Project 071 Predictive Simulation of nvPM Emissions in Aircraft Combustors

Georgia Institute of Technology

Project Lead Investigator

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University Participants

Georgia Institute of Technology (GT)

- P.I.: Prof. Suresh Menon
- FAA Award Number: 13-C-AJFE-GIT-067
- Period of Performance: October 1, 2021 to September 30, 2022
- Tasks:
    1. Kinetic modeling: improvements in the soot kinetic model to predict development of polycyclic aromatic hydrocarbon (PAH) pathways in representative sooting fuels
    2. Nucleation modeling: simulation of PAH rings at a range of relevant flame temperatures to identify key species contributing to the soot nucleation process.
    3. Surface growth and aggregation modeling: reaction-transport-limited growth of soot particle models for cluster–cluster aggregation
    4. (LES): coupling the multi-scale models developed in Tasks 1–3 in a method of moment within LES for soot–turbulence–chemistry interactions with application to canonical flames.

Project Funding Level

Current FAA funding is for a 3-year effort (July 2020 to September 2023), with a request of $500,000 per year from ASCENT (per year). Additional request for funding in year 3 have been submitted. Cost-sharing is provided as follows.

- GT provides cost-sharing for its share of $150,000 per year. The GT point of contact is Kevin Ellis (kevin.ellis@aerospace.gatech.edu).
- Raytheon Technologies Research Center (RTRC) provides cost-sharing of $250,000 per year. Dr. Colket is a consultant in this project with many years of experience in soot modeling. The RTRC contact is John LaSpada (LaSpadJW@RTRC.utc.com).
- The University of Michigan (UM) provides cost-sharing in the amount of $100,000 per year. The UM point of contact is Alexandra Thebaud (thealexi@umich.edu).

Investigation Team

- Prof. Suresh Menon, GT: P.I., Task 4
- Dr. Miad Yazdani, RTRC: co-P.I., Task 3
- Dr. Steve Zeppieri, RTRC: co-P.I., Task 1
- Prof. Angela Violi, UM: co-P.I., Task 2
- Dr. Meredith (Med) Colket, Consultant, RTRC: co-investigator, Task 1

Project Overview

This project is being used to establish a new multiscale approach to predict soot formation in aircraft combustors. A hierarchy of first-principles simulation methods is being used to account for the multiscale physics of the formation and transport of non-volatile particulate matter (nvPM, also called soot in the literature). The final objective is to use this multiscale approach to model the physics in LES of realistic gas turbine combustors. We target and isolate the layers of empiricisms that currently exist, for example, in particle inception models, the roles of precursor species in nucleation, the particle shape assumptions and their impact on surface growth, the sensitivity of predictions to particle size distribution, and the ad hoc coagulation/coalescence mechanisms. The team already has all modeling tools, but a systematic coupling of these tools in a multiscale, multi-physics strategy has yet to be accomplished by any research group. Hence, this study will establish new predictive ability by integrating these capabilities.

The multiscale and multi-physics layers of collaborations among the cost-sharing groups are summarized in Figures 1 and 2, and briefly described herein. The kinetics group at RTRC is conducting a study to understand the role of gas-phase kinetics in predicting important species potentially labeled as soot precursors. The information on reduced kinetics from RTRC is being used by GT and UM to evaluate LES performance and the process of nucleation. In the UM study, the propensity of gas-phase species to form dimers (considered the building blocks of soot inception) under flame conditions is being studied. Identification of soot precursors and the rates of formation of soot nuclei will be the output from these studies. This nucleation rate will be provided to GT to update the source terms associated with nucleation processes through a six-moment method of moment with interpolative coefficients (6-MOMIC) approach, and the information on the structures of these soot nuclei will be provided to RTRC for modeling of surface growth and aggregation processes. Outputs from the aggregation studies in RTRC in the form of global surface growth and aggregation models will then be fed back to GT to update the source term surface growth and aggregation models in the 6-MOMIC approach. Canonical studies are underway at GT to provide information regarding the variations in local conditions, such as pressure, temperature, and local equivalence ratios due to turbulence–chemistry interactions; this information should be useful in each stage of the abovementioned studies. LES studies at GT will also involve modeling the effects of chemistry–soot–turbulence interactions by using advanced subgrid models including the linear eddy mixing (LEM) model. As a project deliverable (at the end of this research effort), the final assessment of both the existing soot model and the improved soot model will be conducted in canonical flame configurations.

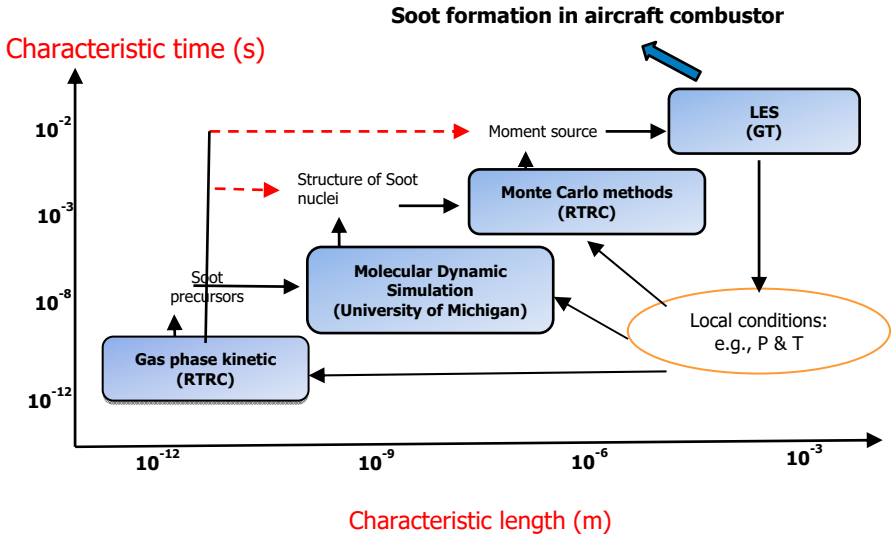


Figure 1. Multiscale collaborative efforts to improve nvPM (soot) predictions.

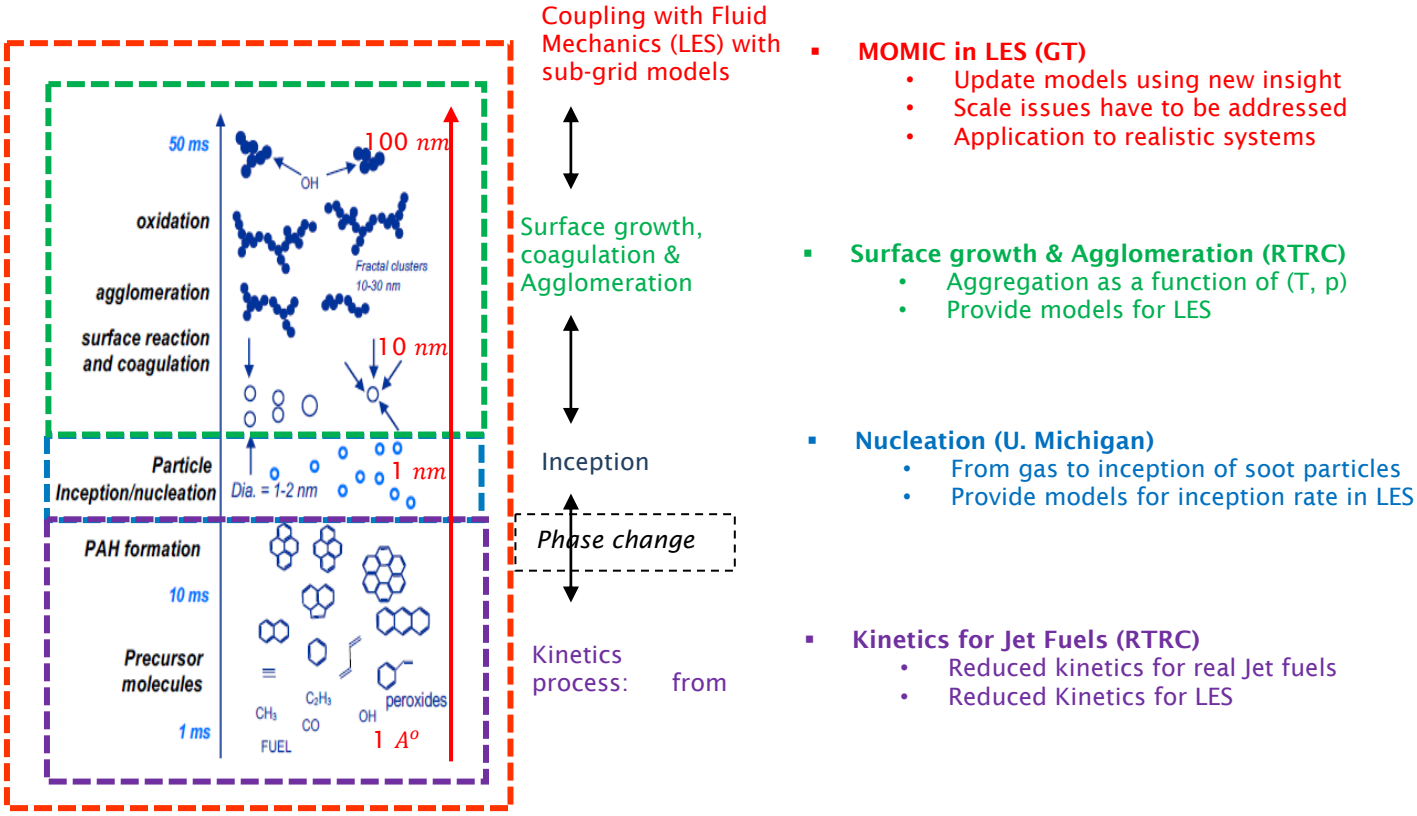


Figure 2. Multiscale collaborative efforts to improve nvPM (soot) predictions.

Task 1 - Kinetics Modeling

Raytheon Technologies Research Center

Objective

The objective of this task is to develop validated, detailed, and reduced chemical kinetic models of parent fuel decomposition and oxidation reactions, with a special focus on fuel rich chemistry, to enable the accurate evolution of PAH/soot precursor formation and incipient soot particle formation, and the evaluation and improvement of reduced-order soot formation models. Year 1 fuel activities focused on ethene, whereas the Year 2 efforts shifted the focus to Jet A fuels, which will be continued in the remainder of the project. The reduced-order kinetic model is also crucial for assessment in Rich-Burn–Quick-Quench–Lean-Burn (RQL) combustors.

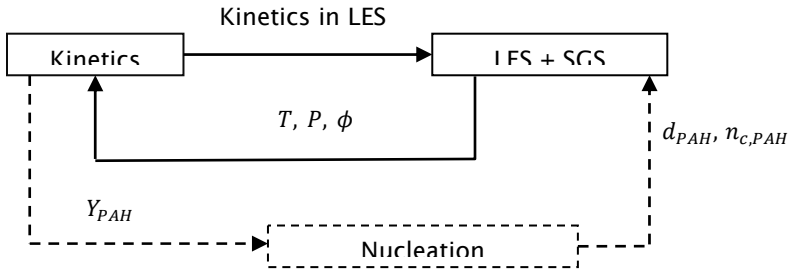


Figure 3. Coupling of LES–kinetics activities.

Research Approach

Reduced-order Model Development for use in LES

The schematic in Figure 3 shows the coupling between the LES, nucleation, and kinetic activities focused during the span of the current Year 2 of the project, where *T*, *P*, and ϕ represent the temperature, pressure, and equivalence ratio of the surrounding gas phase. These background conditions are obtained from the LES studies of canonical premixed and non-premixed configurations. Moreover, PAH represent the polycyclic aromatic hydrocarbon species considered as the soot precursors. The objective of the current kinetic efforts was focused on downselecting the detailed kinetic model of jet fuels and reducing it to a non-stiff version with finite rate expressions for precursor species production determining Y_{PAH}. This kinetic model is to be fed directly into LES studies at GT. The precursor species characteristics (n_{PAH}, d_{PAH}) concentrations will also be input to nucleation studies at UM to assess their ability to form dimers.

In Year 1, published ethene kinetics with varying details of PAH/particulate matter (PM)-related chemistry were assessed. In Year 2, the annual efforts focused on a similar approach for evaluating compact Jet A mechanisms including any PAH chemistry. Although no compact Jet A mechanism with PAH chemistry was identified, two compact mechanisms were selected for evaluation and further study: the Kollrack mechanism (Kollrack,1976) and the HyChem mechanism (Wang et. al., 2018). Essentially, both models implement semi-empirical (i.e., non-Arrhenius) reactions for Jet A. The Kollrack mechanism uses two oxidation/hydrogen abstraction reactions (O_2 and OH) as the initiation reactions. The HyChem model uses seven semi-empirical Jet A reactions: one pyrolysis and six abstraction reactions. The Kollrack mechanism uses product species yielding simple integer stoichiometric coefficients (e.g., C_2H_4 and C_2H_3); in contrast, the HyChem approach uses detailed non-integer stoichiometric coefficients for a wide product spectrum of compounds associated with the fuel decomposition (i.e., C_2H_4, C_3H_6, iC_4H_8, nC_4H_8, C_6H_6, CH_3, CH_4, H, etc.). These non-integer coefficients are derived from canonical (e.g., flow reactor, shock tube, or opposed jet) experiments involving the fuel. The subsequent chemistry involving these fuel intermediate species is the published "USC Mechanism" small-species kinetics model.

On the basis of the above methods, a third mechanism was developed under this program. Earlier analysis within this project indicated that the Wang-Frenklach mechanism (1997) showed good agreement with the results of PM-related experiments using an ethene fueled, well-stirred reactor. Accordingly, following the HyChem approach, semi-empirical Jet A reactions were developed and combined with the Wang-Frenklach ethene kinetics model. This approach provides a heuristic model that is relatively small, handles Jet A fueled systems, and affords PAH/PM-related kinetic information. In this third model, the products associated with the fuel-based reactions are those already in the Wang-Frenklach mechanism: C_2H_4, C_2H_3, C_2H_2, CH_4, CH_3, H_2, H, and C_6H_6. A summary of the above three mechanisms is provided below.

Table 1. Comparison of the number of species, reactions, and PAH chemistry in the above mechanisms.

Mechanism	Fuel Rep.	# Species	# Reactants	PAH Chemistry?
Kollrack	C12H23	21	30	No
HyChem	C11H22	44	202	No
Mod. W-F	C12H23	100	547	Yes

The figures of merit associated with these studies were ignition delay and perfectly stirred reactor (PSR) lean blow-out profiles developed within the National Jet Fuel Combustion Program for the HyChem and Kollrack models. Figure 4 shows the ignition delay times associated with stoichiometric blends of Jet A/air at 30 atm and initial temperatures of 1,000–1,600 K.

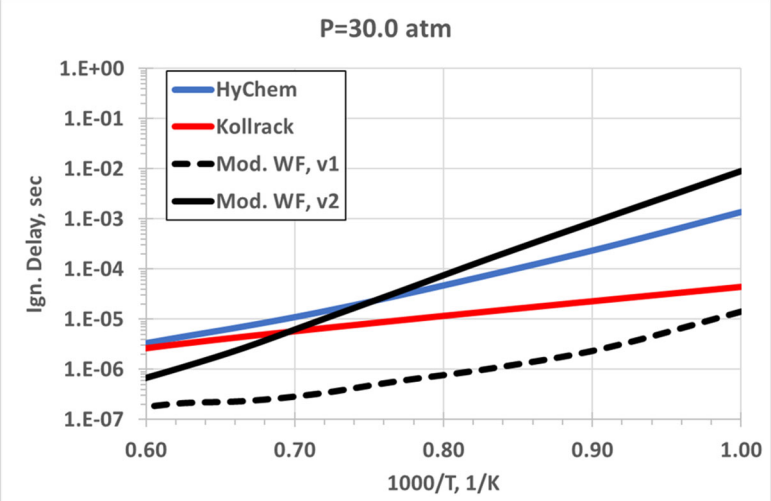


Figure 4. Comparison of ignition delay times associated with the above mechanisms at the indicated conditions.

On the basis of the assumption that the HyChem model is realistic (given that it was developed from exactly such experimental data), the version of the modified Wang-Frenklach model that included benzene as a fuel intermediate species compares well with the HyChem profiles, whereas both the Kollrack and non-benzene modified Wang-Frenklach mechanisms do not trend well with the other two mechanisms.

Similar observations are obtained from the comparisons of each mechanism's PSR lean blow-out profiles (Figure 5). The modified Wang-Frenklach mechanism that includes benzene yields a profile in excellent agreement with the HyChem model, in terms of both the reactor temperature profile and the residence time at blow-out. The modified Wang-Frenklach mechanism that does not include benzene yields a lower reactor blow-out temperature and residence time. The Kollrack mechanism temperature profile is unique and occurs at a considerably lower temperature than the other three mechanisms.

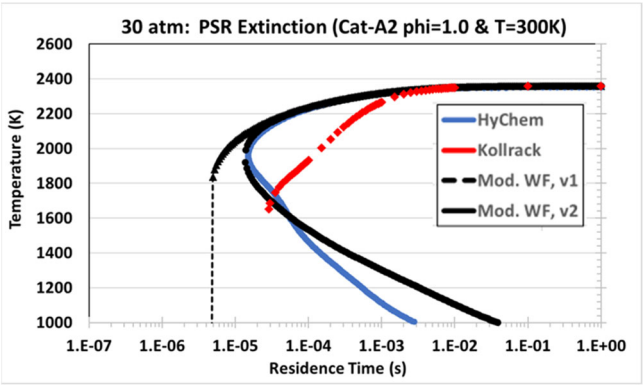


Figure 5. Comparison of PSR lean blow-out profiles associated with the above mechanisms at the indicated conditions.

The final check used the modified Wang-Frenklach mechanism in conjunction with previously developed RTRC reactor network models of RQL combustors and compared the results to experimental data (Figure 6). For a front-end equivalence ratio of approximately 2.25, the modified Wang-Frenklach mechanism predicts a combustor exit smoke number of 32.9. This value is in good agreement with the experimentally obtained smoke number values at similar operating conditions.

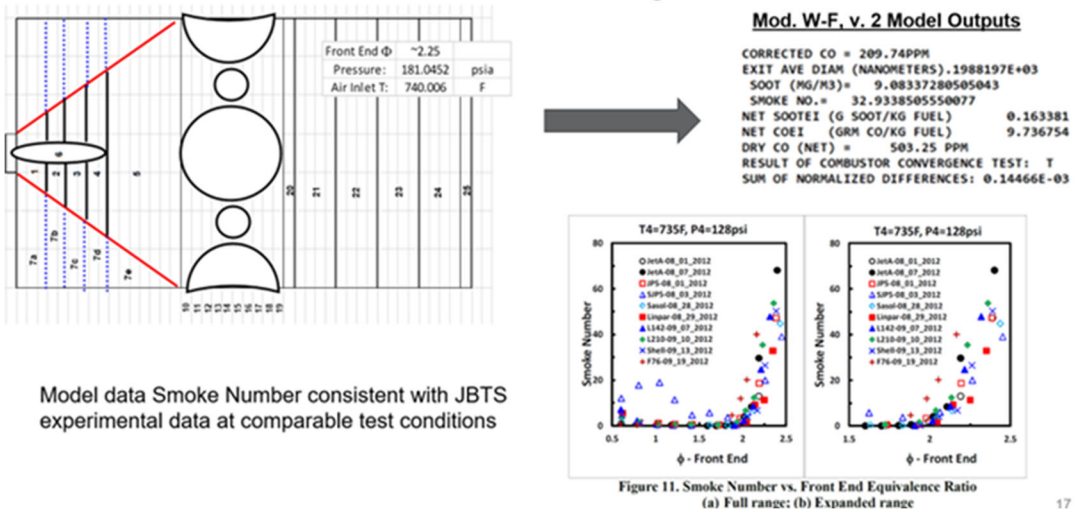


Figure 6. Comparison of combustor exit smoke number values for the indicated RQL combustor operating conditions.

The goal of this year's activities was to determine whether a suitably compact mechanism that incorporates both Jet A and PAH/PM kinetics information could be developed. On the basis of the analysis conducted to date, the construction of such a mechanism appears to be feasible, and efforts in developing such a model (e.g., extending the HyChem mechanism to include PAH chemistry, and adding additional larger intermediate chemistry reactions to the Wang–Frenklach mechanism) will be the focus of the remaining period of the project.

Milestone(s)

Milestone	Planned due date
Assessment of Jet A fuel chemistry	9/30/2022

Major Accomplishments

Framework for assessment of reduced kinetics including PAH pathways

Publications

None.

Outreach Efforts

None.

Awards

None.

Student Involvement

None.

Plans for Next Period

Future efforts will focus on developing a reduced-order Jet A fuel mechanism with validation over a wide range of conditions. The reduced kinetics will be provided to GT, and the information on the gas phase will be provided to UM for nucleation studies.

References

See Task 4 reference list.

Task 2 - Nucleation Modeling

University of Michigan

Objective

The objective of this task is to develop models for nanoparticle inception, a critical step in predicting emissions. This effort bridges the work on gas-phase chemistry (RTRC) with the model for particle growth (RTRC) and provides inputs for the MOMIC model (GT).

Research Approach

Current models for particle inception are unable to reproduce a variety of experimental data, including molecular structure. This work is aimed at developing a predictive model for particle inception that can provide accurate chemical and physical growth pathways for PAHs. Molecular dynamics (MD) simulations are used to study the collisions of PAHs and the formation of aromatic dimers leading to soot inception.

In the last annual efforts, methodology and the MD approach was established for the assessment of the dimer formation stability of different aromatic species. In the current annual effort, we continued the atomistic simulations of these species and higher PAH species, also including oxygen content. Figure 7 shows the structures of gas-phase compounds considered in this study and the results for their homo-dimerization propensity. The data points are broadly clustered in three groups. The first is composed of the compounds I.C (462 u), I.D (462 u), and I.E (460 u), which have the same mass and oxygen/carbon ratios (0.125) and are less stable than I.A (448 u) and I.B (472 u), which constitute the second group. This difference indicates that the presence of oxygen slightly destabilizes the dimers, an effect possibly caused by the increased repulsive electrostatic interactions of oxygenated molecules compared with pure aromatic hydrocarbons. The third group is formed by I.F (402 u), I.G (452 u), and I.H (502 u), which are less likely to dimerize than the other two groups. The low dimer stability of the third group may be due to the presence of a sigma bond, which introduces an internal rotatable bond that interferes with the formation of clusters.

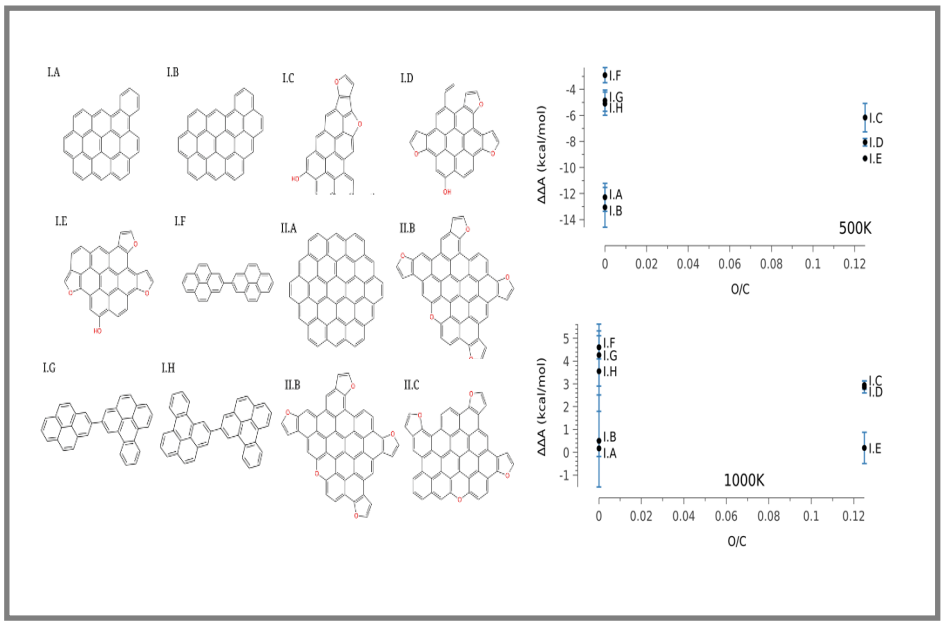


Figure 7. Structural formula of gas-phase compounds (left) and homo-dimerization propensity at 500 K and 1,000 K as a function of oxygen/carbon ratio.

Several conclusions can be drawn from this study. First, the details of the structures of the species that nucleate cannot be ignored. Mounting evidence indicates that the presence of five-membered rings, aliphatic side chains, and oxygenated

groups in soot precursors does not change the nucleation mechanism leading to soot formation. Our results pertaining to both the dimerization propensity and the change in free-energy barriers between monomers and dimers, both of which directly relates to the kinetic rates of dimerization, show otherwise. Second, the effects of shape, presence of oxygen, mass, and rotatable bonds are tightly intertwined, and have different importance as well as diverse temperature dependencies, although they are all dominated by entropic effects at high temperatures. The presence of oxygen affects the dimer propensity by reducing the molecular cohesion due to electrostatic repulsion, although, notably, the force field used in this study cannot capture the effect of molecular polarizability.

These results have been used as a first approach toward the development of predictive trends for quantifying dimer stability. Because of the presence of various intertwined dependencies, we will need a new approach to separate the contributions and their relative importance. Next, we will combine MD simulations with machine learning approaches to identify the main characteristics that drive nucleation and determine the corresponding rates.

Future work will be aimed at improving upon the relationships that we obtained by identifying additional factors influencing dimer stability. This process will involve close feedback with kinetics studies at RTRC to modify the kinetics mechanism in parallel. After the key precursors are identified, the collision rates of these dimers will be provided as inputs in the soot nucleation rates used in the LES-MOMIC code.

Milestone

Milestone	Planned due date
MD assessment of pool of PAH dimers	9/30/2022

Major Accomplishments

MD simulations for pools of PAH rings at a range of flame temperatures

Publications

None.

Outreach Efforts

None.

Awards

None.

Student Involvement

One student at UM is involved in this work.

Plans for Next Period

MD Simulations at UM

Future work will seek to improve upon the relationships that we obtained by identifying additional factors that influence dimer stability. This process will involve close feedback regarding kinetics studies at RTRC to enable modification of the kinetics mechanism in parallel. Once the key precursors are identified, the collision rates of these dimers will be used as inputs in the soot nucleation rates in the LES-MOMIC code.

References

See Task 4 reference list.

Task 3 - Surface Growth and Aggregation Modeling

Raytheon Technologies Research Center

Objective

The objective of this task is to develop a physics-based framework for the prediction of soot particle growth after the inception process. The growth consists of agglomeration due to collisions between the primary particles and surface growth as a result of direct deposition of the precursors on the aggregate. The final aggregate fractal structure and its temporal evolution as a function of local conditions are of interest. This model will provide the morphology characteristics and the growth rate of the particles, which will serve as inputs into the MOMIC formulation.

Research Approach

Soot particles from nucleation stages undergo various surface growth processes and form primary particles. These primary particles are spherical with typical diameters of 1–10 nm. The focus of this effort is to understand evolution of fractal dimension of aggregate since the formation of primary particles to final fractal aggregates through processes of surface growth and aggregation. Experimentally, the structures of these soot particles have been demonstrated to be dependent on the local conditions (e.g., local equivalence ratio). These surface growth process can occur because of heterogeneous reactions of gas-phase precursors on solid soot particle surfaces (reaction limited growth) or through the transport of soot precursors in high-speed flames (transport limited growth). Most of the current growth models account for only reaction limited growth and ignore transport limited growth as well as cluster–cluster interactions, which may be important in aggregate formation. In this work, information on background gas-phase species contributing to soot particles, the structure of the initial soot nuclei, and the local conditions is merged to understand the fundamental processes contributing to the formation of large soot aggregates.

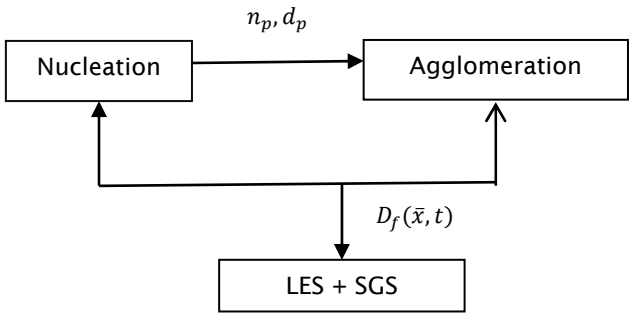


Figure 8. Post-inception growth of soot particles (LES-RTRC-UM coupling).

The coupling between the growth framework being developed at RTRC with MD simulations from UM and the LES study at GT is briefly highlighted in Figure 8. The growth module takes the number density and size distribution of incipient soot particles from nucleation as an input and tracks the growth of such particles along the statistically averaged path lines, which have varying background LES conditions in form of temperature (*T*), pressure (*P*) and local equivalence ratio (ϕ), as detailed below. The output from such studies in the form of the parametrized fractal dimension (D_f) will be fed back into the LES-MOMIC soot approach.

The developed post-inception growth model described in previous annual efforts has been shown to capture the effects of different operating conditions on the growth characteristics of the particles, including size and morphology. The parametric exercise of this model, however, may become computationally prohibitive if a range of operating parameters and combinations therein is swept over. A path line-sampling approach is proposed (schematic in Figure 10), in which the full flow-field (obtained by the LES) is represented through statically sampled path lines over which the operating conditions would be time variant. For example, the temperature and fuel/air ratio would evolve over the course of time as a particle moves from the injector toward the exit of the combustor. This time history is then provided to the growth model, wherein the variation in particle characteristics will be solved over the course of this timeframe for each individual path-line histogram (example output in Figure 9). This information is then fed back into the LES with time–space mapping through the path line coordinates and nearest-point interpolation to provide full spatial representation of growth

information, which is then used in the MOMIC approach. This approach effectively reduces the computational overhead of the growth model to a dozen simulations (assuming that this number of path lines is sufficient to properly sample the combustor flow field), as compared with potentially hundreds in the conventional parametric approach. Of note, this process is considered a one-way coupling approach, in which the macroscopic changes in the flow-field due to the interaction with the soot-particle transport are assumed to have negligible impact on the growth characteristics of the particles.

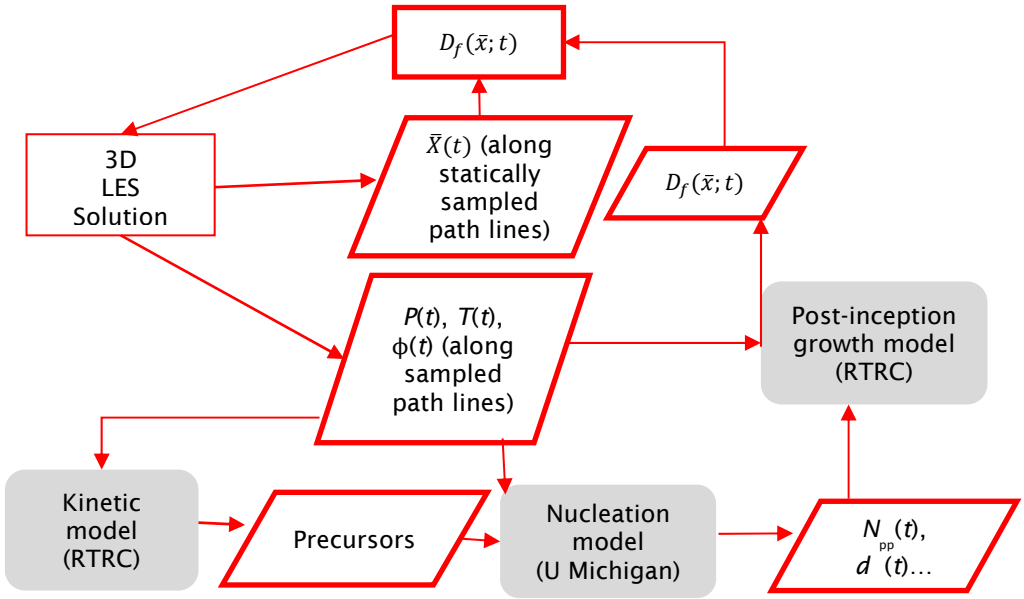


Figure 9. One-way coupled strategy for growth of soot particles.

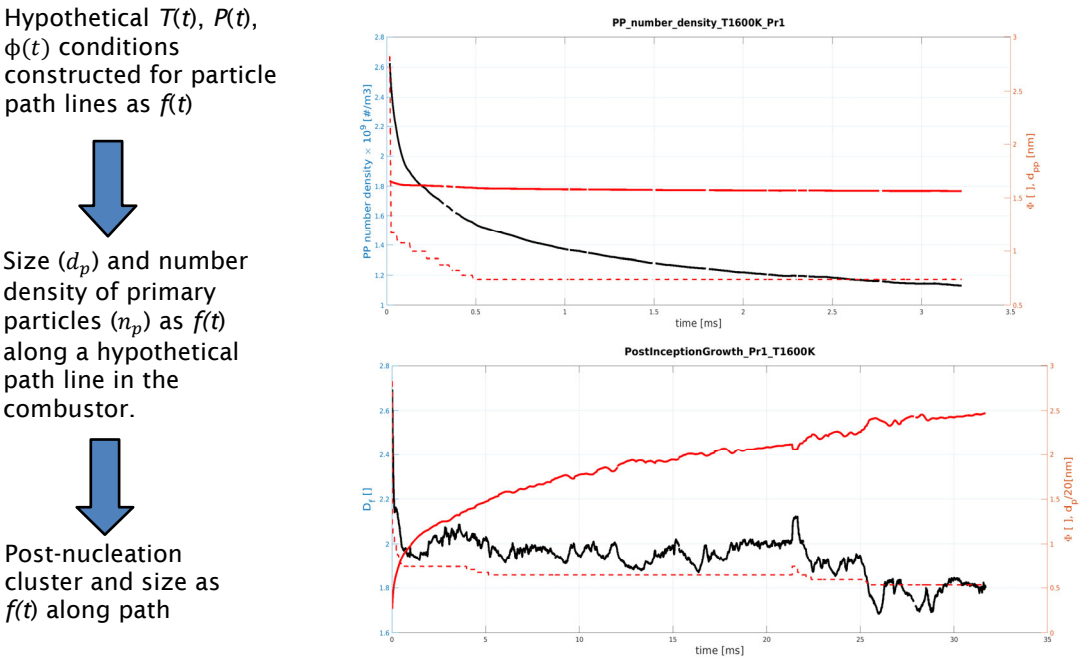


Figure 10. Demonstration of a one-way coupled strategy along the hypothetical LES path-line.

The future work for the remainder of the project uses actual LES simulated data for the premixed and non-premixed flames instead of hypothetical path lines, as demonstrated above.

Milestone(s)

Milestone	Planned due date
One way coupled MC studies of surface growth	9/30/2022

Major Accomplishments

One-way coupled demonstration of using hypothetical LES-like data for soot fractal growth.

Publications

None.

Outreach Efforts

None.

Awards

None.

Student Involvement

None.

Plans for Next Period

The future work for the remainder of the project will use actual LES simulated data for the premixed and non-premixed flames instead of hypothetical path lines, as demonstrated above.

References

See Task 4 reference list.

Task 4 - Large Eddy Simulations

Georgia Institute of Technology

Objective

The objective of this task is to develop a physics-informed LES framework to model soot formation in turbulent reacting configurations of canonical and combustors with practical relevance. The current report discusses the LES-MOMIC and LEMLES-MOMIC frameworks to address large-scale simulations of soot modeling in turbulent reacting flows. The current steps also included a collaboration with the kinetics group to include the effects of various mechanisms within the LES framework, updating the MOMIC framework to incorporate models for key PAH pathways and comparing it with the old MOMIC framework, which does not account for PAH pathways.

Research Approach

LES studies of turbulent sooting problems are very difficult because of the multiscale nature of soot inception, coagulation, and surface growth that must be modeled in a highly turbulent and reactive environment, typically in a complex combustor configuration. Most prior studies have focused on global models that approximate the small-scale physics. Consequently, many available models account for the underlying physics. In contrast, simulations require some approximations, because the computational resources will never meet the simulation requirements. In the current effort, we balance contributing to the prediction of soot formation physics in a realistic gas turbine combustor with the need to obtain high-fidelity, reliable predictions by using advanced models. To achieve this goal, we leverage our past LES capability and upgrade the models by using the results from MD and Monte Carlo (MC) studies. Soot evolution is tracked with MOMIC, wherein the first six moments of the particle size distribution function are used.

The full set of compressible reacting multispecies Navier–Stokes equations cannot be solved directly, because a direct numerical simulation is not feasible for practical applications. For LES, the large-scale flow features are resolved, and subgrid modeling is used for the smaller scales.

The LES governing equations can be written as follows:

$$\frac{\partial \bar{\rho}}{\partial t} + \frac{\partial}{\partial x_i}(\bar{\rho}\,\widetilde{u_\iota}) = 0$$

$$\frac{\partial}{\partial t}(\bar{\rho}\,\widetilde{u_\iota}) + \frac{\partial}{\partial x_j}\left(\bar{\rho}\,\widetilde{u_\iota}\,\widetilde{u_J} + \bar{P}\,\delta_{ij} - \overline{\tau_{\iota J}} + \tau_{ij}^{sgs}\right) = 0$$

$$\frac{\partial}{\partial t}\left(\bar{\rho}\,\tilde{E}\right) + \frac{\partial}{\partial x_j}\left[\left(\bar{\rho}\,\tilde{E} + \bar{P}\right)\widetilde{u_\iota} + \overline{q_\iota} - \widetilde{u_J}\,\overline{\tau_{\iota J}} + H_i^{sgs} + \sigma_i^{sgs}\right] = 0$$

$$\frac{\partial}{\partial t}\left(\bar{\rho}\,\widetilde{Y_k}\right) + \frac{\partial}{\partial x_j}\left[\bar{\rho}\,\widetilde{Y_k}\,\widetilde{u_J} - \bar{\rho}\,\overline{D_k}\,\frac{\partial \widetilde{Y_k}}{\partial x_j} + \Phi_{j,k}^{sgs} + \Theta_{jk}^{sgs}\right] = \bar{\rho}\,\widetilde{\dot{w}_k}$$

$$\frac{\partial}{\partial t}\left(\bar{\rho}\,\tilde{Y}_{soot}\right) + \frac{\partial}{\partial x_j}\left[\bar{\rho}\,\tilde{Y}_{soot}\,\widetilde{u_J} - \bar{\rho}\,\bar{D}_{soot}\,\frac{\partial\,\tilde{Y}_{soot}}{\partial x_j} + \widetilde{V_T}_{soot}\,\tilde{Y}_{soot} + \Phi_{j,k,soot}^{sgs} + \Theta_{jk,soot}^{sgs}\right] = \bar{\rho}\,\tilde{\dot{w}}_{soot}$$

$$\frac{\partial}{\partial t}\left(\bar{\rho}\,\widetilde{M_k}\right) + \frac{\partial}{\partial x_j}\left[\bar{\rho}\,\widetilde{M_k}\,\widetilde{u_J} - \bar{\rho}\,\bar{D}_{soot}\,\frac{\partial\,\widetilde{M_k}}{\partial x_j} + \widetilde{V_T}\,\widetilde{M_k} + \Psi_{j,k}^{sgs} + \Omega_{j,k}^{sgs}\right] = \bar{\rho}\,\tilde{M}_k$$

Here, $\widetilde{u_\iota}$ is the *i*-th filtered velocity, $\bar{\rho}$ is the filtered density, and $\bar{P}$ is the filtered pressure, which is computed from the filtered equation of state. $\tilde{T}$ is the filtered temperature, $\tilde{E}$ is the filtered energy, $\widetilde{Y_k}$ and $\widetilde{Y}_{soot}$ represent the filtered *k*-th gas-phase species and soot mass fraction, respectively, and $\widetilde{M_k}$ represents the *k*-th moments of the particle size distribution function. The details regarding the computations of all these quantities have been described elsewhere (El-Asrag & Menon, 2009) and hence are not discussed herein in specific detail. The filtered heat flux $\bar{q}$ can be supplied to an optically thin radiation model to include effects of radiation. The terms $\tau_{ij}^{sgs}, H_i^{sgs}, \sigma_i^{sgs}, \Phi_{j,k}^{sgs}, \Theta_{j,k}^{sgs}, \Psi_{j,k}^{sgs}$, and $\Omega_{j,k}^{sgs}$ contain the effects of the

subgrid scale on the filtered quantities. Modeling of these terms remains challenging; in addition, the closed system of equations must be solved together in three-dimensional space with temporal accuracy.

In this work, an eddy viscosity type subgrid model with constant coefficients is used to obtain the closure of subgrid momentum stresses and subgrid enthalpy flux. In the LEM formulation, the gas-phase species conservation equations are not spatially filtered as in other LES equations. Instead, the exact unfiltered equations are solved by using a two-scale, two-step Eulerian–Lagrangian approach. First, within each LES cell, the one-dimensional LEM model is used to solve for the scalar fields (species mass fraction, soot mass fraction, LEM temperature, and soot integer moments) along a notional line oriented along the maximum scalar gradient. Second, the subgrid scale fields are convected across the LES cell faces by using a Lagrangian transport approach through the splicing algorithm, which reproduces the effect of large-scale advection of the scalars by the flow field. The resulting scalar fields are then filtered in each LES cell to recover LES-resolved species mass fractions to be used in LES-resolved energy and state equations. The application of LEM for non-sooting ethylene flames was established in the Year 1 annual efforts. Its extension to a MOMIC model with PAH pathways is yet to be demonstrated.

In the current annual period, efforts were focused on incorporating the effects of aromatic precursors on various soot formation processes. The main areas of improvement involved soot nucleation physics based on PAH species, additional surface growth effects due to PAH condensations, and improvements in the structure of aggregates formed. Because Since coagulation of soot particles is already handled in detail in the old-MOMIC approach, no additional improvements are needed or currently planned.

Three variations in the MOMIC model were used:

1) In the last annual effort, the old-GT-MOMIC approach (El-Asrag & Menon, 2009) with moment source terms based on simplified soot kinetics (Leung & Lindstedt, 1991) was used to study soot formation in ethylene-air laminar as well as turbulent flames.
2) In the current annual efforts, the RTRC optimized Lindstedt soot model is implemented within the MOMIC code and is termed the RTRC-MOMIC model hereafter.
   Both MOMIC models 1 and 2 use C_2H_2-based nucleation and surface growth processes for soot particles. The coefficients are fitted to improve soot volume fraction predictions.
3) We also focused on incorporating the effects of aromatic precursors on various soot formation processes and identifying how these inputs can be obtained. This MOMIC model is noted as a new-GT-MOMIC model and is based on the Frenklach MOMIC approach (Brown et al., 1998). This model uses nucleation based on pyrene dimerization as well as a detailed mechanism of hydrogen abstraction–carbon addition for soot growth. It also uses condensation of pyrene dimers on the surfaces of soot particles.

The improvements of the new-GT-MOMIC over the old-GT-MOMIC were assessed by conducting numerical simulations at conditions prescribed in 0D PSR reactor simulations by Vaughn et. al. (1998). As shown in Figure 11, the improved MOMIC approach with additional relevant detailed of PAH species predicts a soot volume fraction closer to the experimental observations.

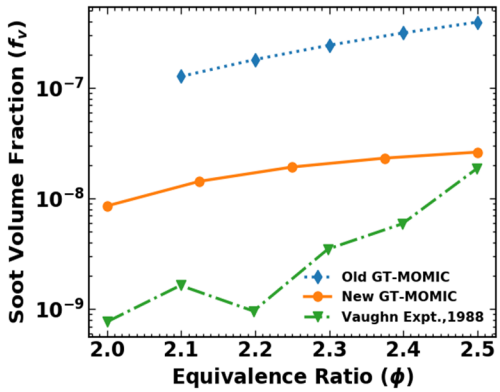


Figure 11. Comparison of old and new GT-MOMIC approaches for rich ethylene flames.

Similarly, all three models are assessed in a 0D PSR of a rich premixed ethylene mixture ($\phi = 2.5$) with P_{ref} = 1 atm, residence time of 0.005 s, and varying reactor temperature from 1,500 K to 2,000 K. The results comparing RTRC-MOMIC and old GT-MOMIC are shown in Figure 12.

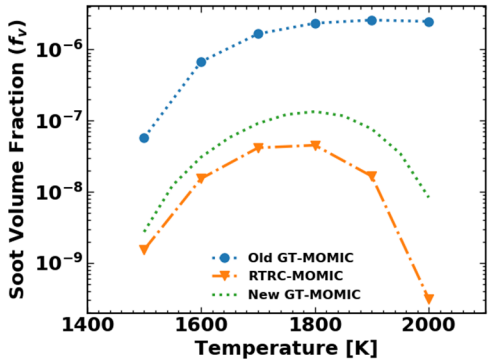


Figure 12. Soot volume fraction dependency on the temperature of the 0D PSR reactor.

As seen in Figure 12, the old GT-MOMIC model predicts higher levels of the soot volume fraction and fails to capture the bell-shaped dependency of the soot volume fraction on the reactor temperature. The RTRC-MOMIC model overcomes these shortcomings, and trends of variation of soot volume fraction with temperature match well with the predictions of the new GT-MOMIC model.

Verification of the New-GT-MOMIC Framework

The new-GT-MOMIC framework (including nucleation, surface growth, and coagulation) is now verified against the Frenklach-MOMIC approach for a range of conditions within the 0D PSR configurations. A detailed 99-species Wang-Frenklach ethylene-air mechanism (1997) is used to account for PAH species formation up to pyrene (and is assumed as the heaviest PAH species undergoing soot inception). The new-GT-MOMIC is verified with respect to the published data (Figure 13).

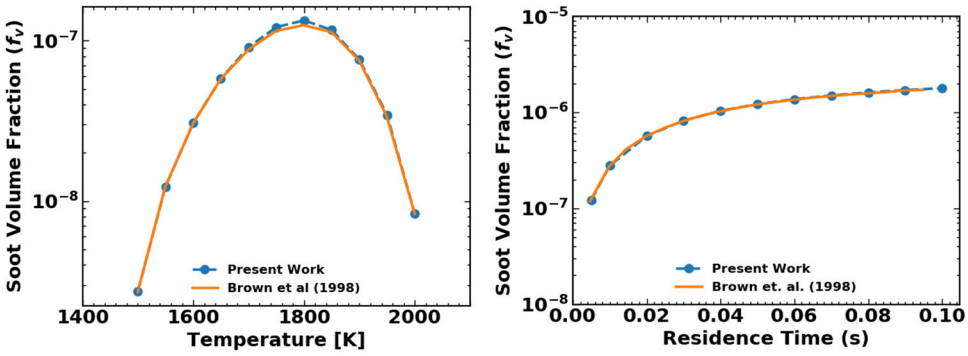
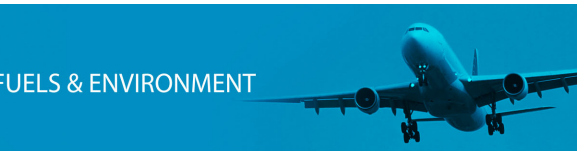


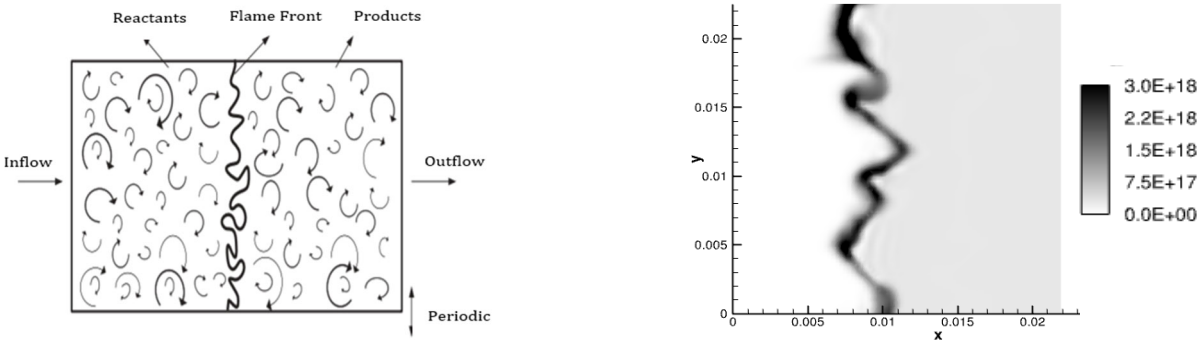
Figure 13. Comparison of the present work with 0D PSR simulations of Brown et al. (1998).

Numerical Simulations of a Premixed Flame of Jet fuels in a Turbulent Environment

The new GT-MOMIC code is coupled with the in-house compressible flow solver LESLIE to study soot formation in reacting simulations: 3D calculations of premixed flames of jet fuels with gasified Jet A fuel are performed. The fuel kinetics is based on the 62-species reduced mechanism developed by Lu and coworkers at the University of Connecticut. The proposed test intends to use the LES-MOMIC model with finite rate chemistry for a premixed flame turbulence interaction problem (El-Asrag et. al., 2007). The schematic for the configuration is shown in Figure 14a. The turbulence level is varied, so that the flame is in the distributed/broken reaction zone regime. The initial flame front is obtained from the laminar premixed flame solution and is specified at the center of the domain, with the left side denoting the reactants and the

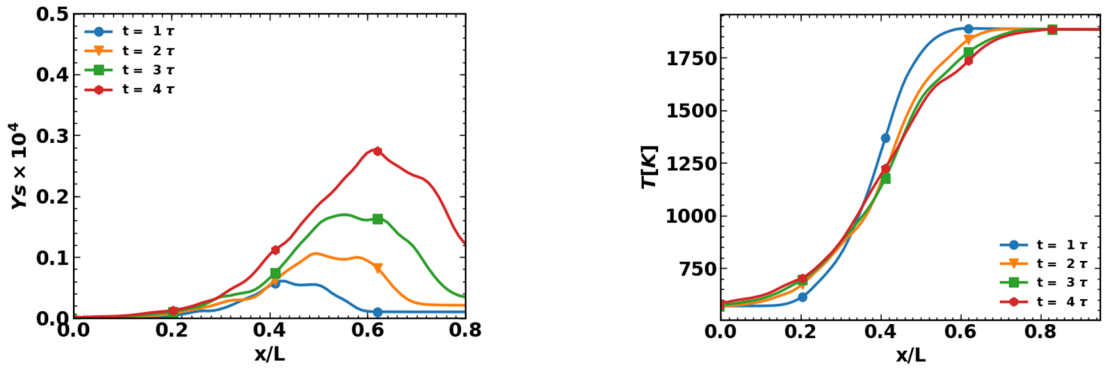


right side denoting the products. The extent of the computational domain is 15 mm × 15 mm × 15 mm in the streamwise *x*, transverse *y*, and spanwise *z* directions. The flow field is initialized by using the von Karman–Pao energy spectrum. Characteristic inflow–outflow conditions are specified in the streamwise direction and are periodic in the other two directions. The LES grid resolution chosen for the simulations is 96 × 96 × 96 LES cells, with the assumption of quasi-laminar chemistry, i.e., no closure for sub-grid turbulence–chemistry interactions is considered. Figure 14b shows the evolving color contour of soot number density in the post flame region. The initial planar flame structure appears severely wrinkled because of the presence of the background turbulence. Figure 15 shows the spatially averaged profiles of the soot mass fraction and temperature along the axial direction. These preliminary results establish the functionality of the first canonical reacting setup of jet fuels that we will be investigating in the current project. Further analysis is underway to understand how the various intermediate processes are interrelated in formation of the final soot particles.



(14a) (14b)

Figure 14. Soot evolution from a rich Jet A/air flame Interacting with background turbulence. (a) Schematic of the problem statement. (b) Contours of soot number density ($1/m^3$) evolution from jet fuel flames.



(15a) (15b)

Figure 15. Spatially averaged contour lines of soot mass fraction (a) and temperature (b) from the jet fuel flame

Numerical Simulations of Temporal Mixing Layers from Jet A Fuel

In this section, 2D calculations of a temporal mixing layer with gasified Jet A fuel are performed, thus providing an ideal non-premixed configuration for the assessment of soot models in which strong coupling between fluid dynamics and chemistry is present. The setup described herein is based on the 3D DNS calculations of an *n*-heptane–air mixture by Atilli et al. (2014). A representative two-dimensional strip (width H = 15 mm) of Jet A fuel (diluted with 85% N_2 by volume) is surrounded by air (21% O_2 and 79% N_2 in volume), as shown in Figure 16. The rectangular computational domain is approximately 94 mm long in a direction parallel (horizontal) to the fuel strip and 105 mm wide in a direction

perpendicular to the fuel strip. Both the oxidizer and the fuel strip are at atmospheric pressure. The air stream is preheated to 800 K to increase the resistance of the flame to the extinction induced by the strain. The computational domain is discretized in a uniform mesh with a total of 1,048,576 cells (1,024 × 1,024), which corresponds to the minimum grid spacing of $\Delta x = \Delta y = 91.5$ micrometers. This resolution is equivalent to the resolutions of DNS studies. Characteristic non-reflecting outflow boundary conditions are imposed in a direction perpendicular to the fuel strip (vertical), and periodic boundary conditions are applied in the other direction. The fuel strip moves to the right at a velocity of 8.74 m/s while the air co-flow moves in the opposite direction at the same speed. For initialization, the temperature and species mass fractions are taken from a flamelet and mapped from mixture fraction space onto the crosswise direction by using a mixture fraction profile:

$$z(y) = \frac{1}{2}\left[\mathrm{erf}\left(\frac{\frac{1}{2}\delta + y}{\sqrt{2\sigma_1^2}}\right) + \mathrm{erf}\left(\frac{\frac{1}{2}\delta - y}{\sqrt{2\sigma_1^2}}\right)\right]$$

Here, y represents the spatial location in a perpendicular direction, the parameter δ controls the width of the fuel strip, and the slope parameter σ_1 controls the slope of the profile. The values of these parameters were taken as 7.5 mm and 1.8. The simulations are conducted by using the 62-species reduced chemical kinetics model for Jet A fuel developed under the National Jet Fuel Combustion Program.

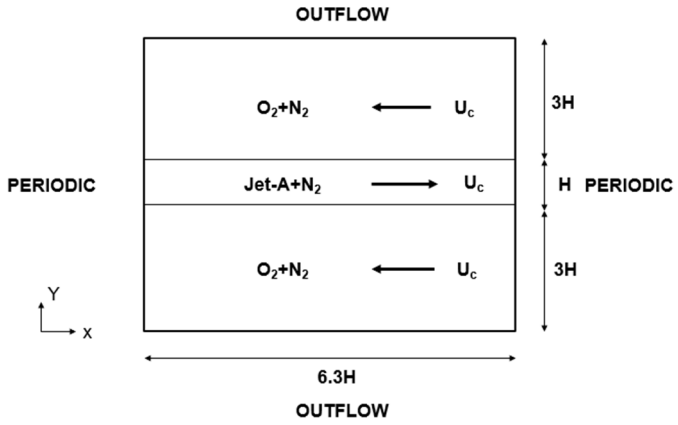


Figure 16. Schematic of the temporal mixing layer of Jet A fuel.

Figure 17 shows the time evolution contours for the Jet A fuel mass fraction at four different times (t = 5 ms, 10 ms, 15 ms, and 20 ms). The mixing layer grows with time, and the simulations are continued until the time reaches 20 ms, when large structures form. As can be seen, the growth of shear layers causes the wrinkling of the flame at the fuel/air interface. Figure 18 shows the contours of evolution of the soot precursor (pyrene), and similar evolution is plotted for the soot number density overlapped with isolines of temperature in Figure 19. The soot number density closely follows the pyrene formation zones in the non-premixed flames, as the nucleation process is assumed to occur through pyrene dimerization. Finally, Figure 20 shows the average profiles of velocity u normalized by characteristic flow velocity U_o, number density of the soot particles formed and the soot mass fraction profiles. The quantitative magnitudes of the soot mass fraction (~10^{-4}) as well as the number density (~10^{12} 1/cm^3) match well with the values obtained by Atilli et. al. (2014).

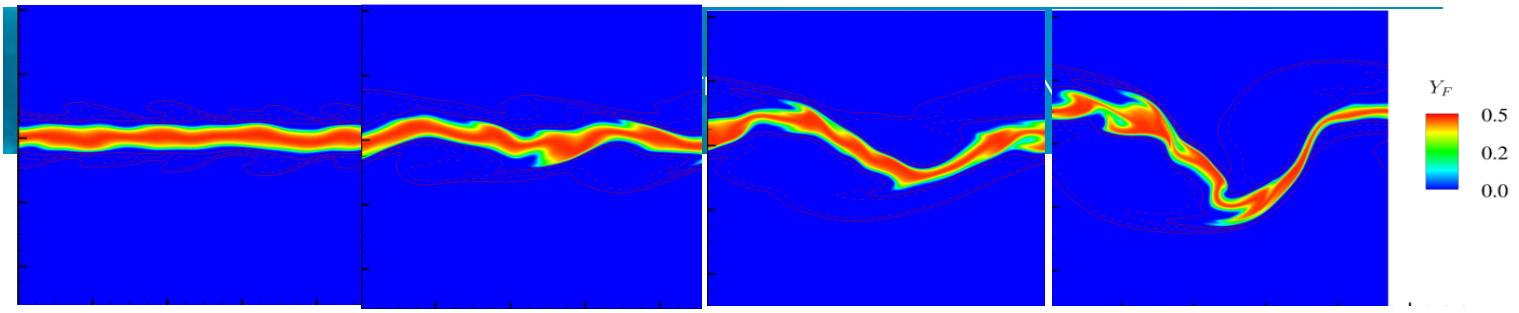


Figure 17. Contours of the Jet A fuel mass fraction overlaid with temperature isolines.

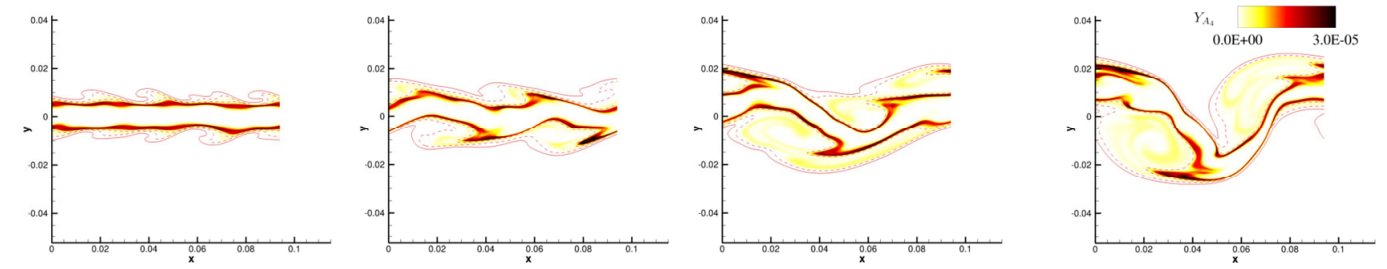


Figure 18. Contours of the pyrene mass fraction overlaid with temperature isolines.

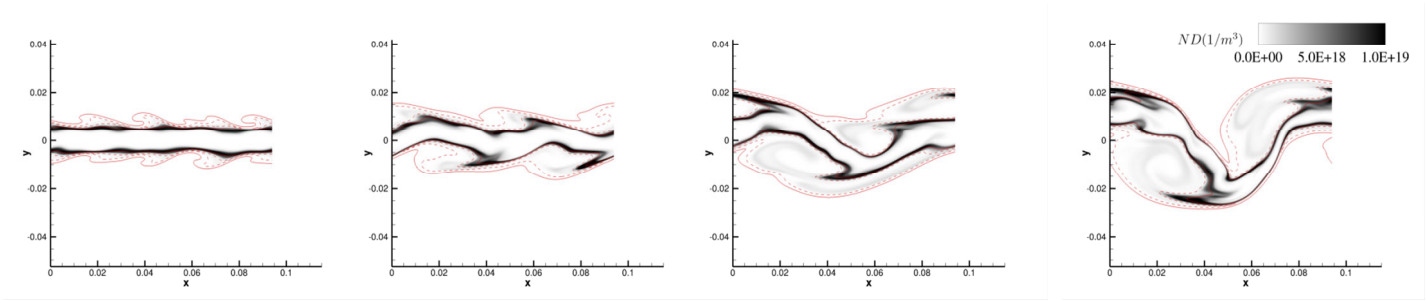


Figure 19. Contours of soot evolution overlaid with temperature isolines; T = 1,600 K (----) and T = 2,000 K (——).

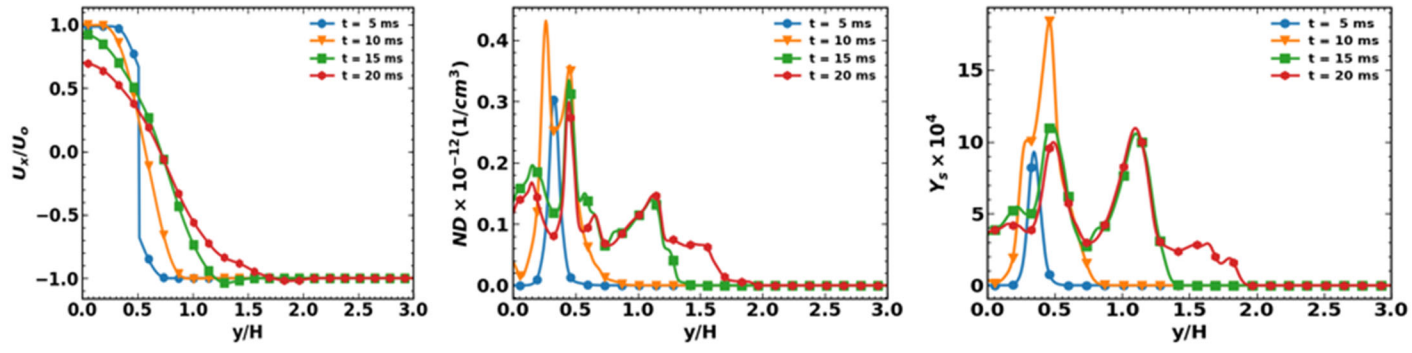


Figure 20. Spatially averaged profiles of velocity, soot number density, and soot mass fractions.

Milestones

Milestone	Planned due date
Improvement of the MOMIC model, identification of inputs and a multiscale strategy for model assessment	9/30/2022

Major Accomplishments

Improvement of the MOMIC framework with PAH pathways, verification against established data, inputs for MOMIC modeling, and simulations of canonical premixed and non-premixed configurations of Jet A fuels

Publications

None.

Outreach Efforts

None.

Awards

None.

Student Involvement

PhD student Shubham Karpe has been assisting in the development of the MOMIC framework within the LES code at GT.

Plans for Next Period

The future work at GT involves incorporating inputs from the abovementioned studies in the existing verified MOMIC framework and assessing any improvements by using the canonical configurations discussed above. The Year 3 efforts will also focus on simulating the RQL combustor rig currently being experimentally investigated under ASCENT FAA Project 70.

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