



Effects of Particle Size on the Ignition of Static CH₄/Air and H₂/Air Mixtures by Hot Particles

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ABSTRACT

Understanding and characterizing the non-homogenous ignition of flammable mixtures by hot particles are important for industry safety. In this study, one-dimensional transient simulations considering detailed chemistry and transport are conducted for the ignition of static methane/air and hydrogen/air mixtures by hot particles with different radii. The objective is to assess the effects of particle size on the ignition delay time for the hot particle-induced ignition process. Unlike the nearly homogenous ignition in shock tubes or rapid compression machines, the non-homogeneous ignition caused by a hot particle depends on not only chemical kinetics but also transport of reactive species. It is found that 1D hot particle ignition is much slower than the 0D homogenous ignition (in which the mixture temperature is the same as the fixed particle temperature) since there is radical loss near the particle surface caused by mass diffusion. The ignition delay time for hot particle ignition is found to strongly depend on the particle radius and temperature. This is interpreted through the change of radical loss with particle radius and temperature. Moreover, to characterize the hot particle ignition, we introduce a Damköhler number which is the ratio between two characteristic times for chemical reaction and mass diffusion. The change of the normalized ignition delay time for 1D hot particle with the Damköhler number is discussed; and a linear relationship is observed for certain conditions.

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Introduction

The hot particles, such as mechanical sparks generated in manufacturing and mining operations, are potential ignition sources for combustible mixtures. Understanding and characterizing the hot particle-induced ignition process and the corresponding critical ignition conditions are important for industry safety. Therefore, in the literature, there are many studies on the ignition of flammable mixtures by hot particles.

Silver (1937) first measured the minimum ignition temperature for spherical hot particles injected into an explosive environment. The minimum ignition temperature was found to decrease with particle diameter and be insensitive to the particle material. Paterson (1939, 1940) measured the minimum ignition temperature at relatively low velocity of the flying particle. Law (1978) derived an explicit ignition criterion using the matched asymptotic analysis with large activation energy and quasi-steady-state assumptions. Beyer and Markus

(2012) investigated the ignition of static flammable mixtures by a stationary hot particle. They found that the minimum ignition temperature weakly depends on the equivalence ratio but strongly depends on the fuel. Recently, the minimum ignition temperature of laser-heated, fixed, small particles with different materials and sizes has been investigated by Roth et al. (2014) and Häber et al. (2017). Roth, Häber, and Bockhorn (2017) proposed an empirical correlation between the minimum ignition temperature and the laminar flame speed. On the other hand, Shepherd and coworkers (Coronel 2016; Coronel et al. 2018a, 2018b; Melguizo-Gavilanes et al. 2017a, 2017b) conducted extensive experimental and numerical studies on the ignition process induced by a hot particle falling into a flammable mixture. They found that the boundary layer development and flow separation are crucial in the ignition process. More recently, Zirwes et al. (2019) have conducted 2D and 3D numerical simulations to investigate the influence of a broad range of relative velocity between the hot particle and the surrounding mixture. They demonstrated that the flow velocity plays an important role in determining the position where the ignition kernel is first formed.

Though there are many studies on the minimum particle temperature required for the successful ignition of flammable mixtures by hot particles, there is little work on the ignition delay time required to achieve thermal runaway. This is partly due to the difficulties in maintaining constant particle surface temperature in experiments. Nevertheless, there are several studies reporting the ignition delay time for the ignition caused by a hot wire/rod/particle with constant surface temperature. Ashman and Büchler (1961) heated a wire to a prescribed temperature through a condenser discharge with a feedback control circuit; and they found that the decrease in wire diameter, wire temperature or gas pressure increases the ignition delay time. Nagata et al. (1994) measured the ignition delay time with different wire surface temperatures and studied the effect of buoyancy. Su, Homan, and Sirignano (1979) conducted 1D transient simulations for the ignition of a combustible mixture by a hot particle and assessed the effects of particle size, initial temperature and equivalence ratio on the ignition delay time. Apart from experimental and numerical studies, Law (1979) and Laurendeau (1982) both derived an explicit correlation for the ignition delay time in a transient ignition process. However, the change of the ignition delay time with the hot wire/rod/particle diameter is still not well understood.

Based on the above discussion, this study aims to assess the particle size effects on the ignition delay time for the transient ignition of a quiescent flammable mixture by a hot spherical particle. Usually, the ignition delay time is measured for a nearly homogenous system using shock tubes or rapid compression machines, in which the ignition is controlled by chemical kinetics. However, the ignition caused by a hot particle depends on not only chemical kinetics but also transport of reactive species. Here we aim to interpret the roles of chemical reaction and mass diffusion during the hot particle-induced ignition process by introducing two corresponding characteristic time scales. One-dimensional simulations for the transient ignition of methane/air and hydrogen/air mixtures by hot particles are conducted; and detailed chemistry and transport are both considered.

Numerical model and methods

The transient ignition process induced by a hot spherical particle at the center of a 1D closed spherical chamber is investigated in this study. The hot particle is characterized by its radius

R and fixed surface temperature T_p . The mixture is initially static at atmospheric pressure (i.e., $P = 1$ atm) and room temperature ($T_u = 300$ K). A large computation domain of $R \leq r \leq 50$ cm is used so that the pressure is nearly constant during the whole ignition process. On the particle surface of $r = R$, an isothermal boundary condition as well as zero flow speed and mass diffusion speed is used. At the outer boundary of $r = 50$ cm, adiabatic and non-reflective boundary conditions are enforced. The boundary conditions are specified as follows:

$$r = R : T = T_p, u = 0, V_k = 0 \quad (1)$$

$$r = 50\text{cm} : \partial T / \partial r = \partial u / \partial r = \partial Y_k / \partial r = 0 \quad (2)$$

where T is the temperature, u is the flow speed, V_k and Y_k are the mass diffusion speed and mass fraction of the k^{th} species. Here the surface reactions are not considered. In their simulations, Häber et al. (2017) found the effect of surface reactions on ignition temperature was on the same order of the experimental uncertainty. They concluded that a reliable quantitative analysis of surface reactions was still impossible. Therefore, similar to previous numerical studies on ignition induced by the hot particles (Beyer and Markus 2012; Coronel et al. 2018b; Melguizo-Gavilanes et al. 2017a, 2017b), here we neglect surface reactions in simulations. This is a shortage of our study, and the effects of surface reactions on particle ignition deserve future studies. The detailed chemical mechanism of Li et al. (2004) is used for hydrogen oxidation and GRI-Mech 3.0 (Gregory et al. 2018) for methane oxidation.

The 1D transient ignition process is simulated using the in-house code A-SURF (Adaptive Simulation of Unsteady Reactive Flow) (Chen, Burke, Ju 2009). A-SURF solves the conservation equations for compressible, multicomponent, reactive flow using the finite volume method. In order to resolve the thin thermal boundary layer and reaction zone in the vicinity of hot particles, the adaptive mesh refinement technique is implemented. The finest mesh is 4 μm and the corresponding time step used is 1 ns. Grid convergence is ensured. A-SURF has been successfully used in previous studies on the ignition and premixed flame propagation (e.g., Faghih and Chen 2017; Li, Gou, Chen 2019; Zhang et al. 2018; Zhang, Gou, Chen 2017). The details on the governing equations and numerical methods can be found in (Chen 2010; Chen, Burke, Ju 2009; Dai and Chen 2015) and thereby are not repeated here.

Results and discussion

The transient ignition process and ignition temperature

The ignition process of stoichiometric CH_4/air mixture initiated by a hot particle with $R = 0.4$ mm and $T_p = 1800$ K is first investigated. Figure 1 shows the temporal evolution of temperature and OH mass fraction distributions during the ignition process in a static stoichiometric CH_4/air mixture with $T_u = 300$ K and $P = 1$ atm. First, a thermal boundary layer develops around the hot particle and the gaseous temperature near the hot particle increases (lines #1~#3 in Figure 1a). Meanwhile, chemical reaction occurs and OH radical is produced and accumulates within the high-temperature region (lines #1~#3 in Figure 1b). Then, at around $t = 6.6$ ms (line #4), thermal runaway occurs: the OH concentration increases dramatically and the peak gaseous temperature becomes slightly higher than the

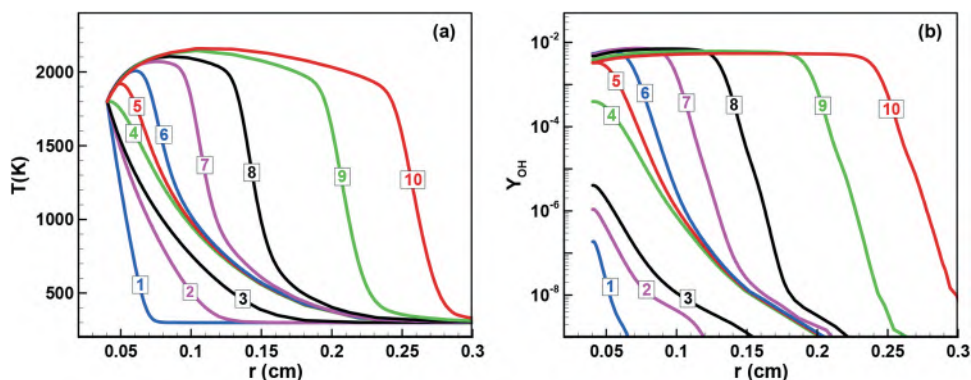


Figure 1. Temporal evolution of (a) temperature and (b) OH mass fraction distributions during the ignition in a static stoichiometric CH_4/air mixture ($T_u = 300\text{ K}$, $P = 1\text{ atm}$) by a hot particle with $R = 0.4\text{ mm}$ and $T_p = 1800\text{ K}$. The time sequence for lines #1–#10 is 0.1, 1.0, 3.0, 6.6, 6.65, 6.67, 6.72, 6.8, 7.0, 7.2 ms.

particle surface temperature, T_p . Finally, a premixed spherical flame is successfully initiated and it propagates outwardly (lines #8–#10 in Figure 1).

Figure 2 shows the temporal evolution of wall heat flux Q and OH mass fraction on the particle surface for hot particles with different temperatures. The wall heat flux is equal to the heat conductivity multiplied by the temperature gradient on the particle surface, i.e., $Q = -\lambda dT/dr$. In the beginning, the hot particle heats up the surrounding mixture and thereby Q is positive. Then, the wall heat flux is shown to drop suddenly and become negative, indicating that heat is conducted from the gaseous mixture to the particle. Meanwhile, the OH mass fraction on the particle surface increases sharply when Q is around zero. Therefore, it is natural to define the ignition delay time τ_{ig} as the time required to reach zero wall heat flux. This means that thermal runaway occurs at the time when the gas temperature starts to exceed particle temperature T_p . This is the well-known Vant' Hoff criterion which was widely used in previous studies (e.g., Kumar 1989; Laurendeau 1982; Law 1978).

Note that the ignition delay time is usually defined for a homogenous system controlled purely by chemical kinetics. Here the ignition delay time is for the non-homogenous ignition caused by a hot particle and it depends on not only chemical kinetics but also transport of reactive species. The comparison between 1D hot particle ignition process and the 0D homogenous ignition process is shown in Figure 3, which depicts the temporal evolution of mass fractions of different species.

To fairly compare with the ignition on the hot particle surface, the gas temperature in the 0D homogenous ignition process is fixed to be same as T_p . In the beginning, OH radical and CH_2O intermediate specie accumulate gradually, which is accompanied by the consumption of a small amount of O_2 . When ignition occurs, the mass fraction of OH suddenly increases to its peak value while the accumulated CH_2O is quickly consumed. For the 0D homogenous ignition process, the ignition delay time is $\tau_{b0D} = 0.15\text{ ms}$, which is about two-order shorter than $\tau_{ig} = 6.8\text{ ms}$ for the 1D hot particle ignition process. Such huge discrepancy in τ_{ig} is attributed to the mass diffusion in the 1D case, in which radicals like OH and H produced around the particle surface diffuse into the surrounding mixtures and thereby prohibit the ignition. Since the radical mass diffusion strongly depends on the

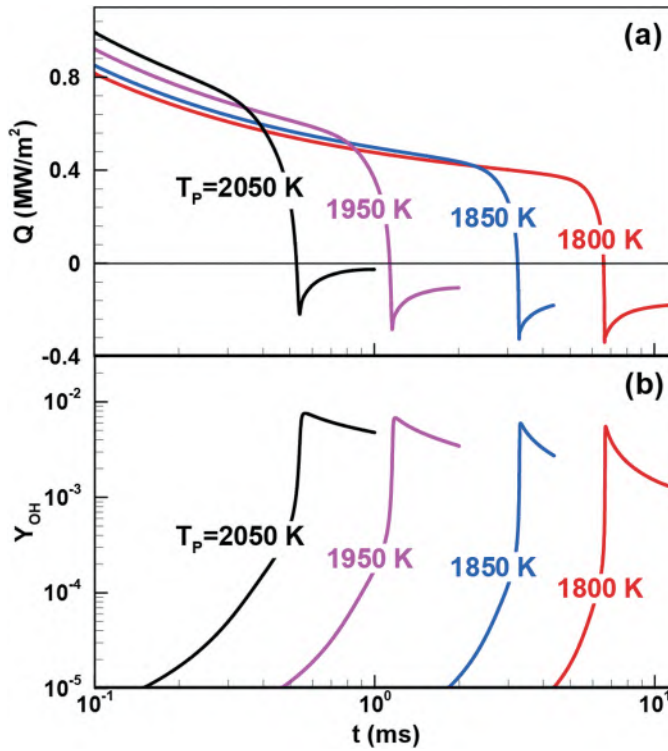


Figure 2. Temporal evolution of (a) wall heat flux (b) OH mass fraction on the particle surface during the ignition in a static stoichiometric CH_4/air mixture ($T_u = 300 \text{ K}$, $P = 1 \text{ atm}$) by a hot particle with $R = 0.4 \text{ mm}$ and different temperatures.

particle size, it is expected that the particle size greatly affects the ignition process, which will be examined in the next sub-section.

Figure 4 plots the ignition delay time τ_{ig} as a function of particle surface temperature T_p . It is observed that τ_{ig} increases exponentially as T_p approaches certain critical value, which is called as the minimum or critical ignition temperature, T_{ig} . This minimum ignition temperature can be extrapolated based on the exponential relationship between τ_{ig} and T_p , which was used by Kumar (1989). The minimum ignition temperatures obtained in our simulations are compared with those from simulations (Beyer and Markus 2012; Häber et al. 2017) and experiments (Roth, Häber, Bockhorn 2017) in Figure 5. It is noted that in the simulation conducted by Häber et al. (2017), the temperature at particle surface was set to increase linearly with time and the minimum ignition temperature was defined as the surface temperature at the time of ignition. Good agreement is achieved for both H_2/air and CH_4/air mixtures. This indicates that the hot particle-induced ignition process can be accurately simulated in the present study.

Effects of particle size

To demonstrate the effects of particle size on the ignition delay time, Figure 6 shows the results for a wide range of particle radius. It is observed that the hot particle-induced

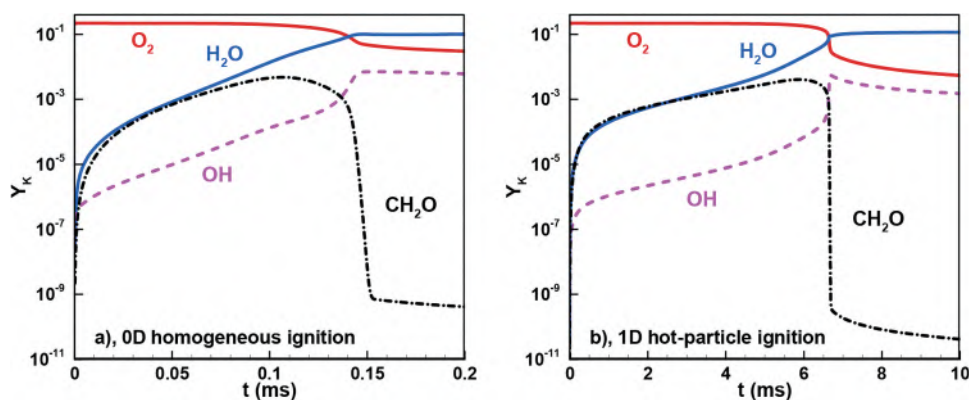


Figure 3. Temporal evolution of mass fractions of different species (a) during 0D homogeneous ignition at fixed temperature and pressure ($T = 1800$ K, $P = 1$ atm); and (b) on the surface of the hot particle with $R = 0.4$ mm and $T_p = 1800$ K. The mixture is CH_4 /air with $\phi = 1$, $T_u = 300$ K and $P = 1$ atm.

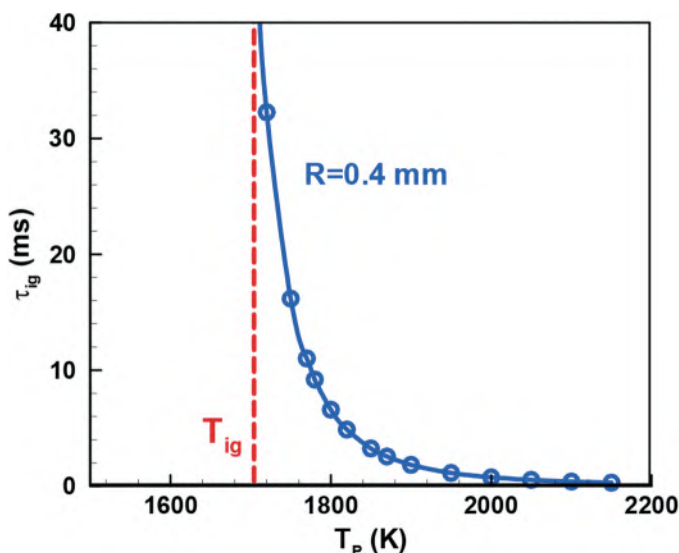


Figure 4. Change of ignition delay time with particle temperature for $R = 0.4$ mm. The mixture is CH_4 /air with $\phi = 1$, $T_u = 300$ K and $P = 1$ atm.

ignition strongly depends on the particle size. With the increase of particle radius, the ignition delay time monotonically decreases, which is consistent with the experimental observation in (Ashman and Büchler 1961) and numerical results in (Su, Homan, Sirignano 1979). When the particle radius is sufficiently small (e.g., $R < 1$ mm), τ_{ig} increases much more rapidly as R decreases. Therefore, this indicates that for each value of T_p , there exists a minimum particle radius below which the mixture cannot be ignited. This minimum particle radius is shown to be below 1 mm and it is difficult to be measured in experiments. On the other hand, τ_{ig} remains to be nearly constant for $R > 4$ mm. In the case of infinitely

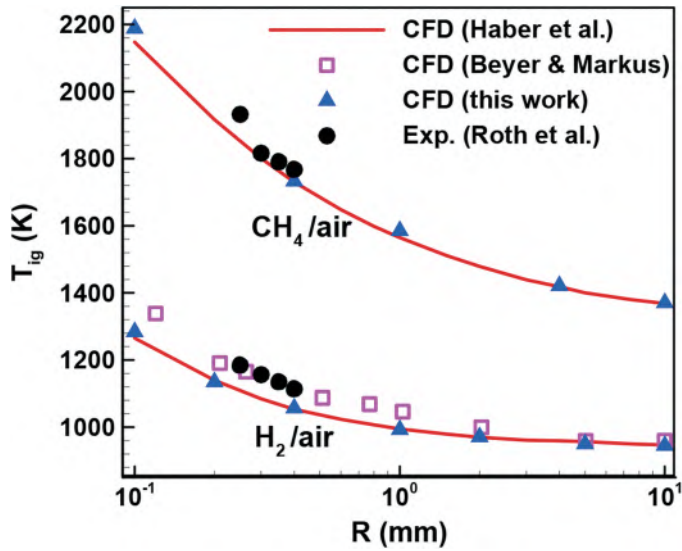


Figure 5. Change of the minimum ignition temperature with particle radius. The mixtures are stoichiometric H_2/air and CH_4/air at $T_u = 300 \text{ K}$ and $P = 1 \text{ atm}$. The experimental data reported by Roth, Häber, and Bockhorn (2017) and simulation results from Häber et al. (2017) and Beyer and Markus (2012) are also shown.

large particle radius, it becomes the ignition caused by a hot planar surface. It is noted that for simplicity, the particle surface temperature is fixed to be constant in this work. However, in practical the particle temperature changes when its radius is very small or the ignition delay time is very long. Considering the change of particle temperature merits future study.

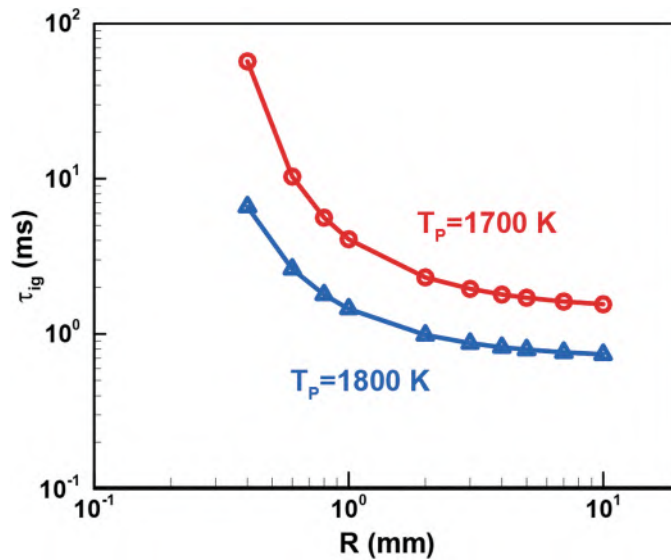


Figure 6. Change of ignition delay time with particle radius for $T_p = 1700 \text{ K}$ and 1800 K . The mixture is CH_4/air with $\phi = 1$, $T_u = 300 \text{ K}$ and $P = 1 \text{ atm}$.

Figure 7 compares the ignition delay time for ignition induced by hot particles with different temperatures and radii. The result for 0D homogenous ignition is shown together for comparison. For a given temperature, τ_{ig} for 1D hot particle ignition is always larger than $\tau_{i,0D}$ for 0D homogenous ignition. For larger particle size, the ignition delay time τ_{ig} becomes shorter and closer to that of 0D homogenous ignition. This is because the radical flux diffusing away from the particle surface is proportional to the inverse of the particle radius. Besides, Figure 7 shows that the sensitivity of the ignition delay time to the temperature (i.e., the slope of each curve) is different for 1D hot particle ignition and 0D homogeneous ignition. This indicates that the effect of radical mass diffusion is much stronger at lower particle temperature T_p . This is reasonable since lower T_p corresponds to longer ignition delay time and thereby there is more time for radicals to diffuse away from the particle surface.

The inhomogeneous ignition process induced by a hot particle is mainly controlled by the chemical reaction and mass diffusion. Thermal conduction is not important since the particle surface temperature is fixed. Therefore, there are two characteristic times: one for chemical reaction, $\tau_{i,0D}$, which is the ignition delay time for the 0D homogenous ignition at a fixed temperature of T_p ; and the other for mass diffusion, τ_d , which is defined as R^2/D_H , where D_H is the mass diffusivity of H radical. H radical is chosen here since it is involved in the most important chain-branching reaction $H + O_2 = O + OH$ and it has the largest mass diffusivity. We introduce a Damköhler number, $Da = \tau_{i,0D}/\tau_d$, which is the ratio between chemical reaction time and mass diffusion time. Figure 8 plots the normalized ignition delay time for 1D particle ignition as a function of the Damköhler number. A good linear relationship between τ_{ig}/τ_d and Da is shown in Figure 8. The results for different particle radii collapse into the black-dashed straight line with slope close to unity in Figure 8, especially for relatively large particles with $R = 4$ and 10 mm. This is consistent with the

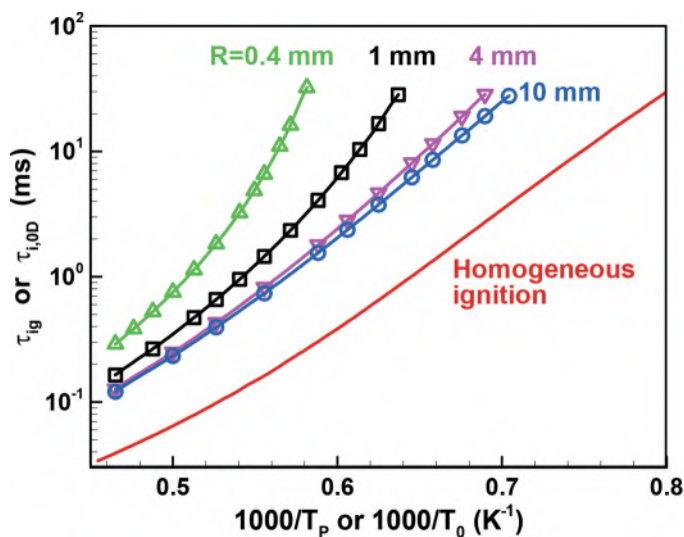


Figure 7. Change of ignition delay time with particle temperature for particles with different radii. The mixture is CH_4/air with $\phi = 1$, $T_u = 300$ K and $P = 1$ atm. The results for homogeneous ignition at constant temperature are shown for comparison.

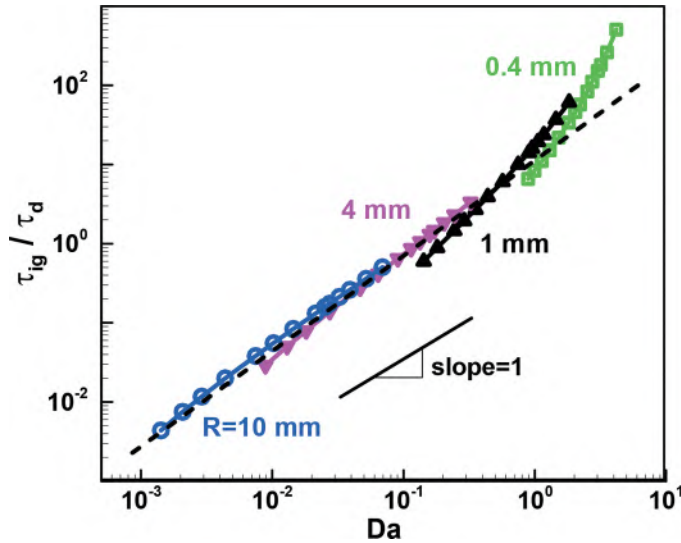


Figure 8. Change of normalized ignition delay time with the Damköhler number for particles with different radii. The mixture is CH_4/air with $\phi = 1$, $T_u = 300$ K and $P = 1$ atm. The results for homogeneous ignition at constant temperature are shown for comparison.

theoretical results obtained in (Law 1979). Law (1979) proposed an expression for the ignition delay time which is proportional to the characteristic chemical reaction time under the assumption that the reaction region is confined to the vicinity of the hot particle surface. For relatively large particles, this assumption holds given that the thickness of the reaction region (i.e., high-temperature region) is much smaller compared to particle radius. Therefore, a linear correlation between τ_{ig}/τ_d and Da is observed in Figure 8. For relatively small particles with $R = 0.4$ mm, there is obvious deviation from the linear relationship. One possible reason is that ignition first occurs at a small distance (denoted as l_{ig}) away from particle surface instead of on particle surface (Melguizo-Gavilanes et al. 2017b; Nagata et al. 1994). For $R \gg l_{ig}$, it is fine to use $\tau_d = R^2/D_H$. However, for small particle radius with R in the same order of l_{ig} , it is improper to use R as the length scale to calculate the characteristic diffusion time τ_d .

The above results are obtained for CH_4/air mixtures. We also assess the effects of particle size on hot particle ignition in a stoichiometric H_2/air mixture. The results are shown in Figures 9 and 10.

Figure 9 compares the ignition delay time for different particle radii and for 0D homogeneous ignition process. Similar to the results for the CH_4/air mixture shown in Figure 7, the ignition delay time for 1D hot particle ignition is always larger than that for 0D homogeneous ignition due to the radical loss caused by mass diffusion in the hot particle-induced ignition process. The smaller the particle radius, the larger the radical loss, and thereby the longer the ignition delay time. On the other hand, there is a large difference between results for CH_4/air and H_2/air mixtures. A linear relationship between $\ln(\tau_{ig})$ and $1/T_P$ is shown for CH_4/air in Figure 7, while $\ln(\tau_{ig})$ is shown to increase nonlinearly with $1/T_P$ for H_2/air in Figure 9. This is because a much broad range of temperature is considered

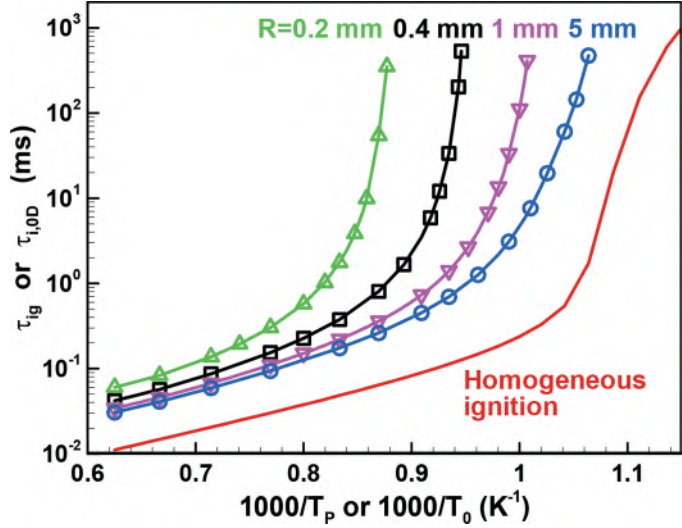


Figure 9. Change of ignition delay time with particle temperature for particles with different radii. The mixture is H_2/air with $\phi = 1$, $T_u = 300 \text{ K}$ and $P = 1 \text{ atm}$. The results for homogeneous ignition at constant temperature are shown for comparison.

for H_2/air . At relatively low temperature, the ignition delay time increases abruptly, similar to the results for 0D homogeneous ignition shown in Figure 9.

Figure 10 shows the change of the normalized ignition delay time τ_{ig}/τ_d with the Damköhler number Da for particles with different radii. The data points for different particle sizes within the relatively high-temperature range lie in the same straight line. However, the data points

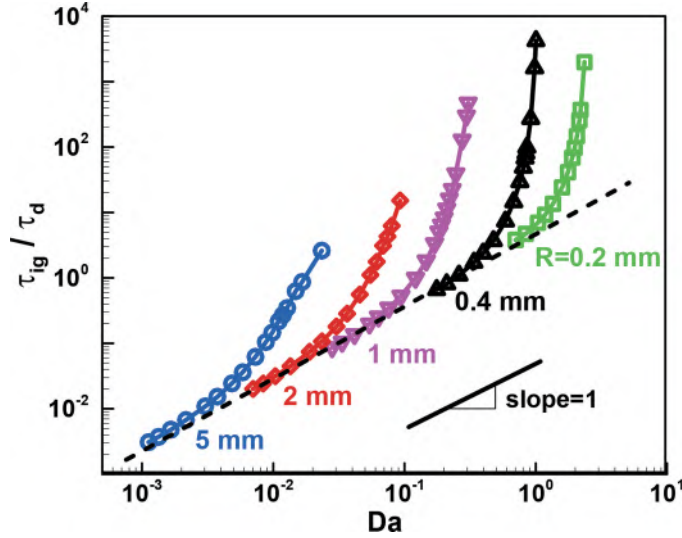


Figure 10. Change of the normalized ignition delay time with the Damköhler number for particles with different radii. The mixture is H_2/air with $\phi = 1$, $T_u = 300 \text{ K}$ and $P = 1 \text{ atm}$.

within the relatively low-temperature range deviate from the straight line even for the large particle with $R = 5$ mm. This large deviation partially comes from the uncertainty on the chemical reaction timescale. Specifically, the homogenous ignition delay time $\tau_{i,0D}$ at the corresponding particle temperature T_p might differ greatly from that at the actual ignition position. It is improper to use $\tau_{i,0D}$ at T_p to represent the characteristic chemical reaction time. Therefore, the distance l_{ig} introduces uncertainty in not only τ_d but also $\tau_{i,0D}$ for the H_2 /air mixture. Besides, the uncertainty in τ_d increases with l_{ig}/R while that in $\tau_{i,0D}$ increases with l_{ig} . This explains why the significant deviation from the straight line in Figure 10 is observed even for large particles when stoichiometric H_2 /air mixture is considered.

Conclusions

One-dimensional transient simulations are conducted for the ignition of static CH_4 /air and H_2 /air mixtures by hot particles with fixed surface temperature. The emphasis is on assessing the effects of particle size on the ignition delay time. Due to the radical loss caused by mass diffusion away from the particle surface, the 1D hot particle ignition is much slower than the 0D homogeneous ignition process. The radical flux diffusing away from the particle surface is proportional to the inverse of the particle radius. Therefore, the smaller the particle radius, the larger the ignition delay time for the hot particle-induced ignition process. Moreover, at lower particle temperature, the ignition delay time becomes longer and there is more time for radicals to diffuse away from the particle surface. Consequently, the effect of radical mass diffusion is much stronger at lower particle temperature. It is found that for the stoichiometric CH_4 /air mixture, the normalized ignition delay time changes linearly with the Damköhler number, which is the ratio of characteristic chemical reaction time to the mass diffusion time. However, for the H_2 /air mixture, the linear relationship only holds for relatively high particle temperature, and significant deviation from this linear relationship is observed for relatively low particle temperature even for the large particles. This is partially due to the uncertainty in the determination of characteristic chemical reaction time in the 1D hot particle ignition process.

As a step toward a better understanding of hot particle-induced ignition, this study considers the simplified case in which the particle surface temperature is fixed and surface reactions are not considered. In addition, here only stoichiometric fuel/air mixtures are studied. In future works, it would be interesting to take into account the change of particle surface temperature and the influence of surface reactions and to consider both fuel-lean and fuel-rich mixtures.

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