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
- PI: Suresh Menon
- FAA Award Number: 13-C-AJFE-GIT-067
- Period of Performance: Oct 1, 2019 – Sep 30, 2020
- Task(s):
    1. Implement high-fidelity method of moments with interpolative closure (MOMIC) model within large-eddy simulation (LES) to account for all physical processes such as nucleation, surface growth & oxidation, coagulation, and aggregation in soot formation.
    2. Evaluate soot/PAH (polycyclic aromatic hydrocarbon) chemistry model developed by Raytheon Technologies Research Center (RTRC) and soot model updates from University of Michigan (UM) and RTRC.

University of Michigan
- P. I.: Angela Violi
- FAA Award Number: 13-C-AJFE-GIT-067
- Period of Performance: One Year
- Task(s)
    1. Use Molecular Dynamics to assess parameters for nucleation process using RTRC's kinetics
    2. Collaborate with RTRC and GT to update MOMIC

Project Funding Level

FAA funding is currently for one-year effort of $500,000/year (Georgia Tech (GT): $150,000, Raytheon Technology Research Center (RTRC): $250,000 and University of Michigan (UM): $100,000). Cost share of $500,000 is also committed by participants.

The project started in late July 20 in GT and early September 2020 in RTRC and UM.

Investigation Team

- Dr. Suresh Menon, Professor, Georgia Institute of Technology, Principal Investigator
- Dr. Miad Yazdani, United Technologies Research Center, Co-Principal Investigator
- Dr. Steve Zeppieri, United Technologies Research Center, Co-Principal Investigator
- Prof. Angela Violi, Professor, University of Michigan, Co-Principal Investigator
- Dr. Meredith (Med) Colket, Consultant, United Technologies Research Center, Co-Investigator

Project Overview

This project will establish a new multi-scale approach to predict soot formation in aircraft combustors. A hierarchy of first principle simulation methods will be used to account for the multi-scale physics of the formation and transport of non-volatile particulate matter (nvPM, also called "soot" in the literature), and then use these methods to model the physics in large-eddy simulations (LES). This project will target and isolate the layers of empiricisms that currently exist, for example in particle inception models, in the role of precursors species in nucleation, particle shape assumptions and their impact on surface growth, the sensitivity of predictions to particle size distribution, and the ad hoc coagulation/coalescence mechanisms. All the relevant modeling tools already exist, but a systematic coupling of these tools in multi-scale, multi-physics strategy has yet to be accomplished by anyone. Hence, this study will establish a new predictive capability by integrating these capabilities.

We report here on the progress over a portion of this year because the project started officially in late July 2020 at GT and in September 2020 at the other team members' institutions. At GT, we are extending the Method of Moments with Interpolative Closure (MOMIC) approach based on first six moments of soot particle size distribution function and have completed some canonical 0D, 1D and 3D tests. The six moment method is considered more accurate compared to the three moment method used earlier and is the most optimal method given the current state of knowledge. The MOMIC model accounts for all soot formation processes such as nucleation, coagulation, surface growth, and aggregation and is coupled with an in-house compressible reacting flow solver, which uses a computational code called LESLIE. The details about the LES-MOMIC approach and corresponding validation cases, along with results, are discussed in this report.

The present MOMIC method uses a simplified four-step Lindstedt soot model (Leung and Lindstedt, 1991) as well as 19-step ethylene-air reduced chemistry (Lu and Law, 2005) which does not account for details about the established aromatic soot precursors (Blanquart et. al., 2009). Therefore, RTRC and University of Michigan (UM) are working on developing improved soot kinetic models as well as a skeletal mechanism that accounts for details about the breakdown of fuel species to soot precursors such as polycyclic aromatic hydrocabons (PAHs). The identification of this detailed chemistry and the reduction to key species are briefly touched upon at the end of this report.

Task 1 – LES-MOMIC of Sooting Flames
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Objective
The objective of this task is to establish the predictive capability of MOMIC within the LES code by simulating canonical sooting test cases.

Research Approach
LES studies of turbulent sooting problems are very difficult due to the multi-scale nature of soot inception, coagulation, and surface growth that have to be modeled in a highly turbulent and reactive environment, typically in a complex combustor configuration. Most past studies have focused on global models that approximate the small-scale physics; as such, there are many models to account for the underlying physics. On the other hand, simulations will require some approximations because the computational resources will never meet the simulation requirements. In the current effort, we balance contributing to the prediction on soot formation physics with a need to obtain high-fidelity and reliable predictions using advanced models. To achieve this goal, we leverage our past LES capability and upgrade the models using the results from molecular dynamics (MD) and kinetic Monte Carlo (KMC) studies. MD studies will be conducted by the UM team in collaboration with the RTRC group that will work on KMC and PAH kinetics tools. These groups outputs will be used to adjust MOMIC at GT.

The current task involves key subtasks that were accomplished in the most recent quarter:
1) Assessment and improvement of the MOMIC method for first six moments of soot particle size distribution function (PSDF). Some optimizations are being performed to improve the performance and accuracy of the code. One-dimensional testcases were performed to verify predictions of the present solver with established results in the literature.
2) Augmentation of the MOMIC library with source terms accounting for agglomeration process.

3) Integration of simplified MOMIC approach within LES framework. Specifically, the objective is to carry the first six moments of the PSDF in the solver and establish a functional architecture for integration of the full-fledged MOMIC model within LESLIE.

4) Testing is performed to assess implementation of the LES-MOMIC model using a premixed flame-turbulence interaction study. The simulations are performed with a C_2H_4/air setup and reduced chemistry that includes the Leung and Lindstedt mechanism for soot formation and growth.

The MOMIC approach involves solving the moment equations in two limiting conditions. In the coalescent limit where the soot particles are assumed to conserve the spherical shape after collision, the equation for the r-th moment M_r is solved either in the subgrid or in the resolved space along with the LES equations. A general form is $dM_r/dt = R_r + C_r + S_r + F_{M_r}$, r = 0,1,2,3...., where M is the moment of the PSDF, and R, C, S, and F are respectively nucleation, coagulation (in the coalescent limit), surface growth, and subgrid turbulent mixing contributions to the r-th moment equation. When soot particles exceed a certain size limit (~27.5 nm), the particles start to aggregate into chain-like structures of fractal dimension D (~1.8). Then the above equations are replaced by another set of moment equations of similar form but with different source terms representing the aggregation rate, the coagulation rate, and the surface growth rate in the non-coalescent limit. The zeroth moment M_0 represents the soot number density N_S defined by the number of soot particles per unit volume of the mixture. The first moment M_1 represents the average total mass of soot particles m_S per unit volume. Thus, the soot mass fraction is $Y_S = M_1/\rho$, where ρ is the mixture density. The average particle diameter is $d_p = (6.0 M_2 / \pi \rho_{soot} M_1)^{1/3}$ (El-Asrag and Menon, 2009, Srinivasan and Menon, 2015).

The model accounts for the different soot formation processes such as nucleation, oxidation, surface growth, coagulation, and aggregation. Soot or nvPM transport by diffusion and thermophoretic forces are included to allow for complete description of the soot physical behavior in a turbulent reactive environment. The energy equation is supplied with an optically thin radiation model to account for the radiation effect by the soot particles. The choice of using the first six moments within the MOMIC approach implies that we solve for the whole moments and interpolate the fractional moments (Frenklach, 2002). Thus, this model is computationally efficient because only six additional equations are solved in the coalescent and nine equations in the non-coalescent limits, and the source terms can be computed using efficient parallel processing approach.

The following 0D,1D, and 3D tests are performed to assess the implementation of the MOMIC model within the code. Results of each case study are provided in a side-by-side figures below the case study descriptions.

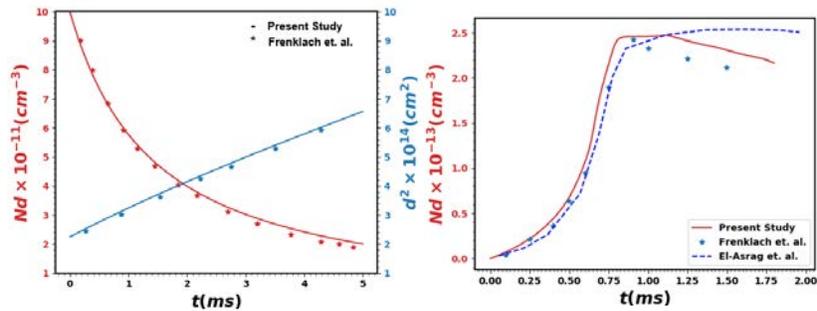**Case Study 1: Verification of Coagulation Source Terms**
The free molecular coagulation model has been validated against one of the tests given by Frenklach and Harris, 1987. Consider a population of soot particles in a closed batch reactor where initially the particles have the same size (i.e., mono-dispersed), and the number density of the particles is 10^{12} cm^{-3}. The pressure and temperature of the reactor are kept constant at 1 atm and 1500 K, respectively. The bulk species comprising the soot particles are assumed as carbon atoms ($m_0 = 3.18 \times 10^{-21}$ g) and the bulk density of the particle core ρ_s is 1.8 g/cm^3. The time profiles of particle number density predicted with the present improved MOMIC library are compared in Figure 1(a) against results predicted by Frenklach and Harris using sectional methods. As can be witnessed from these results, the present study gives good comparison with the results by Frenklach and Harris, 1987 validating the implementation of coagulation source terms.

Case Study 2: Verification of MOMIC with simultaneous nucleation, surface growth, and coagulation
This example, shown in Figure 1(b), is based on results from shock tube data at 1800 K and presented as a verification test in Frenklach and Harris, 1987. Here, it assumed that for this verification study that soot particles with m = 4.784×10^{-22} g and ρ = 1.86 g/cm^3 are formed. We use the parameters/constants and assumptions from Frenklach and Harris (1987), which used a more complex sectional method to validate the model. The time dependent profile of particle inception (nucleation) rate is provided in the literature and therefore can be used for comparison with our more cost-effective MOMIC method. The source terms for the moment equations are calculated based on this nucleation time rate profile. The particles add carbon (C) by surface growth with rate constant k_s = 1×10^{-4} g/cm^{2}s. The particles are assumed to undergo coagulation in free molecular regime. The time profiles of number density evolution of soot particles obtained in the present 6-MOMIC study is compared to the earlier 3-moment (and more approximate) MOMIC method of El-Asrag et. al. (2007). Figure 1(b) shows this comparison. It is clear that the present 6-MOMIC is much more accurate than the 3-MOMIC when compared to the sectional method. A detailed comparison with new data (if available) will be addressed during this effort.

Case Study 3: Verification of agglomeration coupled MOMIC approach for rich laminar premixed ethylene/air flame
Ethylene/air laminar premixed flames, shown in Figure 2, are presented here for the purpose of verification of the MOMIC approach equipped with nucleation, coagulation, surface growth, and aggregation source terms embedded inside the LESLIE code. The test case considered here (Figure 2) is based on experimental configurations of Xu and Faeth, 2001 for laminar premixed sooting flames. The operating pressure considered is 101325 Pa. with the initial temperature of 400 K. The rich laminar 1D C_2H_4/air flame at an equivalence ratio (Φ = 2.93) is considered for the 1D simulations in the domain of 0.05 m in length. The domain is discretized using 500 cells. The inflow is treated as subsonic constant velocity inflow while the outflow is modelled with characteristic subsonic outflow boundary conditions. The solution is initialized with non-sooting flame profiles. The ethylene-air 19 species 15 step reduced mechanism is used to account for chemical pathways modelling the breakdown of fuel to the soot precursor assumed here as acetylene (C_2H_2). The evolution of soot using the MOMIC model is compared against the experimental soot volume fraction as shown in Figure 2 and good agreements are observed.

Case Study 4: Assessment of LES-MOMIC approach for rich turbulent premixed ethylene/air flame
This proposed test intends to use the LES-MOMIC model with the finite rate chemistry for the premixed flame turbulence interaction problem (El-Asrag et. al., 2007). The schematic for the configuration is shown in Figure 3. The critical C/O ratio for ethylene-air premixed flames is 0.6. Therefore, for the current case, C/O ratio is fixed at 0.67 and the turbulence level will be varied so that the flame is in the thin reaction zone (TRZ) 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 x 15 mm x 15 mm in the streamwise x, transverse y, and spanwise z directions. The flow field is initialized using von Karman–Pao energy spectrum. Characteristic inflow-outflow conditions are specified in the streamwise direction while periodic in the other two directions. The LES grid resolution chosen for the simulations is 64 x 64 x 64 number of LES cells with the assumption of quasi-laminar chemistry, meaning that no closure for subgrid turbulence-chemistry interactions is considered. Studies are still underway and will be reported soon.
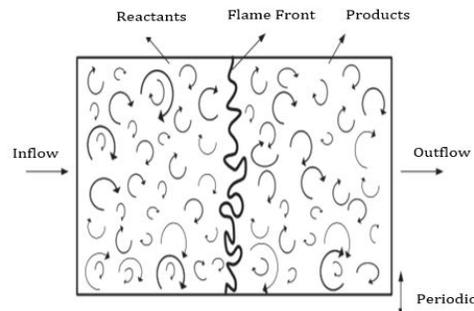

(a) Case 1: Free molecular coagulation   (b) Case 2: nucleation, coagulation & surface growth

Figure 1. Verification of MOMIC Solver using 0D Testcases
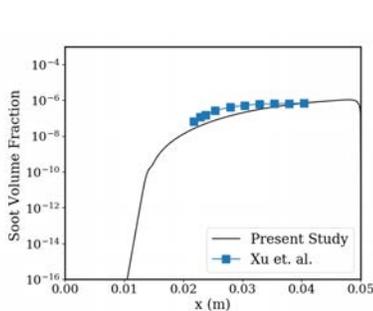
Figure 3: Case 3: Sooting Laminar Premixed Flames   **F**igure 4: Case 4: 3D turbulent premixed flame

In Case 4, rich premixed mixture enters the domain at a speed to approximately balance the forward propagation of the premixed flame and keep it statistically stationary in the middle of the domain, the combustion products leave the domain at the outflow. Inflow also contains turbulent fluctuations that wrinkle the premixed flame and soot formation occurs in the vicinity of this flame.

Task 2 – Kinetic Activities at RTRC

Objective
The goal of the effort is to identify one or two detailed ethylene/PAH kinetic mechanisms that handle both fuel decomposition and soot (PAH) growth reactions sufficiently so that the down-selected mechanism(s) can then serve as a basis for the reduced kinetic model ultimately to be used in the computational fluid dynamics (CFD)-based application.

Research Approach
The research approach involves the assessment of published ethylene/PAH mechanisms, initiated in the current reporting period. In all, seven mechanisms were assembled for analysis:

- Abbel-Bockhorn-Frenklach (99 species/533 reactions)
- Wang-Frenklach (101 species/544 reactions)
- NJFCP Foundational Chemistry submodel with SERDP PAH reactions (166 species/504 reactions)
- NJFCP Foundational Chemistry submodel with KAUST PAH reactions (168 species/818 reactions)
- Full ARAMCO-KAUST C4 mechanism (581 species/3037 reactions)
- CREK Natural Gas mechanism (114 species/1999 reactions)
- UCONN (Lu) Skeletal C2H4 mechanism (33 species/206 reactions)

(The Lu skeletal mechanism serves as a proxy for the reduced 19 species ethylene mechanism currently used at GT, because both mechanisms derive from the same fully detailed mechanism.)

These models have been benchmarked against published ethylene data sets for comparative analysis against both experimental data and relative performance against the various models. Current experimental data has been comprised primarily of shock tube data (Hidaka et al., 1999; Miller and Churchill, 1962; Skinner and Sokoloski, 1960) but comparison to well-stirred reactor, i.e., perfectly stirred reactors (PSR) (Dagaut et al., 1988; Westbrook et al., 1988) datasets have also been completed. The focus of the above analysis has been to assess the computational characteristic times of fuel decomposition and major product yields versus the experimental data. At present, one mechanism that is showing slightly better agreement to the various experiments is the CREK natural gas model, but analysis/comparisons utilizing all the above mechanisms continue.

Milestones

Milestone	Planned Due Date
Establish the framework for MOMIC for LES	12/31/20
Extend the model to agglomeration effects	09/30/20
Simulate the test case(s) using the new MOMIC model	02/27/21
Identification of detailed ethylene-air mechanism	01/30/21
MD and KMC studies of nucleation	03/31/21

Major Accomplishments
- Full inclusion of MOMIC with nucleation, surface growth, coagulation, and aggregation has been accomplished.
- Extensive validation studies with improved MOMIC.
- Basic LES-MOMIC framework is established and tested for a premixed flame turbulence interaction problem.
- Promising detailed kinetic mechanisms for fuel decomposition have been identified.

Publications:
N/A

Outreach Efforts
None

Awards
None

Student Involvement
Shubham Karpe is a PhD student at Georgia Institute of Technology.
Qi Wang is a PhD student at the University of Michigan

Plans for Next Period
MOMIC for CFD Applications (GT, RTRC, UM)
The next steps in this activity will focus on developing postprocessing tools to quantify major features such as rates of different soot formation processes like nucleation, coagulation, and aggregation and how the soot is transported. These results will establish a baseline comparison testbed. The current MOMIC approach focuses on simplified soot model with reduced fuel chemistry. Therefore, improvements can be achieved in these two areas where the involvement of UM and RTRC is critical. Some of the important improvements planned for future work revolve along the following objectives:
1) Involvement of PAH species for soot precursor with inputs from RTRC.
2) Improvements in the nucleation and surface growth reactions and their rates using inputs from UM and RTRC.

Kinetic Assessment of Ethylene/PAH Chemistry (RTRC)
The next steps in this activity will now focus on more fuel-rich experiments, with the purpose of characterizing and assessing the reduced model generation of aromatic compounds (e.g., benzene, styrene, etc.) in order to understand their ability to ultimately characterize PAH production in relevant devices (e.g., aeroengine combustors). Once these fuel-rich comparisons are completed, a mechanism down selection will then be completed

MD and KMC Simulations (UM and RTRC)
The nucleation and surface growth models contain rate parameters that are being revisited with first principal studies using MD to establish the rate constants utilizing the kinetic models developed in RTRC. Then this model will be used in the KMC studies at RTRC to establish growth rate parameters in the inputs for the LES-MOMIC studies at GT.

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