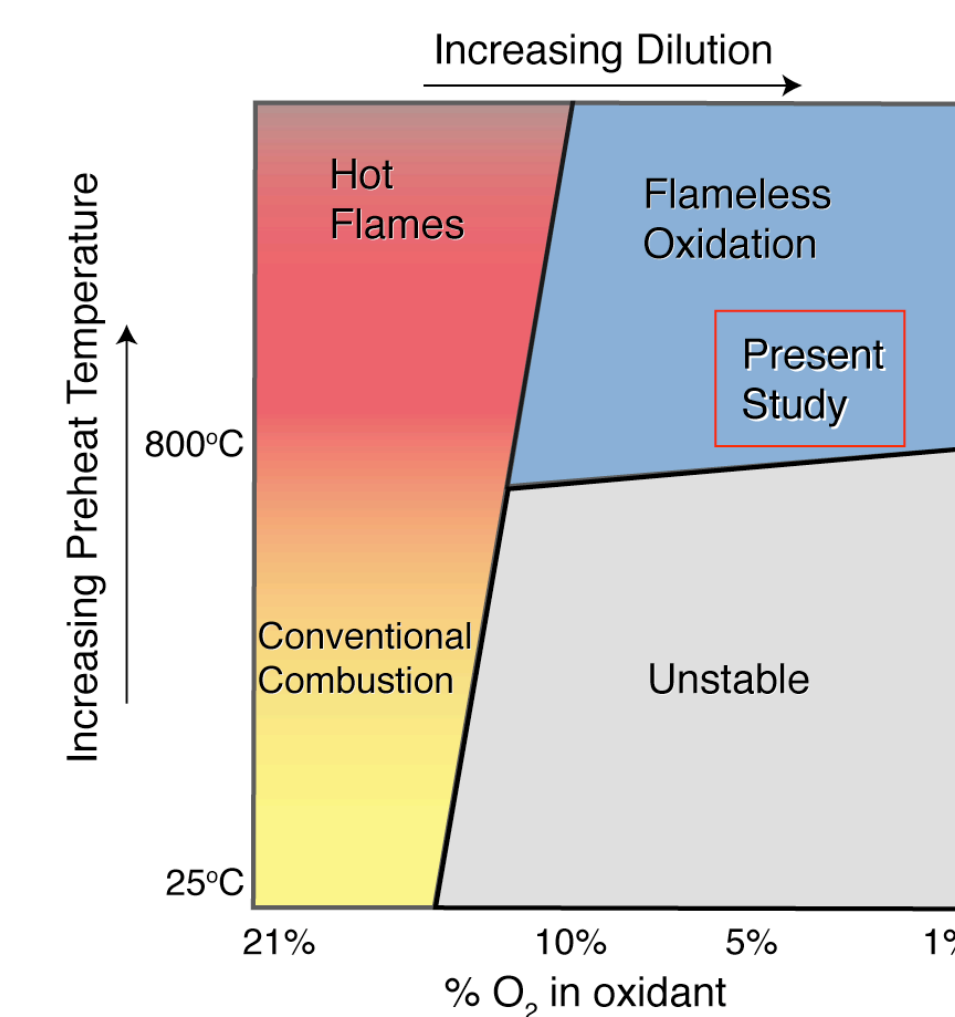


Background

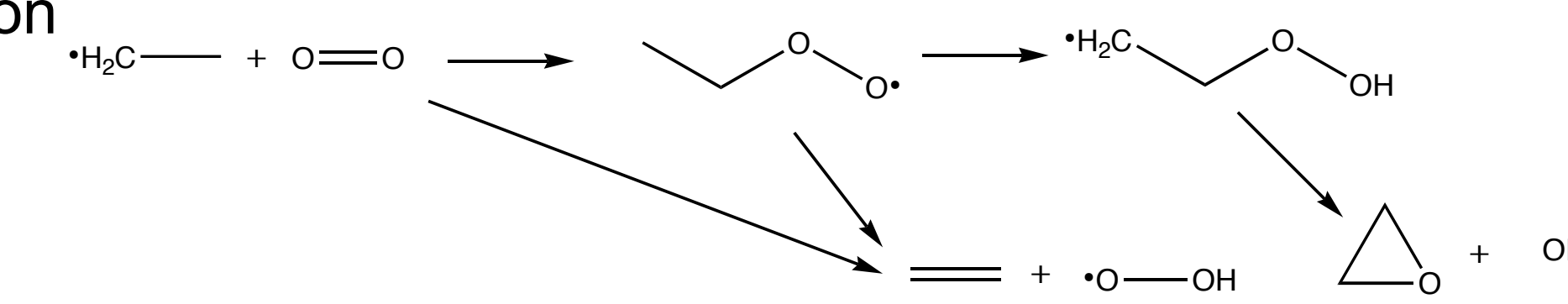
- Hydrocarbon combustion under the conditions of *flameless oxidation*—characterized by well-mixed, dilute, preheated reactants—is a promising technology that can improve thermal efficiency and reduce pollutant and greenhouse emissions.
- Though typically used in industrial furnaces, this process has the potential to deliver improved performance in work-producing devices by matching the time scales for chemical reaction and mechanical expansion.

- Since implementation of flameless oxidation in a gas turbine or homogeneous charge compression ignition engine occurs at pressures much higher than atmospheric, the kinetics of flameless oxidation of fuel-oxidizer mixtures needs to be studied at elevated pressures.
- An improved understanding of autoignition chemistry and refinement of predictive models are essential to the development of these advanced combustion devices



Objectives

- Assess model fidelity and validate experimental techniques
- Acquire experimental data to use in refinement, optimization and reduction of detailed reaction schemes
- Provide high quality experimental data for the development of improved hydrocarbon oxidation mechanisms
- Determine the influence of species from recirculated exhaust gas (e.g., H₂O, CO₂) and from trace concentrations of byproduct species (e.g., NO) on fuel oxidation rates
- Explore the impact of multi-channel reaction pathways on fuel oxidation

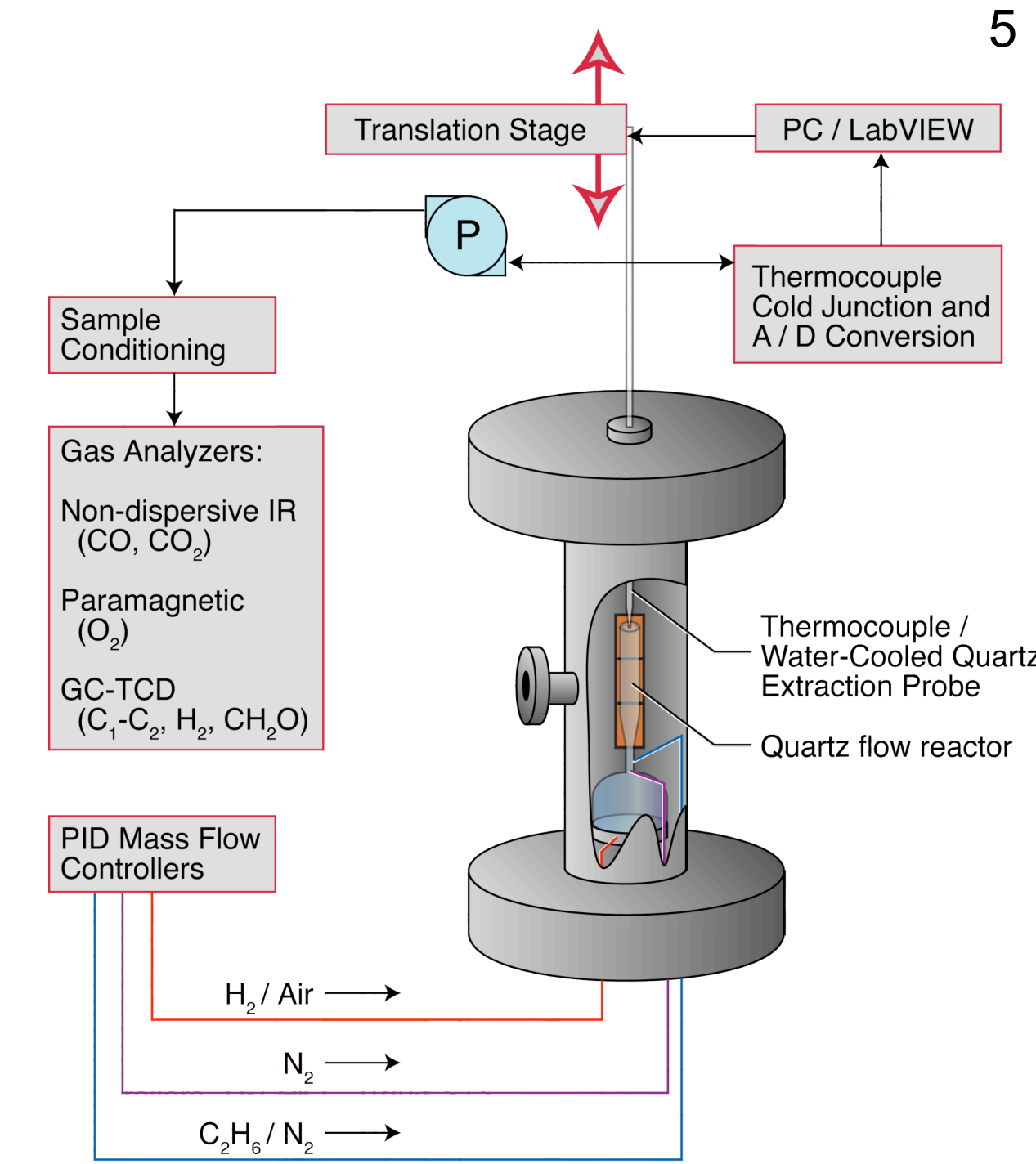


Approach

- Time-evolution of gas temperature and stable species concentration are measured along a nearly adiabatic combustion-driven flow reactor at ambient to elevated pressures and intermediate temperatures.
- Centerline velocity measurements are used to infer the residence time at each measurement location.
- The effect of pressure, temperature and reactant concentration on reaction timescales, heat release rate and intermediate and product species profiles are determined.
- The measured species and temperature profiles are compared to the model predictions using a detailed reaction mechanism.

Experimental Facility

- Flow Reactor**
 - Pressures 1–50 bar
 - Temperatures up to 1100°C
 - Excellent mixing due to 2-stage injection
 - Insulation and heaters provide nearly adiabatic conditions
 - Residence times up to 100 ms
- Thermocouple probe**
 - R-type (Pt - 13%Rh / Pt)
 - 100 mm bead diameter
 - 25 mm SiO₂ coating
 - +/- 5 K accuracy
- Gas Sampling**
 - Extracted and quenched in a water-cooled probe for online analysis
 - Heated sample transfer for condensable species

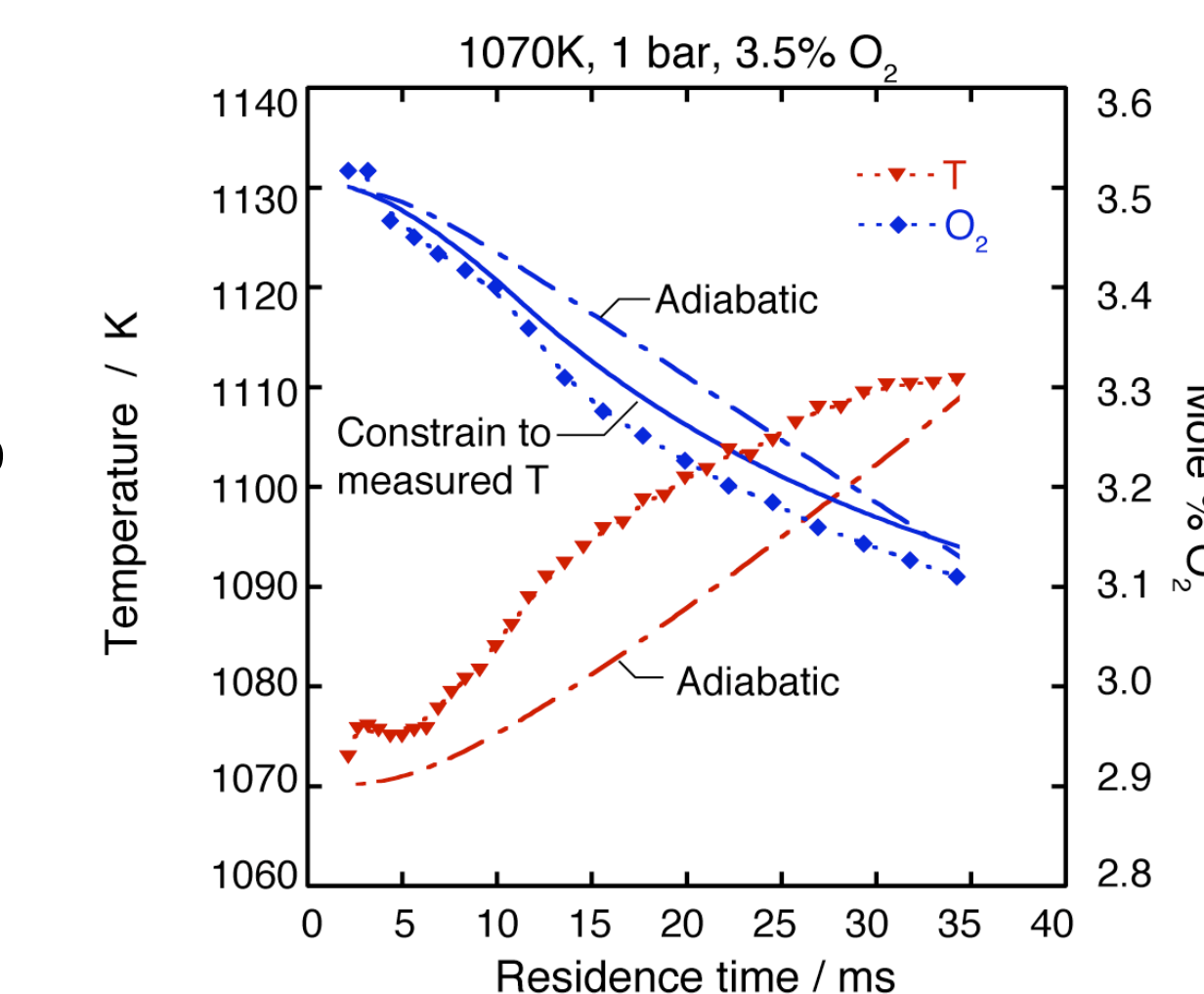


Kinetic Model

- O-D transient reactor model (Chemkin 4.0)
 - Initial conditions set to measured species concentrations at the first well-mixed station
 - Simulation uses measured temperature profiles
 - Mechanism: Naik and Dean, *Combustion and Flame* 145 (2006) 16-37.
 - 2069 elementary reactions among 129 species
 - Rate parameter temperature and pressure dependence for multi-well channels using Chebyshev polynomials
 - Better agreement with experimental data than GRI-Mech 3.0, a standard hydrocarbon combustion mechanism

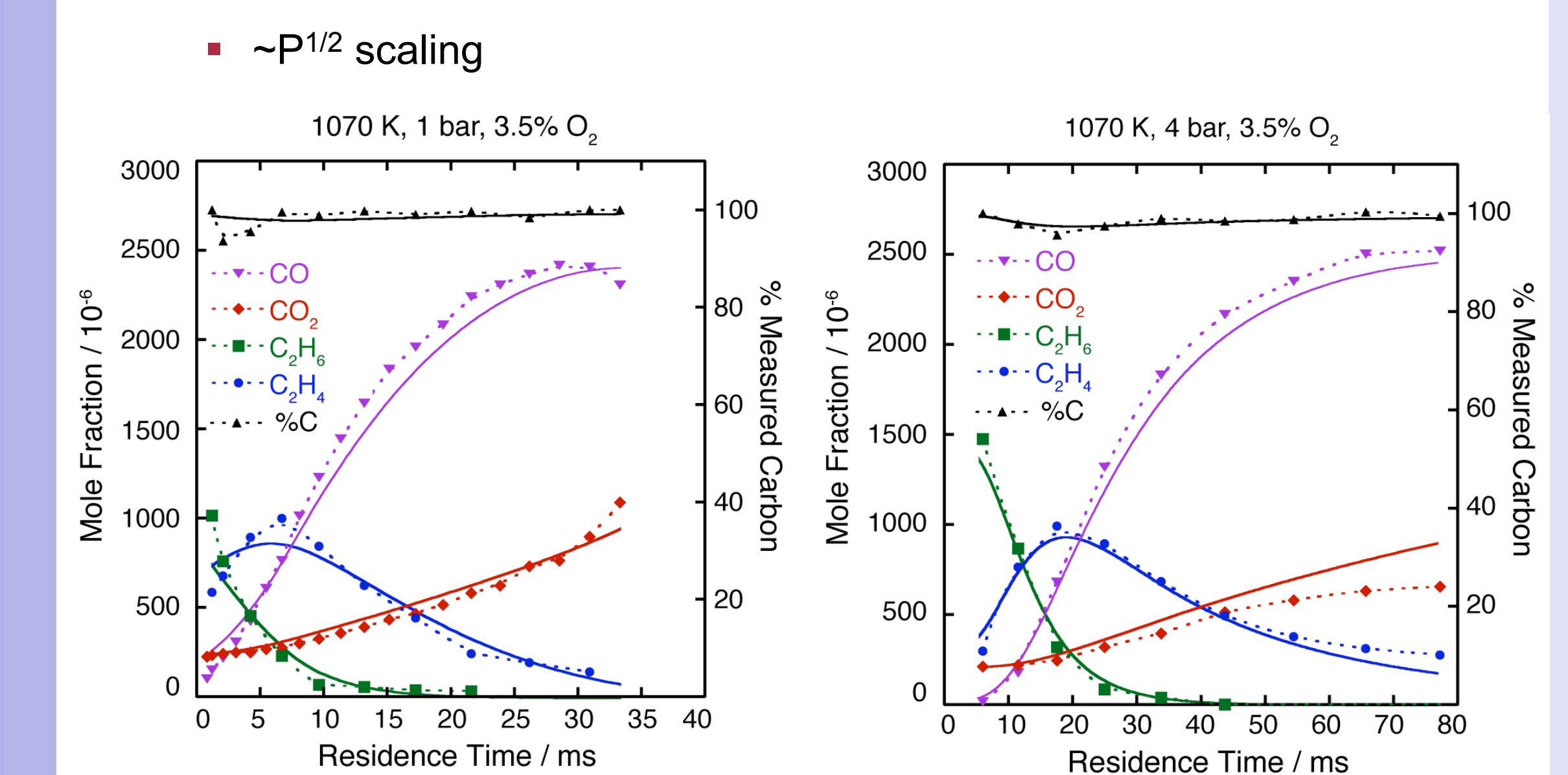
Results

- Experiments have been conducted at 1, 2 and 4 bar with initial temperatures of 1060–1160 K
- Measured heat-release rate differs slightly from the adiabatic prediction, although the time-scales are comparable
- Agreement between measured and predicted species concentrations is improved when the simulation is constrained to the measured temperature profile



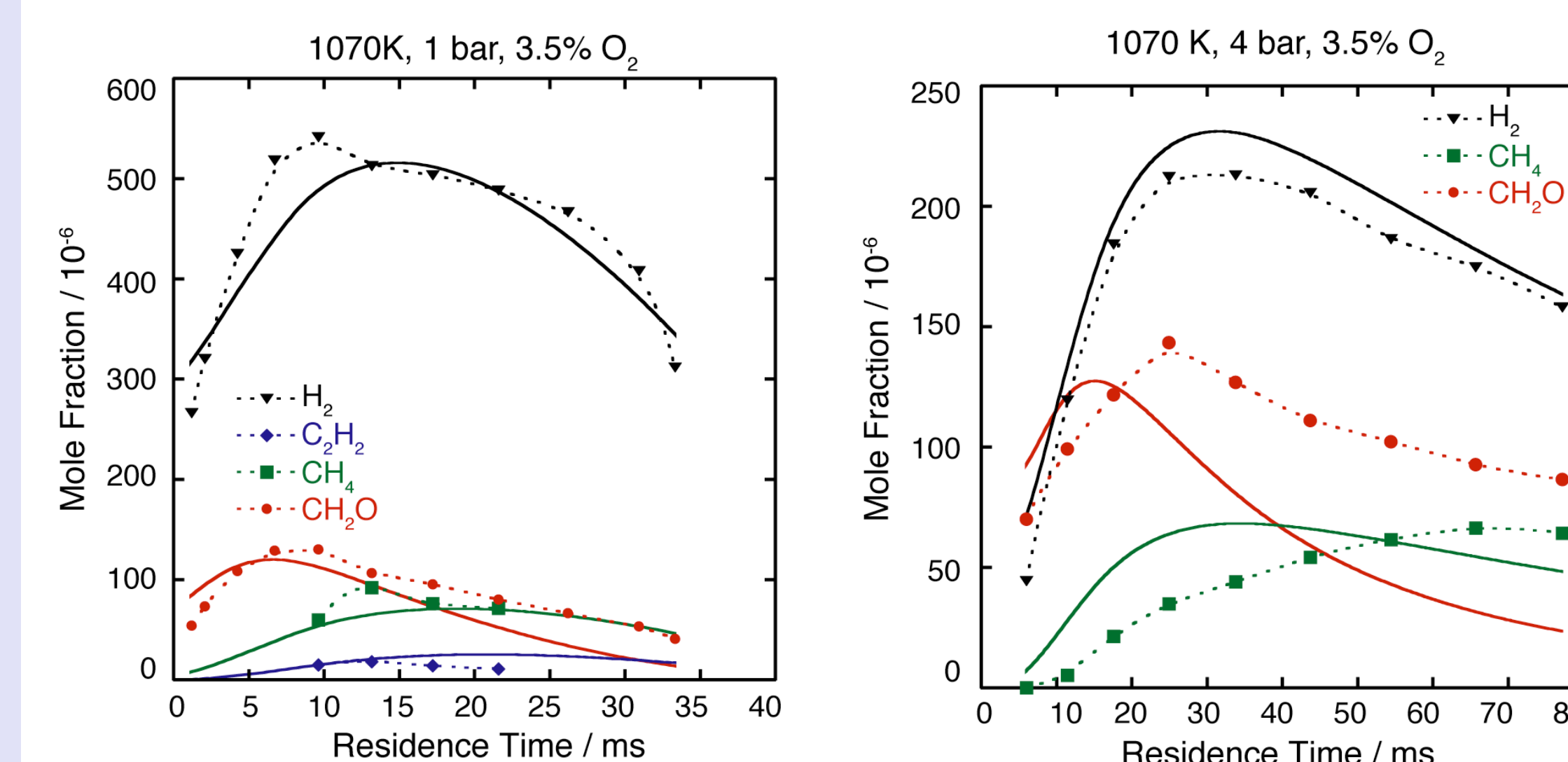
Major Species

- Measured carbon balance closes to better than 5%
- Characteristic reaction time-scales increase with pressure



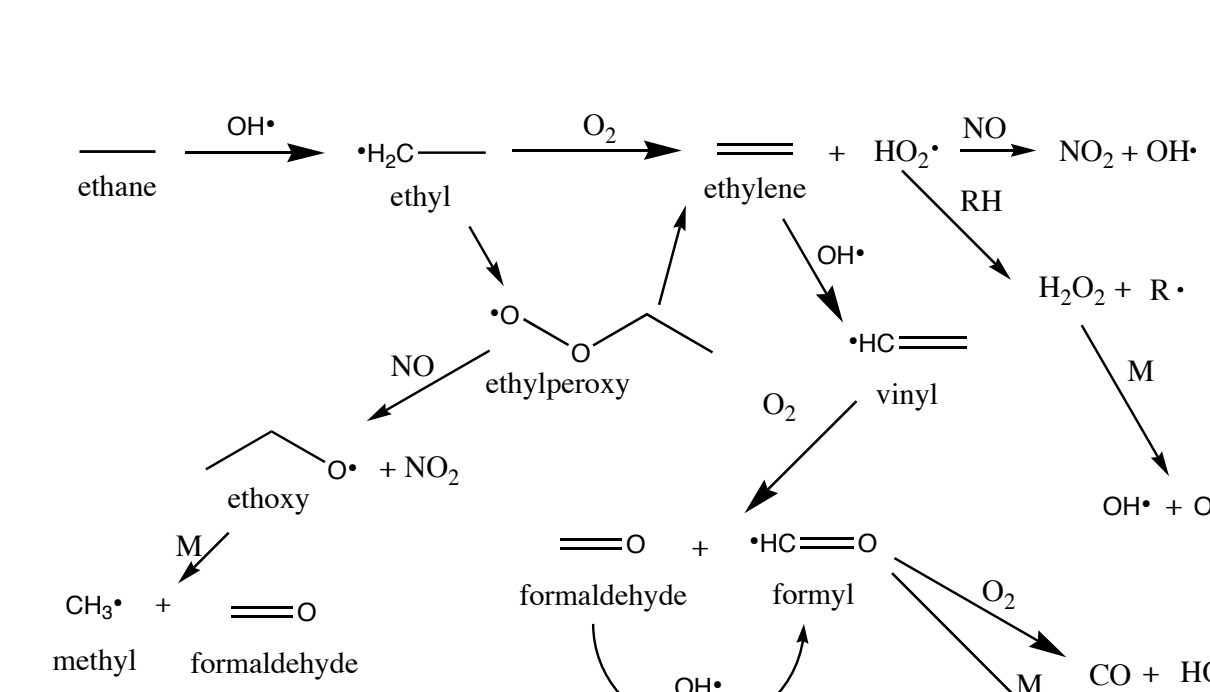
Minor Species

- Moderate levels of H₂, CH₂O and CH₄ with trace concentrations of C₂H₂ are observed
- The model tends to slightly over-predict the rates of CH₄ production and CH₂O removal, especially at higher pressures



Reaction Pathway

- Hydroxyl radicals, generated primarily from reactions involving stable hydroperoxy radical, govern the oxidation of fuel and intermediates
- Radical attack, beta-scission and concerted elimination convert ethane to vinyl radicals
- Vinyl reacts with O₂ to produce CHO+CH₂O, which are subsequently converted to CO and CO₂



Conclusions

- The measured and predicted time-evolution of temperature and species concentration are in relatively good agreement
- The current reaction mechanism accurately predicts the change in reaction time scales due to variations in pressure, temperature and composition
- More than 95% of the carbon present in the unburned fuel can be traced through intermediate species to products
- The presence of NO results in a small enhancement of oxidation rates at early times by contributing to the radical pool
- Opportunities remain for improved accuracy in the measurement and prediction of minor species concentrations

Future work

- Broader range of experimental conditions
 - Higher pressure, lower temperature to further explore the influence of alkylperoxy chemistry
- Optimization and reduction of the current mechanism
- Larger parent fuel (propane, butane)
 - Negative Temperature Coefficient (NTC) behavior
- Oxygenated fuels

