A computational study of soot formation and flame structure of coflow laminar methane/air diffusion flames under microgravity and normal gravity

Armin Veshkini

Department of Mechanical and Industrial Engineering, Ryerson University, Toronto, Ontario, M5B 2K3 Canada. Email Address: a.veshkini@utoronto.ca

Seth B. Dworkin

Department of Mechanical and Industrial Engineering, Ryerson University, Toronto, Ontario, M5B 2K3 Canada. Email Address: seth.dworkin@ryerson.ca

Corresponding Author:

Seth B. Dworkin
Department of Mechanical and Industrial Engineering,
Ryerson University,
350 Victoria St, Toronto, Ontario, Canada, M5B 2K3
Phone: 416-979-5000 x7311
Fax: 416-979-5265
Email: seth.dworkin@ryerson.ca

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A numerical study is conducted of methane–air coflow diffusion flames at microgravity (μg) and normal gravity (1g), and comparisons are made with experimental data in the literature. The model employed uses a detailed gas phase chemical kinetic mechanism that includes PAH formation and growth, and is coupled to a sectional soot particle dynamics model. The model is able to accurately predict the trends observed experimentally with reduction of gravity without any tuning of the model for different flames. The microgravity sooting flames were found to have lower temperatures and higher volume fraction than their normal gravity counterparts. In the absence of gravity, the flame radii increase due to elimination of buoyance forces and reduction of flow velocity, which is consistent with experimental observations. Soot formation along the wings is seen to be surface growth dominated, while PAH condensation plays a more major role on centerline soot formation. Surface growth and PAH growth increase in microgravity primarily due to increases in the residence time inside the flame. The rate of increase of surface growth is more significant compared to PAH growth, which causes soot distribution to shift from the centerline of the flame to the wings in microgravity.

Keywords: laminar diffusion flame, methane–air, microgravity, soot formation, numerical modelling

Introduction

Soot formation plays a crucial role within the context of fire spreading in microgravity, i.e. fire safety in manned spacecraft. Elongated formation residence times due to the absence of buoyancy forces substantially increase concentration of soot in flames under microgravity conditions, enhancing radiation heat transfer and fire spreading. As such, flames established in microgravity rely on empirical models for characterisation of soot formation [1,2]. The emergence of detailed soot models has facilitated the gain of in depth insight on soot production phenomena in flames established in microgravity.

Furthermore, a scenario, which simplifies some aspects of soot generation, is highly desirable for academic study. Soot particles are generated in high temperature, fuel rich regions when burning a variety of fuels. Soot formation is a complex phenomenon, which involves several concurrent chemical and physical processes. Reducing gravity in a coflow diffusion flame is such an example. Elimination of buoyancy simplifies the flow field and simultaneously enhances residence time within the flame. By investigating the structural differences between normal and microgravity flames, an improved understanding of the factors that affect soot formation can be achieved.

Both experimental and numerical methods have been utilized to quantify the effect of gravity on soot formation. Ku et al. [3,4] experimentally investigated soot yield and morphology for various coflow diffusion flames under reduced gravity in the NASA Lewis Research Center's 2.2-s drop tower. In microgravity the soot volume fractions measured using laser extinction increased by a factor 2-4 compared to the same flame under normal gravity. A factor of 2 increase in the measured maximum soot volume fraction and a 40% increase in primary particle diameter at 0g for laminar nitrogen diluted acetylene jet diffusion flames have also been reported by Megaridis et al. [5,6]. The trend of increase in both soot mass and size have been experimentally observed by Walsh et al. [7], Jeon and Choi [8], and Reimann et al. [9,10] using multiple optical measurement techniques. Experimental observations of non-buoyant round laminar jet diffusion flames were made for various jet flames burning in still air by Diez et al. [11] on board the Space Shuttle Columbia to correlate soot volume fraction with estimated mixture fraction for a selection of fuels/pressures. These experiments provide an important foundation for the study of microgravity soot formation.

Simulations of a laminar ethylene-air diffusion flame burning in quiescent air were conducted by Kaplan et al. [12] in reduced gravity. Their unsteady simulations showed a factor of 10 increase in peak soot volume fraction from 1g to μg. More importantly, their simulation did not reach steady-state conditions within 3s of imposing microgravity which is the time period in drop tower experiments. Kong and Liu [13,14], Liu et al. [15], and Charest et al. [16,17] also numerically investigated the influence of gravity on laminar coflow diffusion flames, demonstrating that the μg flame has lower temperatures, thicker soot regions and higher soot volume fractions than the 1g flame. These numerical investigations employed semi-empirical two equation models which based the formation and growth of soot only on acetylene. While these works were major advances in modelling ability, they were performed for conditions different from available experimental data. Therefore, no direct comparison with experiment was available, making it difficult to assess and analyze the computational models.

Recently, Ma et al. [18] measured soot and temperature in a set of methane/air coflow laminar diffusion flames in normal and microgravity on board the International Space Station. From the experiments, the peak soot volume fractions of the microgravity flames were found to be higher than the normal gravity flames by a factor of 4–8. The distribution of peak soot also shifted from the centerline at 1g to the wings at μg. The flame temperature in μg is shown to be lower than its 1g counterpart due to higher radiative loss. The study provides high-fidelity experimental data for model validation.

A detailed numerical model, *CoFlame*, was developed by Thomson, Dworkin, and coworkers [19–26]. It employs a polycyclic aromatic hydrocarbon (PAH) based sectional particle dynamics model, tracking shape and size distribution. CoFlame accounts for all processes that are known to occur in soot formation, including PAH growth, particle inception, surface growth via hydrogen-abstraction-carbon-addition (HACA) and PAH condensation, oxidation, coagulation, fragmentation, gas phase scrubbing, soot particle diffusion, and radiation [27,28]. In the present study, CoFlame is combined with a chemical mechanism by Chernov et al. [25] that includes numerous reaction pathways for formation of PAHs up to five aromatic rings. Additionally, conjugate heat transfer (CHT) between the fluid streams and the fuel tube is modeled to capture the effect of fuel tube preheating [26]. This highly detailed model is employed to predict soot in a set of methane/air coflow laminar diffusion flames [18] in normal gravity and microgravity. The mechanisms by which microgravity affects soot and flame structure are investigated. The present work builds upon the work done by Ma et al. [18], in which the same flame was studied, by linking the solid phase to PAHs up to A5, and considering conjugate heat transfer. Therefore, further insights into the effects of gravity were obtained.

Flame and model description

The burner consists of an annular fuel tube located in the center of a 76mm wide square duct with rounded corners. Details of the burner construction and operation are provided in [18]. Six methane/air flames have been studied under both microgravity and normal gravity. Details are provided in Table 1. The square duct is approximated as a co-annular tube with an identical cross-sectional area (radius r_O=4.288 cm). Soot volume fractions and temperatures were measured using a color-ratio pyrometry approach based on a color DSLR camera [18]. Although most of the discussion will be focused around the first two flames, for the soot models it is essential to illustrate that the model predictions are applicable to a range of conditions. Narrow application of models tends to hinder their development and the value and knowledge obtained from their studies, hence the inclusion of the other data.

CoFlame [29] is adapted to model the flames in this investigation. For the gaseous phase, the fully coupled elliptical conservation equations for mass, momentum, energy, species mass fraction, soot aggregate number densities, and primary particle number densities are solved. The model utilizes the axi-symmetrical nature of the flame, and equations are solved in the two-dimensional (z and r) cylindrical co-ordinate system. All boundary conditions including inlet velocities are set in accordance to the values reported in [18,30]. A detailed description of the governing equations, and solution methodology can be found in [19–26,29]. The Chernov et al. [25] chemical mechanism, being reduced to 94 species and 754 reactions by eliminating methanol chemistry, describes the oxidation of the fuel and the formation of PAHs.

The three heaviest PAH species in the mechanism has been chosen as the PAH species that interact with the soot particles which are benzo(a)pyrene ($C_{20}H_{12}$), benzo(a)pyrenyl ($C_{20}H_{12}$), and benzo(ghi)fluoranthene ($C_{18}H_{10}$). The 5-ring PAHs are used as nucleating species because they are more representative of PAH stacks observed in soot particles [31–33]. The 5-ring model is slightly more versatile. An extensive discussion on this subject can be found in Eaves et al. [29] and Saffaripour et al. [33]. Nucleation is modeled based on the collision of two PAHs in the free-molecular regime as in [33]. The HACA mechanism [34,35] is used to describe soot surface growth. As in previous studies [26,33,36], a constant value of 1.0 is used for the surface site density parameter α, which is a semi-empirical parameter that reconciles the inaccuracies of treating the soot surface like a corresponding PAH. PAH condensation is modelled based on collision theory between PAH molecules and aggregates [37]. All model parameters are consistent with parameters described by Eaves et al. [29], which was validated for soot formation in both methane and ethylene coflow flames. Details of the sectional model and transport equations can be found in [29].

Results and discussion

Temperature is important to soot formation; especially the hydrogen abstraction reaction which has the highest activation energy [37]. Radiation heat losses from soot will influence flame temperature as well. According to Liu et al. [15], these radiation heat losses are more significant at microgravity than 1g and completely alter the structure of the flame.

In order to examine the performance of the model in predicting temperature and demonstrate structural differences between the microgravity and normal gravity flames, computed and measured temperature maps for the F1 and F1g flames are depicted in Figure 1 and radial temperature profiles at three different heights are provided in Figure 2. Ma et al. [18] estimated that measured microgravity soot temperature is underestimated by up to 50K due to variation in range of the soot emission properties (i.e., dispersion exponent). The 50K underestimation is in addition to other uncertainties associated with the color-ratio pyrometry. The soot self-absorption along the optical pathway of collection is not accounted for, which can lead to an additional significant underestimation of the soot temperature. Also, the absorption coefficient fields (or equivalently the soot emissivity fields) at the different wavelengths used by Ma et al. [18] are not directly measured, which means that a model for soot refractive index as a function of wavelength is implicitly required, which can lead to significant discrepancies [38]. Hence, temperature predictions agree with experimental data for both flames. The exception is that temperature is overpredicted on the centerline, which is due to underprediction of soot and resulting radiation. The overprediction of temperature for the microgravity flame is more significant than for the normal gravity flame, consistent with [15]. The maximum temperature difference between the predicted and experimental data for the microgravity flame is twice that of the normal gravity flame. Similar trends in prediction of temperature are observed for the other flames for which results are included in the Supplementary Data.

The flame radius (r_f) is defined as the maximum radius of the 1% CH concentration isopleth [30]. A maximum increase in flame radius due to an absence of gravity is observed in the F1 flame, where r_f is 60% larger than in the F1g flame. For the F2 and F3 flames, r_f is predicted to increase 47% and 21%, respectively. For the normal gravity flames, r_f is relatively constant between the three flames (between 5.1–4.2mm) which is consistent with the experimental and theoretical studies that r_f is proportional to $St^{0.25}$ (St is the Stokes number) [30]. In the microgravity flames however, the average velocity which is the dominant variable in St, is highly dependent on the inlet flow rate. Therefore, the lowest increase in r_f is observed for the F3 flame which has the closest velocity field to its 1g counterpart.

The predicted soot volume fraction maps and the experimental data from [18] for the F1 and F1g flames are demonstrated in Figure 3. The computed maximum soot volume fraction is underpredicted in both 1g and μg by a factor of 10 compared to the experimental data. Although the measured microgravity soot volume fraction is possibly overestimated by up to a factor of 2 due to the uncertainty of the soot emission properties [18], that is not enough to put the predicted peak soot within the range of the uncertainty of the experimental data. The same trend of underprediction of peak soot in comparison to experimental data is found for the remaining flame soot predictions as well, which are depicted in the Supplementary Data. Despite underprediction, the predicted radial distribution of soot particles is in agreement with the experimental data. It should be noted that the soot predictions are in better agreement with experimental data compared to earlier numerical studies of soot in these flames [18] (see Table 2).

Despite the underprediction of soot volume fraction, the model is able to capture the transition of location of peak soot between different flames which was observed experimentally. For the third set of the flames, the absence of gravity causes the peak soot volume fraction to increase but the location of the peak remains on the centerline of the flame. However, for the other two sets, in addition to increases in the maximum soot volume fraction in microgravity, a shift is observed in the location of the peak. The peak soot concentration location moves from the centerline in the 1g flame toward the annular region of the flame (wings) in its μg counterpart. The soot model is accurately capturing this transition for all flames.

Thermal diffusion forces can shift heavy molecules and soot in the coflow diffusion flames towards the centerline, as was shown in Giovangili and coworkers [39,40]. These findings emphasize the importance of including relevant transport phenomena in the model which was incorporated in the soot model for this study. However, the shift observed here is not an artefact of changing transport model parameters. Instead, the shift is between two flames with different gravitational conditions observed experimentally and in the numerical predictions. Since the convection forces due to buoyancy are much stronger in the 1g flame it would lessen the relative effect of diffusion forces on the shift compared to the μg flame. Therefore, the diffusion forces are having a much stronger effect in pushing the particles towards the centreline. Despite this effect, the soot is formed mostly on the wings. Hence, the shift cannot be attributed solely to the diffusion forces although they are present and are responsible for part of the shift towards the centerline in all of the coflow diffusion flames.

In addition, the predictions show a factor of 10 increase in soot volume fraction in microgravity which is in good agreement with the experimental.

A sophisticated soot and PAH simulation too the Coflame code includes state-of-the-art soot formation processes and PAH growth pathways. The Coflame code has been validated against experimental data for an axi-symmetric pipe experiencing a sudden expansion, and substantial data available for ethylene–air and methane–air diffusion flames [29], high pressure [41], partially premixed combustion [19], diluted flames, and different burner geometries [21]. In addition, the soot model has been validated against experimental data for several premixed flames [32,42]. The facts that the validation here has been extended to six different flames and the model is able to correctly capture the trend of increase of soot due to micro-gravity and the transition of soot maximum concentration location from wings toward centerline, in addition to previous extensive validation of the Coflame code, allow further insights to understand soot formation in microgravity.

Predicted soot peak primary particle diameter, primary particle number density, and number of particles per aggregate along the pathline of maximum soot in the annular region, and along the centerline of the flames are summarized in Table 3. The particle pathline follows the particle trajectory and includes the thermophoretic velocity. In the F3g flame, the experiments and model prediction shows the soot to be formed in a very narrow area in the vicinity of the centreline, causing the wings to be irrelevant to this analysis. In general, the model predicts an increase in particle diameters in microgravity. However, the rate of increase is higher on the wings than on the centerline which is also consistent with the trend of increase in soot volume fraction. The increase in the particle diameter observed is within the range that is experimentally observed by Ku et al. [3] and Reimann et al. [9].

An interesting trend is observed for the number density of particles. On the wings, the number density remains constant as the gravity disappears. Number density of particles is primarily controlled by the nucleation process [29,43]. Nucleation is assumed to be a function of collision rate between PAHs [37] which is proportional to temperature and PAH concentrations. Predicted nucleation on the wings is highest downstream of the fuel tube near the flame front where the temperature is close to its peak. The fact that number density remains unchanged indicates that the cumulative effect of different parameters is such that nucleation remains constant. One explanation is that since the nucleation process in this region is fast, the increase in the residence time due to microgravity has an insignificant effect on the nucleation rate. On the centerline, an increase in number density of particles is predicted as the buoyancy is eliminated, which can be attributed to the increase of PAH concentration (see Figure 5) as well as residence time. Primary particles per aggregate (n_p) increases by reducing buoyancy both on the wings and on the centerline. Since the number density of particles is also increased on the centerline, the effect of increasing residence time in microgravity conditions is more pronounced on n_p.

Mass based contributions of different growth and oxidation processes for the F1 and F1g flames along the wings and centerline are presented in Figure 4. This figure illustrates the integrated contributions of different processes to soot mass, with respect to time along the particle pathline. The horizontal axis indicates the height up to which the values are time-integrated. This set of data can be used to better understand the formation pathways of soot and interpret the mechanism for the shift of the location of peak soot from the centerline to the wings in microgravity. A common pattern among all cases is that soot mass yield starts with the nucleation of soot particles. However, the PAH contribution to soot mass does not prevail further downstream. The halt of PAH addition before entering the oxidation zone is a consequence of PAH depletion. After the inception stage, HACA growth comes into effect and continues all the way through the oxidation zone where all soot mass is depleted by O_2 and OH oxidation. Since these are non-sooting flames, the mass production of HACA + PAH addition balance the mass consumption via oxidation downstream of the flame front.

From comparisons of contributions of different processes in 1g and μg, the following observation is made. On the wings, the soot mass growth is dominated by HACA, accounting for 89% of peak soot mass in 1g and 93% in μg condition. In microgravity on the wings, the contribution of HACA increased by a factor of 12 while the PAH contribution to soot mass increased by a factor of 7. On the centerline a different behaviour is observed; HACA is still responsible for most of the soot mass. In normal gravity, 93% of the peak soot mass is gained through HACA growth. Under microgravity the balance for soot yield is shifted more toward PAH addition. PAH addition accounts for 36% of the total soot mass in the F1 flame on the centerline. In comparison to 1g, the mass addition by PAH is increased by a factor of 11 while HACA only increased by a factor of 4. From these observations, it can be concluded that slower increase in HACA on the centerline is the reason for the shift of peak soot location from the centerline in the F1g flame to the wings in the F1 flame.

Figure 5 illustrates the variation of acetylene, benzene, and pyrene mole fractions, which are all soot precursors, along the wings and centerline of the F1 and F1g flames. As presented in Table 4, the carbon addition step in HACA for soot and PAHs is most often considered to be by acetylene and the hydrogen abstraction to be via H radical [28,34,35]. Therefore, studying acetylene behaviour in the flame can be used to comprehend HACA growth. The C_2H_2 levels in the F1g flame are comparable on the wings and on the centerline. By comparing these results with the HACA contribution to soot mass in Figure 4, it can be concluded that the balanced concentration of C_2H_2 on the wings and centerline translates to balanced HACA growth in these regions.

However, in the absence of buoyancy acetylene concentrations decrease both on the wings and on the centerline while HACA increases considerably. Hence, the increase of HACA is not a direct result of increasing C_2H_2. Instead, HACA is increased due to more consumption of C_2H_2 which leads to lower levels of C_2H_2 under microgravity. As presented in Table 4, the reactions with highest rate for HACA are the C_2H_2 addition and H abstraction. H radical, often the bottleneck of the HACA growth [21,29], also displays a similar behaviour; the increase in HACA growth as a result of prolonged residence time comes at the cost of consumption of H and C_2H_2 from the gas phase.

Benzene and pyrene represent the behaviour of small aromatics and moderate PAH species in the flame, respectively. The model predictions suggest that as gravity reduces, the concentration of benzene and pyrene increase everywhere but not at the same rate. The increase in aromatic compounds on the centerline is more significant than on the wings. Peak benzene mole fraction is increased by 71% on the centerline while it increases by only 14% on the wings. Also, these results show that the relationship between benzene and pyrene is not direct. Aforementioned growth in benzene concentration results in a 150% increase of pyrene on the wings and a factor of 5.6 increase in pyrene concentration on the centerline. The enormous increase in heavier PAH species on the centerline is also one of the reasons for PAH addition becoming more dominant on the centerline.

Finally, to leverage the power of the numerical model, a sensitivity analysis is conducted to quantify the effect of HACA soot growth rate and residence time on soot formation. The soot particle surface reactivity parameter (α) [21] which directly influences HACA, is increased from 1 to 8. While these values are unphysical, they allow further understanding on needed areas of model development. The predicted soot volume fraction in the F1 flame with various α along the wings and centerline are depicted in Figure 6. The results with $\alpha = 1$ represent the results provided in previous sections and the rest shows the effect of enhancing HACA surface growth. On the wings, the soot volume fraction monotonically increases with α. With $\alpha = 8$, enough soot is produced to overcome the previous underprediction. These results show that there is a potential to investigate new surface growth venues to upgrade soot model predictions on the wings. Contrarily, on the centerline, soot mass becomes insensitive to α due to depletion of H radical. Although appreciable amounts of C_2H_2 are present in the mixture, due to a lack of hydrogen-abstraction, soot particles are unable to absorb them. These results reiterate previous study conclusions which suggested that the underprediction of centerline soot is a result of PAH chemistry [23,36]. Thus, it is necessary to revisit PAH growth pathways and invest in introducing novel growth pathways for PAHs.

In flame F1g, when α is increased from 1 to 8, the peak soot location moves toward the wings. However, with α=8, the model overpredicts soot volume fraction by an order of magnitude, suggesting that in that case the model is not correct. Moreover, with α=8 the difference between the soot volume fraction on the wings and centerline of the F1g flame is not drastic, as it is for the μg flame.

The absence of gravity causes an increase in the residence time in the flame. This increase in residence time as has been shown in Figure 1 is accompanied by a change in temperature field both in the temperature magnitude and distribution. To separate the influence of temperature and residence time, a numerical experiment has been conducted in which the temperature field has been fixed to the F1 flame temperature but the gravity has been set to 1g. This configuration will impose a residence time close to the F1g flame. The result is a complete disappearance of soot particles everywhere in the flame. A substantial reduction of PAH precursors (propargyl and *i*-C_4H_5) is caused by the lack of formation of PAHs. As a result, no soot is formed in this configuration. This experiment alludes that the temperature field distortion as a result of the absence of gravity has a negative effect on soot formation. In conclusion, the increase of soot formation as a result of an increase in residence time is so significant that it can overcome the temperature effect and leads to an order of magnitude increase in soot formation.

Conclusions

Laminar, sooting coflow methane–air diffusion flames at microgravity and normal gravity are numerically simulated using a detailed PAH-based sectional soot model. The trends observed in the experimental data are captured by the model. The flame temperature in microgravity is shown to be lower than its 1g counterpart with experimental data. Computations of both 1g and μg flames yield agreement in flame shape with experimental results. As in other investigations, soot volume fraction is underpredicted. The peak soot volume fraction in microgravity flames was found to be higher than in normal gravity by a factor of 10. Primary particle diameters and number of particles per aggregate were found to increase in the absence of gravity.

Experimental data shows a shift of distribution of peak soot from the centerline of the flame at 1g to the wings at μg. The computational model successfully captured this soot migration and it was attributed to a considerable increase in HACA surface growth on the wings in microgravity.

Soot formation is seen to be surface growth dominated in the studied flames, while the significance of PAH growth increases on the centerline for microgravity flames. HACA and PAH growth increase along the wings and centerline of the flames with elimination of gravity due to increases of the growth residence time. The HACA growth on the wings was found sensitive to the soot surface reactivity parameter whereas soot on the centerline is shown to be insensitive to it due to reduced H radical concentration.

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References

[1] Legros G, Torero JL. Phenomenological model of soot production inside a non-buoyant laminar diffusion flame. Proc Combust Inst 2015;35:2545–53. doi:10.1016/j.proci.2014.05.038.

[2] Legros G, Fuentes A, Rouvreau S, Joulain P, Porterie B, Torero JL. Transport mechanisms controlling soot production inside a non-buoyant laminar diffusion flame. Proc Combust Inst 2009;32:2461–70. doi:10.1016/j.proci.2008.06.179.

[3] Ku JC, Griffin DW, Greenberg PS, Roma J. Buoyancy-induced differences in soot morphology. Combust Flame 1995;102:216–8. doi:10.1016/0010-2180(95)00108-I.

[4] Greenberg PS, Ku JC. Soot volume fraction maps for normal and reduced gravity laminar acetylene jet diffusion flames. Combust Flame 1997;108:227–30. doi:10.1016/S0010-2180(96)00205-2.

[5] Megaridis CM, Griffin DW, Konsur B. Soot-field structure in laminar soot-emitting microgravity nonpremixed flames. Symp Combust 1996;26:1291–9. doi:10.1016/S0082-0784(96)80347-X.

[6] Konsur B, Megaridis CM, Griffin DW. Soot aerosol properties in laminar soot-emitting microgravity nonpremixed flames. Combust Flame 1999;118:509–20. doi:10.1016/S0010-2180(99)00021-8.

[7] Walsh KT, Fielding J, Smooke MD, Long MB. Experimental and computational study of temperature, species, and soot in buoyant and non-buoyant coflow laminar diffusion flames. Proc Combust Inst 2000;28:1973–9. doi:10.1016/S0082-0784(00)80603-7.

[8] Jeon B-H, Choi JH. Effect of buoyancy on soot formation in gas-jet diffusion flame. J Mech Sci Technol 2010;24:1537–43. doi:10.1007/s12206-010-0406-4.

[9] Reimann J, Will S. Optical diagnostics on sooting laminar diffusion flames in microgravity. Microgravity - Sci Technol 2005;16:333–7.

doi:10.1007/BF02946001.
[10] Reimann J, Kuhlmann S-A, Will S. Investigations on Soot Formation in Heptane Jet Diffusion Flames by Optical Techniques. Microgravity Sci Technol 2010;22:499–505. doi:10.1007/s12217-010-9204-y.
[11] Diez FJ, Aalburg C, Sunderland PB, Urban DL, Yuan Z-G, Faeth GM. Soot properties of laminar jet diffusion flames in microgravity. Combust Flame 2009;156:1514–24. doi:10.1016/j.combustflame.2009.04.006.
[12] Kaplan CR, Oran ES, Kailasanath K, Ross HD. Gravitational effects on sooting diffusion flames. Symp Combust 1996;26:1301–9. doi:10.1016/S0082-0784(96)80348-1.
[13] Kong W, Liu F. Effects of Gravity on Soot Formation in a Coflow Laminar Methane/Air Diffusion Flame. Microgravity Sci Technol 2009;22:205–14. doi:10.1007/s12217-009-9175-z.
[14] Kong W, Liu F. Numerical study of the effects of gravity on soot formation in laminar coflow methane/air diffusion flames under different air stream velocities. Combust Theory Model 2009;13:993–1023. doi:10.1080/13647830903342527.
[15] Liu F, Smallwood GJ, Kong W. The importance of thermal radiation transfer in laminar diffusion flames at normal and microgravity. J Quant Spectrosc Radiat Transf 2011;112:1241–9. doi:10.1016/j.jqsrt.2010.08.021.
[16] Charest MRJ, Groth CPT, Gülder ÖL. A numerical study on the effects of pressure and gravity in laminar ethylene diffusion flames. Combust Flame 2011;158:1933–45. doi:10.1016/j.combustflame.2011.02.022.
[17] Charest MRJ, Groth CPT, Gülder ÖL. Effects of gravity and pressure on laminar coflow methane–air diffusion flames at pressures from 1 to 60 atmospheres. Combust Flame 2011;158:860–75. doi:10.1016/j.combustflame.2011.01.019.
[18] Ma B, Cao S, Giassi D, Stocker DP, Takahashi F, Bennett BA V., et al. An experimental and computational study of soot formation in a coflow jet flame under microgravity and normal gravity. Proc Combust Inst 2015;35:839–46. doi:10.1016/j.proci.2014.05.064.
[19] Chernov V, Zhang Q, Thomson MJ, Dworkin SB. Numerical investigation of soot formation mechanisms in partially-premixed ethylene–air co-flow flames. Combust Flame 2012;159:2789–98. doi:10.1016/j.combustflame.2012.02.023.
[20] Zhang Q, Guo H, Liu F, Smallwood GJ, Thomson MJ. Modeling of soot aggregate formation and size distribution in a laminar ethylene/air coflow diffusion flame with detailed PAH chemistry and an advanced sectional aerosol dynamics model. Proc Combust Inst 2009;32:761–8. doi:10.1016/j.proci.2008.06.109.
[21] Veshkini A, Dworkin SB, Thomson MJ. A soot particle surface reactivity model applied to a wide range of laminar ethylene/air flames. Combust Flame 2014;161:3191–200. doi:10.1016/j.combustflame.2014.05.024.
[22] Zhang Q, Thomson MJ, Guo H, Liu F, Smallwood GJ. A numerical study of soot aggregate formation in a laminar coflow diffusion flame. Combust Flame 2009;156:697–705. doi:10.1016/j.combustflame.2008.10.022.
[23] Dworkin SB, Zhang Q, Thomson MJ, Slavinskaya NA, Riedel U. Application of an enhanced PAH growth model to soot formation in a laminar coflow ethylene/air diffusion flame. Combust Flame 2011;158:1682–95. doi:10.1016/j.combustflame.2011.01.013.
[24] Zhang Q, Thomson MJ, Guo H, Liu F, Smallwood GJ. Modeling of Oxidation-Driven Soot Aggregate Fragmentation in a Laminar Coflow Diffusion Flame. Combust Sci Technol 2010;182:491–504. doi:10.1080/00102200903463050.

[25] Chernov V, Thomson MJ, Dworkin SB, Slavinskaya NA, Riedel U. Soot formation with C1 and C2 fuels using an improved chemical mechanism for PAH growth. Combust Flame 2014;161:592–601. doi:10.1016/j.combustflame.2013.09.017.

[26] Eaves NA, Thomson MJ, Dworkin SB. The Effect of Conjugate Heat Transfer on Soot Formation Modeling at Elevated Pressures. Combust Sci Technol 2013;185:1799–819. doi:10.1080/00102202.2013.839554.

[27] Bockhorn H, editor. Soot Formation in Combustion. vol. 59. Berlin, Heidelberg: Springer Berlin Heidelberg; 1994. doi:10.1007/978-3-642-85167-4.

[28] Haynes BS, Wagner HG. Soot formation. Prog Energy Combust Sci 1981;7:229–73. doi:10.1016/0360-1285(81)90001-0.

[29] Eaves NA, Zhang Q, Liu F, Guo H, Dworkin SB, Thomson MJ. CoFlame: A refined and validated numerical algorithm for modeling sooting laminar coflow diffusion flames. Comput Phys Commun 2016. doi:10.1016/j.cpc.2016.06.016.

[30] Cao S, Ma B, Bennett BA V., Giassi D, Stocker DP, Takahashi F, et al. A computational and experimental study of coflow laminar methane/air diffusion flames: Effects of fuel dilution, inlet velocity, and gravity. Proc Combust Inst 2015;35:897–903. doi:10.1016/j.proci.2014.05.138.

[31] Wang H. Formation of nascent soot and other condensed-phase materials in flames. Proc Combust Inst 2011;33:41–67. doi:10.1016/j.proci.2010.09.009.

[32] Veshkini A, Eaves NA, Dworkin SB, Thomson MJ. Application of PAH-condensation reversibility in modeling soot growth in laminar premixed and nonpremixed flames. Combust Flame 2016;167:335–52. doi:10.1016/j.combustflame.2016.02.024.

[33] Saffaripour M, Veshkini A, Kholghy M, Thomson MJ. Experimental investigation and detailed modeling of soot aggregate formation and size distribution in laminar coflow diffusion flames of Jet A-1, a synthetic kerosene, and n-decane. Combust Flame 2014;161:848–63. doi:10.1016/j.combustflame.2013.10.016.

[34] Appel J, Bockhorn H, Frenklach M. Kinetic modeling of soot formation with detailed chemistry and physics: laminar premixed flames of C2 hydrocarbons. Combust Flame 2000;121:122–36. doi:10.1016/S0010-2180(99)00135-2.

[35] Frenklach M, Wang H. Detailed modeling of soot particle nucleation and growth. Symp Combust 1991;23:1559–66. doi:10.1016/S0082-0784(06)80426-1.

[36] Eaves NA, Veshkini A, Riese C, Zhang Q, Dworkin SB, Thomson MJ. A numerical study of high pressure, laminar, sooting, ethane–air coflow diffusion flames. Combust Flame 2012;159:3179–90. doi:10.1016/j.combustflame.2012.03.017.

[37] Appel J, Bockhorn H, Wulkow M. A detailed numerical study of the evolution of soot particle size distributions in laminar premixed flames. Chemosphere 2001;42:635–45. doi:10.1016/S0045-6535(00)00237-X.

[38] Legros G, Wang Q, Bonnety J, Kashif M, Morin C, Consalvi J-L, et al. Simultaneous soot temperature and volume fraction measurements in axis-symmetric flames by a two-dimensional modulated absorption/emission technique. Combust Flame 2015;162:2705–19. doi:10.1016/j.combustflame.2015.04.006.

[39] Giovangigli V. Multicomponent transport in laminar flames. Proc Combust Inst 2015;35:625–37. doi:10.1016/j.proci.2014.08.011.

[40] Dworkin SB, Smooke MD, Giovangigli V. The impact of detailed multicomponent transport and thermal diffusion effects on soot formation in

ethylene/air flames. Proc Combust Inst 2009;32:1165–72. doi:10.1016/j.proci.2008.05.061.
[41] Eaves NA, Veshkini A, Riese C, Zhang Q, Dworkin SB, Thomson MJ. A numerical study of high pressure, laminar, sooting, ethane-air coflow diffusion flames. Combust Flame 2012;159. doi:10.1016/j.combustflame.2012.03.017.
[42] Kholghy MR, Veshkini A, Thomson MJ. The core-shell internal nanostructure of soot - A criterion to model soot maturity. Carbon N Y 2016;100. doi:10.1016/j.carbon.2016.01.022.
[43] Smooke MD, Long MB, Connelly BC, Colket MB, Hall RJ. Soot formation in laminar diffusion flames. Combust Flame 2005;143:613–28. doi:10.1016/j.combustflame.2005.08.028.

Tables

Table 1 Burner geometry and inlet boundary of the simulated laminar diffusion flames

Flame des.	Gravity	Velocity(cm/s)		Fuel tube ID (mm)
		Fuel	Air	
F1	0	46	18	3.23
F1g	1g	46	18	3.23
F2	0	169	18	1.59
F2g	1g	169	18	1.59
F3	0	169	40	1.59
F3g	1g	169	40	1.59

Table 2 Comparison of maximum soot volume fraction predicted by Coflame model, and Ma et al. [18] with experimental data [18].

Flame	Coflame	Ma et al.	Exp.
F1	0.5	0.2	4.4
F1g	0.05	0.18	0.5
F2	0.2	0.16	2.0
F2g	0.02	0.12	0.4
F3	0.1	0.12	1.3
F3g	0.01	0.09	0.4

Table 3 Predicted maximum soot primary particle diameter (D_p), number density of primary particles (N_p), and number of particles per aggregate (N_p/N) along the pathline of maximum soot on the wings (W) and the centerline (C) of a set of methane flames under normal and microgravity conditions.

Flame	D_p (nm)		N_p (#/cm^3)		N_p/N	
	W	C	W	C	W	C
F1	23	17	5.E10	1.E11	12	26
F1g	9	9	5.E10	7.E10	7	9
F2	18	14	4.E10	8.E10	10	20
F2g	9	8	4.E10	5.E10	5	6
F3	11	11	5.E10	7.E10	9	14
F3g	–	9	–	4.E10	–	4

Table 4 HACA–based soot surface growth and oxidation reactions [34], $k = AT^{b}e^{-E_a/RT}$.

No.	Reaction			A $\left(\frac{cm^3}{mol.s}\right)$	b	E_a $\left(\frac{kcal}{mol}\right)$
S1	C_{soot}–H+ H	$\Longleftrightarrow$	$C_{soot}^{\bullet}$ + H_2	4.2×10^{13}	0.0	13.0
S2	C_{soot}–H+ OH	$\Longleftrightarrow$	$C_{soot}^{\bullet}$ + H_2O	1.0×10^{10}	0.73	1.43
S3	$C_{soot}^{\bullet}$ + H	$\longrightarrow$	C_{soot}–H	2.0×10^{13}	0.0	0.0
S4	$C_{soot}^{\bullet}$ + C_2H	$\longrightarrow$	C_{soot}–H+ H	8.0×10^{7}	1.56	3.8
S5	$C_{soot}^{\bullet}$ + O_2	$\longrightarrow$	2CO + product	2.2×10^{12}	0.0	7.5
S6	C_{soot}–H+ OH	$\longrightarrow$	CO + product		γ_{OH}=0.13	

List of figure captions

Figure 1. Computed temperature (K) isopleths and measured soot temperature (K) from [18] under (left) normal gravity and (right) microgravity for a CH_4 flame with 3.23 mm ID nozzle and average fuel speed of 46 cm/s.

Figure 2. Computed temperature (K) radial profiles and measured soot temperature (K) from [18] at three different heights above the burner under (left) normal gravity and (right) microgravity for a CH_4 flame with 3.23 mm ID nozzle and average fuel speed of 46 cm/s.

Figure 3. Comparison of soot volume fraction maps (ppm) with measured soot volume fractions (ppm) from [18] under (left) normal gravity and (right) microgravity for a CH_4 flame with 3.23 mm ID nozzle and average fuel speed of 46 cm/s.

Figure 4. Total mass contribution by HACA surface growth, PAH addition (nucleation and condensation), and the amount of soot mass depleted by oxidation for a soot particle travelling (top) along the pathline of maximum soot on the wings, and (bottom) along the centerline for a CH_4 flame with 3.23 mm ID nozzle and average fuel speed of 46 cm/s under normal and microgravity.

Figure 5. Acetylene, benzene and pyrene mole fractions as a function of height above the burner along the pathline of maximum soot on the wings and along the centerline for a CH_4 flame with 3.23 mm ID nozzle and average fuel speed of 46 cm/s under normal and microgravity.

Figure 6. Predicted soot volume fraction (ppm) using various surface reactivity parameter as a function of height above the burner (top) along the pathline of maximum soot on the wings and (bottom) along the centerline for a CH_4 flame with 3.23 mm ID nozzle and average fuel speed of 46 cm/s under microgravity.