Hot-gas ignition of non-premixed methane flames in the presence of inert particles

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Abstract

A detailed numerical study was conducted on the ignition of non-premixed atmospheric CH_4/air flames containing inert Al_2O_3 particles in counterflow configurations. The gas injection temperatures of the fuel-side, the air-side, or both sides were increased separately until ignition was observed. The coupled conservation equations were solved for both phases along the stagnation streamline, with detailed descriptions of gas-phase chemical kinetics, molecular transport, and radiative heat transfer. The reactant injection temperature, strain rate, orientation of particle seeding, and injection particle number density and temperature were varied. Results showed that particles could drastically modify the temperature field of the gas phase and with it the ignition temperature. It was found that when the particle number densities are relatively high, the velocity field is also affected due to the momentum exchange between the two phases. Thus, the local strain rates and ignition temperatures are modified. No table differences were observed among the three configurations considered, namely heated air-side, heated fuel-side, and simultaneously heated air- and fuel-sides due to the different temperature fields generated. The controlling chemical pathways were found to depend not only on whether particles are present or not but also on whether the particles were injected from the air- or fuel-side. When particles are injected from the hot air-side rather than the cold fuel-side, lower ignition temperatures were observed. It was also determined that ignition occurs more readily in air-side seeding compared to fuel-side seeding as the particle number density increases. The effect of the particle injection temperature on hot-gas ignition was also assessed. It was shown that the ignition process is controlled by the competition between heating and cooling of the gas by the particles in cases where only one reactant stream is heated and that the conditions under which the particles assist or retard ignition cannot be readily determined without performing a detailed analysis of the problem.

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1. Introduction

Combustible mixtures can ignite when brought into contact with any heat source of sufficient magnitude. The resulting ignition may be desirable or it may present a safety hazard. In a typical premixed ignition scenario, hot air (or some other gas) is brought into contact with a gaseous fuel/oxidizer mixture. For non-premixed flames, ignition occurs upon contact of the two reactant streams with one or both of them being heated to high enough temperature (e.g., [1,2]).
However, if particles are present, the ignition conditions may be notably modified. In fact, if the particles are large and hot enough they can ignite cold mixtures by themselves [3]. In previous work [4–6], the authors have focused on the opposite problem of flame extinction in dusty flow counterflow configurations, both experimentally and numerically. More recently, this work was extended to the problem of ignition by hot inert particles, comparing and contrasting it with pure hot-gas ignition [2]. However, the effects on hot-gas ignition of particles that are cold or heated (but are not hot enough to cause ignition on their own) have not yet been investigated.

The general understanding of reacting dusty flows has great importance considering the common applications of such flows at various fields, from propulsion (e.g., Al, B, and Mg) to power generation (coal), to hazards such as explosions of grain silos and sawdust storerooms in lumber mills.

The presence of particles can lead to complex phenomena in reactive flows. The dynamic behavior of both gas and particle phases can be affected by the Stokes drag, phoretic, and gravitational forces that are exerted on the particles. Likewise, significant temperature differences that may develop between the two phases due to the large thermal inertia of the particles can substantially affect the thermal behavior of both phases. For the case of reacting particles, many chemical interactions between the two phases are further complicated by the interactions between gas-phase and surface reactions.

Past studies on reacting dusty flows were conducted in rather complex configurations such as, for example, fluidized beds and tubes, and from these studies, global behaviors were quantified. However, the numerous interactions among the various processes in such systems can mask the fundamental physics. But, a detailed understanding of the dynamics and structure of such flows can be best achieved by using the counterflow configuration in which the physico-chemical processes in both phases can be systematically assessed (e.g., [7–11]).

Thus, a numerical study was carried out to investigate the effects of chemically inert particles on hot-gas ignition of nitrogen-diluted non-premixed methane/air flames in counterflow configurations. The current study is distinct from the authors' previous work [2] in which ignition was achieved by increasing the particle temperature; in other words, the ignition source was the particles themselves. In this study, ignition is achieved by increasing the temperature of the gas phase in the presence of particles, and the particles are not hot enough to serve as the ignition source. The effects of reactant temperature, strain rate, orientation of particle seeding, and injection particle temperature and number density were considered.

The results were used to obtain physical insight into the mechanisms that control the ignition response of the gas phase.

2. Numerical approach

A set of equations, valid for both phases along the stagnation streamline of a counterflow established between two opposed burners, has been developed by the authors [12]. The burners are separated by a distance $L$. Figure 1 illustrates the schematics of all configurations considered, and it should be noted that in all cases, the particles are injected from the bottom burner. The momenta of both jets were kept constant throughout the calculations to keep the gas stagnation plane (GSP) at the center of the domain. In all figures that follow, the bottom and top burners correspond to the left and right of the spatial domain, respectively.

The code includes a quasi-one-dimensional set of equations for the gas phase similar to the one in [13], except that terms are included to account for the dynamic and thermal interactions between the phases. The equations for the particle phase were formulated for a single inert particle, as the number densities are assumed to be small enough so that particles are unlikely to interact with one another. The equations contain all known and pertinent dynamic and thermal interactions between the two phases and include a conservation equation for the evolution of the particle number density. The details are given in [12]. The kinetics of the gas phase was described by the GRI 3.0 mechanism [14]. The code was integrated with the CHEMKIN [15] and Transport [16] subroutine libraries.

In all simulations, a 1:3 (mole-ratio) $\text{CH}_4 + \text{N}_2$ mixture was injected against an air jet either from the bottom or the top burner. The injection veloc-

![Fig. 1. Schematics of the counterflow configurations considered in the simulations.](image-url)
ity of both jets was $u_{g, inj} = 100$ cm/s. Al$_2$O$_3$ particles with 60 μm diameter were seeded from the bottom burner with an injection velocity of $u_{p, inj} = 400$ cm/s to ensure that they pass through the ignition zone and reach the top burner, thus passing out of the system. (Smaller particles injected at lower velocities may repeatedly reverse direction each time they encounter a counter-flowing stream and pass through the ignition zone many times. These flow reversals further complicate the problem (e.g., [5]) and will be considered in a future investigation). The particles were injected with temperatures of $T_{p, inj} = 300, 700, \text{and } 1100$ K. The injection particle number density, $n_{p, inj}$, was also varied from virtually particle-free cases ($10$ part/cm$^3$) to heavily loaded cases ($9000$ part/cm$^3$) in various calculations. Ignition was achieved by increasing the gas injection temperature, $T_{g, inj}$, of either the fuel-side, or the air-side, or both sides simultaneously. The $T_{g, inj}$ of the hot stream/streams at the ignition state was taken as the ignition temperature, $T_{ign}$, i.e., $T_{ign} = T_{g, inj}$.

The ignition response shows a turning-point behavior as $T_{ign}$ increases (e.g. [1,2]). This singular behavior was captured by introducing a one-point continuation [17,18] in the original code [12]. In effect, this technique reverses the dependent and independent variables of the problem in which configuration, the “S-curve” is single-valued.

3. Results and discussion

Figure 2 depicts the variation of the ignition temperature, $T_{ign}$, with the injection particle number density, $n_{p, inj}$, for configurations of Figs. 1A–C. The particle loading was varied from almost particle-free (10 part/cm$^3$) to heavily loading (1000 part/cm$^3$) conditions. In all configurations, particles were injected at $T_{p, inj} = 300$ K.

The addition of cold particles into the flowfield significantly affects the ignition temperature for all configurations, regardless of whether the particles are injected from the hot or cold jet. For example, the addition of particles with $n_{p, inj} = 1000$ part/cm$^3$ leads to an increase in $T_{ign}$ by as much as 600 K. It is also apparent that much higher $T_{ign}$’s are required to attain ignition for configuration (c) (i.e., when the air is injected cold), compared to configurations (a) and (b).

Reaction path analysis was performed for states (3), (4), and (5) shown in Fig. 2. These are the ignition states for configurations (a), (b), and (c) respectively, for $n_{p, inj} = 1000$ part/cm$^3$. First, it was surprisingly observed that the consumption of CH$_4$ mainly occurs by reactions with H rather than by reactions with OH, which is by far the most common case for both premixed and non-premixed particle-free ignition (as well as for hot-particle ignition [2]). This is mainly due to the very different temperature fields that are developed in the presence of the particles. For configuration (b), for which the temperatures of both sides are simultaneously increased to induce ignition, the cold (i.e., $T_{p, inj} = 300$ K) particles cool the gas resulting in a “well”-shaped temperature profile shown in Fig. 3. Thus, the temperature around the gas stagnation plane, GSP, is reduced, compared to the injection values, to about 900 K. Under these conditions, this cooling around the GSP results in the displacement of the ignition kernel towards the higher temperatures on the fuel side, and thus, the ignition process is limited by the diffusion of O-containing species. As H is abundant close to the hot fuel-side nozzle ($\sim 1700$ K), CH$_4$ is consumed via H + CH$_4$ → CH$_3$ + H$_2$ (R52 in GRI 3.0) rather than the more common OH + CH$_4$ → CH$_3$ + H$_2$O (R97).

It is apparent that for all configurations in which the gas around the GSP is cooled by the

![Fig. 2. Variation of ignition temperature, $T_{ign}$, with particle number density, $n_{p, inj}$, for configurations (a), (b), and (c).](image)

![Fig. 3. Spatial variations of gas phase temperature, $T_g$, for configurations (a) and (b), for $n_{p, inj} = 10$ and 1000 part/cm$^3$.](image)
particles, the radicals generated in the hot zones close to the nozzles need to survive while they diffuse “long” distances through “colder” gases to encounter species that are transported by the opposing jet. This limits the rates of the reactions that typically control the radical production in particle-free hot-gas ignition processes, in which there is no mechanism for cooling the gas around the GSP and the radicals do not have to diffuse “long” distances to effectively react.

For all three states (3), (4), and (5), H radicals are mainly produced by:

\[ \text{CH}_4(\pm M) \rightarrow \text{H} + \text{CH}_3(\pm M) \quad (R51) \]

\[ \text{C}_2\text{H}_4(\pm M) \rightarrow \text{H} + \text{C}_2\text{H}_4(\pm M) \quad (R73) \]

\[ \text{CH}_3 + \text{CH}_3 \rightarrow \text{H} + \text{C}_2\text{H}_5 \quad (R157) \]

For these states, more than 99% of the carbon-containing species’ reaction chains pass through R73. As C2H5 is produced in reaction R157, which is also one of the main H producers, C2H5 stands out as a central species in the ignition process.

It was also found that the main branchiong reaction \( \text{H} + \text{O}_2 \rightarrow \text{O} + \text{OH} \) (R37) is not contributing significantly to OH radical production. In fact, in none of the states (3), (4), and (5), does R37 account for more than 25% of the total OH generation (instead of the 80–90% that is typical for a vigorously burning flame).

The reason why the \( T_{\text{ign}} \)’s are noticeably higher for configuration (c) (hot fuel/cold air) is that for that case, the ignition kernel will reside in a zone of high temperature, deep within the fuel side of the GSP. For ignition to occur, \( \text{O}_2 \) must diffuse upstream into the kernel. Hence, the relatively low diffusivity of \( \text{O}_2 \) hinders the ignition process.

In Fig. 2, the dependence of \( T_{\text{ign}} \) on \( n_{\text{p, inj}} \) for configurations (a) and (b) seems to be parallel at first glance, but actually the slope is significantly milder for configuration (b); note that the difference between ignition temperatures for \( n_{\text{p, inj}} = 10 \text{ part/cm}^3 \) (between states (1) and (2)) is double that for \( n_{\text{p, inj}} = 1000 \text{ part/cm}^3 \) (between states (3) and (4)).

Figure 3 depicts the gas phase temperature profiles for states (1) through (4). Comparing the profiles for \( n_{\text{p, inj}} = 10 \) and 1000 part/cm³, it is seen that for configuration (b) where both jets are injected at the same temperature, increasing \( n_{\text{p, inj}} \) cools the gas phase so that the temperatures around the GSP are significantly reduced.

Further examination of Fig. 4 reveals that as \( n_{\text{p, inj}} \) increases, the ignition kernel shifts towards the top burner, i.e., towards higher temperatures and lower strain rates, for both (a) and (b), but more so for (b). Thus, at higher \( n_{\text{p, inj}} \)’s ignition is more readily obtained for configuration (b) than for configuration (a). Because of the particle cooling of the gas phase in configuration (a), \( n_{\text{p, inj}} = 1000 \text{ part/cm}^3 \), it is easier to ignite at locations where the strain rates are lower. In contrast, for the \( n_{\text{p, inj}} = 10 \text{ part/cm}^3 \) situations (where the particles have minimal effect on the gas temperatures), both (a) and (b) ignite at similar strain rates.

It was also found that as the number density increases further (i.e., \( n_{\text{p, inj}} > 2000 \text{ part/cm}^3 \)), the GSP shifts significantly towards the top burner due to the excessive drag exerted by the particles on the gas, which likewise displaces the ignition kernel towards the top burner.

The reaction pathways of particle-free and seeded \( (n_{\text{p, inj}} = 1000 \text{ part/cm}^3) \) cases were also compared for a \( T_{g, \text{ign}} = 1218 \text{ K} \) in configuration (b). This is the ignition temperature of the particle-free case, but, due to particle cooling, is lower than the \( T_{g, \text{ign}} = 1700 \text{ K} \) ignition temperature for the seeded case.

As mentioned earlier, particle-free and seeded cases differ as CH₄ reacts with OH to form CH₃, in particle-free cases as opposed to reacting with H in seeded flows. For the particle-free case with
$T_{g,\text{inj}} = 1218$ K, CH$_3$ is converted mainly to C$_2$H$_6$, C$_2$H$_5$, CH$_2$O, and CH$_3$O, and the temperatures around the GSP are high enough to keep the CH$_4 \rightarrow$ C$_2$ path $[\text{CH}_4 \rightarrow \text{CH}_3 \rightarrow (\text{C}_2\text{H}_6, \text{C}_2\text{H}_5) \rightarrow \text{C}_2\text{H}_4 \rightarrow \text{C}_2\text{H}_3 \rightarrow \text{C}_2\text{H}_2]$ active. In the seeded case, however, temperatures are notably reduced around the GSP, and the CH$_4 \rightarrow$ C$_2$ path is not activated; at lower temperatures, the CH$_4$ conversion slows down, thus affecting the CH$_3$ pool. Subsequently, CH$_3$ reacts to mainly produce CH$_2$O following a rather simpler kinetic path at this temperature (1218 K) as opposed to the ignition state (i.e., $T_{g,\text{inj}} = 1700$ K) that was considered earlier.

The H radical production occurs through very different reactions in the two cases. For the particle-free case, H is produced by R56, CH$_3$O (+M) $\rightarrow$ H + CH$_2$O (+M) and R10, CH$_3$ + O $\rightarrow$ H + CH$_2$O. In the seeded case, 100% of H is produced by R51, CH$_4$ (+M) $\rightarrow$ H + CH$_3$ (+M), 99.9% of OH is produced from R154, CH$_3$ + O$_2$ $\rightarrow$ OH + CH$_2$O, and the contribution of the main chain branching reaction is negligible to the OH production.

Figure 5 depicts the ignition temperatures obtained by injecting hot air against a cold fuel mixture. Two cases were studied, namely configuration (a) where particles are supplied from the (cold) fuel-side, and configuration (f) where they are injected from the (hot) air-side. All results shown in Fig. 5 were obtained by injecting particles with $T_{p,\text{inj}} = 700$ K (compared to the $T_{p,\text{inj}} = 300$ K used previously), to also assess the effect of particle injection temperature. It can be seen that ignition occurs at lower temperatures when particles are injected from the hot air-side rather than the cold fuel-side. This trend becomes more pronounced as $n_{p,\text{inj}}$ increases.

To provide insight into the results shown in Fig. 5, the H radical mass fraction profiles are shown in Fig. 6 for $n_{p,\text{inj}} = 800$ part/cm$^3$ and an air-side gas injection temperature of $T_{g,\text{inj}} = 1575$ K; note from Fig. 5 that this is the ignition temperature for configuration (f) but not for configuration (a). This choice was intentionally made so that comparison can be made for the same conditions. The results of Fig. 6 indicate that the ignition kernels develop on the air-side for both configurations (recall that the GSP is located near $x = 0$). However, at the ignition temperature for configuration (f), the magnitude of the H radical pool for (a) is two orders lower compared to (f).

To understand why this occurs, the gas and particle phase temperature distributions are plotted in Fig. 7. It is observed that for configuration (f), where particles are injected with the hot air, the air raises the particle temperature to 1000 K in the vicinity of the ignition kernel (located at about $x = -0.18$ cm). In contrast, in configuration (a), the cold fuel/inert mixture reduces the particle temperature to 650 K at the ignition kernel (located at about $x = 0.17$ cm). Thus, while the temperature difference between the gas and particle phases at the ignition kernel for configuration (f) is only about 250 K, it is more than 650 K for configuration (a), leading to a more extensive cooling of the gas phase. As a result, the local gas phase temperature within the ignition kernel for configuration (a) drops to a value of about 60 K lower than the corresponding temperature in configuration (f). The strong Arrhenius dependence of the reaction rates on temperature explains the observed difference between the radical pools.

Figure 8 depicts the gas and particle phase temperatures for configuration (d) where heated particles are injected with cold air against a hot fuel/inert-gas mixture entering at $T_{g,\text{inj}} = 1950$ K. The particles were injected with $n_{p,\text{inj}} = 1000$ part/cm$^3$ and at three different temperatures,
It was observed that when the particle injection temperature $T_{p, \text{inj}}$ is increased the air temperature notably increases as a result of the heat exchange between the two phases. Thus, one may predict that increasing the $n_{p, \text{inj}}$ of the heated particles (although not hot enough to cause ignition by themselves) can facilitate ignition under conditions for which ignition would not be possible in particle-free flows. The extra heating of the air-stream results in the displacement of the ignition kernel closer to the air-side, enhancing the OH radical generation. Although the above argument appears to be physically sound at first, one should also notice from Fig. 8 that when the particles cross the GSP and eventually enter the (hot) fuel-side, they actually cool the gas phase because, locally, their temperature is lower than the gas. Thus, the H radical generation, which occurs on the fuel-side and is vital for the OH generation on the air-side, is adversely affected by the increase in particle number density. As a result cooling of the fuel-side, the ignition kernel is displaced closer to the fuel nozzle, which could counteract the effect of heating the air-side. In fact of the results of Fig. 9, which depicts the effect of $n_{p, \text{inj}}$ for 1100 K particles, demonstrate that increasing the $n_{p, \text{inj}}$ from 1000 to 9000 part/cm$^3$ leads to an increase in the air-side temperature of about 600 K (at about $x = 0.1$ cm). However, this occurs at the expense of the fuel-side temperature, which drops by 500 K (at about $x = 0.4$ cm). Thus, it is not possible to ignite the gas, despite the increased number of heated particles, and the effect of addition of heated particles into the gas phase on the ignition process cannot be assessed a priori.

4. Concluding remarks

A numerical study was carried out to determine the effects of inert particles on hot-gas ignition of non-premixed methane/air flames diluted in various counterflow configurations. The effects of reactant temperature, strain rate, orientation of particle seeding, and injection particle number density and temperature were investigated.

The results revealed that the presence of particles significantly alters the temperature field and modifies the ignition temperature. It was also found that for high particle number densities, the velocity field is significantly modified due to the momentum exchange between the two phases. In turn, the strain rates are modified locally, significantly altering the ignition temperatures. Also apparent differences were observed in the ignition process.
response of the three configurations for which the hot gas is injected from the fuel-side, the air-side, or both sides, because of the differences in the resulting temperature fields. Chemical pathways were found to be notably different not only compared to particle-free cases but also to the other seeded configurations studied.

The differences between particle injection from the air- and the fuel-side were investigated. It was determined that when particles are injected from the hot air-side rather than the cold fuel-side, ignition was achieved at lower temperatures. It was also observed that for the larger particle number densities, ignition happens more readily in air-side seeding compared to fuel-side seeding. The effects of particle injection temperature on hot-gas ignition were also studied. It was shown that particles locally heat and cool the gas at different locations in the flowfield. Thus, the ignition behavior is determined by the competition between the favorable effect of particles on cold side of the domain in which they add heat and their adverse effect on the heated side of the domain from which they remove heat. As a result, the effect of heated particles on hot-gas ignition cannot be determined without performing a detailed analysis.

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References


Comment

Mitchell Smooke, Yale University, USA. I noticed that the particles cause the gas phase temperature to increase in the domain with the result that the flame no longer is adiabatic at the boundaries. Given this observation, what happens to the axial velocity near the boundaries assuming that the similarity variable is set to zero at the jet exits?

Reply. The problem formulation, originally taken from Ref. 1, is not restricted to adiabatic boundaries. Assuming zero radial gradient of the radial velocity v at the nozzle exit, i.e. \( \frac{\partial v}{\partial r} = 0 \) where \( G = \frac{\partial v}{\partial r} = v/r \), will not necessarily imply zero axial gradient of the axial velocity u. This is because in the mass conservation equation the axial velocity response is controlled by the gradient of the mass flux \( \rho u \), as shown below for a steady state problem: \( 2\rho G + \frac{\partial (\rho u)}{\partial x} = 0 \)

Typically, in most problems the boundaries are adiabatic and a zero radial gradient of v corresponds to a zero axial gradient of u. However, this should not be generalized and it is not the case in our problem, as the axial velocity gradient at the nozzle exit is controlled by the gradient of the gas density in order to conserve mass. Additionally, we would like to comment that assuming non-zero values of G at the boundaries, the effect on the derived physics of ignition would not be of the first order.

Reference