Effects of Exhaust Gas Recirculation (EGR) on Turbulent Combustion Emissions in Advanced Gas Turbine Combustors with High Hydrogen Content (HHC) Fuels

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National Energy Technology Laboratory University Turbine Systems Research Program

Thanks: Chris Reuter, Prof. Sang Hee Won

# Contents

- Kinetic mechanism development and validation including non-equilibrium reaction of HCO prompt dissociation in high pressure CH<sub>2</sub>O flames with CO<sub>2</sub> dilution.
- H<sub>2</sub>O/CO<sub>2</sub> kinetic effects on turbulent flame speeds/structures of CH<sub>4</sub>/air mixtures with H<sub>2</sub>O/CO<sub>2</sub> dilution using RATS Burner
- New design of Axisymmetric Reactor-Assisted Turbulent (ARAT) Burner for High Turbulence with EGR.

1. Kinetic study of non-equilibrium prompt dissociation of HCO at elevated pressures with CO<sub>2</sub>

Major oxidation pathway of HHC fuels

 $C_2H_6, CH_4 \rightarrow CH_2O \rightarrow HCO \rightarrow H+CO \rightarrow CO_2, H_2O$ 



# Why CO<sub>2</sub>? HCO+M=H+CO R2a HCO+O<sub>2</sub>=HO<sub>2</sub>+CO R2b



CO<sub>2</sub> affects the competition between the two key reactions

- Formaldehyde (CH<sub>2</sub>O)/CO<sub>2</sub> flames are excellent targets to study HCO prompt reactions and CO<sub>2</sub> effect, but have not been reported in the literature
  - Formaldehyde flame sampling has been done at very low pressure (~30 torr)<sup>1-4</sup>
  - Formaldehyde has been studied in shock tubes<sup>5-9</sup> and flow reactors<sup>10,11</sup>
- We reports the *first flame speed measurements* at high pressure and with CO<sub>2</sub>.
- 1. J. Vandooren et al. (1986) 7. G. Friedrichs et al. (2004)
- 2. M.C. Branch et al. (1991) 8. G. Friedrichs et al. (2002)

9. S. Wang et al. (2013)

10. P. Glarborg et al. (2003)

11. S. Hochgreb et al. (1992)

- 3. A.R. Hall et al. (1957)
- 4. V. Dias et al. (2012)
- 5. Y. Hidaka et al. (1993)
- 6. V. Vasudevan et al. (2005) 12. J. Santner et al. (2015)

# How do we create CH<sub>2</sub>O and HCO?

- Formaldehyde is toxic and difficult to generate in its pure form due to polymerization
- Use trioxane to generate formaldehyde *in situ*
- Formaldehyde (CH<sub>2</sub>O) forms before radical pool
  - > 99% of trioxane is consumed through thermal decomposition
- These flames are governed by the same chemistry as formaldehyde flames



J. Santner, F.L. Dryer, Y. Ju, Proc. Combust. Inst. 34 (2013) 719–726.

# **High Pressure Flame Experiments**



# Experimental conditions of pressurized trioxane flame speed measurements with/without CO<sub>2</sub>

Case	Equivalence ratio	Trioxane	02	CO2	N <sub>2</sub>	Не	Т <sub>f</sub> (К)
Lean	0.7	0.0454	0.1946	0.0000	0.5846	0.1754	1900
Ultra-lean	0.34	0.0454	0.4000	0.0000	0.3792	0.1754	1882
Rich	1.4	0.0889	0.1904	0.0000	0.5544	0.1663	2000
Lean with CO2	0.7	0.0491	0.2103	0.2000	0.2201	0.3206	1900
Rich without CO2	1.4	0.0943	0.2021	0.2000	0.1266	0.3770	2000

$$\begin{array}{c} X+CH_2O=HCO+XH \quad (equilibrium) \qquad (R_{1a}) \\ X+CH_2O=H+CO+XH \quad (non-equilibrium, Prompt) \qquad (R_{1b}) \end{array}$$
   
Why Lean? 
$$\begin{array}{c} HCO+M=H+CO \qquad R_{2a} \\ HCO+O_2=HO_2+CO \qquad R_{2b} \end{array}$$

### Flame speeds and radical pool

Ultra-lean:  $\Phi=0.34$ 

#### H+O2=O+OH H+O2+M=HO2+M

#### HCO+M=H+CO+M (R2a) HCO+O2=CO+HO2 (R2b)



- Mole fractions of O, OH, and total radical pool in the ultra-lean cases is larger than that in the lean case, because of increase of H+O2=O+OH,
- While H radical's mole fraction is lower than that in the lean case, causing a decrease of system reactivity directly, because of the increase of R2a/R2b branching ratio.

### HCO prompt dissociation on H production at 4 atm (HP Mech-Prompt)



HCO Prompt dissociation effect increases with increasing O2 mole fractions. 9

### Flame speeds and reaction sensitivity

#### •HCO+M=H+CO+M (R2a) •HCO+O2=HO2+CO (R2b)



### HP-Mech with prompt HCO reactions has better predicted flame speeds.

### **Effect of CO<sub>2</sub> dilution on HCO prompt reactions**



HCO+M=CO+H+M

 HP Mech with HCO prompt reactions has a good prediction of flame speed on fuel lean conditions but over-predict considerably at fuel rich conditions with CO<sub>2</sub>.

### 2. Turbulent flame speeds/structure with $H_2O/CO_2$ dilutions

#### Reactor-assisted turbulent slot (RATS) burner

10 cm by 1 cm rectangular slot

- Methane/air pilot flame ٠
- OH PLIF for flame imaging

#### Experimental study:

- $CH_{4}/Air$  flames with  $CO_{2}$  and  $H_{2}O$  dilution
- Moderately intense turbulence (up to  $Re_{\tau} \sim 170$ )
- Dilution at either variable  $T_{ad}$  or constant  $T_{ad}$



			CH₄/Air	$T_0 = 450 \text{ K},$	p=1 atm, $U$	/ = 15 m/s
		_ [	φ	Dilution	$T_{ad}(\mathbf{K})$	$S_{L}$ (cm/s)
$Dilution = \frac{X_{Diluent}}{1}$	temperature – dilution"		1.0	None	2304	70.6
$X_{Fuel} + X_{Air} + X_{Diluent}$		ך [	1.0	20% H <sub>2</sub> O	1992	28.4
			1.0	15% CO <sub>2</sub>	2014	25.9
		r	0.9	$(N_2 \text{ only})$	2025	40.9
Add N. for each case	"Constant flame temperature dilution"		0.9	10% H <sub>2</sub> O	2025	40.0
Add $N_2$ for each case $\leftarrow$		4	0.9	10% CO <sub>2</sub>	2025	34.6
until I <sub>ad</sub> is matched			0.9	20% H <sub>2</sub> O	1822	20.5
			0.9	20% CO <sub>2</sub>	1822	16.8

# Determination of $S_T$



- (1) Find inner flame surface, set unburned region to c = 0 and burned to c = 1
- (2) Create average of progress variable for entire run (500 images)
- (3) Trace c = 0.1 contour to find perimeter
- (4) Evaluate turbulent flame speed ( $S_T$ ) through mass conservation
- More consistent than previous method of using PDF of flame perimeters

# Scaling of Dilution Effects

- Damkohler's original scaling:  $S_T/S_L = (u'/S_L)^{0.5} (l/l_F)^{0.5}$
- Modify using Sc = 1 and  $l_F = \alpha/S_L$
- Modified Damköhler scaling :

$$S_{T,diluted} = S_{T,standard} \left(\frac{S_{L,dil}}{S_{L,std}}\right) \left(\frac{Re_{T,dil}}{Re_{T,std}}\right)^{0.5} \left(\frac{Le_{dil}}{Le_{std}}\right)^{-0.5}$$
Flame speed Turbulence Thermo-diffusivity contribution contribution

# Variable flame temperature with Dilution



- Effects of increased dilution are apparent even at a glance
- Clear lengthening of the flame can be seen at higher dilution levels

# Variable $T_{ad}$ Dilution

$CH_4/Air, T_0 = 450 \text{ K}, p = 1 \text{ atm}, U = 15 \text{ m/s}$					
φ	Dilution	$T_{ad}(\mathbf{K})$	$S_{L}$ (cm/s)		
1.0	None	2304	70.6		
1.0	20% H <sub>2</sub> O	1992	28.4		
1.0	15% CO <sub>2</sub>	2014	25.9		

Variable  $T_{ad}$ 

- Large reduction in  $S_T$  for both  $CO_2$  and  $H_2O^{-\frac{3}{2}}$
- CO<sub>2</sub> dilution is noticeably stronger



$$S_{T,diluted} = S_{T,standard} \left(\frac{S_{L,dil}}{S_{L,std}}\right) \left(\frac{Re_{T,dil}}{Re_{T,std}}\right)^{0.5} \left(\frac{Le_{dil}}{Le_{std}}\right)^{-0.5}$$
Scaling Measured  
20% H<sub>2</sub>O  $\downarrow 60\% \uparrow 0.2\% \uparrow 2.1\% \longrightarrow \downarrow 59\% \downarrow \sim 59\%$   
15% CO<sub>2</sub>  $\downarrow 63\% \uparrow 6.0\% \uparrow 4.0\% \longrightarrow \downarrow 59\% \downarrow \sim 55\%$   
 $\uparrow S_T/S_L$ 

Thermal effect is the largest contribution with diultions.



# Constant Flame temperature with Dilution: $T_{ad} = 2025 \text{ K}$



• Effects of increased dilution are much more nuanced at fixed  $T_{ad}$ 

# Constant $T_{ad}$ =2025 K with Dilution

$CH_4/Air, T_0 = 450 \text{ K}, p = 1 \text{ atm}, U = 15 \text{ m/s}$					
φ	Dilution	$T_{ad}(\mathbf{K})$	$S_L$ (cm/s)		
0.9	$(N_2 \text{ only})$	2025	40.9		
0.9	10% H <sub>2</sub> O	2025	40.0		
0.9	10% CO <sub>2</sub>	2025	34.6		

Constant  $T_{ad}$ 

- Water has almost no discernible effect
- Slight (~12%) drop in  $S_T$  for 10% CO<sub>2</sub>



• Scaling analysis:

$$S_{T,diluted} = S_{T,standard} \left( \frac{S_{L,dil}}{S_{L,std}} \right) \left( \frac{Re_{T,dil}}{Re_{T,std}} \right)^{0.5} \left( \frac{Le_{dil}}{Le_{std}} \right)^{-0.5}$$
Scaling Measured  
10% H<sub>2</sub>O  
10% H<sub>2</sub>O  
10% CO<sub>2</sub>  

$$V^{2.1\%} \uparrow 0.0\% \downarrow 0.3\% \longrightarrow \downarrow 2.3\% \downarrow \sim 0\%$$
  

$$V^{-0\%} \downarrow 15\% \uparrow 3.9\% \uparrow 1.3\% \longrightarrow \downarrow 11\% \downarrow \sim 12\%$$

**Chemistry turbulence diffusion** 

At the same flame temperature, chemistry plays a dominant role

# Flame Surface Density

 We can also examine the turbulent flame brush as a whole through the flame surface density:

$$\Sigma(c) = \frac{1}{n} \sum_{i=1}^{n} \frac{L_i(c)}{A_i(c)}$$
, n: num. of images

- As an example,  $\Sigma(c = 0.45)$  is the combined length of all the flame edges in the region between c = 0.4 and c = 0.5, divided by the area of the region and the number of images
- $\Sigma(c)$  decreases by 15-25% for variable flame temperature dilution
- At constant  $T_{ad}$ :
  - $\Sigma(c)$  nearly collapses for  $CO_2$  addition due to opposing increases in flame brush size and flame wrinkling
  - Water slightly reduces  $\Sigma(c)$  in the upper region of the brush (c > 0.6)





C. Cohé et al., Proc. Combust. Inst. 32 (2009) 1803-1810

# Constant Flame temperature with Dilution: $T_{ad} = 1822 \text{ K}$



0% Dilution  $T_{ad} = 1822 \text{ K}$  20% H<sub>2</sub>O Dilution  $T_{ad}$  = 1822 K 20% CO<sub>2</sub> Dilution  $T_{ad} = 1822$  K

# $T_{ad} = 1822 \text{ K Dilution}$

$CH_4/Air, T_0 = 450 \text{ K}, p = 1 \text{ atm}$				
φ	Dilution	$T_{ad}(\mathbf{K})$	$S_L$ (cm/s)	
0.9	$(N_2 \text{ only})$	1822	23.2	
0.9	20% H <sub>2</sub> O	1822	20.5	
0.9	20% CO <sub>2</sub>	1822	16.8	

- Water dilution is impactful beyond what the scaling predicts
- Is this an actual trend caused by turbulence-chemistry interactions?



$$S_{T,diluted} = S_{T,standard} \left(\frac{S_{L,dil}}{S_{L,std}}\right) \left(\frac{Re_{T,dil}}{Re_{T,std}}\right)^{0.5} \left(\frac{Le_{dil}}{Le_{std}}\right)^{-0.5}$$
Scaling Measured  
20% H<sub>2</sub>O  $\qquad \downarrow 12\% \qquad \uparrow 0.1\% \qquad \downarrow 0.2\% \longrightarrow \downarrow 12\% \qquad \downarrow \sim 29\%$   
20% CO<sub>2</sub>  $\qquad \downarrow 27\% \qquad \uparrow 7.2\% \qquad \uparrow 2.7\% \longrightarrow \downarrow 20\% \qquad \downarrow \sim 25\%$ 

# $T_{ad}$ = 1822 K Dilution

- At 20% dilution, CO<sub>2</sub> and H<sub>2</sub>O produced similar S<sub>7</sub> values (approximately 25% lower than the undiluted case)
- However, the CO<sub>2</sub> flame surface density is noticeably less
- This is primarily due to differences in the area between c contours (larger for CO<sub>2</sub>) rather than differences in flame wrinkling





# Scaling of $S_T$ for All Conditions



- Power fit to current data:  $S_T/S_L = 2.3(u'/S_L)^{0.42}$
- Kobayashi fit for  $CH_4$ :  $S_T/S_L = 2.9(u'/S_L)^{0.38}$

H. Kobayashi, Exp. Therm. Fluid Sci. 26 (2002) 375-387

$$S_{T,diluted} = S_{T,standard} \left(\frac{S_{L,dil}}{S_{L,std}}\right) \left(\frac{Re_{T,dil}}{Re_{T,std}}\right)^{0.5} \left(\frac{Le_{dil}}{Le_{std}}\right)^{-0.5}$$

3. A new design of Axisymmetric Reactor-Assisted Turbulent (ARAT) Burner for Turbulent Flame Studies at Higher Reynolds Numbers and with EGR Effects

Co-flow Axisymmetric Reactor-Assisted Turbulent (CARAT) Burner

- 15 mm central jet diameter (CH<sub>4</sub>/Air)
- Inner pilot flame (have tested CH<sub>4</sub>/Air)
- Outer vitiated co-flow (15cm in diameter, tested for  $CH_4$ /Air and  $H_2$ /Air)



# ARAT vs RATS

For same flow rates (450 LPM air, 47 LPM CH<sub>4</sub>,  $\phi = 1.0$ ,  $T_{exit} = 300$  K)

RATS (100 mm by 10 mm)

- *U* = 8.3 m/s
- *Re<sub>bulk</sub>* = 9500
- $Re_T = 172$
- T<sub>env.</sub>=300 K



ARAT (15 mm diameter)

- *U* = 47.0 m/s
- $Re_{bulk} = 44000$
- *Re<sub>T</sub>* ≈ 1758
- T<sub>coflow</sub>=up to1900 K
- High higher turbulent intensity
- More turbulent flame regimes accessit
- •Capable of more extreme conditions w EGR effects due to vitiated co-flow

# Conclusions

- The high pressure flame speeds of CH<sub>2</sub>O flames are measured for the first time.
- HCO prompt reaction affects the burning velocity and radical pool of flames. The updated HP-Mech improved the flame speed prediction.
- CO<sub>2</sub> dilution strongly affects the radical production via HCO(+M) =H+CO(+M) reaction. Existing models are not able to predict the high pressure flame speeds on CH<sub>2</sub>O flames with CO<sub>2</sub> dilution.
- In addition to the thermal effect of  $H_2O/CO_2$  dilution, the chemistry effects also significantly reduce the turbulent flame speeds of  $CH_4$ /air flames.
- The lower the flame temperature, the higher the kinetic effects of  $H_2O/CO_2$  dilution on flame speeds.
- At lower flame temperature, the kinetic effect  $H_2O$  is increased and comparable to that of  $CO_2$ .

### **Reaction sensitivities with CO<sub>2</sub> dilution**

Fuel lean  $\Phi=0.7, 20\% CO_2$ 

Fuel Rich  $\Phi=1.4$ , 20% CO<sub>2</sub>



- Sensitivity of HCO+M=CO+H+M increases in fuel rich conditions with CO<sub>2</sub>
- Sensitivity of H+O<sub>2</sub>+M=HO<sub>2</sub>+M and CO+OH=CO<sub>2</sub>+H decrease in fuel rich conditions with CO<sub>2</sub>
- Suggesting that collisional energy transfer in HCO+M=CO+H+M may not be well characterized for CO<sub>2</sub>

#### Validation of previous experiments at 1atm



• The new experiments have reduced uncertainty due to high speed imaging improvement

J. Santner, F.L. Dryer, Y. Ju, Proc. Combust. Inst. 34 (2013) 719–726.

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# **Outline of the Presentation**

- Yiguang Ju Chemical kinetics with EGR effects, Reactor Assisted Turbulent Slot (RATS) burner studies at atmospheric pressure
- Bob Lucht and Jay Gore: Premixed Axisymmetric Reactor Assisted Turbulent (PARAT) burner, CARS, OH PLIF, and PIV Measurements
- Michael Mueller Advanced numerical modeling of the RATS and PARAT burners

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- PhD student Dong Han, postdoctoral research associate Aman Satija
- Jupyoung Kim for help with PIV measurements, Hasti Veeraraghava Ragu for help with PARAT burner development
- DOE Program Manager Mark Freeman

# **Atmospheric Pressure PARAT Burner Studies**

- Measurements performed in CH<sub>4</sub>/air/CO<sub>2</sub> flames with same adiabatic flame temperature and Re=10,000, but different levels of CO<sub>2</sub> addition, flames 1, 2, and 3 had 0%, 5%, and 10% CO<sub>2</sub> addition
- Equivalence ratio adjusted to maintain same adiabatic flame temperature
- Dual-pump CARS measurements of temperature, O<sub>2</sub>, and CO<sub>2</sub> performed to characterize flame structure
- High-speed (4 kHz) OH PLIF was performed to characterize the flame structure
- Stereo PIV was performed to characterize boundary conditions for advanced numerical simulations

# Experiment, RATS Burner

#### • <u>Reactor Assisted Turbulent Slot burner (RATS burner)</u><sup>1</sup>

- Heat large flow rates (1000 LPM) up to 700 K with CO2/H2O/N2 dilutions
- ~ 55 cm heated length, 100 × 10 mm exit cross-section ( $D_H \approx 18$  mm)
- Two turbulence generators<sup>2,3</sup>, homogeneous isotropic turbulence confirmed by hot-wire anemometry
- High Reynolds number (Re<sub>bulk</sub> > 10,000)



<sup>1</sup>S. H. Won, B. Windom et al, Combust. Flame 161 (2014) 475-483. <sup>2</sup>Coppola, G., and Gomez, A., *Experimental Thermal and Fluid Science*, Vol. 33, 2009, pp. 1037-1048. <sup>3</sup>Venkateswaran, P. *et. al., Combustion and Flame., 158, 2011, 1602-1614* 

## **PARAT Burner**

Piloted axisymmetric reactor assisted turbulent (PARAT) burner



# **PARAT Burner**



 $H_2$  piloted  $CH_4$ /air flame specification

- CO<sub>2</sub> addition increases the visible flame length
- More local extinctions are observed with increasing of CO<sub>2</sub> additions

#### Visible Images at 0.5 ms exposure time



### **PARAT Burner**

### H<sub>2</sub> piloted CH<sub>4</sub>/air Flame specification

Flame #	1	2	3
Reynolds number		$10000\pm50$	
Adiabatic Temperature(K)		$2030\pm 50$	
Equivalence ratio	$\textbf{0.80}\pm\textbf{0.02}$	$\textbf{0.84} \pm \textbf{0.02}$	$\textbf{0.89} \pm \textbf{0.02}$
CO <sub>2</sub> % by total mass	0.0	$\textbf{5.0}\pm\textbf{0.1}$	$\textbf{10.0} \pm \textbf{0.1}$
Lewis number	0.99	0.98	0.97
Laminar flame speed (cm/s)	33.7	29.7	24.9

Flames are designed to minimize thermal and transport effects on NO formation

# **PARAT Burner: Centerline Temperature Profiles**

Flame #	Flame length x/D*	Unburned reactant length x/D	Flame brush thickness Δx/D**
1	5.17	1.39	3.78
2	6.28	1.39	4.89
3	7.39	1.39	6

\*Flame length is defined as the distance from burner exit to the location  $\overline{C} = 0.99$  $\overline{C}$  is the mean progress variable

\*\*Flame brush thickness is defined as the width from leading edge  $\overline{C} = 0.01$  to trailing edge  $\overline{C} = 0.99$ 



# **PARAT Burner: Centerline Temperature Histograms**



### **PARAT Burner: Radial Temperature Profiles**



# PARAT Burner: Comparison with Wrinkled Flame Theory



# **PARAT Burner: 4 kHz OH PLIF Measurements**



## **PARAT Burner: 4 kHz OH PLIF Measurements**



### **PARAT Burner: 4 kHz OH PLIF Measurements**



x/D

44

### **PARAT Burner: PIV Measurements, Flame 1**



With upper turbulence generator plate

### **PARAT Burner: PIV Measurements, Flame 1**



Without upper turbulence generator plate

## **PARAT Burner: IR Imaging Measurements**



0% CO2 10% CO2 4.38 ± 0.08 μm spectral region

# PARAT Burner: Comparison of Temperature Profiles from CARS and IR Emission Measurements



# PARAT Burner: Comparison of Temperature Profiles from CARS and IR Emission Measurements



# **PARAT Burner Studies: Conclusions**

- Dual-pump CARS temperature measurements reveal significant differences in structure for flames 1, 2, and 3 ,even though the measured peak mean temperatures are virtually the same for all three flames. Bimodal structure of histograms clearly evident in the inner flame.
- OH PLIF imaging reveals major differences in flame structure for flames 1, 2, and 3. As the percentage of added CO<sub>2</sub> increases, the occurrence of isolated pockets of unreacted premixed gas in the flame brush increases, as does the apparent size of these gas pockets.
- Stereo PIV was performed to characterize boundary conditions for advanced numerical simulations, analysis still in progress.
- Comparison of CARS and IR imaging show consistent temperatures for all three flames.

# PARAT Burner Studies Future Work

- High speed CH PLIF, simultaneous OH and CH PLIF
- Continued interaction with Princeton on numerical modeling of the PARAT burner
- Operation of the PARAT burner at high pressure, initial experiments were performed at high pressure last year
- NOx, CO emission measurements from the highpressure for comparison with numerical modeling

# Cross-sectional View of PARAT Burner into the Windowed High-Pressure Test Rig



# **PARAT Burner Operation in HP Test Rig**

#### P & ID



#### **Control panel**



#### H<sub>2</sub> pilot flame



Natural gas flame, Re=100,000,  $\psi$ =0.9

