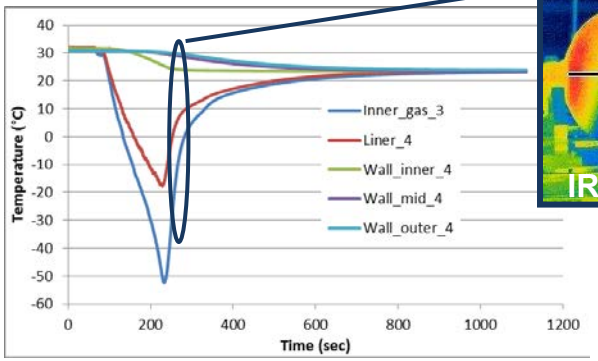
✓ Initial fill and release data collected for model validation

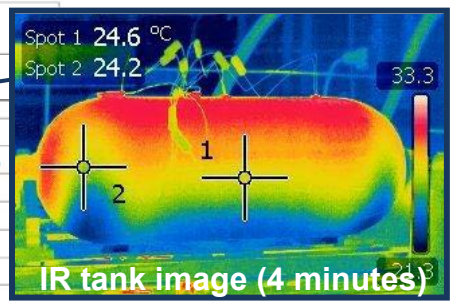
Test #	Initial pressure (psi)	Final pressure (psi)	Fill time (sec)
1	20	1500	60
2	20	1500	90
3	20	1500	150
4	20	1500	300
5	20	1000	90
6	**20**	**2000**	**90**
7	145	2000	90
8	290	2000	90
9	725	2000	90

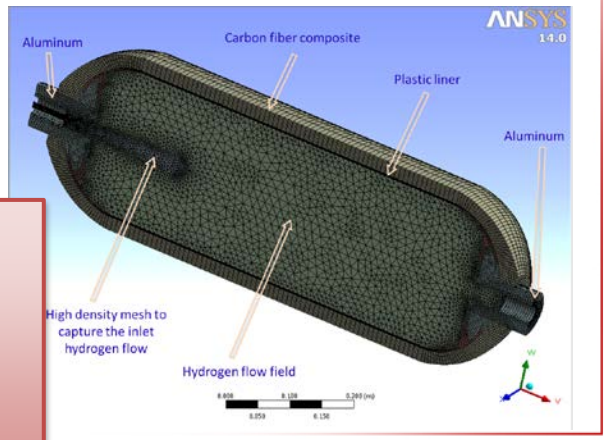
Test #	Initial pressure (psi)	Final pressure (psi)	Release rate (g/sec)
1	1300	20	1.5
2	1300	20	0.75
3	1300	20	1.0
4	**1300**	**20**	**1.9**
5	1700	20	0.4
6	1700	20	1.5
7	1700	20	0.75











Model Results:

- ✓ Model completed, currently in validation
- ✓ *Goal:* results available for SAE Interface Group (J2601) discussion; Sept 2012
- ✓ *Goal:* Comparison with other research and industrial datasets in support of 2015 vehicle deployment

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Broad Approach to Risk Reduction

Risk, as defined in the objectives, is quantified by coupling validated physical modeling with stochastic scenario frequencies.

FY12 Research Activities

Stochastic scenario frequencies: incident data, environmental/human factors, system design/mitigation

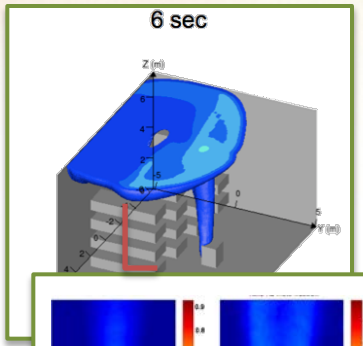
Dispersion
- Permeation
- Buoyant creeping flow
- **Turbulent jet**
- Volumetric rupture

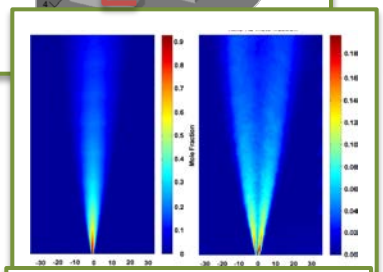
Ignition
- Ignition mechanism
- **Mixture ignitability**
- Ignition delay/location
- **Sustained light-up**

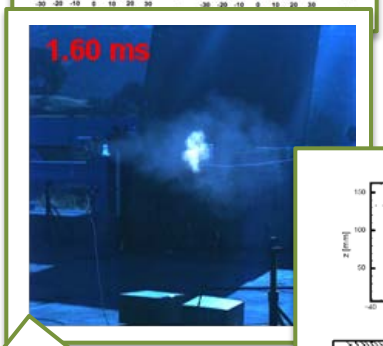
Hazard
- **Flame radiation**
- Overpressure (deflagration/detonation)
- O_2 dilution/depletion

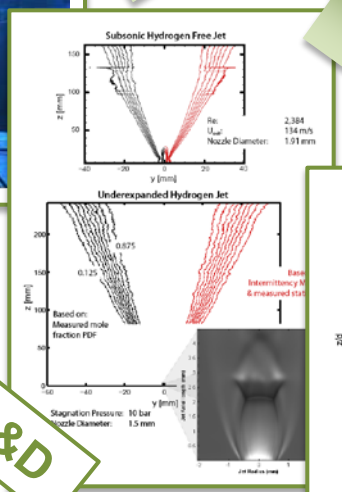
Risk

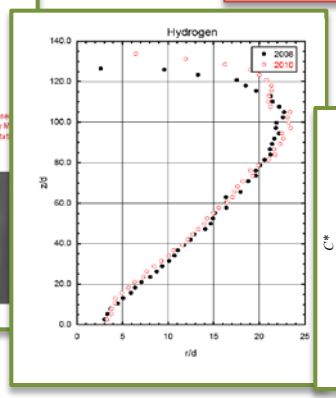
Harm
- Burns
- Lung damage
- Shrapnel wounds
- Building collapse

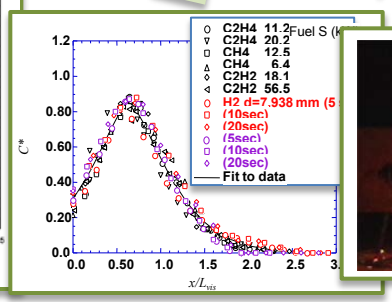


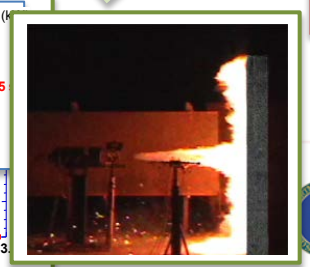












SNL H_2 SCS R&D

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Fundamental Hydrogen Release Behavior

Isaac Ekoto (PI)
SNL
Experiments

Adam Ruggles
SNL
Experiments

Partners and Collaborators:
Université du Québec à Trois-Rivières (CAN)

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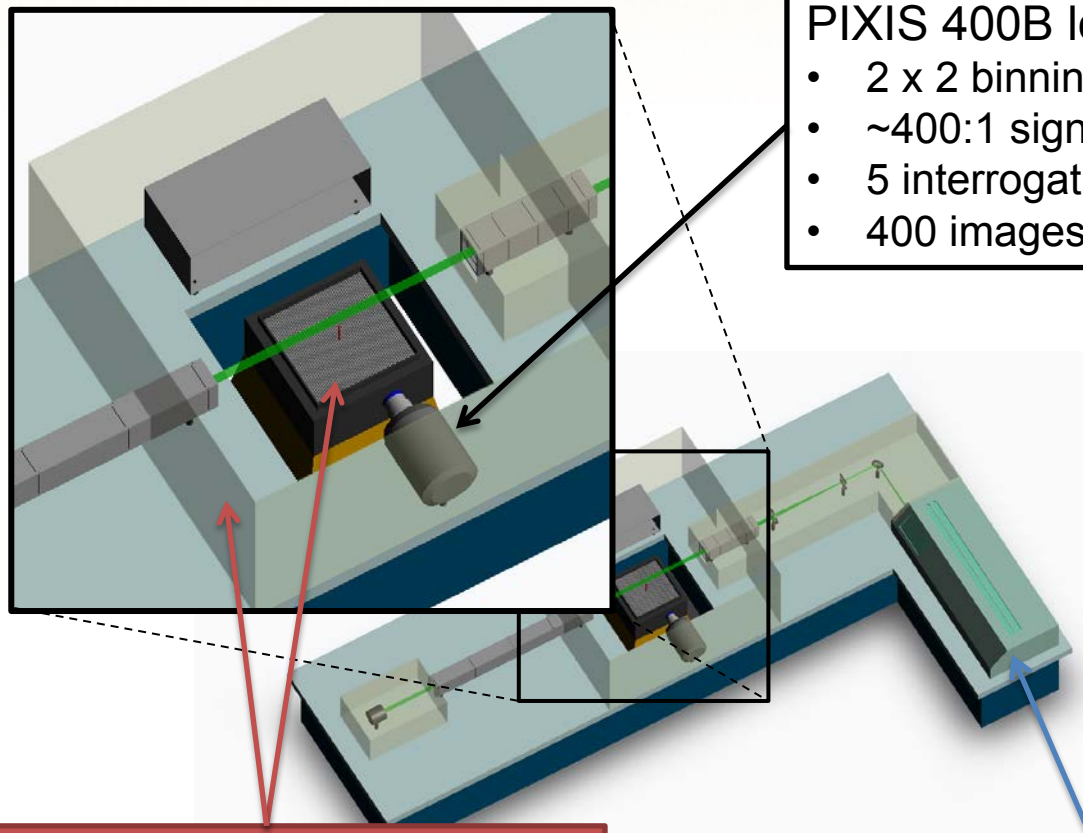
Approach:

Turbulent Jet Experiment

Scalar field of a momentum driven, turbulent H_2 jet was examined via high-resolution Planar Rayleigh Scatter Imaging (PLRS).



PIXIS 400B low noise CCD Camera

- 2 x 2 binning for 3.94 pix/mm resolution
- ~400:1 signal-to-noise
- 5 interrogation regions (37 x 125 mm^2)
- 400 images per interrogation region

$$r_0 = 0.95\ mm$$
$$L_{pipe} = 250\ mm$$
$$Q = 100\ lit/min$$
$$Fr_{den} = 1170$$

Air co-flow & barriers to minimize impact of room currents

High power injection seeded ND: Yag laser (1 J/pulse, 532 nm)

Additional diagnostics include Particle Image velocimetry (PIV), Laser Doppler Velocimetry (LDV) and OH Laser Induced Fluorescence (LIF).

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Accomplishment:

Detailed Concentration Statistics Acquired

Radial statistics collapse when plotted against normalized radial coordinates.

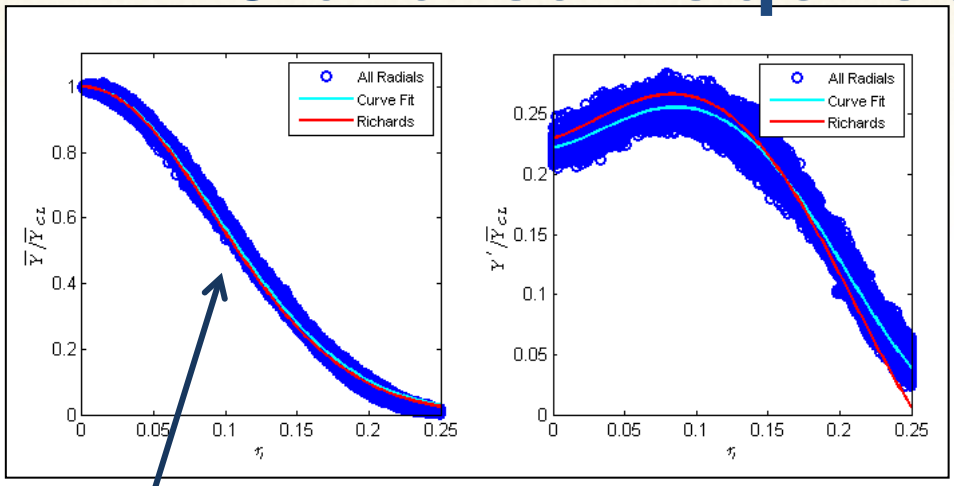
$$\bar{Y} = f(\bar{Y}_{CL}, \eta);\ Y' = g(\bar{Y}_{CL}, \eta)$$

Where,

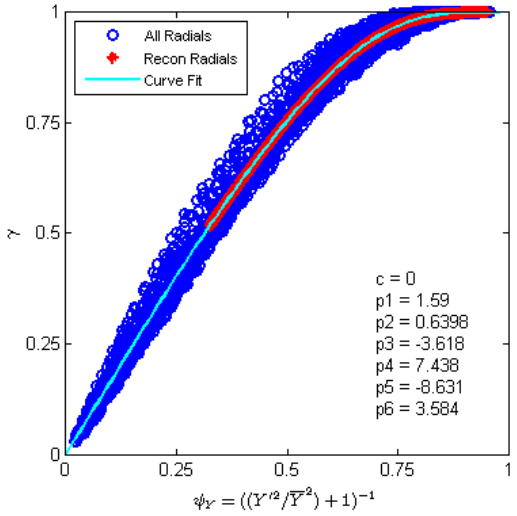
$\eta = \frac{r^*}{(r - z_{0j})}$; normalized radial coordinate

z_{0j}: momentum virtual origin

Richards and Pitts, 1993



✓ Collapsed statistics slightly deviate from traditional models, which impact ignitability predictions embedded in QRA models



Non-linear correlation observed between intermittency and the ratio of the 1st and 2nd statistical moments.

- Contradicts linear relationship often assumed in CFD modeling approaches
- Results impact PDF distribution prediction

✓ A more suitable intermittency correlating parameter is based the ratio of conditioned and non-conditioned mean statistics

✓ Intermittency likewise influences ignitability predictions in QRA models

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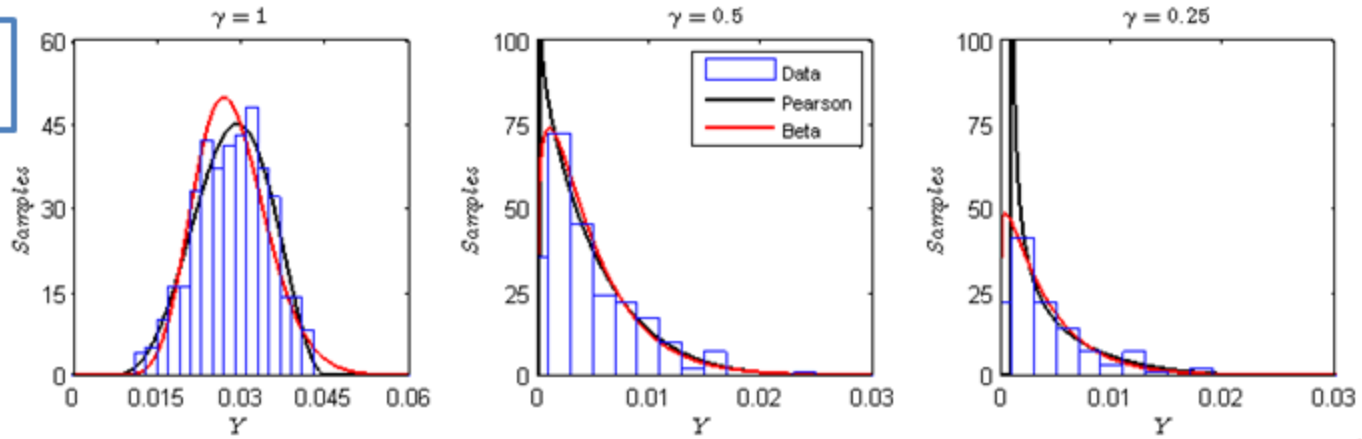
Accomplishment:

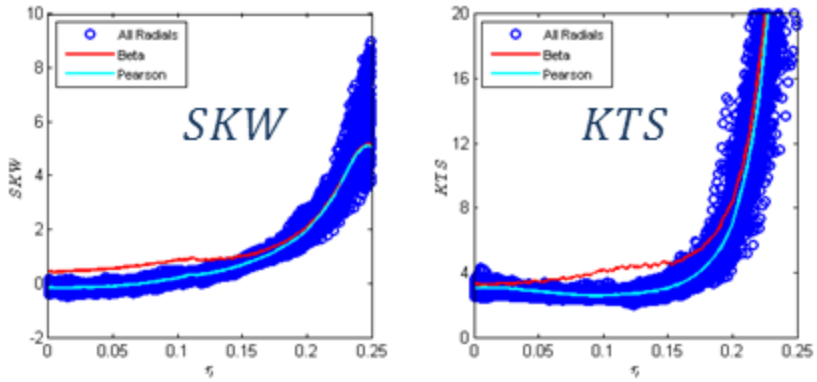
Mixture Ignitability

- Mixture ignitability is a function of the mass fraction distribution (PDF) and jet intermittency.
- PDF is commonly reconstructed from Beta function distributions.

$$P_C(Y_{H2}) = \frac{Y_{H2}^{\alpha-1}(1-YH_2)^{\beta-1}}{B(\alpha,\beta)}, \text{ where } \bar{Y} = \frac{\alpha}{\alpha+\beta}, \text{ and } \overline{Y'Y'} = \frac{\alpha\beta}{(\alpha+\beta)^2(\alpha+\beta+1)}$$

Conditioned probability that neglects $\gamma = 0$ values





- Skewness & Kurtosis are measures of the goodness of measured to modeled PDFs
- Beta fit often worse than a Gaussian fit

✓ Created benchmark data to evaluate optimum distribution functions, that are used to model mixture ignitability

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High-Source Pressure Hydrogen Release Behavior

Isaac Ekoto (PI)
SNL
Experiments

Adam Ruggles
SNL
Experiments

Partners and Collaborators:
Commissariat à l'Energie Atomique (FR)
Joint Research Centre (NL)

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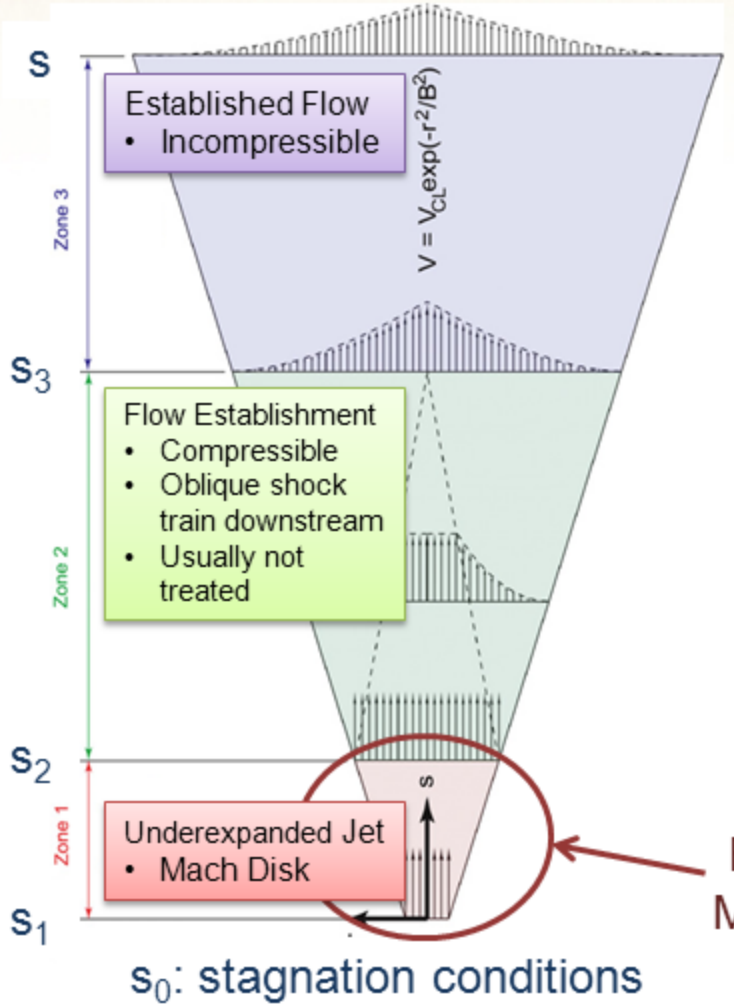
Choked Flow Releases

- Notional nozzle models represent complex underexpanded jet shock structure with an equivalent jet exit diameter.



Notional Nozzle Models

Model	Conservation Equations				Critical Assumptions
	Mass	Momentum	Energy	Entropy	
Birch et al. (1984) Ruggles & Ekoto (2012)†	x		x		$T_2 = T_0$ V_2 = sonic
Ewan & Moodie (1986) Ruggles & Ekoto (2012)†	x		x		$T_2 = T_1$ V_2 = sonic
Molkov (2008)	x		x		V_2 = sonic S_1 (Abel Noble) S_2 (Ideal gas) $T_2 \approx T_1$
Birch et al. (1987) Schefer et al. (2007)†	x	x	x		$T_2 = T_0$ V_2 supersonic
Yüceil & Ötügen (2002) Ruggles & Ekoto (2012)†	x	x	x		V_2 supersonic
Harstad & Bellan (2006) Winters & Houf (2008)†	x	x	x	x	All fluid passes through Mach disk

† Original model updated with Abel-Noble hydrogen equation of state:

$$p = Z\rho RH_2T;\ Z = (1 - b\rho)^{-1}$$

Validation datasets are still needed

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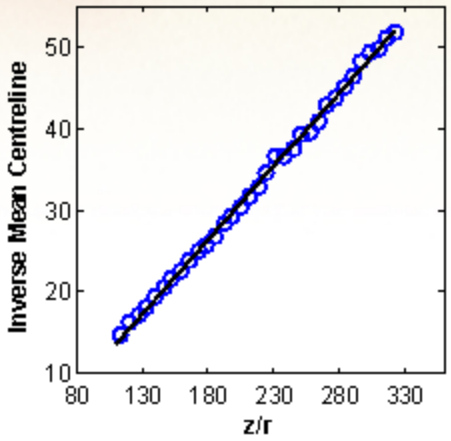
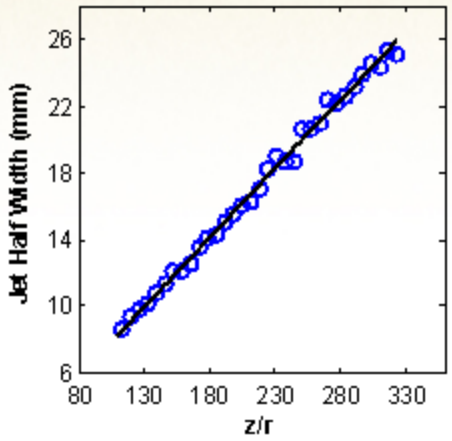
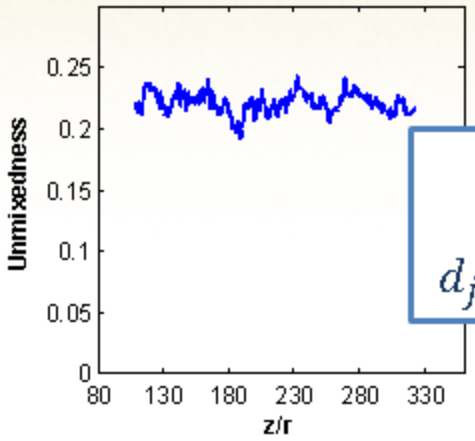
Accomplishment:

Notional Nozzle Concept Validated



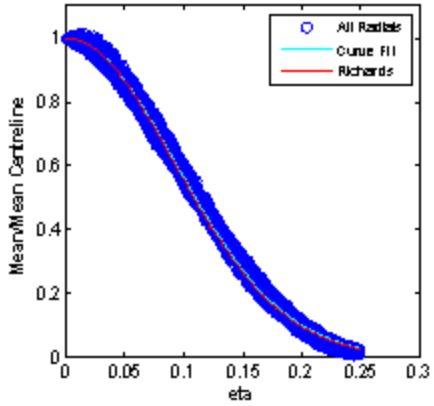


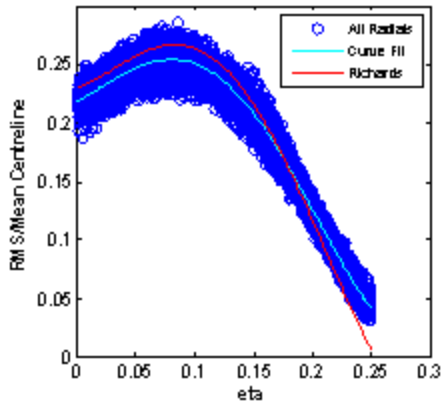


$$\frac{p_0}{p} = 10;$$
$$d_{jet} = 1.5 \text{ mm}$$

Constant centerline decay, jet spreading, and unmixedness observed.

Mass weighted effective diameter ($d^* \equiv d_{eff}\sqrt{\rho_{eff}/\rho_{air}}$) fit to the data so that mass fraction decay rates matched incompressible values.





Radial statistics collapsed when measured d^* was used to normalize radial coordinates.

- ✓ Acquired benchmark d^* values for validation of current notional nozzle models and facilitate development future model
- ✓ Notional Nozzle models are integral to dispersion predictions from QRA models that inform NFPA 2 code development

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Sustained Light-Up Probability

Isaac Ekoto (PI)
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Experiments

Adam Ruggles
SNL
Experiments

Partners and Collaborators:

Sandia National Laboratories

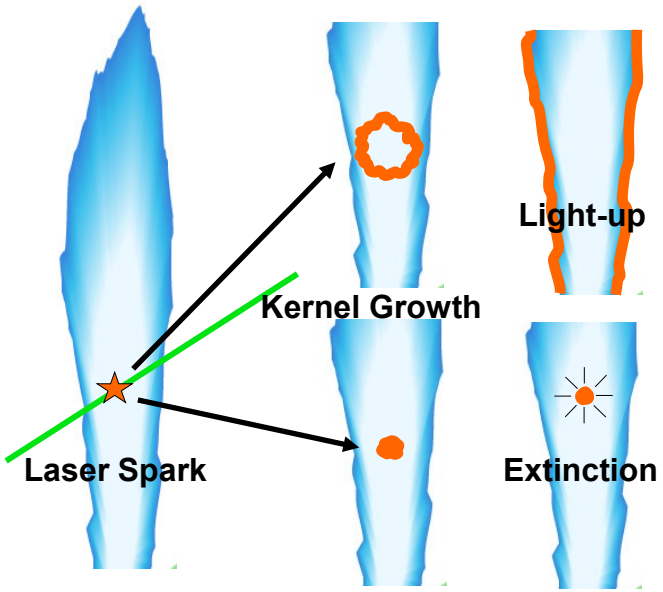
Accomplishment:

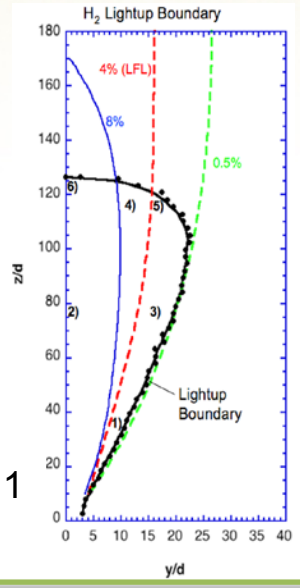
- ✓ Determined flame light-up boundaries from localized ignition sources e.g., ESD ignition

Improved Mixture Measurement

Flame light-up: Mixture ignitability is a necessary but insufficient criterion

Two possible pathways from ignition kernel development:

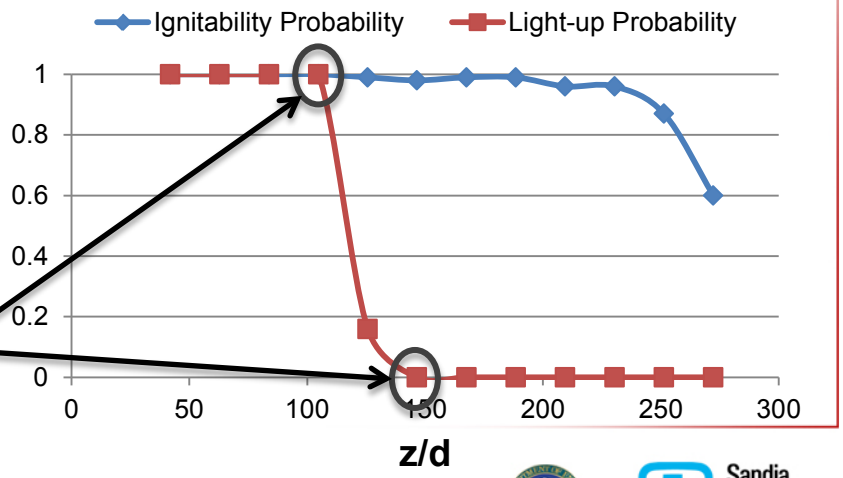




Schefer et al., 2011

Test Case:
1.91 mm tube (unchoked)
100 lit/min flow rate

High-speed visualization



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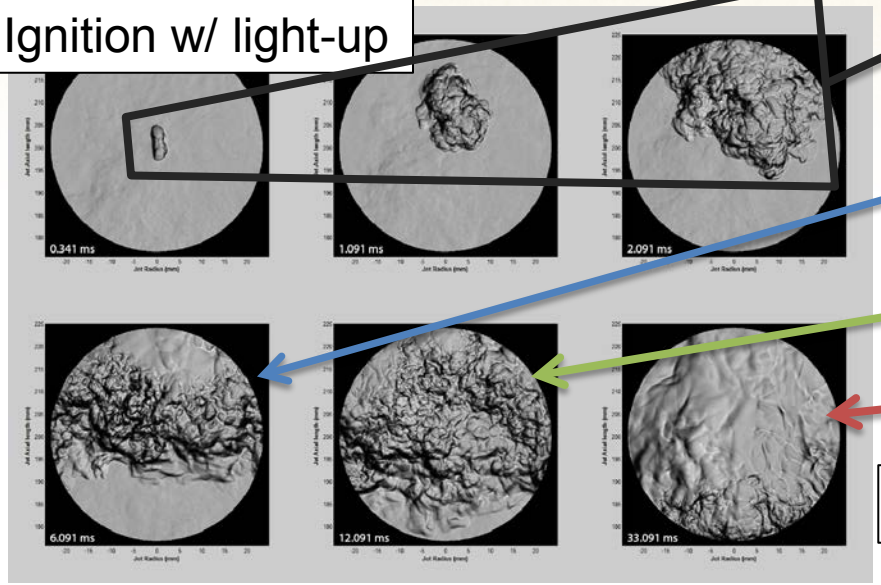
Accomplishment:

Flame Light-Up Imaging

- ✓ High-speed flame ignition imaging elucidates sustained light-up mechanisms.

Ignition w/ light-up



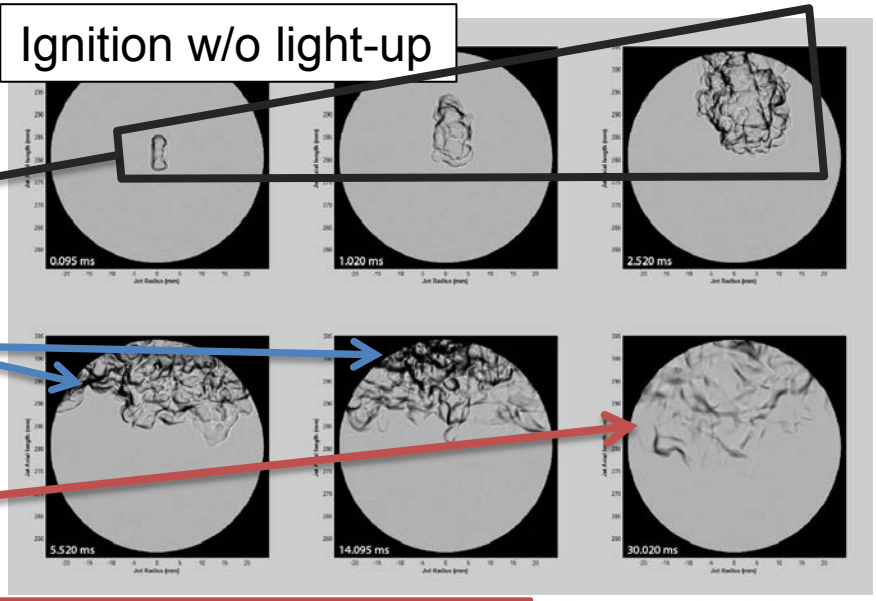
Rapid volumetric ignition kernel growth

Kernel forms into turbulent flame front

Flame front broadens

Front **overcomes** flow convection

Ignition w/o light-up



Slower kernel growth

Thinner, less turbulent flame front

Front **overcome** by convection

- ✓ Imaging observations indicate sustained flame light-up correlates with 1D flame front propagation speeds.

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Flame Radiation

Isaac Ekoto (PI)
SNL
Experiments

Adam Ruggles
SNL
Experiments

Partners and Collaborators:
Air Products and Chemicals Inc.

Sandia National Laboratories

Previous Accomplishment:

Radiation Experiments

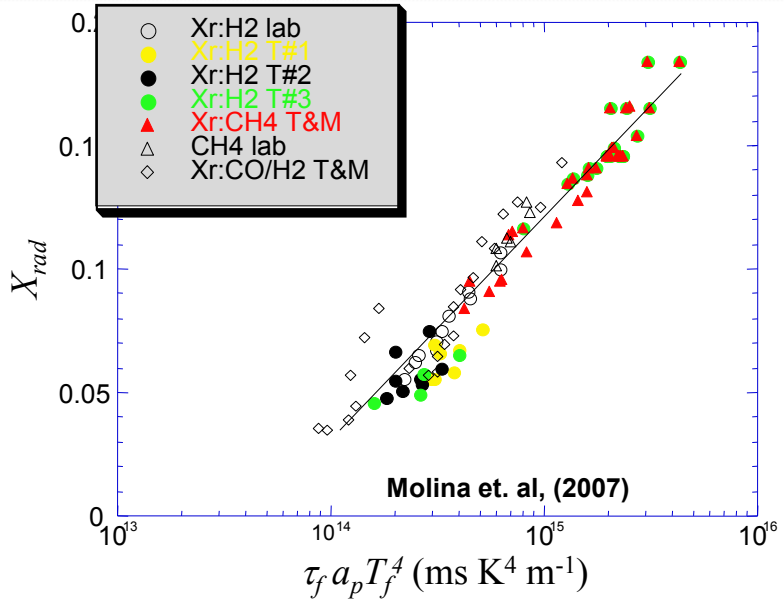
H_2 Flame radiation significantly lower than corresponding HC flames.

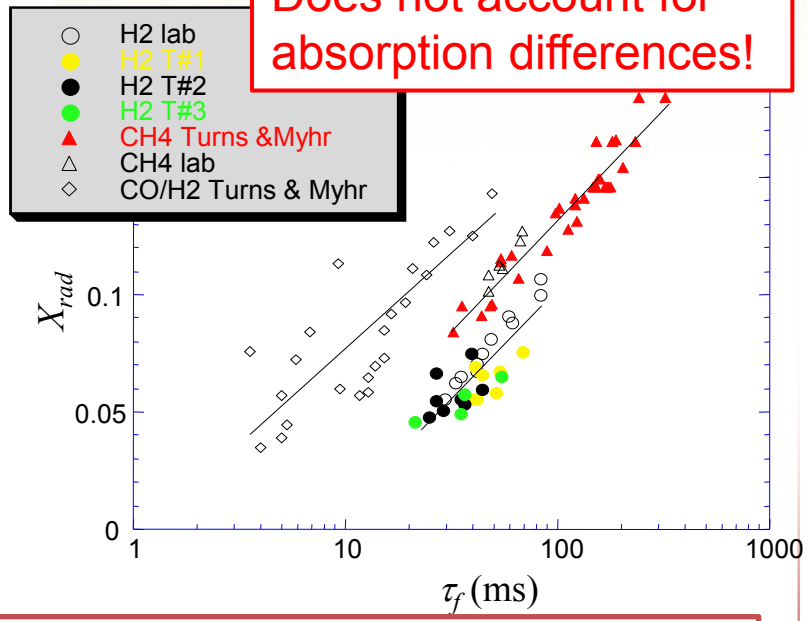
$$X_{rad} = \frac{S_{rad}}{\dot{m}_{fuel}\Delta H_c}$$

$$\tau_f = \frac{\rho_f W_f^2 L_f y_s}{\dot{m}_{fuel}}$$

X_{rad} = radiant fraction
S_{rad} = surface emissive power
$\dot{m}_{fuel}$ = fuel mass flow rate
ΔH_c = heat of combustion
τ_f = flame residence time
ρ_f = flame density
W_f, L_f = flame width/length
y_s = stoichiometric mixture fraction

Turns & Myhr, (1991)





Does not account for absorption differences!

Universal correlation developed for small & mid-sized flames, regardless of fuel gas type, where:

$$S_{rad} \propto a_p T_f^4 \tau_f$$

a_p = plank-mean absorption
T_f = adiabatic flame temperature

Does developed correlation hold for larger scale hydrogen jet flames?

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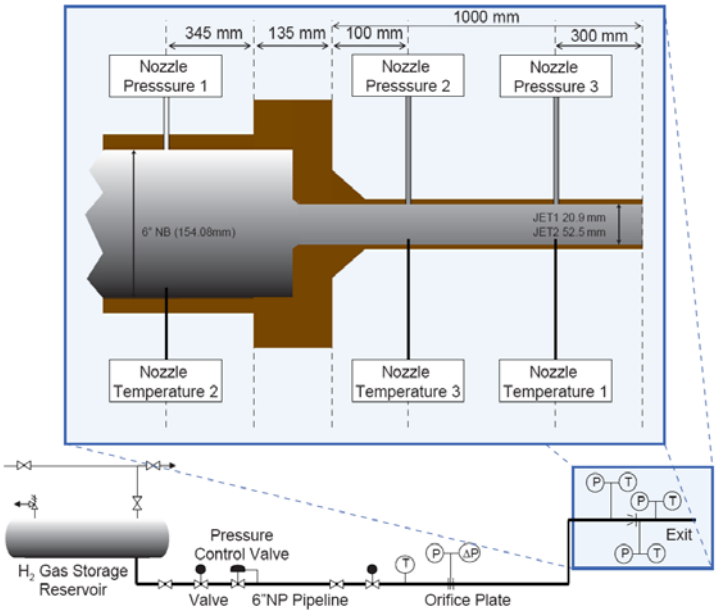
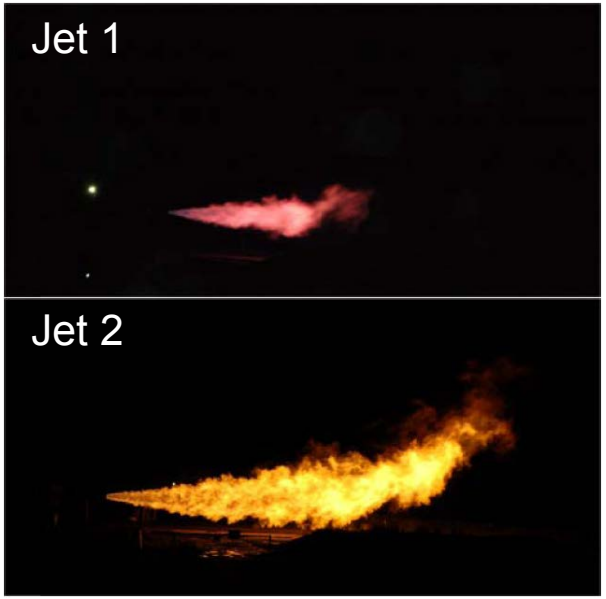
Accomplishment: Applied Radiation Science

✓ Worked with industrial partner to apply SNL flame radiation models to understand large-scale flame radiation behavior

Jet	d_j [mm]	[kg/s]	L_f [m]	p_0 [barg]	T_0 [K]	RH [%]	T_{amb} [K]	p_{amb} [mbar]	U_{wind} [m/s]	[°]
1	20.9	1.0	**17.4**	59.8	308.7	94.3	280	1022	2.84	68.5
2	52.5	7.4	**48.5**	62.1	287.8	94.5	280	1011	0.83	34.0

Measurements performed by Advantica (2008)





Measured heat fluxes ~40% higher than predicted values

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International and Industry R&D Collaborations

General release, ignition, and light-up phenomena for non-buoyant H_2 leaks

Pierre Benard & Boris Chernyavsky, Université du Québec à Trois-Rivières, Canada

- Detailed Large Eddy Simulations (LES) used to elucidate fundamental release behavior and relevant ignition and light-up mechanisms. (IEA, IPHE, ISO and NFPA)

Characterization of near-nozzle shockwave behavior and compressible shear layer growth from compressed H_2 releases

Sergey Kudriakov & Alexey Velikorodny, Commissariat à l'Energie Atomique (CEA), France

- Detailed LES to model the interplay between complex shockwave and flow phenomena (e.g., Görtler vortices in the barrel shock) from high source pressure releases. (IEA, IPHE, ISO and NFPA)

Reduced order modeling of complex H_2 release phenomena

Daniele Baraldi, Joint Research Centre (JRC), Netherlands

- Reduced order simulations (i.e., 2-equation Reynolds Averaged Navier-Stokes turbulence modeling) of compressed H_2 leaks that rely on validated notional nozzle models for jet-exit boundary conditions. (IEA, IPHE, ISO, NFPA and Industry Hazard Analysis)

Large-scale flame radiation measurements and modeling

Jimmy Li & Leonard Creitz, Air Products and Chemicals Inc., Allentown PA

- Evaluation and improvement of current radiative heat flux predictive methodologies for large-scale hydrogen jet flames. (ISO, NFPA and Industry Hazard Analysis)

Multidimensional modeling of Type IV fast-fill

Jianjun Ye, Institute of Process Equipment, Zhejiang University, P. R. China

- Multidimensional modeling of SNL fast fill experiments to validate fill model protocol development (GTR, ISO TC 197, SAE J2601, and CSA HGV 4.3).

H2CAN/Sandia collaboration on Hydrogen Safety

- Sandia hosted hydrogen safety workshop - April 11-12
- Workshop Goal: Coordinate hydrogen safety efforts between H2 CAN and US Programs

- Strong alignment of efforts identified at ICHS hosted by Sandia in Sept 2011
- Further reinforced by IEA Task 31 meeting in Jan 2012
  - Identified several near-term risk and behavior collaborative topics
  - Research roadmap presented during IEA Task 31 meeting – April 2012

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Future Work

- FY12
  - Acquire fundamental turbulent diffusivity measurements for **unreacting** flow fields to support flame light up model development
  - Work with NFPA 2 to finalize indoor refueling requirements
  - Enhance current notional nozzle model approaches using acquired data as validation benchmark
  - Help incorporate mitigation credit table into NFPA, ISO codes
  - With collaborators, validate burst pressure ratio performance test
  - With collaborators, analyze large-scale jet flame data
  - Participate in IPHE round robin fast-fill test and model validation activities
- FY13
  - Acquire fundamental turbulent diffusivity measurements for **reacting** flow fields for light-up model development
  - Develop fundamental light-up model framework based on turbulent flamelet concepts
  - Update radiative heat flux model for large-scale jet flames
  - Develop characteristic heat flux predictive model for pipeline ruptures

Summary

- Sandia R&D program develops the scientific basis for national and international codes and standards development
  - Enables deployment of hydrogen systems and infrastructure
- Fast-fill hydrogen fueling of Type IV tanks were experimentally examined in support of SAEJ2601, GTR and other RCS.
- Characteristics of predictive choked flow dispersion models were examined against Sandia generated validation datasets.
  - Deficiencies identified & updated models are under development
- Qualitative high-speed ignition imaging elucidated potential sustained flame light-up mechanisms.
  - Light-up boundaries for choked flow releases were experimentally mapped
  - Flamelet models can be used to predict light-up boundaries–experimental apparatus needed to measure relevant flow and combustion variables was constructed
- Measured radiative heat fluxes from large-scale H_2 flames were compared against model predictions.
  - Deficiencies identified and model improvements are ongoing
- Engaged international & industry collaborators to address complex R&D and RCS issues

Technical Back-Up Slides

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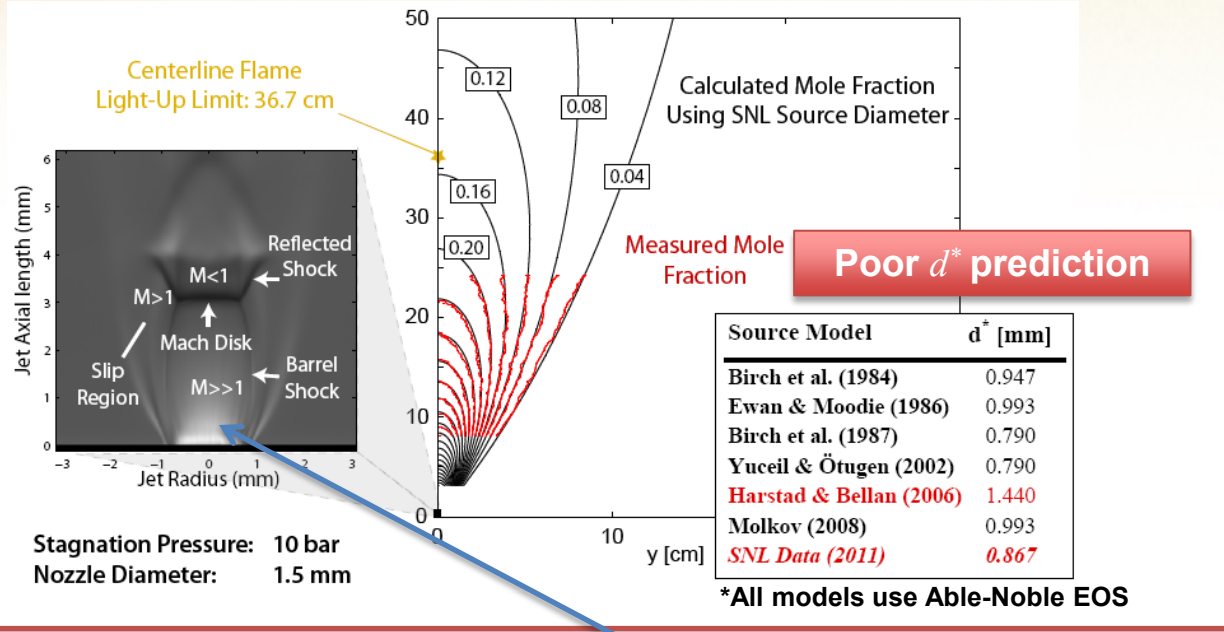
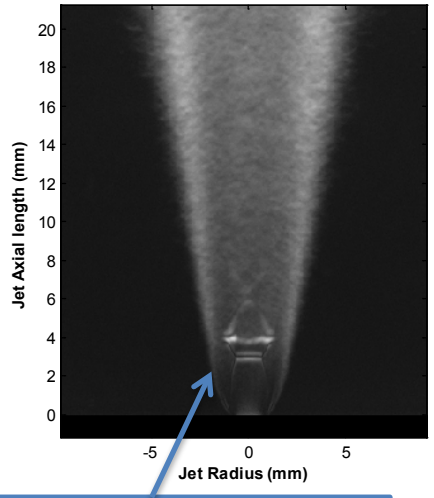
Excellent agreement observed between computed & measured mole fraction statistics if measured d^* was used.



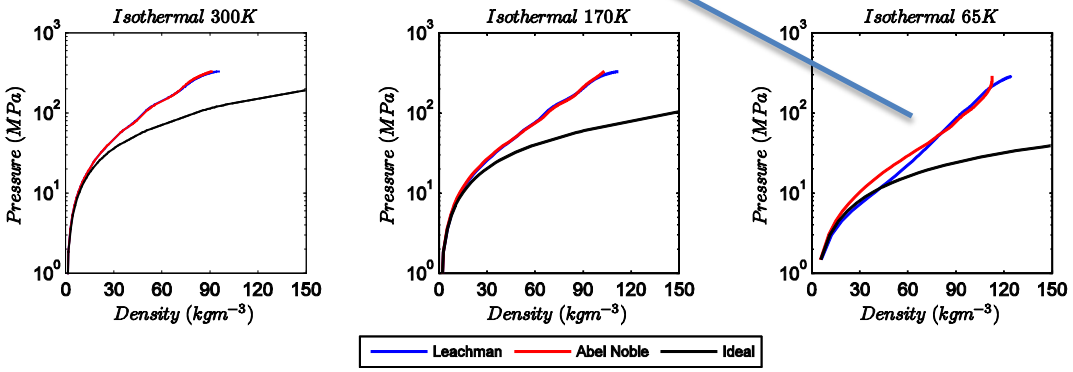
Poor d^* prediction

Source Model	d* [mm]
Birch et al. (1984)	0.947
Ewan & Moodie (1986)	0.993
Birch et al. (1987)	0.790
Yuceil & Ötugen (2002)	0.790
Harstad & Bellan (2006)	1.440
Molkov (2008)	0.993
SNL Data (2011)	*0.867*

*All models use Able-Noble EOS



Mixing in the slip region?



Abel-Noble EOS
- Works well at ambient *T*
- Cryogenic states poorly predicted (present in barrel shock; T < 70 K)

What is the best way to incorporate these refinements?
Is something else missing?

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Fundamental Hydrogen Release Behavior

Isaac Ekoto (PI)
SNL
Experiments

Adam Ruggles
SNL
Experiments

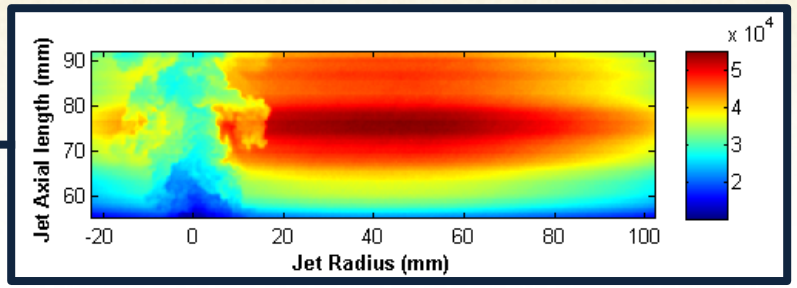
Partners and Collaborators:
Université du Québec à Trois-Rivières (CAN)

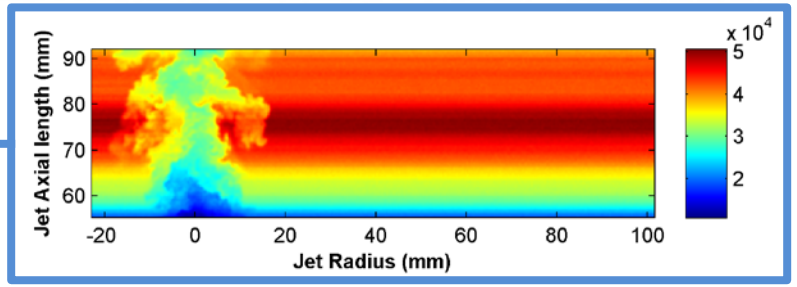
Sandia National Laboratories

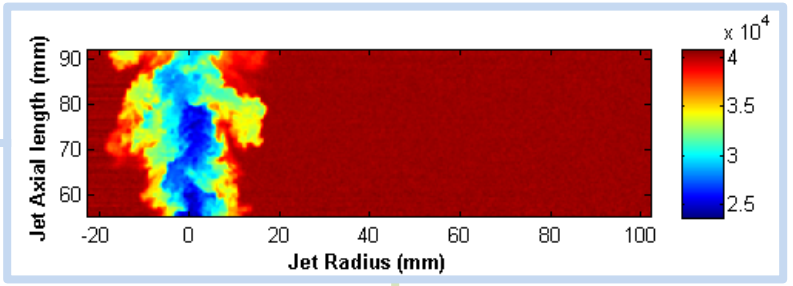
Raw signal intensity corrections used to create a quantitative concentration image

R:	Raw image
E_B:	Electronic bias
B_G:	Background luminosity
p_F:	Laser power fluctuation
O_R:	Camera/lens optical response
S_B:	Background scatter
S_t:	Laser sheet profile variation
I:	Corrected intensity

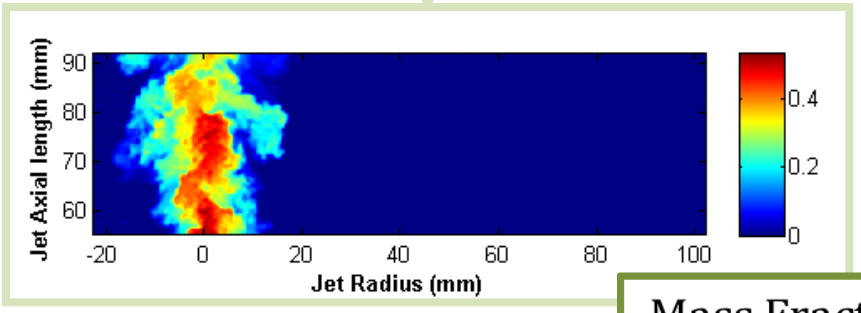
$$R = p_F \cdot O_R \cdot (I \cdot S_t + S_B) + E_B + B_G$$

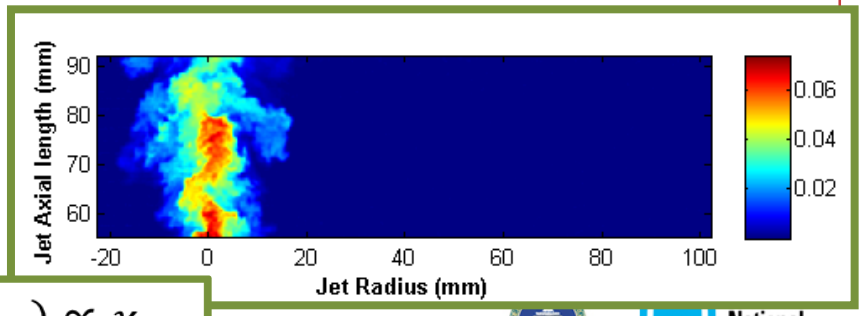






Mole Fraction $(\chi_{H2}) \propto I$





Mass Fraction $(Y_{H2}) \propto \chi_{H2}$

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Incompressible free-jets:

$$\frac{1}{\bar{Y}_{CL}} = K_c \frac{z - z_{0Y}}{r^*} \Rightarrow r^* = K_c \frac{z - z_{0Y}}{\bar{Y}_{CL}}$$

Where;

$\bar{Y}_{CL}$: centerline mass fraction

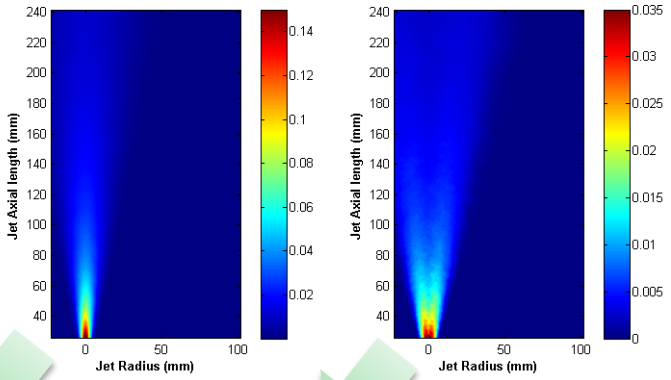
$r^* \equiv r_0 \sqrt{\frac{\rho_{jet}}{\rho_{air}}}$: mass weighted effective radius

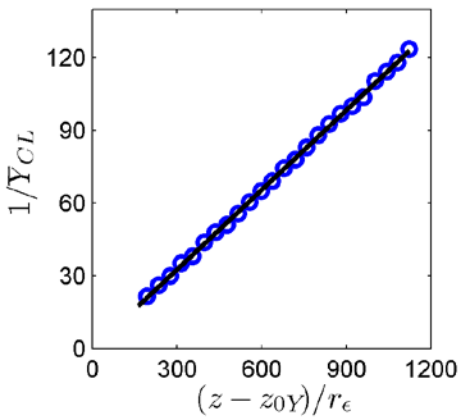
K_c: centerline decay rate constant

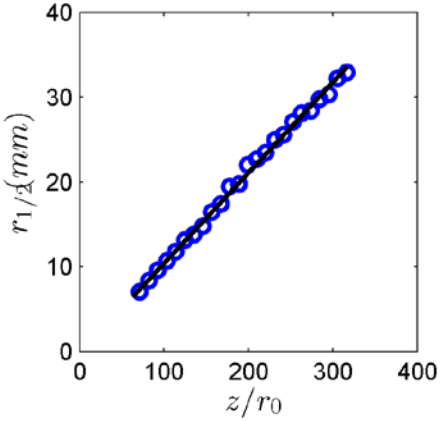
z: axial coordinate

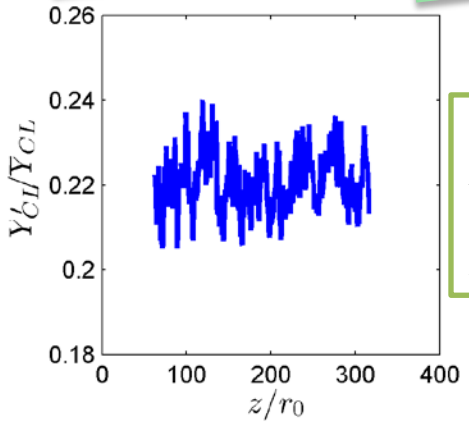
z_{0Y}: mass fraction virtual origin

Seamless reconstruction of mass fraction statistics from stitched together interrogation regions









$r_0 = 0.95\ mm$
$L_{pipe} = 250\ mm$
$Q = 100\ lit/min$
$Fr_{den} = 1170$

- Constant centerline decay (= 0.105) and jet spreading rates (0.113)
- Constant unmixedness ($\equiv Y'_{CL}/\bar{Y}_{CL} = 0.22$)

Centerline constants agree very well with literature reported values.

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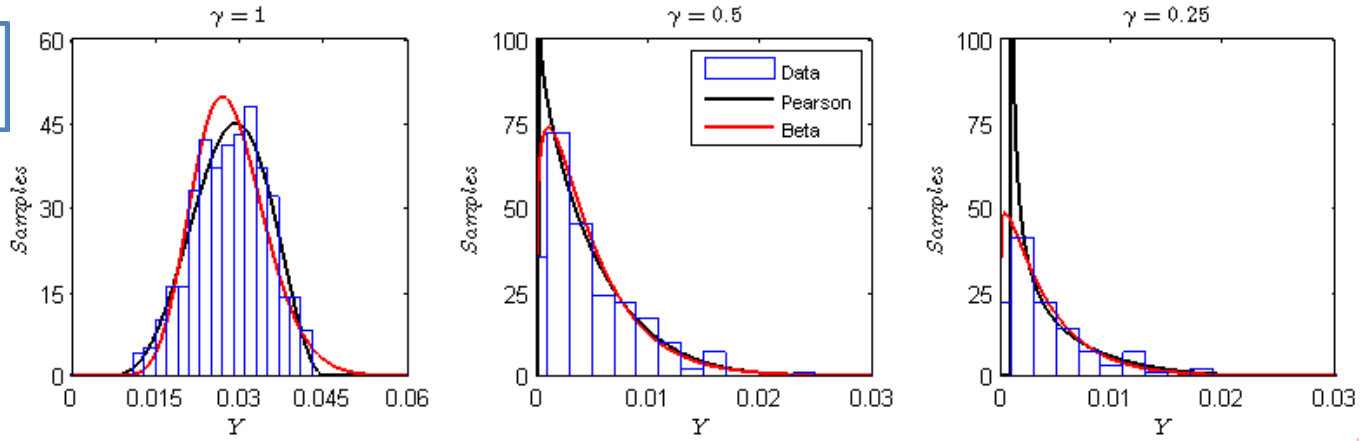
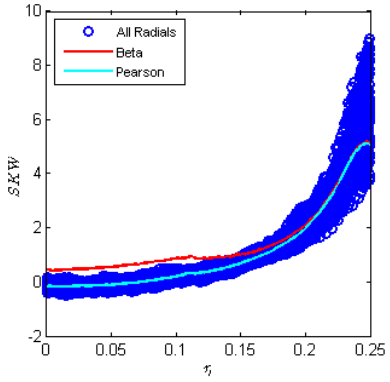
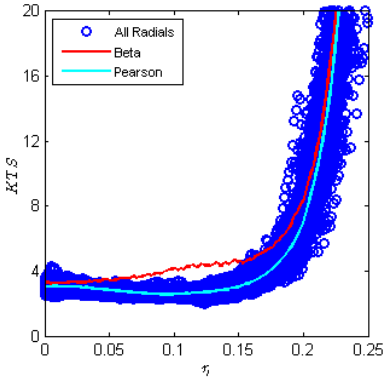
Measured PDFs were compared to Beta function distribution produced from measured 1st and 2nd order statistical moments.

$$P_C(Y_{H2}) = \frac{{Y_{H2}}^{\alpha-1}(1-YH_2)^{\beta-1}}{B(\alpha,\beta)}, \text{ where } \bar{Y} = \frac{\alpha}{\alpha+\beta}, \text{ and } \overline{Y'Y'} = \frac{\alpha\beta}{(\alpha+\beta)^2(\alpha+\beta+1)}$$

Conditioned probability that neglects $\gamma = 0$ values







Skewness & Kurtosis values for $\eta < 0.15$, deviate from measured values.

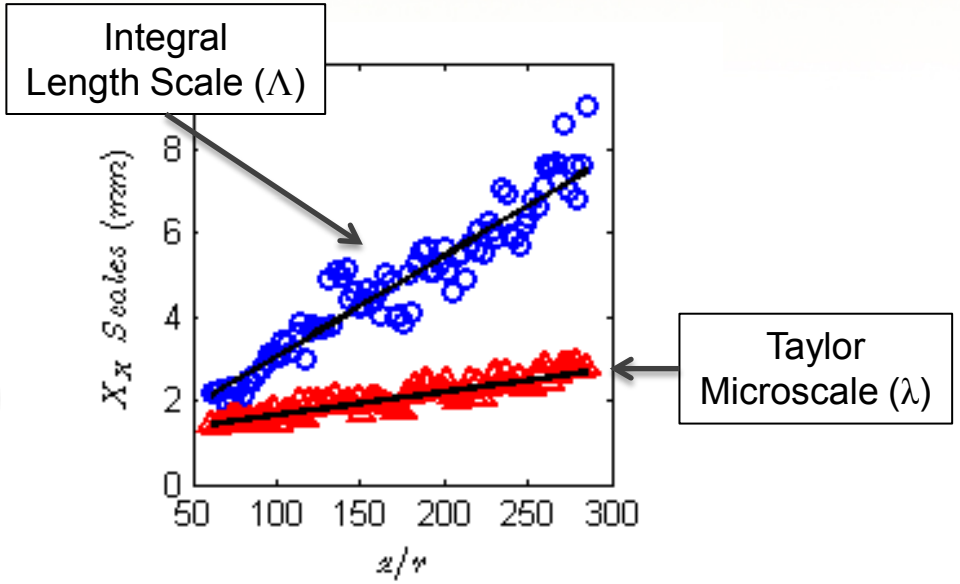
- Beta fit often worse than a Gaussian fit

What is a more suitable distribution function?
Is a 3rd PDF needed to account for a superlayer?

Along w/ conventional statistics (**mean/rms, PDFs, intermittency**), 2D high res. imaging enables **turbulent length scale** measurements.

$$\Lambda = \int_0^\infty \frac{\overline{Y_{H2}(x)' Y H_2(x+dx)'}}{\overline{Y_{H2'} Y_{H2'}}} dx$$

$$\frac{1}{\lambda^2} = -\frac{1}{2}\frac{\partial^2}{\partial x^2}\left(\frac{\overline{Y_{H2}(x)' Y H_2(x+dx)'}}{\overline{Y_{H2'} Y_{H2'}}}\right)$$



Are non-reacting flow integral length-scales suitable for turbulent flame speed determination in light-up models?

Can energy density spectra be reproduced form the integral length scale, or is an integral time scale needed instead?

New diagnostic capability that **enables direct turbulent diffusion measurements** was designed & constructed in FY12.

Along with molecular diffusion, D, turbulent diffusion, D_{turb}, controls mixing.

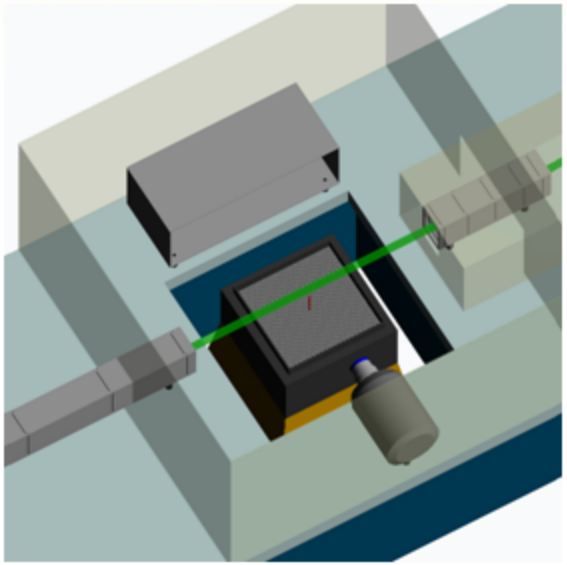
For unreacting H_2/air mixtures mixture density is:

$$\rho = Y_{H_2}\rho_{H_2} + \left(1 - Y_{H_2}\right)\rho_{air}$$

and,

$$D_{turb} \equiv \frac{1}{\nabla \bar{Y}_{H_2}} \cdot \left[\overline{\boldsymbol{u}' Y_{H_2}'} + \frac{\bar{\boldsymbol{u}}}{\bar{\rho}} \overline{\rho' Y_{H_2}'} + \frac{\bar{Y}_{H_2}}{\bar{\rho}} \overline{\rho' \boldsymbol{u}'} + \frac{1}{\bar{\rho}} \overline{\boldsymbol{u}' \rho' Y_{H_2}'} \right]$$

Direct measurements possible only through coupled velocity and concentration statistics



$\boldsymbol{u}$: Particle Image Velocimetry

Y_{H_2}: Acetone laser fluorescence

- *PIV seed particles interfere w/ Rayleigh scatter*

System construction complete; 1st measurements in Q4FY12.

How will these measurements compare to detailed LES?

Sustained Light-Up Probability

Isaac Ekoto (PI)
SNL
Experiments

Adam Ruggles
SNL
Experiments

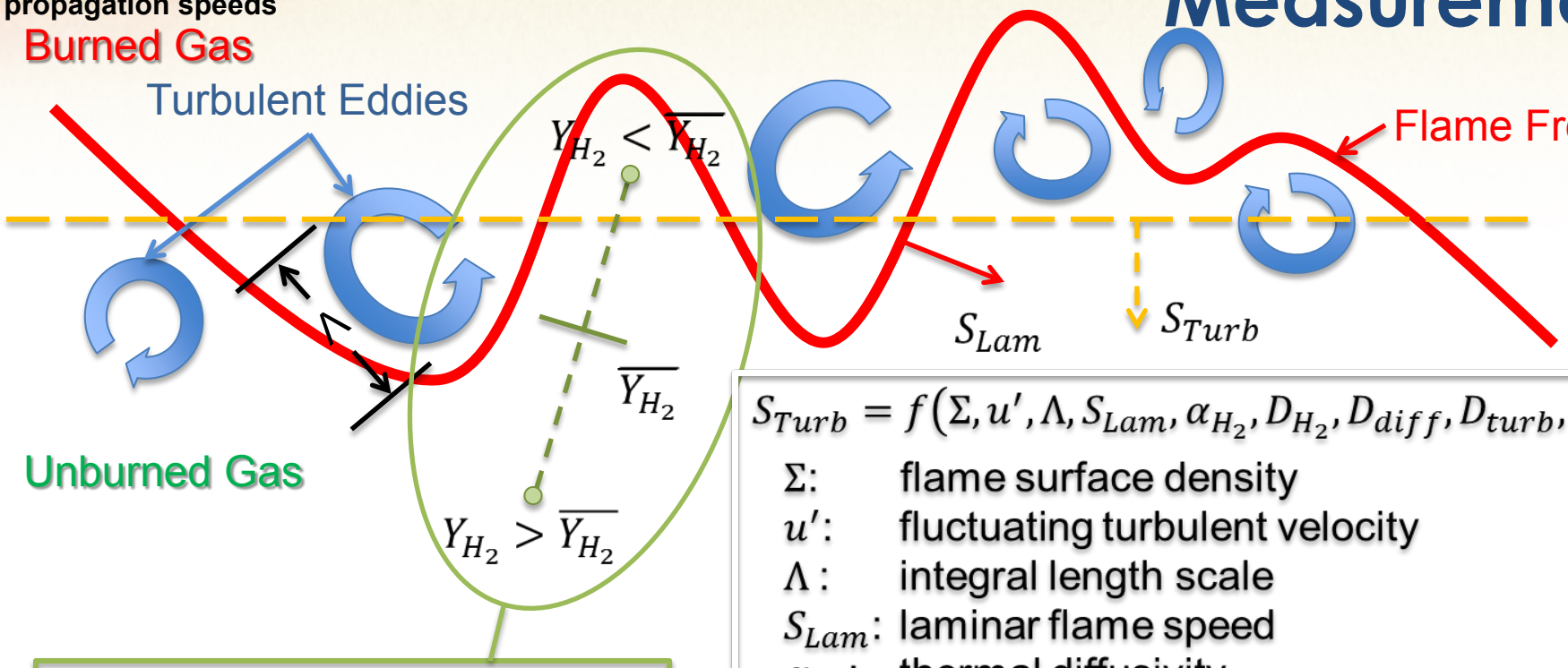
Partners and Collaborators:

Sandia National Laboratories

Accomplishment:

Improve Mixture Measurement

Flamelet models can be used to predict these turbulent flame front propagation speeds



H_2 preferentially diffuses toward reaction front (differential diffusion)
- Increased flame front distortion

Driscoll, 2007

$$S_{Turb} = f(\Sigma, u', \Lambda, S_{Lam}, \alpha_{H_2}, D_{H_2}, D_{diff}, D_{turb}, q, c)$$

- Σ: flame surface density
- u': fluctuating turbulent velocity
- Λ: integral length scale
- S_{Lam}: laminar flame speed
- α_{H_2}: thermal diffusivity
- D_{H_2}: molecular diffusivity
- D_{diff}: differential diffusion
- D_{turb}: turbulent diffusivity
- q: heat release
- c: reaction progress variable

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Fast Fill Modeling

Terry Johnson(PI)
SNL
Experiments

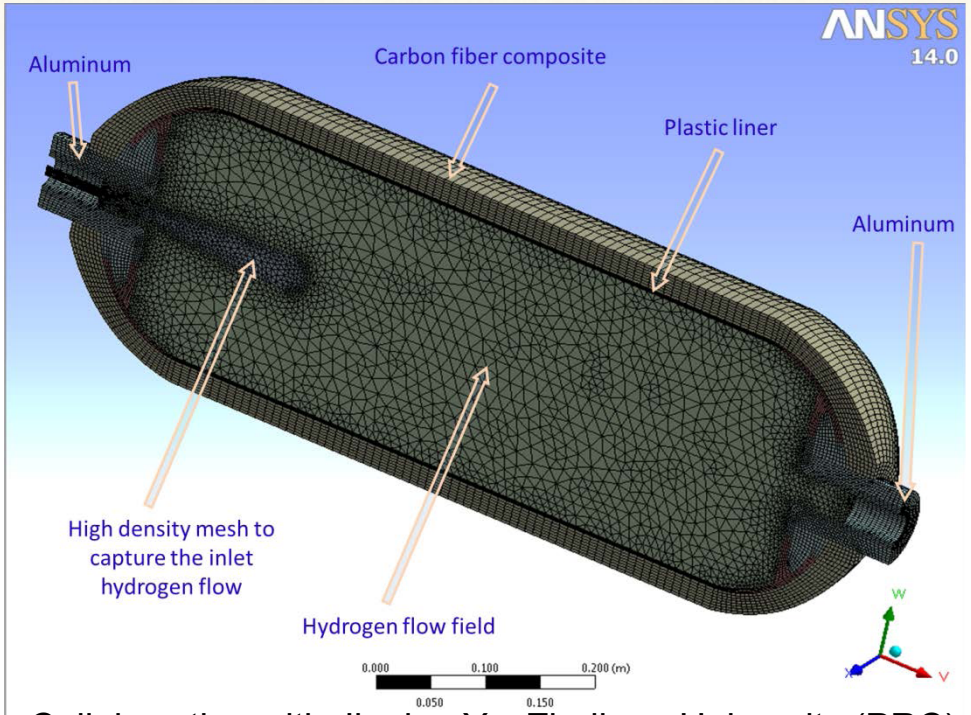
Jianjun Ye
Zhejiang University, PRC
CFD Modeling

Partners and Collaborators:
Lincoln Composites and Zhejiang University, PRC

Sandia National Laboratories

Tank CFD model under development to compare against validation datasets.

- Ansys Fluent V14.0
- 3D to capture buoyancy effects
- Structured grid for the tank
- Unstructured flow grid
- NIST real gas state modeling
- Coupled fluid dynamics & heat transfer



Collaboration with Jianjun Ye, Zhejiang University (PRC)

Future Work

- Broaden test matrix to include different tank geometries & types
- Upgrade test facility to accommodate 70 MPa fast filling experiments
- Create generalized from validation data as a comparative standard for developing refueling protocols

Laboratories