

Pressure and Radiation Effects on the Dynamics of Hot and Cool Diffusion Flames

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1 Introduction

Due to the importance of low-temperature chemistry in the ignition of many hydrocarbons, cool flames continue to be studied in great detail. Cool flames are the direct result of a successful first-stage ignition event, and the strength of the cool flame stage is closely correlated to the likelihood of engine knock [1]. The presence of propagating cool flames in a mixture can significantly shorten the ignition delay time between the first and second stages of ignition compared to what would be expected in a homogeneous reactor [2]. Furthermore, at near-limit fuel concentrations, cool flames can possess broader flammability limits than those of classical hot flames [3, 4].

Much of the recent interest in cool flame research can be attributed to the discovery of cool diffusion flames aboard the International Space Station [5]. In this study, large *n*-heptane droplets were observed to undergo radiative flame extinction into quasi-steady cool flames. Subsequent numerical simulations confirmed that the cool diffusion flames surrounding the droplets were supported by low-temperature reactions in the negative temperature coefficient regime [6-8]. Further microgravity experiments have confirmed the existence of quasi-steady cool diffusion flames for other *n*-alkanes [9] and even primary reference fuel mixtures [10]. In ground-based counterflow experiments, it has been shown that steady cool diffusion flames can be established for large *n*-alkanes [11-13] as well as dimethyl ether [14, 15].

Despite the growing number of cool diffusion flames studies performed at atmospheric pressures, very few investigations have taken place at elevated pressures closer to engine-relevant conditions. Farouk et al. examined *n*-heptane droplets at 3 atm which oscillated between a hot flame stage and cool flame stage for multiple cycles [16]. More recently, Deng et al. studied the ignition and extinction of dimethyl ether cool diffusion flames at pressures up to 3 atm [17]. However, the modeling efforts in these studies, while insightful, were primarily restricted to the conditions of the complementary experiments. Much is still unknown about the dynamics of cool diffusion flames at elevated pressures.

The objective of this current study, therefore, is to understand how cool diffusion flames behave under various conditions at elevated pressures. Direct comparisons will be made between atmospheric cool

flames and high-pressure cool flames. The addition of diluents will be shown to have a substantial impact on the dynamics and regimes of near-limit flames at elevated pressures. Finally, the effects of radiation on the behavior of high-pressure cool diffusion flames will be examined.

2 Numerical Modeling

The numerical calculations in this study involve a one-dimensional counterflow flame geometry with a modified arc-length continuation method [18]. The fuel side, located at the origin, has a fixed boundary temperature of 550 K. The oxidizer side, located at a separation distance of 2.25 cm, is set to 300 K. Computations are performed at pressures ranging from 1 atm to 12 atm. For calculations that include radiation, an optically thin radiation model is assumed. The Plank mean absorption coefficients for CH₄, CO₂, H₂O, and CO are computed using a statistical narrow-band model [18].

Dimethyl ether (DME) is chosen as the fuel for this study due to its strong low-temperature chemistry and its relatively compact detailed chemical kinetic models compared to those of similarly reactive *n*-alkanes. The chemical kinetic model chosen for this study is the AramcoMech 1.3 model [19]. It consists of 124 species and 766 reactions and, mostly importantly, contains the reactions relevant to low-temperature DME oxidation. The model's previous competency in predicting DME cool flames can be seen in [15].

3 Results

It is known from previous studies that the influence of pressure on hot flame structure can be essentially neutralized when considering the pressure-weighted strain rate [20]. Figure 1a shows that, for fixed $p \cdot a = 455$ atm/s, the hot diffusion flame profiles collapse onto each other. In other words, the change in thermal and mass diffusivity due to the twelve-fold increase in pressure is offset by the twelve-fold decrease in the strain rate. The differences in the maximum temperature are primarily due to slightly higher reactant leakage at lower pressures near the extinction limit. However, this similarity does not hold nearly as well for cool flames, as seen in Figure 1b. Even though the 3 cool diffusion flames are each at $p \cdot a = 195$ atm/s, there is not a one-to-one relationship between the influence of pressure and the influence of strain rate. In particular, the fuel-side temperature distributions (on the left side of the flame) show particularly substantial deviation as the pressure changes.

A closer look at the detailed flame structure reveals further insight into the differences between the hot

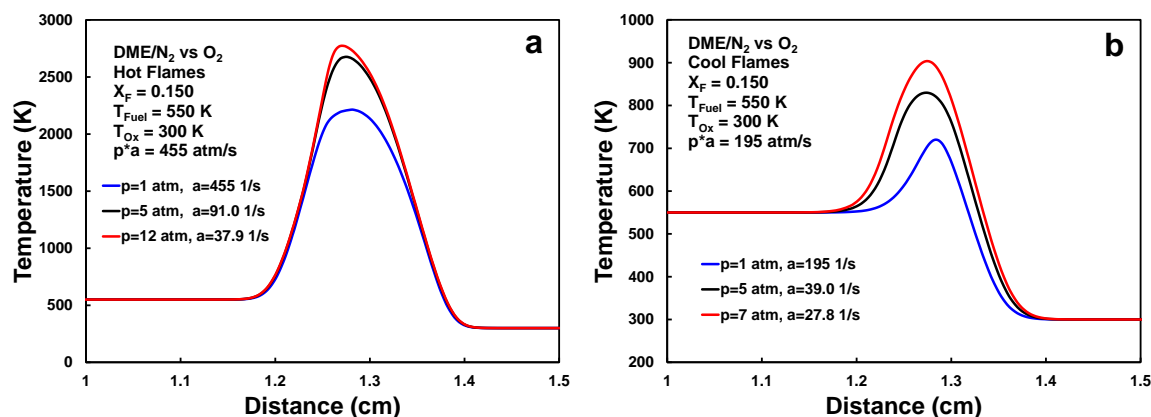


Figure 1. Temperature profiles for dimethyl ether (a) hot diffusion flames and (b) cool diffusion flames at constant pressure-weighted strain rate.

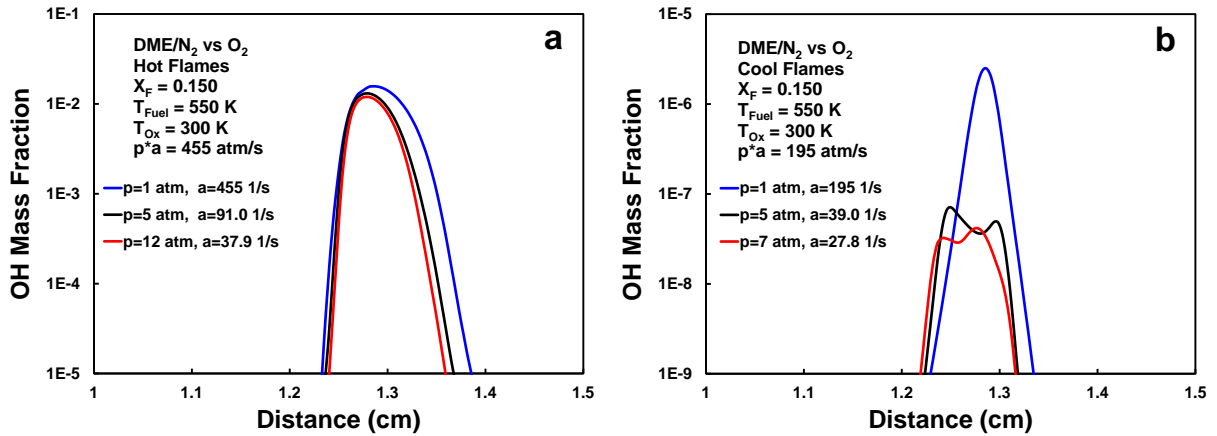


Figure 2. OH radical profiles for dimethyl ether (a) hot diffusion flames and (b) cool diffusion flames at constant pressure-weighted strain rate.

flames and cool flames. The OH radical profiles for the hot flames in Figure 2a again show similar (though not quite identical) structures at constant pressure-weighted strain rate. The magnitudes are very similar, being on the order of a percent mass fraction, and the spatial distributions match decently well, particularly on the fuel side. On the other hand, Figure 2b indicates that the cool flames OH profiles are entirely different when the pressure is increased. Not only is the maximum OH mass fraction an order of magnitude higher for the $p = 1$ atm case, but the elevated pressure cases actually show multiple peaks, indicating a drastically different flame structure. High-pressure conditions, therefore, affect cool diffusion flames quantitatively and qualitatively different than hot diffusion flames.

The flames in Figures 1 and 2 are calculated using pure oxygen for the oxidizer, as is common for many experimental counterflow cool flame setups [11-13, 15]. However, in real engines, air is almost universally used as the oxidizer. The effect of diluting the oxidizer stream from pure oxygen to air can be seen in Figure 3. While there are two stable flame branches in the pure oxygen case (a hot flame branch and a cool flame branch), a third intermediate branch emerges when N₂ dilution is accounted for. In Figure 3, point B lies on the “mild flame” branch, which is distinct from the hot flame branch of point A and the cool flame branch of point C. The existence of a steady “mild flame” may be difficult to demonstrate experimentally since its stability region is narrow, is at relatively low strain rates, and falls between the ignition limit of the cool flame branch and the extinction limit of the hot flame branch [21]. However, it is quite possible that such a “mild flame” may be present in a quasi-steady state form in an unsteady turbulent combustion environment. In any case, it is clear from Figure 3 that a shift to near-limit conditions due to diluent addition causes a drastic change in the shape of the classic S-curve, both in the emergence of the “mild flame” branch and the decrease of the hot flame extinction limit below the cool flame extinction limit.

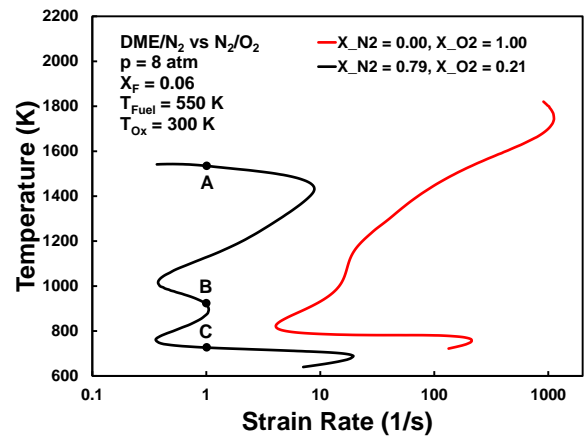


Figure 3. S-curve calculations indicating the effect of N₂ dilution on high-pressure dimethyl ether flames.

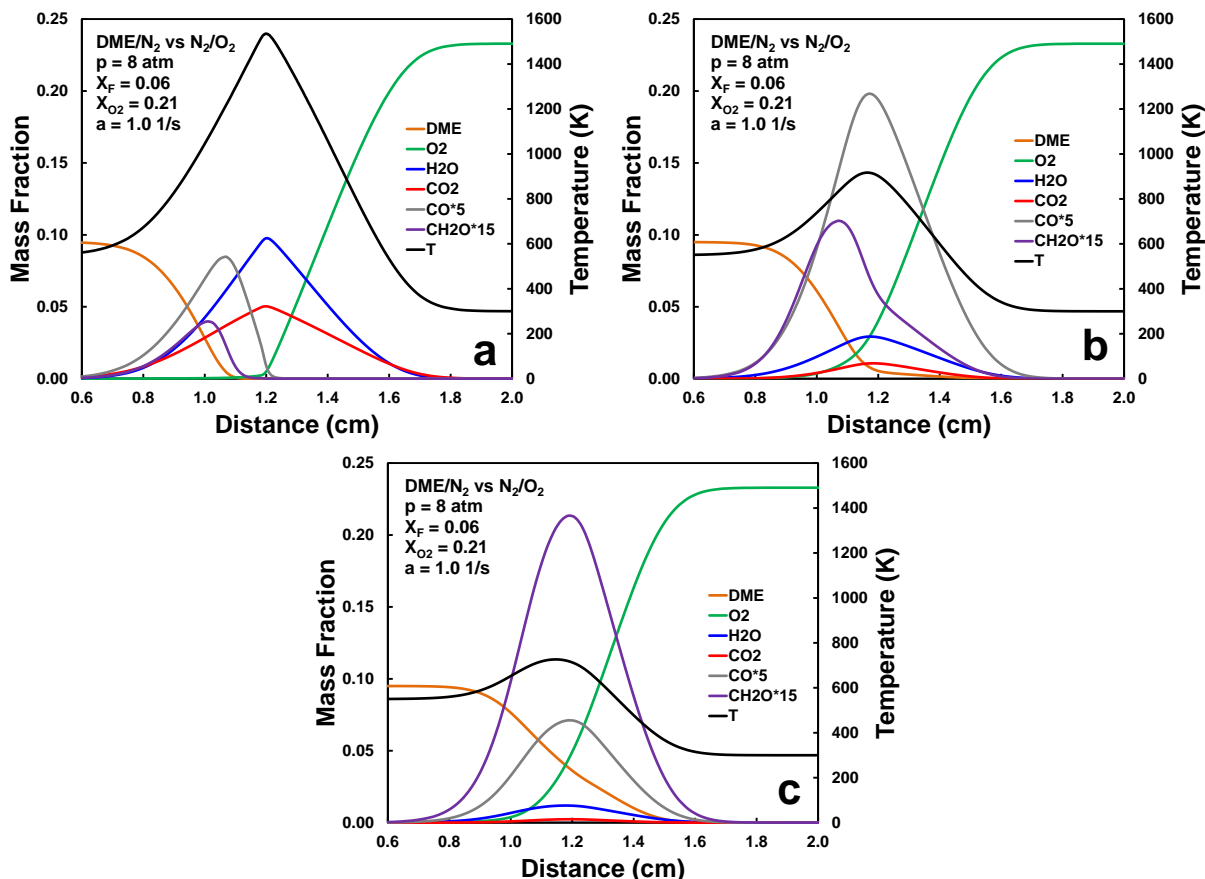


Figure 4. Spatial profiles of the temperature and species distributions for a (a) hot flame, (b) “mild flame,” and (c) cool flame (corresponding to points A, B, and C in Figure 3, respectively) at a pressure of $p = 8$ atm.

The flame structures of the three stable flames obtained with N_2 dilution at a strain rate of $a = 1.0$ 1/s are shown in Figure 4. These solutions correspond to points A, B, and C in Figure 3. The hot flame in Figure 4a exhibits the usual structure, with a maximum temperature of 1535 K, significant H_2O and CO_2 production, and very little leakage of DME or O_2 through the reaction zone. The cool flame in Figure 4c also shows the typical low-temperature indicators—it possesses a flame temperature of 726 K, extreme reactant leakage, and substantially higher CH_2O production than the hot flame. However, the unusual “mild flame” in Figure 4b is unlike either the hot flame or the cool flame. While the hot flame is driven by the oxidation of CO to CO_2 , and the cool flame is governed by the oxidation of DME into CH_2O , the “mild flame” appears to be sustained by the oxidation of CH_2O into CO . This is made possible by OH production from H_2O_2 decomposition, which replaces the degenerate low-temperature chain-branching sequence that forms OH in the cool flame. Moreover, the temperature (917 K) is not high enough for high-temperature reactions such as $H + O_2$ to become dominant. Therefore, the “mild flame” is sustained by an intermediate-temperature chemistry sequence distinct from that of either the hot flame or cool flame.

Since heat loss from thermal radiation is known to be an important factor at near-limit conditions [18], the effect of radiation on the dynamics of DME diffusion flames at elevated pressure is depicted in Figure 5. While the addition of radiation at constant $X_{O_2} = 0.30$ has only a small effect on the hot flame extinction limit and the cool flame extinction limit due to the high strain rates involved, the “mild flame” and low

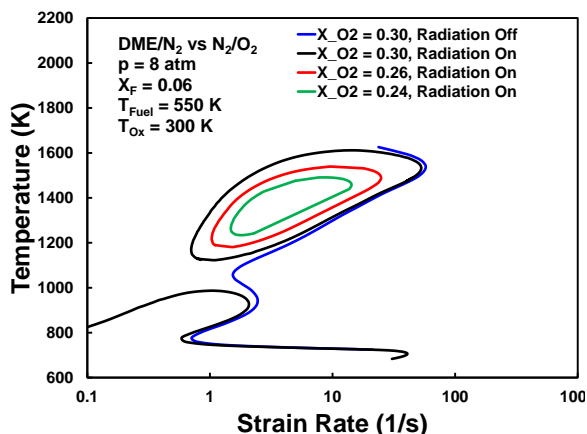


Figure 5. Effect of radiation on the flame dynamics of DME diffusion flames at a pressure of $p = 8$ atm.

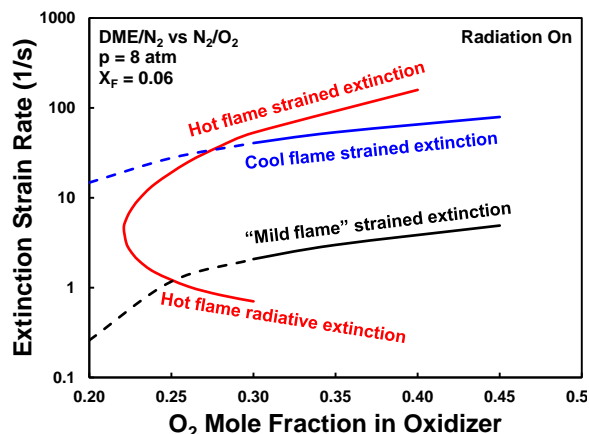


Figure 6. Schematic of radiative DME diffusion flame extinction modes at a pressure of $p = 8$ atm. Dashed lines indicate the assumed trend.

strain-rate hot flame regions are substantially affected. A new radiative extinction limit for the hot flame appears—that is, there is now a minimum strain rate at which the hot flame can be sustained before radiation losses overwhelm the chemical heat release. As a result, the stability of the “mild flame” is greatly increased. In fact, the only flames that can exist below a strain rate of $a = 0.5$ 1/s at $X_{O_2} = 0.30$ are these “mild flames.” It appears, therefore, that high-pressure DME hot flames do not radiatively extinguish into cool flames, as was seen for the *n*-heptane flames in [5], but into “mild flames.” This is likely due to the strong intermediate chemistry of DME, as DME can decompose directly into CH_2O in a manner that *n*-alkanes cannot.

As the dilution is increased further, Figure 6 shows that radiation losses eliminate the hot flame entirely, as the radiative extinction limit eventually intersects the normal strained extinction limit. For oxygen mole fractions less than $X_{O_2} = 0.22$, therefore, only cool flames and “mild flames” are stable. It is presumed (though not clarified by calculations) that the “mild flame” would have its own radiative extinction limit at lower O_2 mole fractions. Beyond an even lower O_2 mole fraction, only cool flames would be stable.

4 Conclusion

The effects of elevated pressure and radiation on dimethyl ether diffusion flames have been examined numerically. It is seen that the similarities in hot flames at constant pressure-weighted strain rate do not extend to cool flames due to changes in the nature of low-temperature chemistry across different pressures. At elevated pressures, N_2 diluent addition to the oxidizer is shown to shift the extinction limits of the hot flame dramatically and enables the formation of a steady “mild flame” branch sustained by intermediate-temperature chemistry. Computing these high-pressure diffusion flames with radiative losses included further increases the stability of the “mild flame” branch due to the presence of a hot flame radiative extinction limit.

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