Growth mechanism for soot primary particles in recirculating hydrocarbon flames

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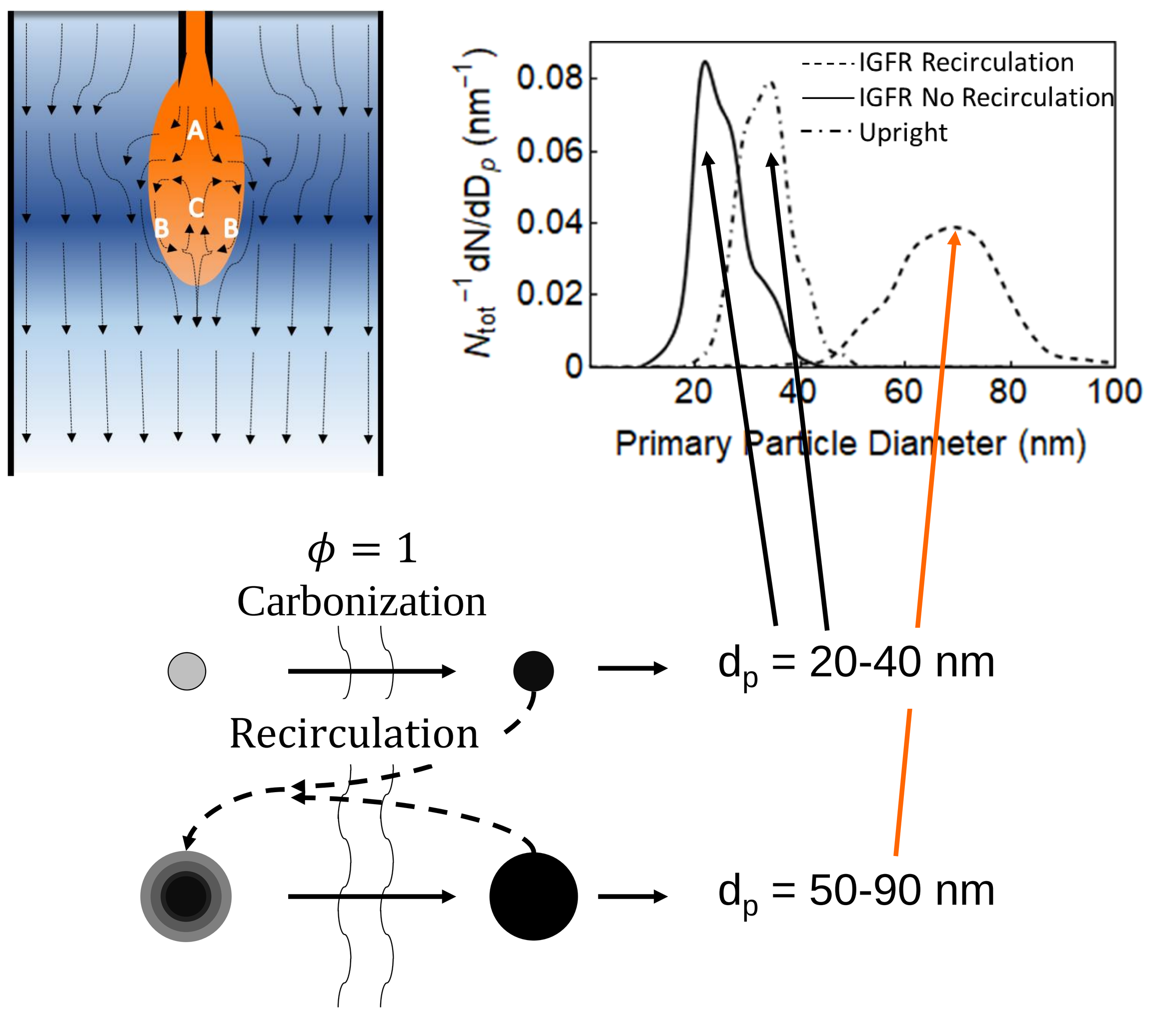
ABSTRACT

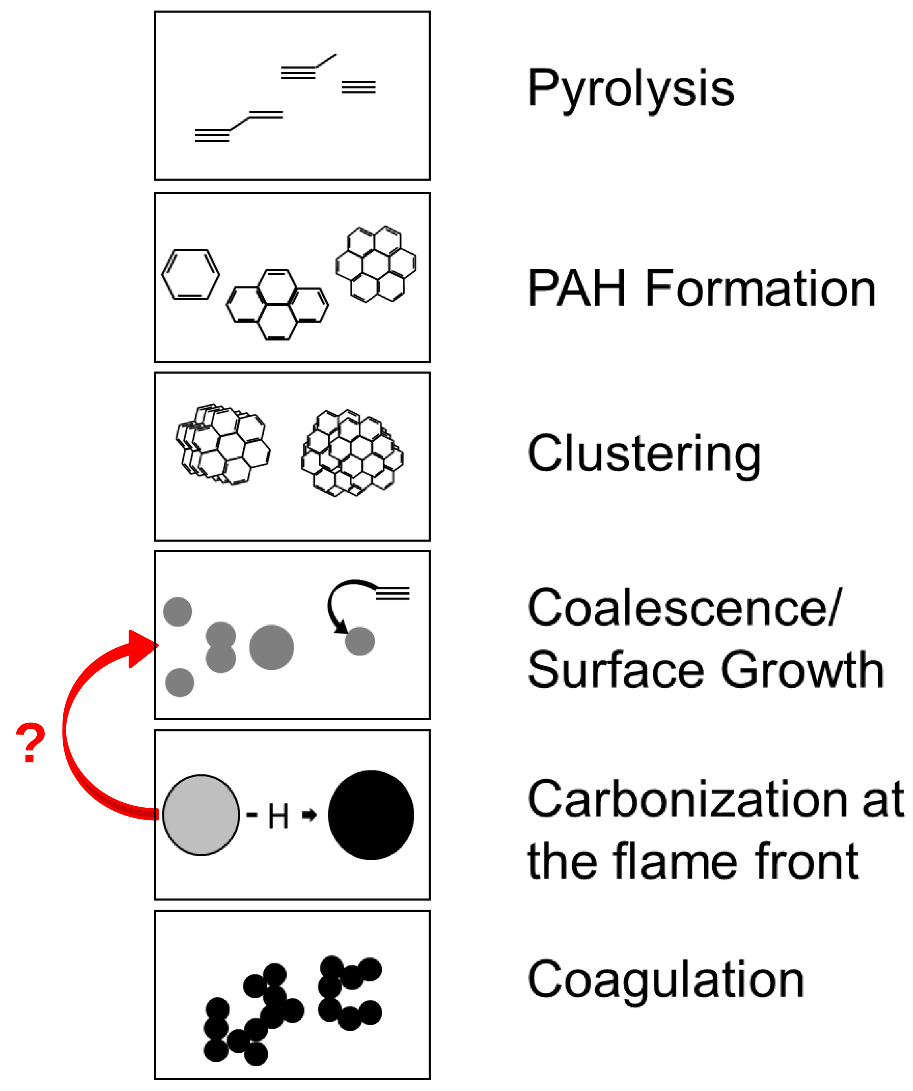


INTRODUCTION & BACKGROUND

Combustion sources are estimated to contribute greater than 25% of total PM2.5 levels in some locations and are detrimental to human health including asthma, cardiovascular neurological diseases, and cancer. Also, the role of combustion generated material in climate modeling is filled with uncertainty due to unknowns in particle characteristics. Both areas create a necessity for information on chemical and physical characteristics of PM2.5.

Research on soot attempts to understand formation, but a lack of modeling accuracy on the transition region from gas phase to solid fractals persists. Soot primary particle diameter (d_p) provides important information on the transition from gas and liquid precursors to solid state fractal aggregates.

Soot Formation Process



Project Objectives

Hypothesis: Repeated particle exposure to the gas and liquid growth species in the fuel rich zone after particle carbonization in the flame front leads to surface growth by formation of liquid hydrocarbon film on the surface followed by solidification of particle due to hydrogen abstraction.

1.Experimentally observe and measure flame conditions leading to particle growth with gaseous C_xH_y fuels

2. Analyze size and structure of nanoparticles

3.Model IGFR to gain insight on variables that play a major role in particle growth

4.Distinguish between recirculating and other flame conditions on particle formation

IGFR DESIGN

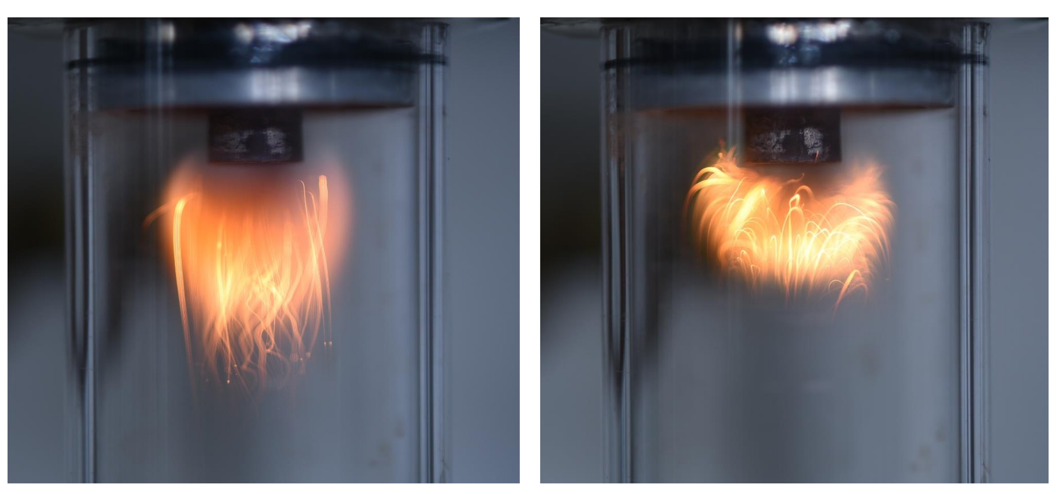


Figure 1. Hydrogen diffusion flames with tracer particles injected to visualize pathlines (streaks). Left: High fuel inlet with no recirculation. Right: Flow moves axially from the nozzle until convective and buoyant forces drive the particles to recirculate and move upwards towards the fuel inlet.

Experimental

***Table 1.** Experimental conditions for CH_4 diffusion flame*

	Upright	IGFR with no Recirculation	IGFR with Recirculation
CH_4 Flow Rate (lpm)	0.13	0.24	0.13
Air Flow Rate (lpm)	1.8	3.3	3.3
Reactor Orientation	upright	Inverted	Inverted
Flame Length (mm)	80.5	60.0	27.2
Residence Time (s)	0.06-0.01	0.3-0.8	0.5-3.0

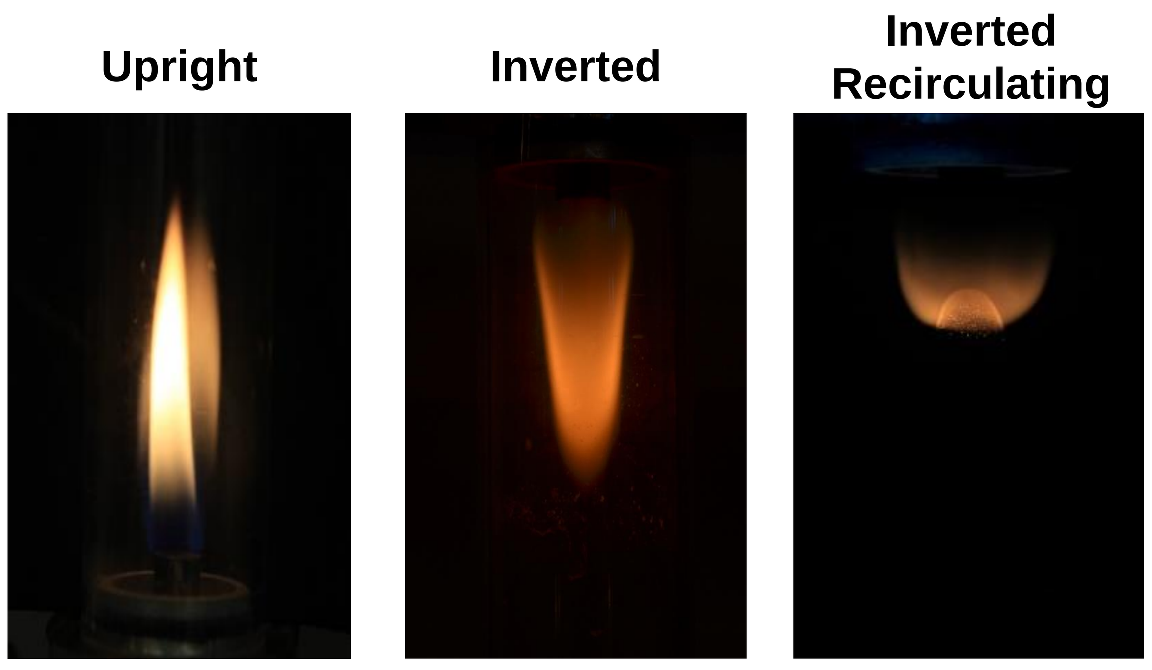


***Figure 2**. Three methane diffusion flames studied. Soot at the base of the recirculating flame is observed flowing from the flame front back inside of the high temperature fuel rich region.*

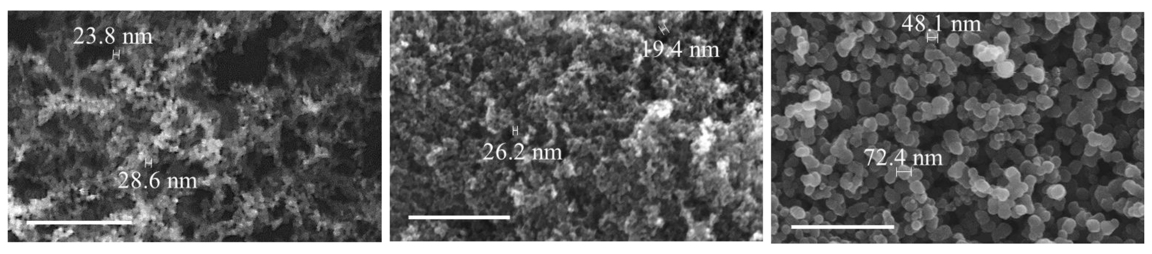


***Figure 3.** SEM images of soot from the three flames shown in Figure 2. Recirculation increases d_p for soot particles. Scale bars represent 500 nm.*

***Table 2.** Morphology of soot collected from the above methane diffusion flames*

Diameter (nm)	33.4±3.9	25.2 ±4.1	68.0±7.9
Number of Particles	150	240	354
Fractal Dimension	1.68-1.69	1.68-1.70	1.69-1.70

$$N = kf\left(\frac{R_g}{d_p}\right)^{d_f} \qquad (1)$$

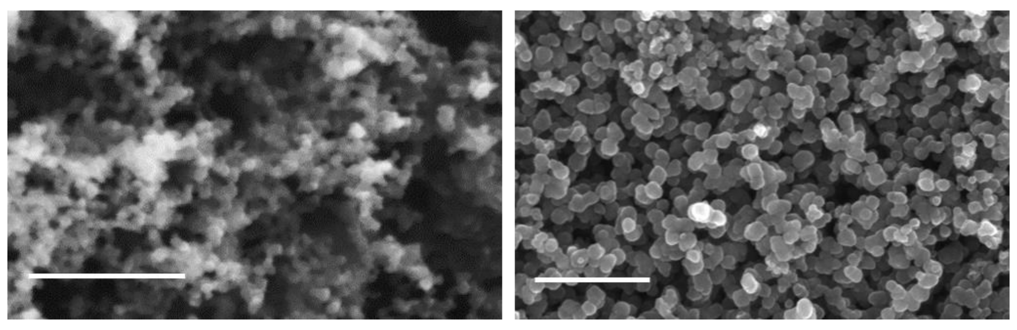


***Figure 4**. Ethylene diffusion flame soot. A recirculating flame creates fractals with a consistently larger d_p. Left: SEM image of soot produced in upright ethylene flame with d_p in typically reported range. Right: SEM image of soot produced in ethylene IGFR recirculating flame.*

***Table 3.** Morphology of soot collected from ethylene diffusion flame*

Ethylene	IGFR	Upright
Diameter (nm)	39.3 ± 5.5	53.7 ± 8.4
Number of Particles	403	150
Fractal Dimension	1.66-1.67	1.58-1.61

NUMERICAL

COMPUTATIONAL FLUID DYNAMICS

- Two dimensional axi-symmetric CFD modeling provides gas flow, temperature, and species profiles in the combustor
- Basic chemistry to be expanded with additional CRN

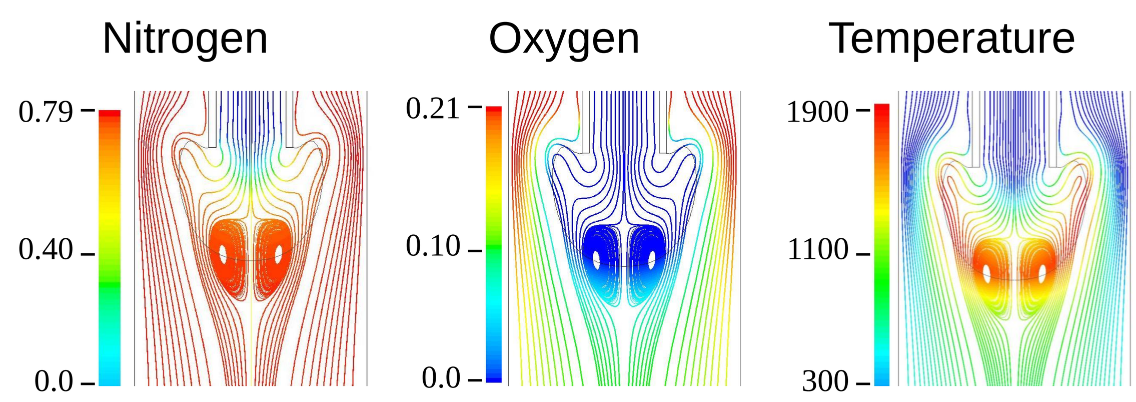


***Figure 5**. Pathlines of the gaseous flow inside the IGFR recirculating flame. Left: Nitrogen mole fraction profile. Middle: Oxygen mole fraction profile. Right: Temperature profile. Temperature includes heat radiation from soot production. Peak flame temperature is 1920 K.*

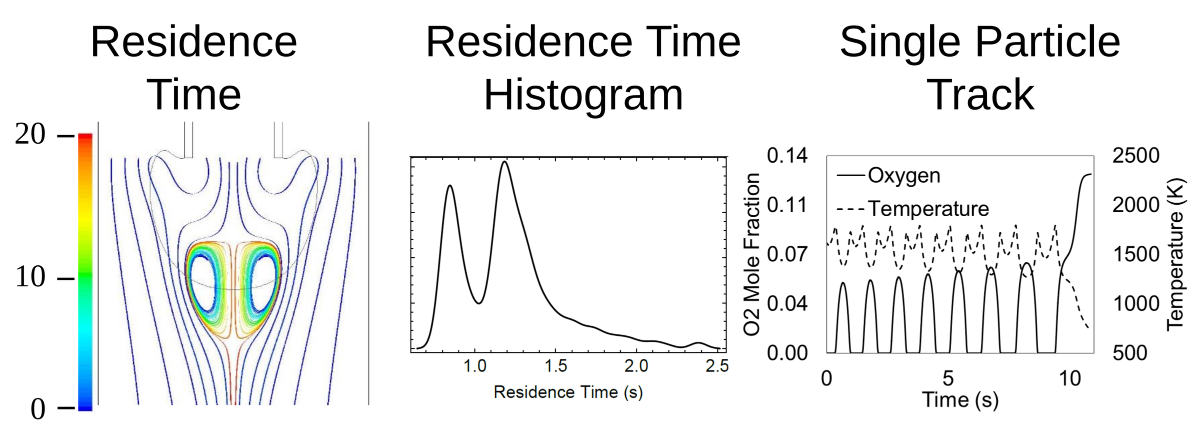


***Figure 6**. Left: CFD showing recirculating pathlines and resulting residence time. Particles in the entrainment have residence times up 20 seconds. Middle: Probability distribution of residence times. Right: The surrounding oxygen and temperature of a single particle entrained for 10 seconds inside the recirculation region. The particle oscillates between rich and lean regions of the reactor.*

CHEMICAL REACTION NETWORK

- CRN based on CFD results and experimental observations for the qualitative representation of growth species
  - Recirculation region from fuel lean to fuel rich, nitrogen profile, stagnation point at flame front, and residence time
- Ability to tweak results to match observable results
- Composed of two Perfectly stirred reactors (PSRs) which assume diffusion is infinitesimally small compared to chemical kinetics
- Chemical kinetics follow Arrhenius relationships for C1 and C2 diffusion flames

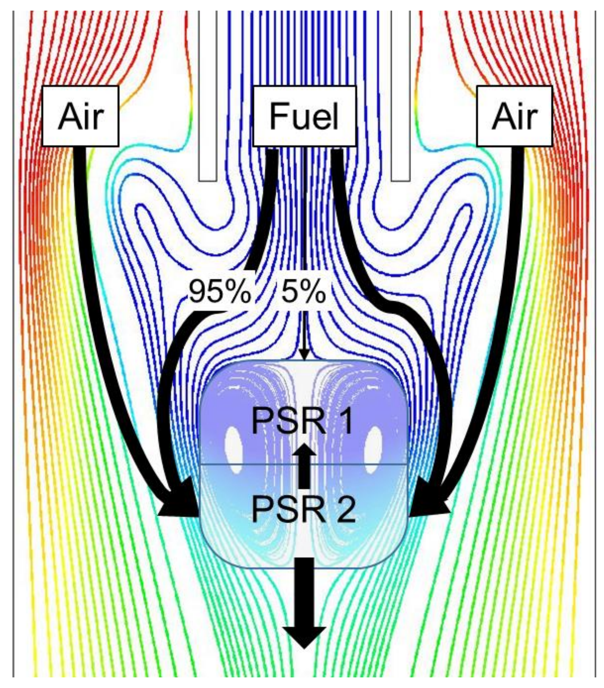


***Figure 7**. CRN created from experimental observations and CFD simulations. The PSR is composed of three main components, a PSR modeling the fuel rich region conducive to primary particle growth, a second PSR at stoichiometric conditions that carbonizes and solidifies soot particles, and a recirculation region that carries soot from the flame front back inside of the fuel rich region.*

***Table 4.** Comparison of CFD and CRN results to verify the accuracy of the CRN model.*

	CFD	CRN [1]	CRN [2]
Fuels flow rate (slpm)	0.10	0.10	0.10
Growth Region Volume (cm^3)	1.59	1.39-1.95	1.41-1.97
Growth Region Temperature (K)	1604	1630-1620	1674-1666
Recirculation mass flow rate (g/s)	9.36E-4	9.35E-4	9.35E-4
Nitrogen Mole Fraction	0.5 -0.70	57.9 – 58.0	57.8-58.9

***Table 5.** Resulting mole fraction of important growth species inside PSR1 for two mechanisms*

	CRN [1]	CRN [2]
Mole Fraction C_2H_2	1.23E-02	1.29E-02
Mole Fraction A1	5.01 E-05	4.22E-04
Mole Fraction A4	2.57E-06	1.37E-05

CONCLUSIONS

•Recirculation of soot particles from oxygen rich to fuel rich regions leads to larger d_p

•Re-exposure to growth region inside flame front with C_2H_2 and PAHs after carbonization and restructuring at the flame front is the reason for larger d_p

•Recirculation increases d_p for multiple gaseous hydrocarbon fuels (methane, ethylene, propane) and changes soot morphology.

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