On the Role of Soot and Radiative Heat Flux for Flame Spread Along Solid Fuels

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Abstract: This study presents the influence of \( \text{O}_2/\text{N}_2 \) oxidizer blends on soot formation and radiative heat flux (RHF) in PMMA-Air slab burner experiments. RHF is measured using two-color pyrometry along with flame reconstruction techniques. Far-field \( \text{O}_2 \) concentrations are varied in the range of 17 – 28\% in experiments and the overall flame temperatures, soot volume fraction (SVF), and RHF are found to increase. These observations are further analyzed using numerical solutions to diffusion flamelets. A semi-empirical two-equation soot formation model is coupled with the flamelet solver along with a fully-coupled discrete ordinates radiation solver. \( \text{O}_2 \) concentrations are varied from 15 – 100\%. Flame temperatures are found to increase with \( \text{O}_2 \) concentrations. Soot volume fraction initially increases, peaking at 45\% \( \text{O}_2 \) concentration and then decreases with further increase in \( \text{O}_2 \) concentration. Regardless of soot behavior, RHF from flamelets is found to increase with \( \text{O}_2 \) concentrations. The field averaged RHF from slab burner experiments are found to be in very good agreement with the RHF from 1D flames at low \( \text{O}_2 \) concentrations. Furthermore, the RHF values are also compared to Tewarson et al.\textsuperscript{[1]} values extrapolated using radiation corrected Spalding B-number equation and are found to be consistent. These results enable one to correlate RHF predictions from flamelets to those observed in multi-dimensional flame spread experiments.

Keywords: Radiative Heat Flux, Slab burner, Two-color pyrometry, Flamelets, Solid fuel combustion

1. Introduction

Solid fuel boundary layer combustion has a wide range of applications from fire safety (flame spread in both gravity and microgravity)\textsuperscript{[1–6]} to propulsion (hybrid rockets)\textsuperscript{[7–11]}. Heat transfer from the flame to fuel surface drives the flame spread and fuel regression in these reacting boundary layer applications. In hybrid rockets, convective effects are dominant over radiation at high oxidizer mass flux, \( G \), and at lower \( G \), radiation dominates\textsuperscript{[10,11]}. In microgravity flame spread, due to the absence of buoyancy effects, flame spread is widely governed by radiation heat transfer from the flame to fuel\textsuperscript{[6]}. For flame spread in gravity, buoyancy induced convective effects are equally important as the radiation heat transfer\textsuperscript{[12]}. Soot radiation is the leading contributor in the overall radiation as compared to gas radiation. The key challenges in understanding the overall radiative effects is to understand the soot formation processes as a function of the oxygen concentration in the oxidizer and its effect on the incident radiative heat flux.

Measuring the radiative heat flux (RHF) from the flame to fuel is possible using water-cooled Schmidt-Boelter gauges but the measurements are limited to single point and are often challenging especially in very high temperature environments and small geometry fuels. Recent advances in
the development of non-intrusive diagnostic using two-color pyrometry and laser induced incan-
descence methods allow accurate flame temperature and soot volume fraction (SVF) measurements
[11, 13–17]. Two-color pyrometry diagnostic is further extended to measure flame to fuel surface
RHF using two orthogonal images of the flame and reconstructing an approximate 3D flame hull
[16, 17]. These non-intrusive diagnostic allow for spatial and temporal measurements of the overall
flame spread and enable detailed understanding of the underlying theories and model validation.

Soot formation in hydrocarbons is phenomenologically described using four main processes -
1) nucleation, 2) particle surface growth, 3) soot oxidation, and 4) agglomeration [18, 19]. The
overall flame temperatures and oxygen concentration are the leading sources driving the soot for-
mation process. Higher oxygen concentrations lead to higher flame temperatures and increased
soot nucleation and surface growth rates. However, it is not very clear how the increased oxygen
concentrations will affect the soot oxidation and consequently the overall radiative heat flux (RHF)
from the flame to fuel surface. Multiple studies have focused on understanding the soot processes
and its dependence on the concentration of oxygen in both fuel stream and oxidizer stream leading
to a non-monotonic behavior of soot [20–22]. However, very few studies have focused on the over-
all effect of this non-monotonic behavior on the soot radiation and the total RHF from the flame to
fuel surface. Wang et al. studied the effects of jet velocities and increasing oxygen concentrations
in co-flow turbulent jet diffusion flames for methane, propane, and blend of the fuels [23]. They
experimentally measured the SVF and observed that SVF initially increases with increasing oxy-
gen concentration, peak at $\sim 40\%$ and then decreases with further increase in oxygen. The radiant
fraction, $\chi_{rad}$, was found to increase with increasing oxygen concentration [23].

Multiple studies have focused on understanding the role of oxygen concentration on the radia-
tion heat transfer in flame spread over Poly(methyl methacrylate) (PMMA), polypropylene, and
polyoxymethylene solid fuels [1, 24, 25]. Most of these studies have focused on lower levels of $O_2$
concentrations going as high as 45% with a primary aim of understanding the flammability limits
of PMMA at lower $O_2$ concentrations [25]. All these studies found that in general fuel regression
rate, mass loss rate, and heat fluxes incident on the fuel decreased with decreasing oxygen concen-
trations [1, 24, 25]. Ailbert et al. also found that heat flux exhibits linear oxygen-concentration-
dependent behavior at low concentrations [25]. However, none of these studies focused on higher
oxygen concentrations above 40% on PMMA or any other solid fuels. Furthermore, there are not
many studies that focused on the role of soot formation in solid fuels and its effect on RHF with
increasing oxygen concentrations.

The main objective of this study is to establish a correlation of the flame to fuel surface RHF
to multidimensional flame spread over solid fuels through 2D slab burner experiments and 1D
diffusion flamelets. The influence of soot formation-oxidation processes on RHF in PMMA-Air
combustion with increasing oxygen concentration is obtained through the diffusion flamelets. A
two-equation semi-empirical acetylene-based soot formation model is coupled with the flamelet
solver along with an $S_8$ discrete ordinates radiation solver. The oxygen concentrations are varied
in the range of 15% – 100% (pure oxygen) and the SVF and flame to fuel radiative heat flux is
numerically studied. 2D slab burner experiments are conducted for PMMA-Air by varying $O_2$ in
the range of 17 – 28%. A recently developed non-intrusive flame to fuel surface RHF diagnostic
[16] is utilized to measure the flame temperatures, SVF, and RHF. The field-averaged RHF
measurements from slab burner are correlated to those obtained from 1D flames. Additionally,
the RHF extrapolated by Tewarson et al. using the B-number equation are also compared to both
experimental measurements and 1D flame solution and are shown to be in very good agreement.
2. Experimental Setup

Figure 1 is the 2D slab burner front and top views showing the solid fuel location, glow plug at the leading edge to ignite the fuel, oxidizer feed pipe, and borosilicate glass windows on the top and sides to provide the optical access. The combustion chamber is made of stainless steel plates at the top and the bottom and provides a square cross section of 2.54 cm. The chamber is 15.24 cm long and is fed by a 2.54 cm inner diameter stainless steel pipe. The feed pipe is 1.83 m long consistent to that in [26] to provide a fully developed flow. The borosilicate glass on the sides and top provide optical access to the combustion chamber. Two Nikon DSLR cameras are positioned at 50 cm from the fuel surface to capture the top view and the front view of the slab burning to obtain two-color pyrometry measurements. The camera radiometric self-calibration is performed as described in [16]. PMMA solid fuel ∼ 1 cm wide, 1 cm high and 7 − 10 cm long with a 45° bevel at the front face to promote flame attachment is used. The oxidizer utilized is a mix of $O_2$ and $N_2$ gas with varying $O_2$ concentrations of $X_{O_2} = 17\%, 21\%, 24\%$, and $28\%$. Both the gas flows are controlled using Brooks Instruments mass flow controller and are adjusted to provide a flow velocity of 15 cm/s at the entrance of the chamber. The corresponding oxidizer mass flux is $G = 0.012 \text{ kg/m}^2 \text{ s}$. Low $G$ is specifically set to promote radiation heat transfer.

![Fuel Slab](image1.png)

**Figure 1**: The 2D slab burner setup (a) front view, (b) top view.

3. Modeling

The aim of this study is to establish a correlation for the flame to fuel RHF measured in slab burner to 1D steady-state diffusion flames. Towards that extent, the modeling aspect of this study focuses on numerical solution of steady-state diffusion flame formed by reacting interfaces between the fuel pyrolyzates and the incoming oxidizer. The PMMA-Air diffusion flames model developed for reacting interfaces similar to that in Ref. [17] is utilized. The numerical model is briefly presented here. Assuming steady-state conditions, the conservation equations for 1D gas phase reacting interfaces reduce to only species and energy conservation given as,

\[
\frac{\partial}{\partial x} \left( \rho \frac{\partial Y_i}{\partial x} \right) + \dot{w}_i = 0 \quad (1a)
\]

\[
\frac{\partial}{\partial x} \left( k \frac{\partial T}{\partial x} + \sum \rho \frac{\partial Y_i}{\partial x} h_i \right) - \frac{\partial}{\partial x} \left( \rho V_T h \right) - \sum \dot{w}_i h'_{f,i} - \kappa \left( 4 \sigma T^4 - G_{irr} \right) = 0 \quad (1b)
\]

where, $\rho$ is the density, $k$ is the thermal conductivity, $T$ is the temperature, $Y_i$ is the mass fraction of the $i^{th}$ species in the mixture, $\dot{w}_i$ is the mass consumption or production rate of the $i^{th}$ species, $h_i$ is the enthalpy of the $i^{th}$ species, $h$ is the total enthalpy, $h'_{f,i}$ is the heat of formation of the $i^{th}$ species, and $V_T$ is the thermophoretic velocity associated with soot thermophoresis. Thermal
equilibrium among the phases is assumed. Diffusivity, $D$, is assumed constant for all the species to simplify Eq. (1b) and unity Lewis number is assumed. The last term in Eq. (1b) is the radiation source term that accounts for both soot and gas radiation (absorption and emission) in which, $\sigma$ is the Stefan-Boltzmann constant, $G_{irr}$ is the irradiation and $\kappa$ is the total absorption coefficient.

A 1D discrete ordinates method (DOM) with an $S_8$ approximation for ordinate directions in a 1D plane parallel slab bounded by two diffusely emitting infinitely long isothermal plates is modeled to obtain the radiation source term [27]. Scattering effects are neglected and the walls are assumed to be black. Additional details are provided in Ref. [17]. A phenomenological two-equation semi-empirical soot formation model developed by Lindstedt group is utilized in this study [18, 28]. The soot reactions and rates from Ref. [17] are employed. The soot formation model is validated by comparing the temperature and soot volume fraction profiles for ethylene-air diffusion flames with previous experimental and numerical studies in Ref. [17] and is not repeated here.

In this study, solution of 1D diffusion flames is obtained by initially separating the fuel boundary (right of the domain) and oxidizer boundary (left of the domain) by a domain length of 1 cm (approximately the boundary layer thickness from experiments) and solving Eq. (1) until the soot volume fraction, $f_v$, reaches steady state. The oxidizer boundary is at 300 K and pressure of 1 atm. The fuel boundary for PMMA, ($C_5O_2H_8$), is more complex and is described in the form of basic hydrocarbon species defined in GRI Mech 3.0 [29] using Canetra’s multi-phase equilibrium description at PMMA decomposition temperature [17, 30].

4. Results

4.1 Experimental Results

Figure 2 shows the front view images of the flame over PMMA solid fuel at four different oxygen concentrations of 17%, 21%, 24%, and 28% in the oxidizer stream. The snapshots are shown at approximately ~ 5 s after ignition when the flame appears to be steady. Flame brightens with increasing the oxygen concentration suggesting increase in temperatures. Additionally, it can be seen that the overall flame size/thickness grows with increasing $O_2$ concentration. The flame to fuel heat feedback increases with increasing $O_2$ concentrations due to increased temperatures further increasing the mass blowing. However, the increased heat feedback promotes more burning of the fuel vapors from the pyrolysis of PMMA and heat transfer dominates the mass blowing effect, increasing the flame boundary height and bringing it closer to the fuel surface.

Figure 3 shows the flame field temperatures obtained using two-color pyrometry approach described in [11, 16]. The flame temperatures range from 1043 – 2287 K across the oxygen concentrations studied showing a high dynamic range of variation. The average temperatures for $O_2$ concentrations of 17%, 21%, 24%, and 28% are found to be 1338 K, 1450 K, 1488 K, and 1562 K, respectively. The maximum temperatures are in the range of 1800 – 2300 K. Field averaged flame temperatures are found to increase with time as the $O_2$ concentration is increased. The 28% case has a longer soot residence time and thus is conducted for a longer time period than the other three cases until the soot layer develops to steady state.

The soot volume fraction profiles corresponding to the flame snapshots are shown in Fig. 4. The soot concentrations are found to be lower inside the flame and gradually increasing outward of the flame. The maximum soot volume fraction is found around the periphery of the flame. The field averaged SVF values are found to be 1.4 ppm, 2.15 ppm, 2.33 ppm, and 3.48 ppm, respectively, for the four oxygen concentrations. The maximum soot volume fractions range from 9 – 40 ppm.
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Figure 2: Boundary layer flame over PMMA fuel for $G = 0.012 \text{ kg/m}^2 \cdot \text{s}$ at different oxygen concentrations of - (a) 17%, (b) 21%, (c) 24%, (d) 28%.

Figure 3: Two-color based pyrometry flame temperature profiles at four different $O_2$ concentrations of - (a) 17%, (b) 21%, (c) 24%, (d) 28%.

Figure 4: Two-color pyrometry based soot volume fraction profiles at four different $O_2$ concentrations of - (a) 17%, (b) 21%, (c) 24%, (d) 28%.
4.2 Numerical Results

Figure 5 shows the peak temperature and SVF for increasing $O_2$ concentration in PMMA-Air 1D diffusion flames. $O_2$ concentration is varied in the range of 15 – 100% for the numerical simulations. Temperatures are found to increase with increasing oxygen while, SVF initially increases up to 45% oxygen and then decreases. Overall the flame thickens with increasing oxygen and the soot layer thickness reduces.

Figure 5: PMMA-Air 1D flames with varying oxygen concentrations - (a) peak temperatures, (b) peak SVF.

Figure 6: Soot formation-oxidation process leading reaction rates - (a) surface growth rates, (b) soot oxidation by $O_2$.

To better understand the soot behavior, soot reaction rates are investigated. Fig. 6 shows the surface growth and soot oxidation by $O_2$, the leading reaction rates of soot formation and oxidation processes. The soot formation processes - nucleation and surface growth are always near the fuel domain while the soot oxidation processes are always near the oxidizer domain of the 1D flames. Additionally, the nucleation and surface growth rates are active in the temperature range of 1500 – 2100 K while the soot oxidation is triggered only above 2100 K. Since above 45% oxygen, the peak flame temperatures are very high, soot oxidation rates increase and flame thickens. The oxidation rates move from the oxidizer side to the fuel side of the domain and thus reduce the overall effect of surface growth. This reduces the overall soot layer thickness further decreasing the peak SVF.
4.3 Radiative Heat Flux

Figure 7 shows the flame to fuel surface field RHF obtained from experiments using two-color pyrometry based non-intrusive diagnostic \[11, 16\] corresponding to the four flame snapshots in Fig. 2. As the oxygen concentration increases from 17% to 28%, the incident RHF from the flame to fuel surface increases. The peak RHF ranges from $15 - 220 \text{ kW/m}^2$. The RHF accounts for both gas radiation and soot radiation. Gas radiation is obtained by using the empirical emissivity for $CO_2$ and $H_2O$ using stoichiometric mole fractions \[16\]. Since the temperatures and the SVF increase with increasing oxygen concentration in the low regime, the RHF increases. The field averaged RHF for the four cases are 8, 18, 28, and 38 kW/m$^2$.

![Figure 7: Two-color pyrometry based flame to fuel radiative heat flux for varying $O_2$ concentrations of - (a) 17%, (b) 21%, (c) 24%, and (d) 28%.](image)

Figure 8a shows the RHF obtained in 1D diffusion flames for the range of $O_2$ concentrations studied. The RHF increases with increasing $O_2$ concentration and is maximum at 100% oxygen. A wide range of heat flux is obtained from as low as 8 kW/m$^2$ for 15% $O_2$ to about $≈ 1.5 \text{ MW/m}^2$ with pure oxygen. The results are also compared with the field averaged non-intrusive RHF measurements from Fig. 7 (blue squares). The results obtained by Tewarson et al. (red circles) \[1, 12\] by extrapolating their experimental data using the Spalding’s B-number definition corrected for radiation are also shown and are found to be in very good agreement with both the non-intrusive measurements and those obtained from 1D steady state flame simulations. The inset plot shows the zoomed in comparison of the RHF for oxygen concentration of 15 – 50% to provide a better comparison between the three data sets. For the $O_2$ concentrations of 17% – 30%, the RHF exhibits a linear relationship consistent to that reported in Ref. \[25\]. The radiant fraction, $\chi_{rad}$, is computed as the ratio of the RHF to the chemical energy from the fuel. The chemical energy from the fuel is obtained by integrating the chemical source energy over the flame domain. The resulting $\chi_{rad}$ is shown in Fig. 8b. The radiant fraction is found to have a similar trend as the RHF. The maximum radiant fraction of 44% is obtained for pure oxygen case. The non-monotonic SVF behavior does not affect the RHF. Both the trends observed for the RHF and the radiant fraction are consistent to those reported in Ref. \[23\] for methane and propane turbulent jet diffusion flames. One of the possible explanations for this is that the flame temperatures consistently increase with increasing $O_2$ concentrations and reach as high as 3100 K. Furthermore, the flame thickens with increasing oxygen concentration essentially increasing the path length of radiation and re-absorption of the radiative intensity. The RHF at dry air (21% $O_2$) is 27 kW/m$^2$ consistent to those measured in PMMA-Air flame spread experiments \[16, 17, 24, 31, 32\].
This result shows that steady-state 1D diffusion flames provide very good estimate of the total radiative heat transfer from the flame to fuel in comparison with those obtained in 2D slab burner and can be utilized in developing fire spread and fuel regression theories of solid fuels. This result is promising as it allows a direct correlation between the RHF from 1D diffusion flames and the fuel surface RHF observed in 2D slab experiments. Future work will explore this correlation for other solid fuels like paraffin wax, hydroxyl-terminated polybutadiene, epoxy, polypropylene, etc.

![Graphs showing radiation in PMMA-Air diffusion flames over varying O₂ concentrations](image)

Figure 8: Radiation in PMMA-Air diffusion flames over varying O₂ concentrations showing - (a) comparison of experimental field average radiative heat flux to 1D diffusion flames and to those from Tewarson et al. [1], (b) radiant fraction from 1D diffusion flames.

5. Conclusions

The influence of oxygen concentration in the oxidizer of a 2D slab burner on the flame to fuel surface radiative heat flux is studied. Radiation heat transfer promotes the regression rate in hybrid rockets and is driven by the amount of soot formed during the combustion. Experimental measurements of flame to fuel radiative heat flux are obtained using a recently developed non-intrusive two-color pyrometry based diagnostic in low oxidizer mass flux PMMA combustion. The oxygen concentrations in experiments are varied from 17% – 28% and the radiative heat flux is found to increase. The role of soot formation and impact on radiation is further analyzed through numerical solutions of 1D steady-state diffusion flames. The simulations are conducted by varying the oxygen concentrations in the range of 15% – 100%. The flame temperatures are found to increase with increasing oxygen. Peak soot volume fractions initially increase up to 45% oxygen concentration and are then found to be dropping. The flame thickens with increasing oxygen and the soot layer is found to reduce in thickness. The soot reaction rates study show that the soot oxidation reactions are initially near the oxidizer side and move towards the fuel side with increasing oxygen thus reducing the peak soot values. This non-monotonic behavior of soot does not affect the radiative heat flux. The radiative heat flux and radiant fraction increases with increasing oxygen concentration primarily because of the increasing temperatures. The field averaged non-intrusive heat flux measurements are found to be in good agreement with the numerical solution of 1D flames and with the values reported in literature. The radiative heat flux obtained from the 1D flames can serve as a baseline estimate in developing fire spread and solid fuel regression theories.
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