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Argonne NATIONAL LABORATORY

RAPID COMPRESSION MACHINE (RCM) STUDIES TO UNDERSTAND AUTOIGNITION FUNDAMENTALS

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Project ID # ACE054
FY2019 DOE Vehicle Technologies Program Annual Merit Review
Advanced Combustion Engine R&D
Tuesday, June 11, 2019
Program Managers: Gurpreet Singh, Michael Weismiller

OVERVIEW

Timeline

- Project started FY 2011
- Project directions and continuation reviewed annually, and in FY 2019 VTO Advanced Light-Duty Combustion Consortium

Barriers

- Lack of fundamental knowledge of advanced combustion engine regimes
- Lack of modeling capability for combustion, and emissions control

Budget

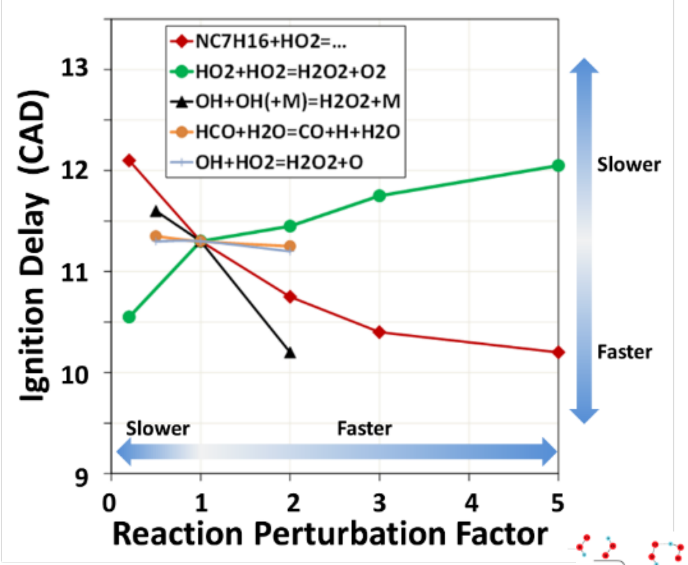
- Project funded by DOE / VTP
  - → FY 2017 funding: $410 k
  - → FY 2018 funding: $310 k
  - → FY 2019 funding: $360 k

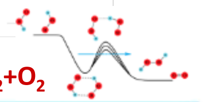
Partners

- ANL – Lead, Goldsborough (PI)
- LLNL – gasoline surrogate model, simulation tools
- TCD – surrogate methodologies
- SNL – gasoline engine data (RD5-87)
- International RCM Workshop

OBJECTIVES AND RELEVANCE TO DOE

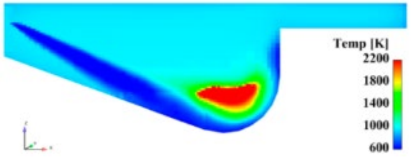
- Acquire fundamental data, and help develop / validate / refine chemical kinetic and relevant models for transportation-relevant fuels (conventional and future gasolines, diesels and additives) at conditions representative of modern / advanced combustion regimes, leveraging collaborations with researchers across the broader community.
- Predictive simulations with such models, which require low associated uncertainties, could be utilized to overcome technical barriers to advanced compression ignition schemes, and achieve required gains in engine efficiency and pollutant reductions.

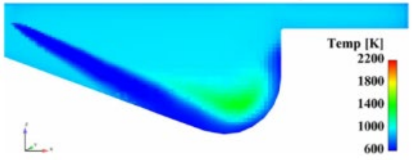




doi:10.1021/jz400874s

HO_2+HO_2=H_2O_2+O_2





PROJECT MILESTONES

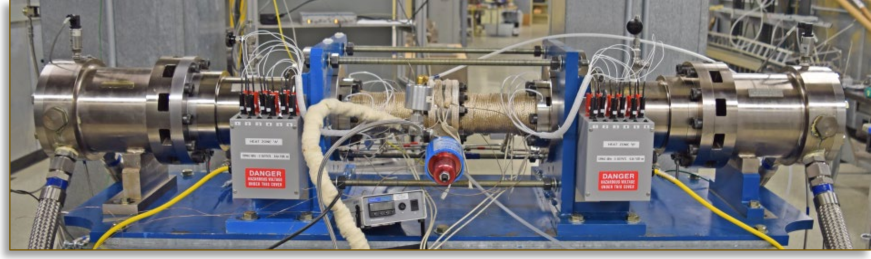
FY 2019

Task	Milestone	Status
1	Experimentally quantify chemical exothermicity (LTHR/ITHR/HTHR) of PRF90, and identify trends across a wide range of engine-relevant conditions, comparing measurements to kinetic model predictions.	✓
2	Acquire autoignition measurements for representative, branched olefin (2M2B) spanning ranges of temperature, pressure, and stoichiometry.	✓
3	Acquire autoignition measurements of high RON / high Sensitivity, binary blends of branched olefin + aromatic (2M2B + TOL), to probe antagonistic/synergistic trends.	✓
4	Acquire autoignition measurements for multi-component surrogate blends to mimic 'neat', and ethanol-blended gasolines (E0–E30); Evaluate, quantify performance of surrogate formulation approaches.	ongoing
5	Acquire autoignition measurements for CRC AVFL31b project (four full boiling-range gasolines). [Data to be presented after permission granted by CRC.]	ongoing
6	Coordinate RCM Workshop 2nd Characterization Initiative, CFD activities, and organize 4th International RCM Workshop (Dublin, IRELAND).	✓

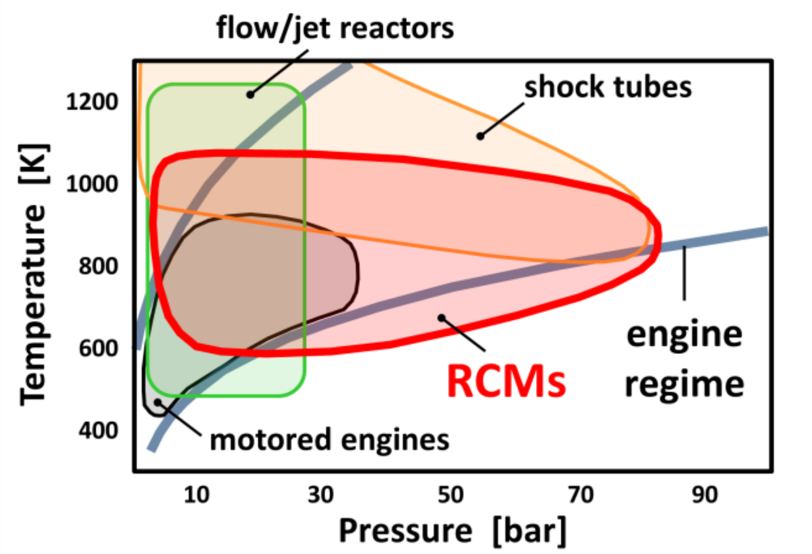
PROJECT APPROACH

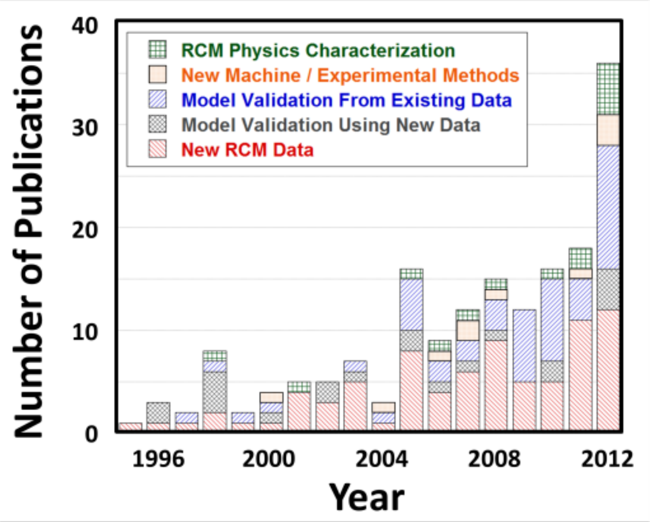
PROJECT APPROACH

Rapid Compression Machine



- Utilize ANL's twin-piston RCM to acquire autoignition data



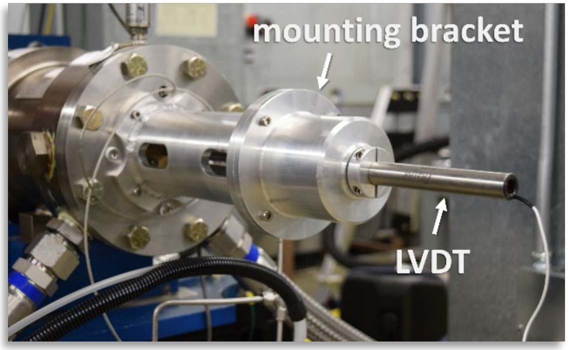


- Employ novel data analysis tools and advanced diagnostics
  - Physics-based, reduced-order system model;
  - New, integrated physical gas sampling capabilities to better probe chemistry.
- Synergistically improve kinetic models using analysis techniques (e.g., UQ/GSA) and detailed calculations/measurements of sensitive processes (e.g., individual reaction rates)

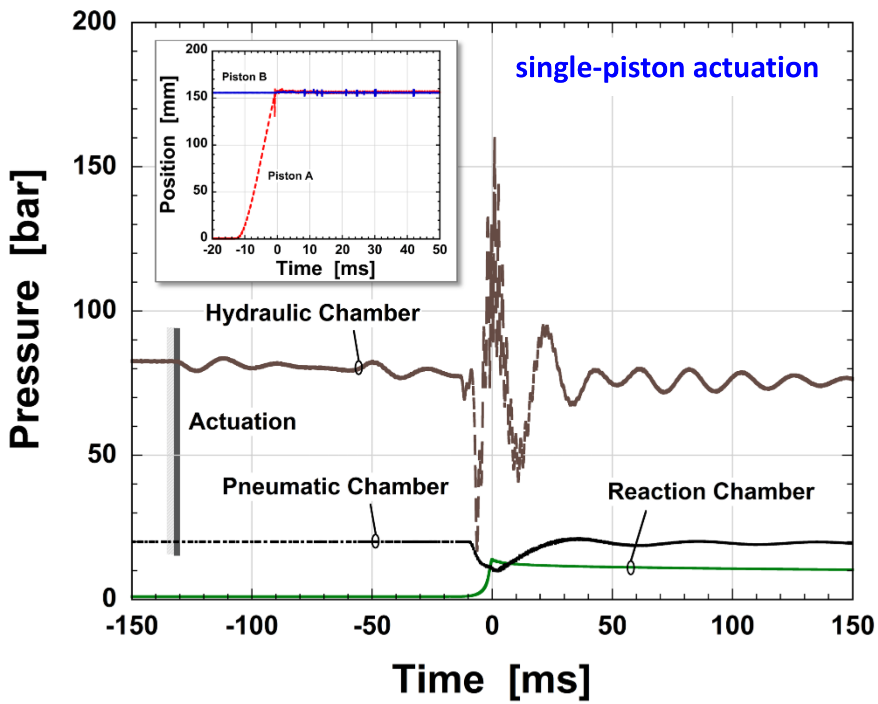
TECHNICAL ACCOMPLISHMENTS / PROGRESS

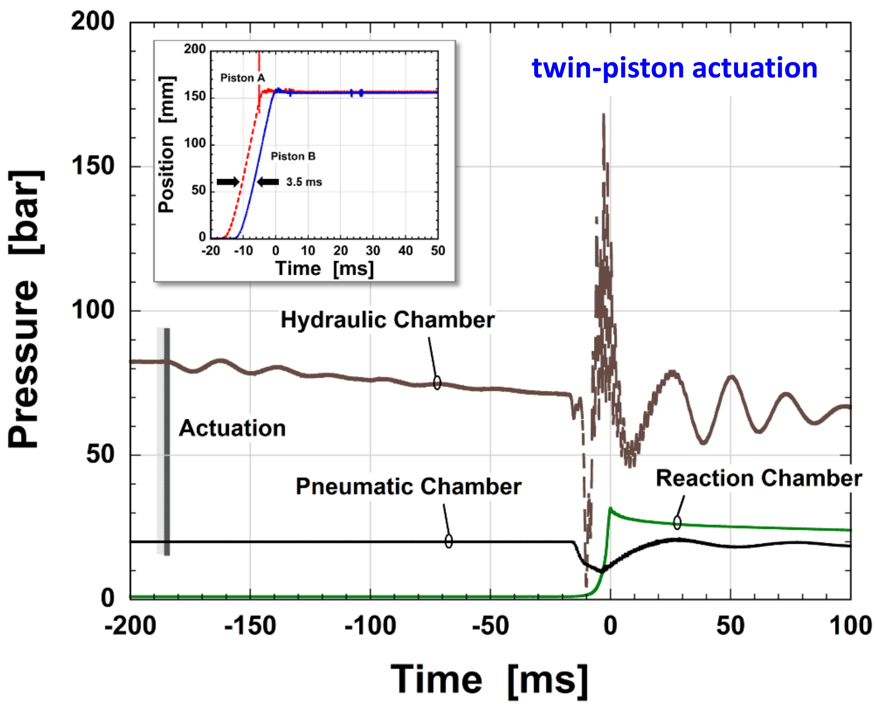
TECHNICAL ACCOMPLISHMENTS / PROGRESS

Twin-Piston Rapid Compression Machine



- Modifications, upgrades implemented since FY 2018 AMR to improve measurement capabilities
  - LVDT dynamic tracking of pistons; dynamic measurement of pneumatic, hydraulic pressures
    - machine operating performance to be enhanced, e.g., piston synchronization
    - Physics of in-cylinder (reaction chamber) processes can be better understood
    - Heat release analyses to be augmented using measured, dynamic geometry



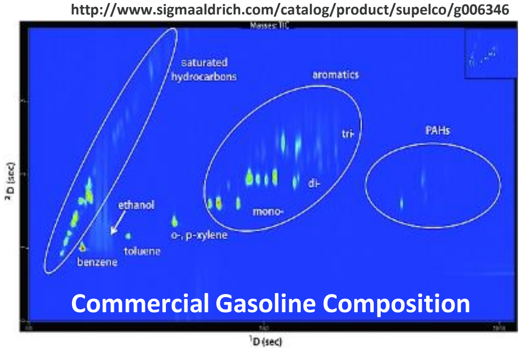


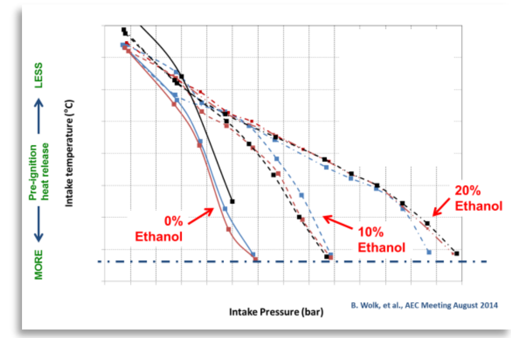
TECHNICAL ACCOMPLISHMENTS / PROGRESS

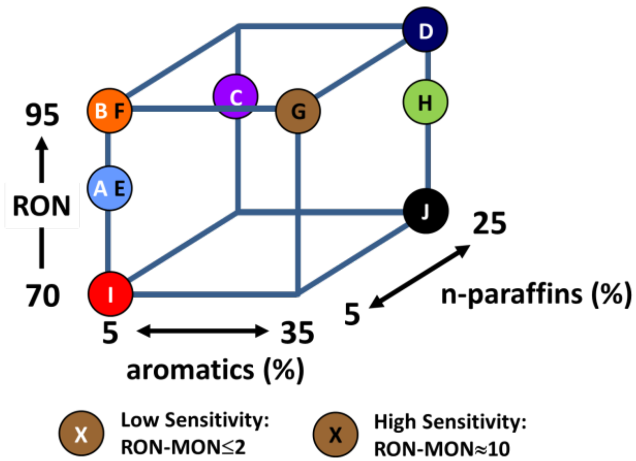
Gasoline, Components and Surrogate Blends

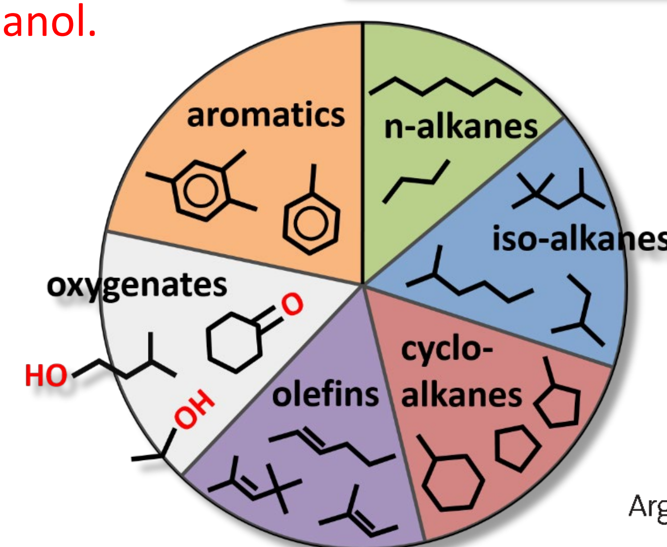
- Predictive modeling of combustion to guide design
  - Gasoline is complex, and compositionally variant
    - → How do these features affect autoignition behavior, especially phenomena at engine-relevant, low to intermediate temperatures (T = 600–1100 K)?
    - → How can real fuels be represented by multiple-component (3-10) formulations?
    - → Well-characterized data are needed to compare autoignition behavior of real, full boiling range fuels with surrogates, including individual components, blends of these, and mixtures with ethanol.

http://www.sigmaaldrich.com/catalog/product/supelco/g006346





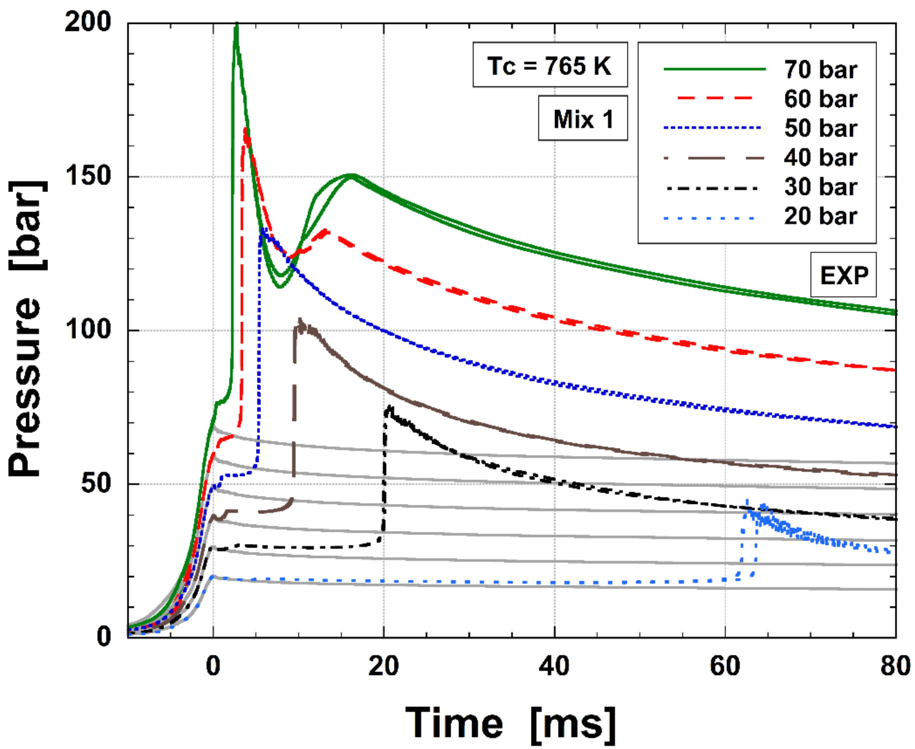


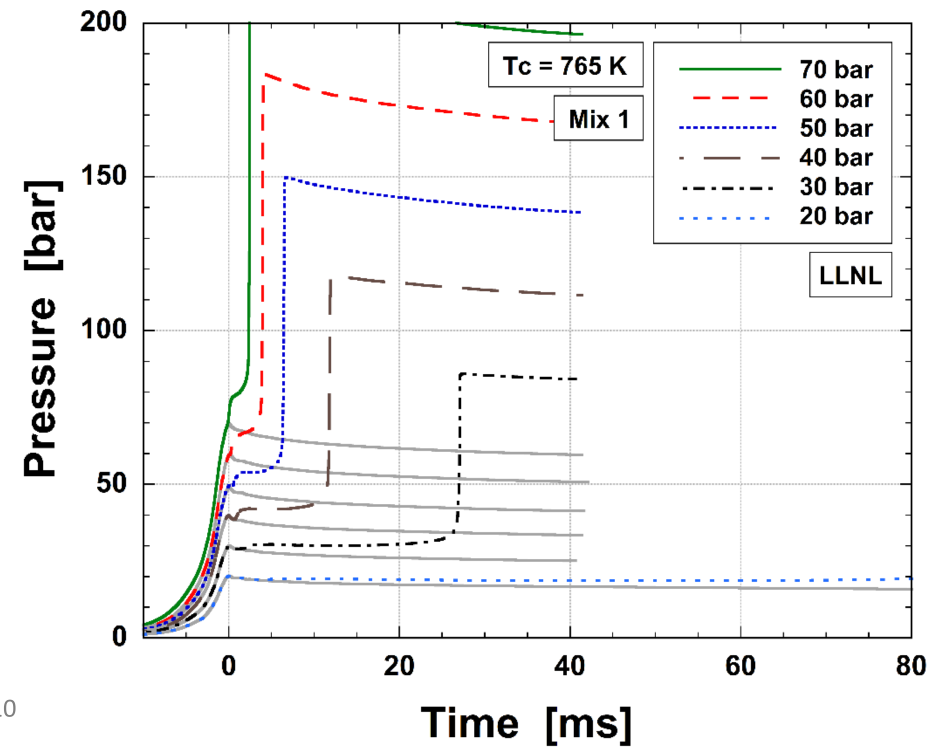


TECHNICAL ACCOMPLISHMENTS / PROGRESS

Task 1 – Chemical Exothermic Behavior (PRF90)

- Quantifying autoignition, heat release processes is critical towards understanding fuel-engine interactions, predicting engine performance
  – Measurements conducted with PRF90 over wide ranges of compressed temperature (Tc) and pressure (Pc), with ignition delay times, as well as chemical exothermicity quantified;
  – Comparisons made against most-recent, LLNL gasoline surrogate model.

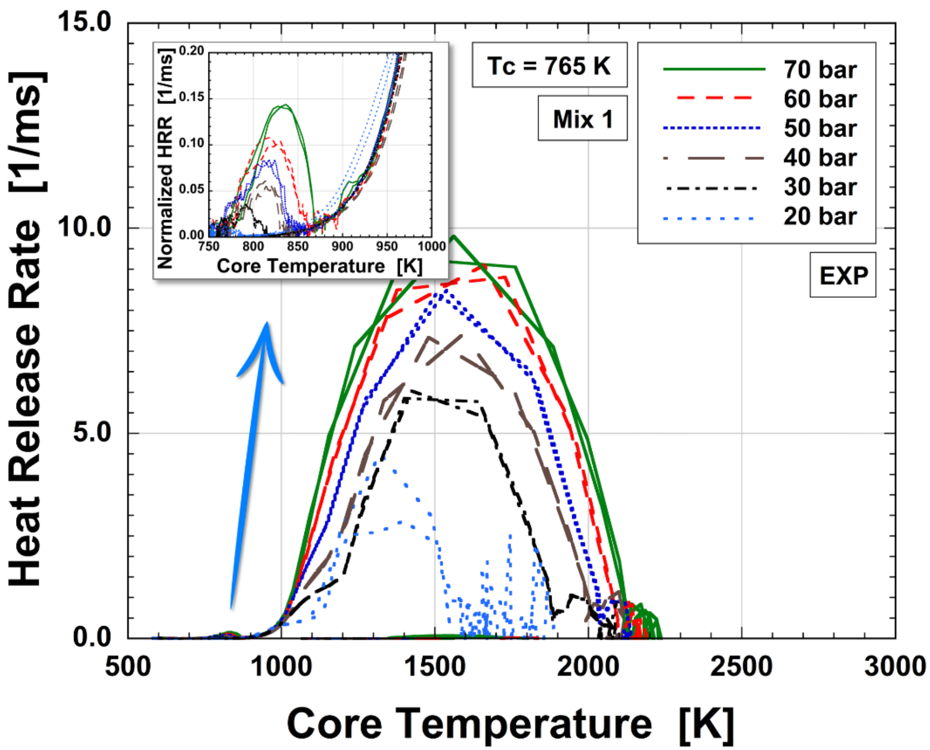


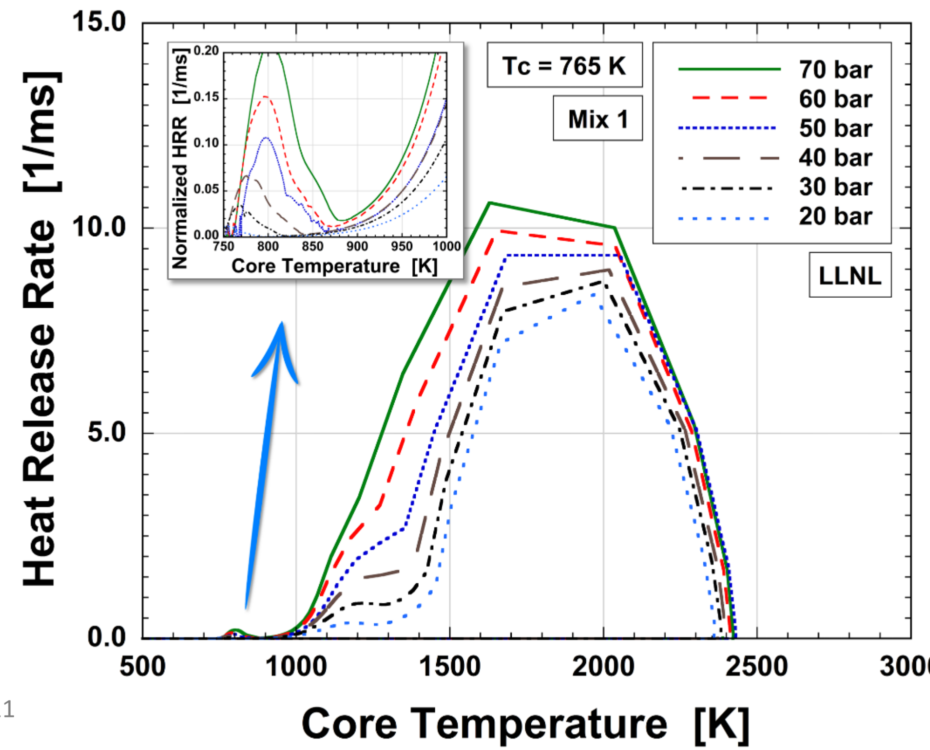


TECHNICAL ACCOMPLISHMENTS / PROGRESS

Task 1 – Chemical Exothermic Behavior (PRF90)

- ‘Adiabatic core’ model employed to facilitate calculation of heat release rates, reacting temperatures through autoignition process
  - Low-, intermediate-, and high-temperature heat releases identified, with shifts observed across Tc, Pc; slow oxidation detected at tail end of process;
  - Similar features observed in model, though discrepancies exist for all three phases of predicted exothermicity.

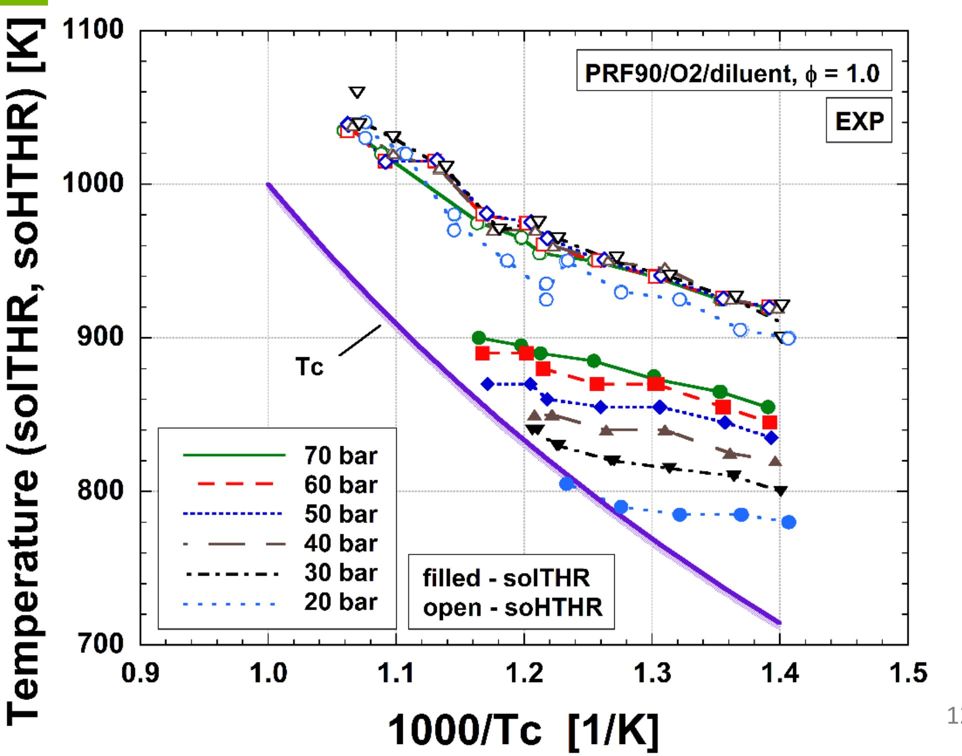


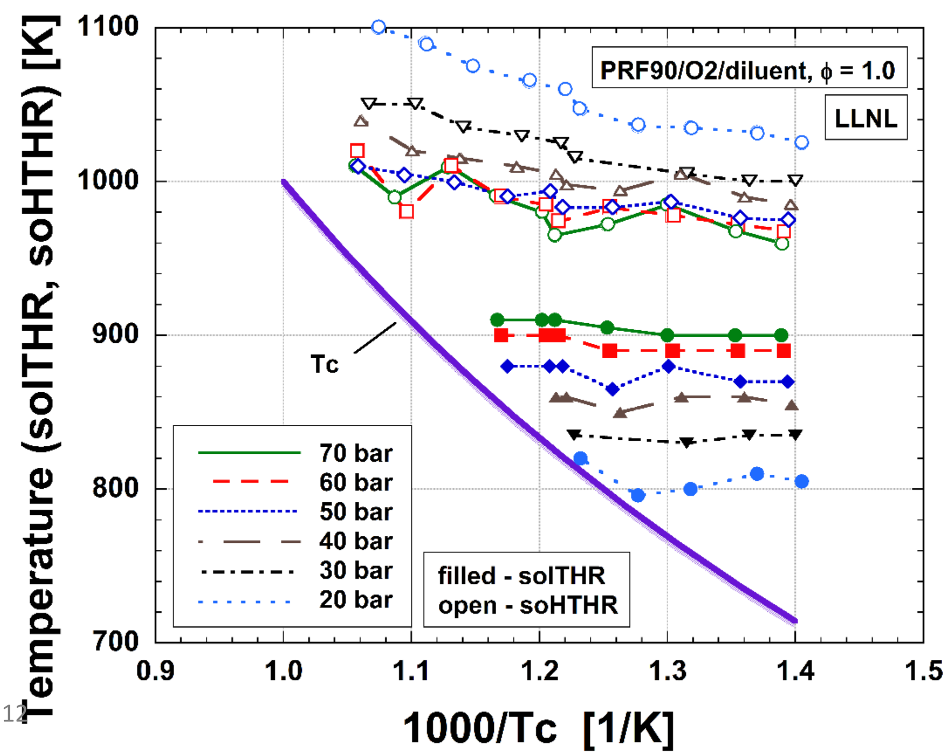


TECHNICAL ACCOMPLISHMENTS / PROGRESS

Task 1 – Chemical Exothermic Behavior (PRF90)

- Transitions between three exothermic phases demarcated to quantify trends, compare extents of exothermicity with model
  - Temperatures at starts of ITHR, HTHR exhibit clear trends across Tc, Pc;
    - Experiments indicate little dependence of TsoITHR on Tc, and TsoHTHR on Pc;
    - Model indicates similar TsoITHR behavior, but greater dependency of TsoHTHR on Pc; efforts ongoing to understand differences.

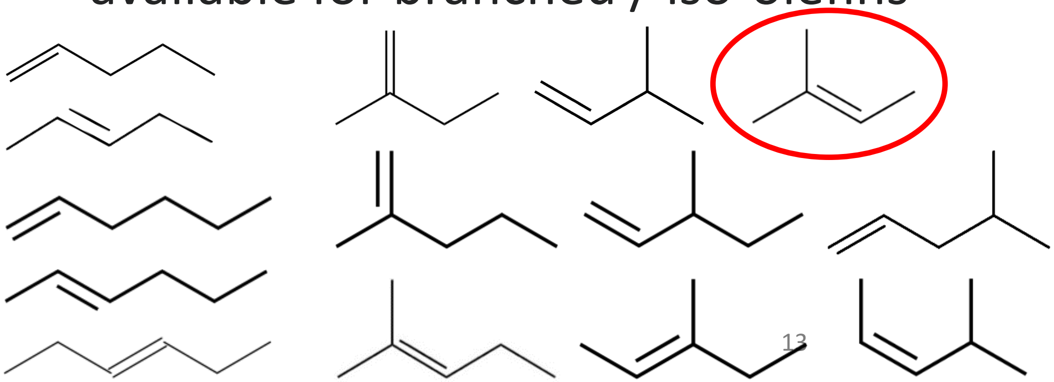




TECHNICAL ACCOMPLISHMENTS / PROGRESS

Task 2 – Measurements of gasoline-representative olefin (2M2B)

- Commercial gasolines typically contain olefins
  - Low, to substantial content depending on source (2–20% vol./vol.)
  - Wide range of linear and branched structures can be present
- Olefins can impart beneficial fuel characteristics
  - Improvement to knock resistance, especially at lower concentrations
  - Substitute for aromatics (within regulated limits)
- Olefins are combustion intermediates of other fuel structures
  - Understandings can be applied to many sub-mechanisms
- Linear olefins have been investigated for some time, but far less data / fewer models available for branched / iso-olefins



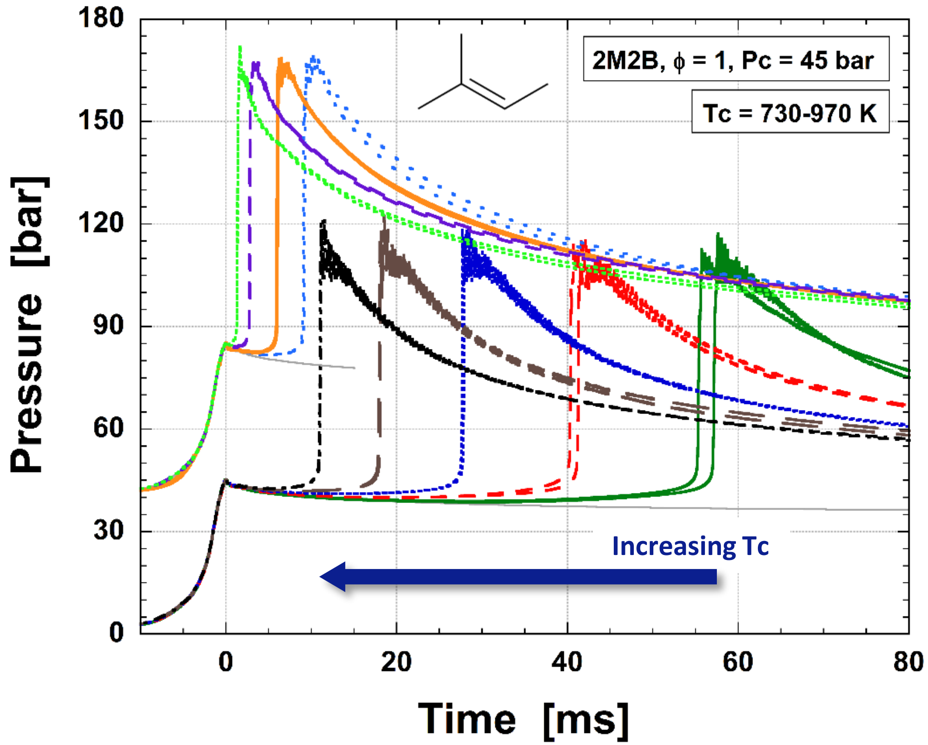
Representative gasoline content

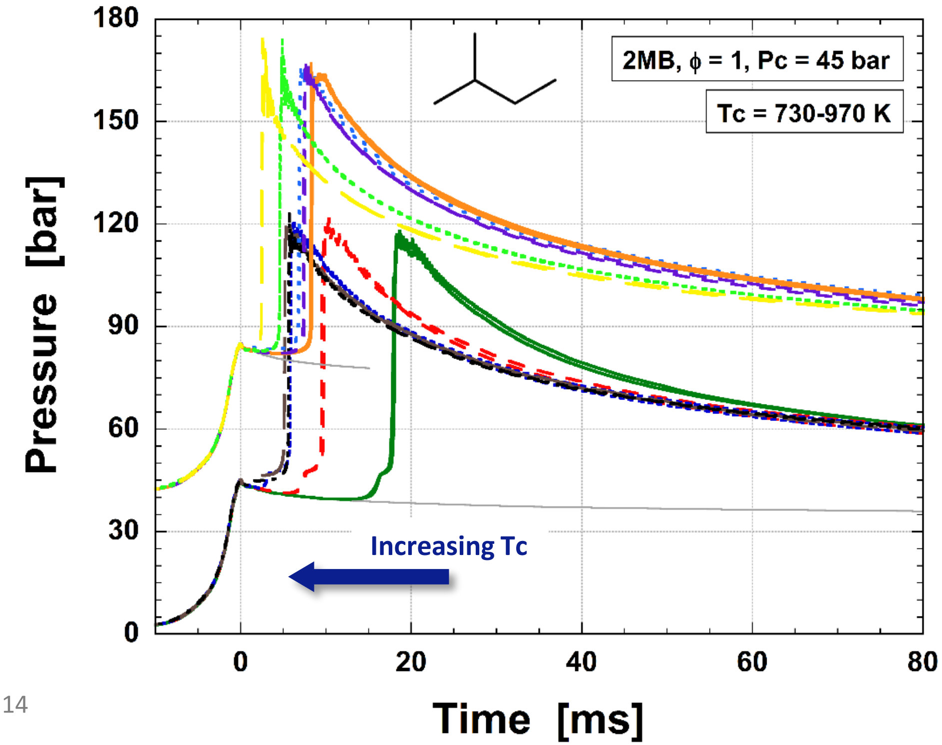
	%-mol	RON	MON
Linear			
1-pentene	7.5%	90.9	77.1
2-pentene	11.5%		
1-hexene	0.7%	76.4	63.4
2-hexene	3.7%	92.7	80.8
3-hexene	2.2%	94.2	80.1
Branched			
2-methyl-1-butene	7.5%	100.2	81.9
3-methyl-1-butene	1.4%		
2-methyl-2-butene	20%	97.3	84.7
2-methyl-1-pentene	2.3%	94.2	81.5
3-methyl-1-pentene	0.6%	96.0	81.2
4-methyl-1-pentene	0.4%	95.7	80.9
2-methyl-2-pentene	5.0%	97.8	83.0
3-methyl-2-pentene	2.9%	97.2	81.0
4-methyl-2-pentene	1.5%	98.8	83.5

TECHNICAL ACCOMPLISHMENTS / PROGRESS

Task 2 – Measurements of gasoline-representative olefin (2M2B)

- Tests conducted at 730–970 K (covering low-, NTC- and intermediate-temperatures), Pc = 25, 45 bar, and ϕ = 0.5, 1.0, 2.0
  - comparison to measurements of 2MB highlight impacts of 2M2B C=C bond
- 2M2B is much *less reactive* at lower Tc; slightly *more reactive* at higher Tc
- 2MB exhibits two-stage ignition at low-temperature

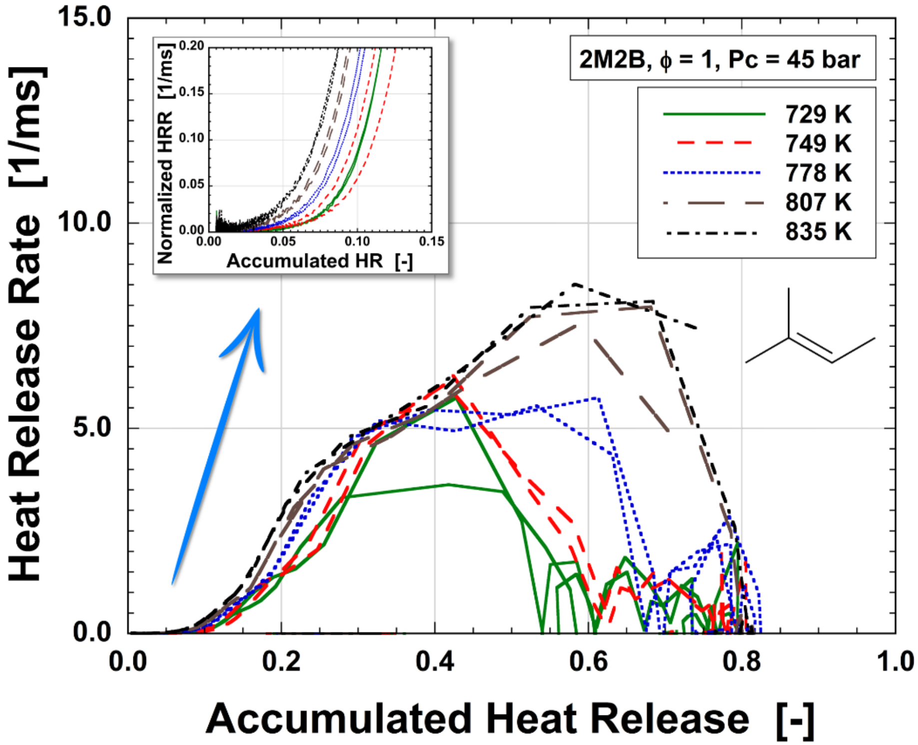


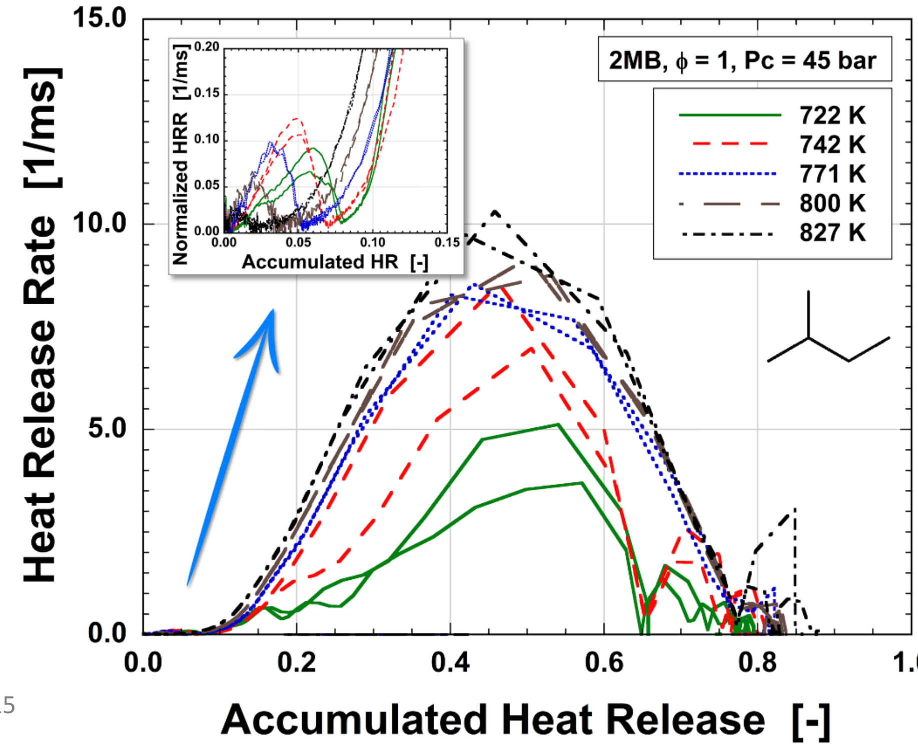


TECHNICAL ACCOMPLISHMENTS / PROGRESS

Task 2 – Measurements of gasoline-representative olefin (2M2B)

- Heat release analyses reveal similarities / differences between 2M2B and 2MB
  - 2M2B exhibits no LTHR, though magnitude of ITHR (i.e., near transition to HTHR, @0.1/ms) appears comparable;
  - HTHR phase for 2M2B and 2MB exhibit similar peak heat release rates, as well as slow oxidation periods at tail ends of ignition (also seen at Tc > 840 K).

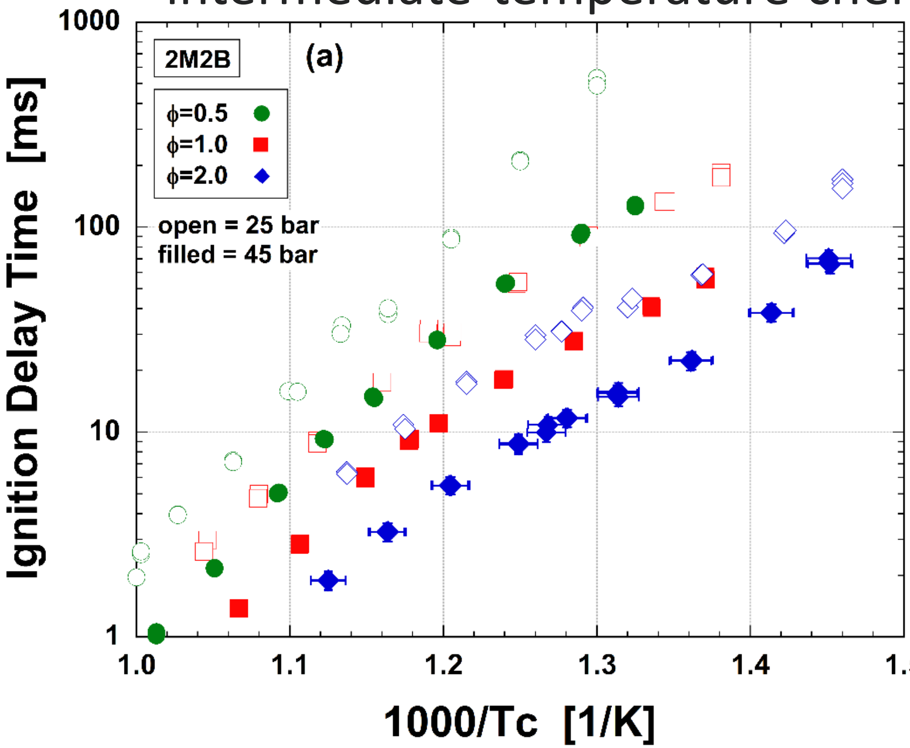


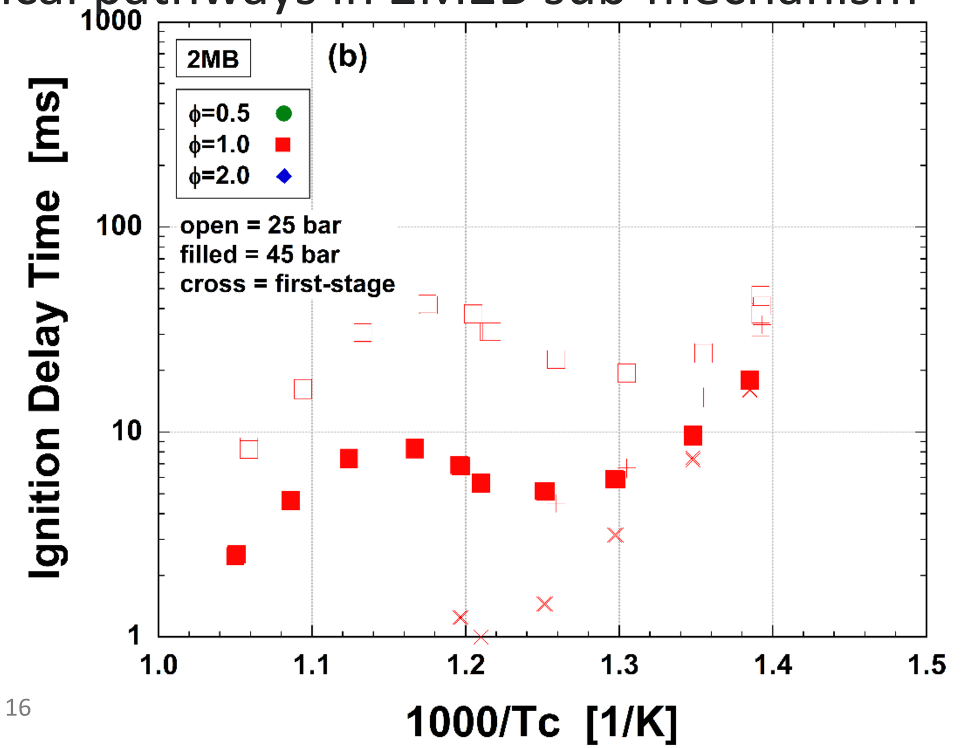


TECHNICAL ACCOMPLISHMENTS / PROGRESS

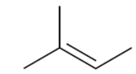
Task 2 – Measurements of gasoline-representative olefin (2M2B)

- Temperature plots highlight non-Arrhenius behavior observed for 2M2B, but without NTC; 2MB exhibits typical alkane trends
  - At lower Tc, 2M2B is *less reactive* than 2MB; at higher Tc, 2M2B is *more reactive* than 2MB.
- Collaboration ongoing with LLNL to develop / validate NTC-, intermediate-temperature chemical pathways in 2M2B sub-mechanism

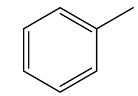


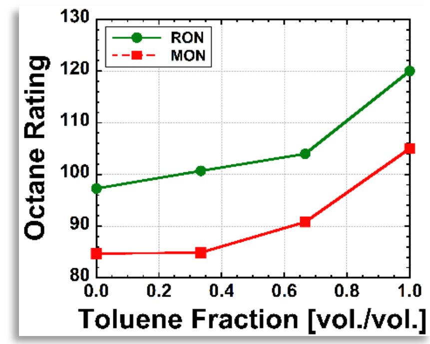


TECHNICAL ACCOMPLISHMENTS / PROGRESS



Task 3 – Measurements of high RON/S blend (2M2B + TOL)



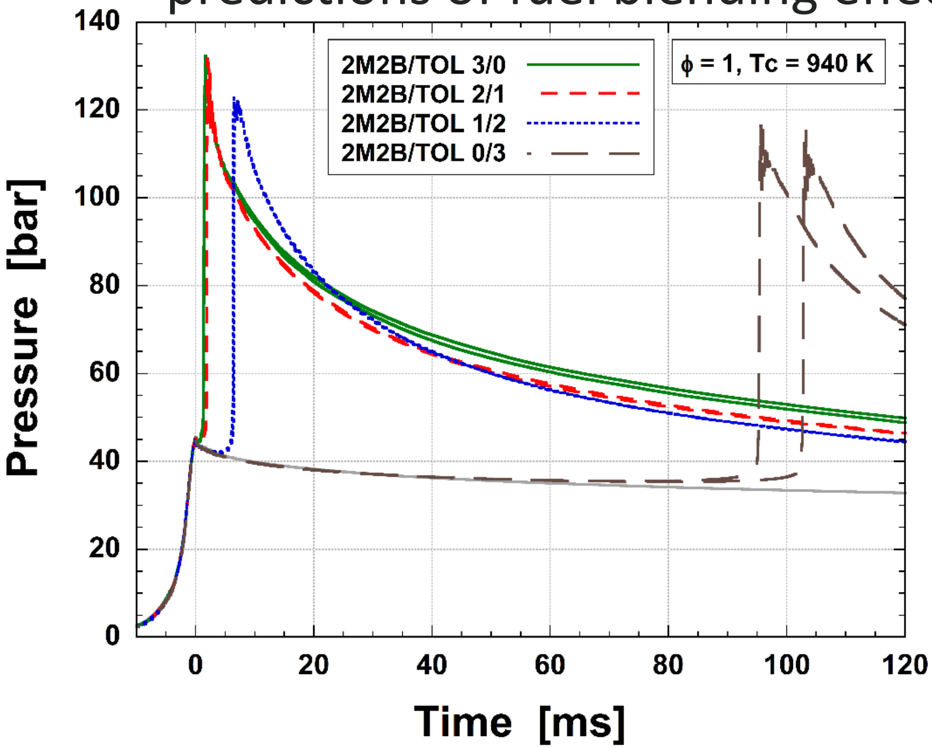


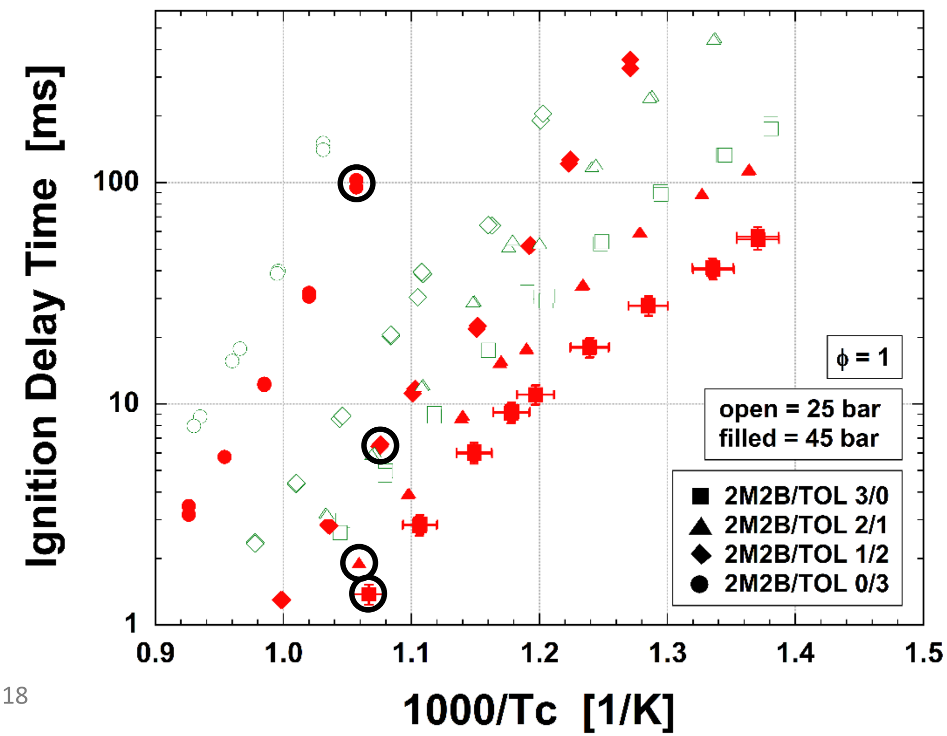
- Mono-aromatics and olefins are major constituents of commercial gasoline fuels
  - Chemical structures can impart beneficial fuel characteristics such as improved knock resistance (high RON), and octane sensitivity (high S)
- Interactions between fuel components are critical to understand, providing bases for interpreting fuel-engine interactions
  - Measurements are needed to quantify interactions, and validate chemical kinetic modeling
- New experiments conducted in FY 2019 with blends of 2-methyl-2-butane + toluene (2M2B + TOL) at 2:1 and 1:2 ratios
  - Tc = 700–1100 K; Pc = 25, 45 bar; ϕ = 0.5–2.0

TECHNICAL ACCOMPLISHMENTS / PROGRESS

Task 3 – Measurements of high RON/S blend (2M2B + TOL)

- Data indicate, like octane ratings, highly non-linear blending effects between these two fuel components, across all Tc, Pc and fuel loadings
  - Autoignition processes (at these conditions) are driven by reactivity of 2M2B, with toluene having little (e.g., radical-scavenging) effects.
- Collaboration ongoing with LLNL to properly capture chemical kinetic predictions of fuel blending effects.



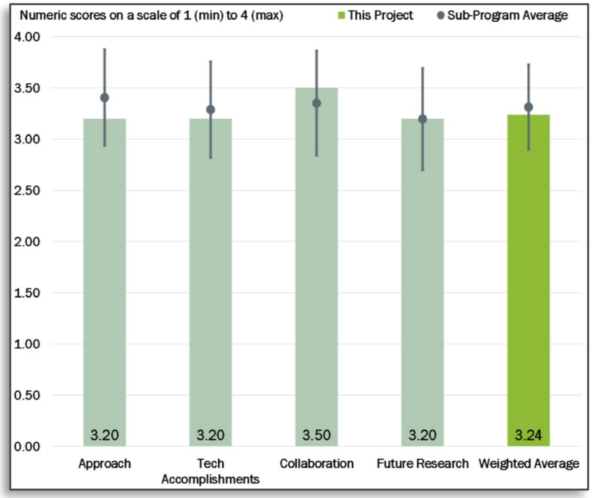


TECHNICAL ACCOMPLISHMENTS / PROGRESS

Task 6 – International RCM Workshop

- Data from many facilities used for kinetic model development
  - RCMs and shock tubes cover a wide range of engine-relevant conditions
- 2nd Characterization Initiative ongoing to better understand and quantify facility influences, and platform-to-platform differences
  - To-date, contributions from 13 facilities worldwide using ethanol/'air', T_c = 780–1100 K, P_c = 20, 40 bar; non-reacting / reacting test measurements
  - Further confirm primary causes of machine-to-machine differences
  - Are there ways to modify design / operating protocol for more consistency?
  - Identify / promote 'Best Practices' for reporting / archiving datasets (collaborate with EU COST SmartCats Program WG4)
- Computational Fluid Dynamics Initiative
  - Enhance understandings of machine-to-machine, test-to-test differences
  - Explore challenges with advanced diagnostics implementation
    - Intrusive (e.g., physical sampling) and non-intrusive (laser-based)
  - Identify / promote 'Best Practices' for modeling of RCM experiments

RESPONSE TO REVIEWER COMMENTS



- This project addresses VTO technical barriers and supports several other VTO projects. The approach is clearly laid out, it is properly managed, with progress demonstrated from last year. The RCM work is useful for establishing kinetic fundamentals for gasoline-representative fuels that transpire at engine-relevant conditions. An impressive amount of experimental data has been produced to study ethanol effects, the behavior of full boiling range gasoline and intermediate temperature chemistry. The work establishes a base of scientific data needed to develop models and experiments for improvement to engine efficiency.
- Excellent coordination all around across multiple subjects and levels, with extensive sharing of information within the international community. The RCM Workshop is a great move and contribution. It will be interesting to see how computational calculations can be used to aid understanding of facility-to-facility measurement differences. It is good to see shock tube measurements included as reference for understanding / validation.
- Proposed future work supports VTO goals of improving kinetic mechanisms and reducing uncertainties in ignition characteristics for conventional and advanced fuels, with benefit across multiple other VTO projects. Work should also target single, gasoline-representative fuel molecules, along with capability to understand interactions within mixtures. Additional interaction with US universities is encouraged, and funding should be increased, if possible, to facilitate this.
  - The project team at ANL actively coordinates with the LLNL groups focused on chemical kinetic modeling, and advanced numerical tools. This extensive collaboration is leveraged to ensure that RCM experimental campaigns target fuels and conditions needed to address surrogate model deficiencies (such as branched alkenes (iso-olefins), and blending interactions in FY19), and thus model improvement. These interactions are ongoing, with flexibility used in test definitions.
  - Attempts are made in FY19 to initiate / extend collaborations with US universities

COLLABORATIONS

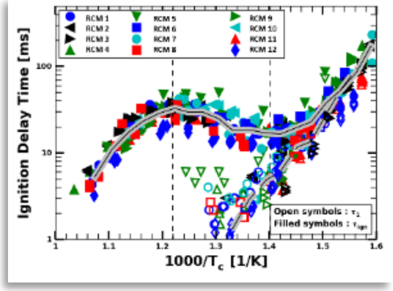
Ongoing Interactions (Inside / Outside VTO)

- DOE Working Groups: share data at meetings of AEC MOU, Co-Optima teams
- CRC FACE Working Group: participate in meetings; testing of AVFL-20 fuels
- ANL: gasoline engine data-sharing; physical gas sampling
- LLNL/Polimi: kinetic model development / validation; formulation of gasoline surrogates; ToolKit development / testing
- SNL: LTGC engine data with RD5-87; tests with surrogate molecules
- Trinity College Dublin: functional-group basis for surrogate formulation
- RCM Workshop: facility-to-facility influences; 'Best Practices' (data reporting/ archiving standards)
- Other organizations: NUI Galway (kinetic models, similar fuel testing); Université Lille 1 Sciences et Technologies (similar fuel testing); Vrije Universiteit Brussel (CFD, reduced-order physical models); ETH-Zürich (DNS of RCM / autoignition processes); Cal State LA (scaling analyses); University of Illinois Chicago (gas sampling/analysis)

COLLABORATIONS

Community-wide Activities

- ANL-led, International RCM Workshop to better understand autoignition chemistry, turbulence-chemistry interactions, etc. using RCMs
  - Participation includes experimentalists, modelers, theoreticians
  - Establishing consensus for ‘Best Practices’
    - → Approaches for reporting / analyzing / comparing data
    - → Approaches for simulating the experiments
    - → Uncertainty quantification for experiments and modeling
    - → Overlap with other experimental devices (e.g.,shock tubes)



  - CFD (MZM, RANS, LES, DNS) to explore measurement challenges
  - 4th Workshop held 27 July 2018 at Trinity College Dublin, IRELAND (in conjunction with Int. Combustion Symposium, TNF, ISF, Flame Chemistry…)

- Working Group formed to expand collaborations to Rapid Compression Expansion Machine laboratories
- Development of online forum for data-sharing, technical knowledge exchange, announcements, etc.

REMAINING CHALLENGES / BARRIERS

- Understanding and representing the autoignition characteristics of full boiling range fuels, blending with ethanol, etc., via multiple-component (3-10) surrogate mixtures requires improved capabilities to formulate surrogates, considering new methods and surrogate components.
- Improvements to gasoline surrogate model require deeper understandings of mechanism behavior, and uncertainties associated with low temperature chemistry pathways of base model.
- Ignition delay time and preliminary heat release are integrated metrics for ignition chemistry, constraints exist with their utility; additional diagnostics, like heat release rates, measurements of chemical intermediates, etc., could improve development / validation efforts.

PROPOSED FUTURE WORK

FY 2019 and beyond

- Proposed future work is subject to change based on funding levels
- Physical testing of bi-, and multi-component surrogates, leveraging interactions with LLNL / others, to improve robustness of formulations
  - Quantify chemical kinetic interactions between components (like olefins-aromatics), surrogate blends, and with ethanol;
  - Utilize novel techniques / targets to select component molecules, blending ratios, including blends with ethanol.
- Conduct RCM tests to quantify effects of constituents within exhaust gas recirculation (EGR)
  - Coordinate with SNL to target LTGC engine conditions (T, P, ϕ, EGR) using a multi-component surrogate, and RD5-87 (E10 certification gasoline);
  - Coordinate with LLNL to formulate and test surrogate blends for RD5-87.
- Design/fabricate gas sampling apparatus; couple with newly acquired GCxGC.

PROPOSED FUTURE WORK

FY 2019 and beyond

- Conduct additional tests with CRC AVFL-20 fuels
  - Coordinate with MIT, AVFL-31 committee to better understand / interpret measurements across RCM and spark-ignition engine platforms considering knock-limited performance
- Continue coordination of International RCM Workshop
- Extend UQ/GSA to additional targets such as reaction intermediates, heat release rates

SUMMARY

- Objective
  - Acquire data, validate / improve models for transportation-relevant fuels
- Project Approach
  - Utilize ANL's RCM and novel analysis tools, leverage expertise of DOE-funded researchers to synergistically improve predictive models
- Technical Accomplishments / Progress
  - Quantified chemical exothermicity of PRF90 across wide ranges of Tc and Pc, identifying discrepancies with kinetic model predictions;
  - Acquired data for market-representative olefin, and olefin/aromatic blends;
  - Organized 4th RCM Workshop, coordinated working group activities.
- Collaborations
  - National labs, universities and industry; International RCM Workshop
- Future Work
  - Testing with gasoline surrogate components, blends and full boiling range gasolines across engine-relevant (T, P, ϕ, EGR) conditions;
  - Advances / improvement in UQ/GSA; integration of gas sampling system.

THANK YOU