

Article

Ignition Thresholds and Flame Propagation of Methane/Air Mixtures Ignited via Radiatively Heated Inert Particles

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Abstract: The prevention and evaluation of explosions requires suitable standards of measurement. As such, for this study two ignition thresholds, the ignition temperature and the minimum ignition irradiance were selected as the assessment criteria. These ignition threshold values were experimentally determined by heating stationary inert silicon carbide particles via thermal radiation with a large spot size in order to ignite quiescent methane-air fuel mixtures. A high-speed Schlieren camera was used to capture the progression of the formation and propagation of the flames throughout the experiments. The results of the experiments show that the irradiance and temperature threshold are directly and inversely proportional to the particle size, respectively. Furthermore, the irradiance and temperature thresholds have similar tendencies within the flammability limits; wherein, the minimum value corresponds to fuel mixtures at a stoichiometric ratio, and increases as the equivalence ratio shifts toward the flammability limits. Irradiance thresholds, though, are more sensitive to changes in equivalence ratio than temperature. The temperature histories of the heated particle determined that when the irradiance is lower than its ignition threshold value, the heated particle-fuel mixture system will arrive at a thermal equilibrium, rather than ignition, due to the inability of the particle to reach the ignition temperature. This study also found that longer ignition times will result in a more drastic deformation of the flame fronts caused by natural convection.

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1. Introduction

Optical radiation, with sufficient energy, is an ignition source for flammable gases in various working environments; wherein three laser ignition mechanisms for fuel mixtures are: (1) ignition by local temperature increase [1,2] or photochemical processes [3,4] that are caused by the fuel mixture directly absorbing the radiation; (2) ignition by plasma which was formed via focused laser radiation [5]; and (3) radiatively heated particle ignition, which is the heating of particles via continuous-wave lasers that subsequently induce ignition of the fuel mixture. The previously inexplicable overpressure and high rate of combustion of unconfined dust explosions can be attributed to the radiatively heated particle ignition mechanism; wherein, the explosion can be spread and intensified by ignition kernels that form ahead of the flame front and are capable of igniting larger and larger volumes of the fuel-air mixture [6,7]. This work intends to study the radiatively heated particle ignition mechanism because it is substantially under-researched and is significant for improving the understanding of dust explosion mechanisms.

Previous research on heated particle ignition has advanced along two avenues: utilization of either stationary or moving heated particles to ignite fuel mixtures. Usually the laser is used as a thermal radiation source to heat the particles, and the energy threshold refers to the lowest laser energy required for ignition. In contrast to sparks or other vectors of energy transfer that are used to ignite fuel mixtures, the means of energy transfer for

radiatively heated particle ignition are thermal conduction and radiation from the heated particles. Dubaniewicz Jr. et al. [8] found that the energy threshold of heated coal particles is higher than that of heated iron-oxide particles. Hills et al. [9] observed that the energy threshold decreases as the particle size decreases when a heated single coal particle is used to ignite hydrogen-air fuel mixtures. Welzel et al. [10] indicated that the energy threshold rapidly decreases as the mixture temperature increases when using a single oxidized iron particle to ignite ether air-fuel mixtures at varying temperatures. Dubaniewicz Jr. et al. [11] conducted analyses of laser ignition experiment results and concluded that power is a good standard of measurement for minimum ignition energy when the spot size is smaller than 300 μm ; whereas irradiance is a better standard of measurement when the spot size is larger than 300 μm .

The application of sufficient energy will allow fuel mixtures to be heated to the ignition temperature. Many researchers focus their research on the ignition temperature because it is the determinant factor for whether ignition can occur. Beyer and Markus [12] performed stationary heated particle ignition experiments using inert particles and n-pentane-air fuel mixture. The results of the experiments indicate that ignition temperature is not sensitive to changes in the fuel mixture equivalence ratio; however, the temperature increases as particle size decreases. Silver [13] shot two types of heated particles to ignite coal-gas-air, n-pentane-air, and hydrogen-air fuel mixtures and asserts that particle size and particle surface temperature are the two main factors that are instrumental in determining whether ignition occurs. A comparison of the results of stationary and dynamic ignition, which correspond to [12,13], respectively, indicates that the moving particles have a higher ignition temperature than the stationary particles. Paterson [14,15] expanded on Silver [13] by varying the velocity of the particles and determined that, as particle velocity increases, so too does the ignition temperature. Beyrau et al. [16,17] supplemented the research on stationary heated particle ignition by utilizing various types of particles to conduct experiments and found that chemical reactions increase the difficulty of ignition for fuel-rich mixtures. Furthermore, the particle chemical reactions were also found to significantly affect ignition temperature, minimum ignition energy and ignition time.

Interest in studying the propagation of the flame is being stoked by the improvements in simulation techniques and the development of photography technology. Melguizo-Gavilanes et al. [18] simulated the ignition caused by the falling of a heated particle from nitrogen into a combustible fuel mixture medium, and concluded that the position of the ignition kernel shifts from the separation region to the area below the particle as the initial temperature of the particle increases. Häber et al. [19] created 2D simulations for stationary inert heated particle experiments to analyze the effect of equivalence ratio on the position of the ignition kernel. The reaction zone was shown to be broader for fuel-rich mixtures, and the ignition kernel always formed above the heated particle at the location with the highest temperature. Coronel et al. [20] captured the ignition of fuel mixtures by falling heated particles using a Schlieren camera, and simulated the ignition temperature as well as the importance of differential diffusion. To the best of the authors' knowledge, researchers have yet to publish studies that capture detailed ignition processes for ignition by stationary heated inert particles.

In actual explosions, the thermal radiation generated by the dust explosion is very broad, and suspended particles in front of the flame are usually aggregated to form particle clusters under the action of shock waves. However, previous studies were limited to the ignition of a single particle under a small spot size. Therefore, this paper studies the ignition threshold of methane ignited by particle clusters under a large spot area. The objectives of the study are to determine: (1) the minimum ignition energy and temperature that are required for the ignition of methane-air fuel mixtures by heated inert silicon carbide particles at varied particle size; (2) the distribution and trends of ignition threshold

of energy and temperature at varying equivalence ratios; and (3) position of ignition kernels and factors that affect the propagation of flame fronts through qualitative observation and analyses, respectively.

2. Experiment Methods

A cylindrical chamber, with both an inner height and diameter of 200 mm, was used for the ignition experiment. The circular top and bottom faces of the chamber were both sealed with quartz glass to facilitate the observation of the experiment progression using a 3000 fps Schlieren camera. A window, with dimensions of 30×50 mm, was opened on the side of the chamber to allow for the passage of both the heating and temperature measurement lasers. The heating laser was produced using a semiconductor and the temperature measurement laser was produced by a two-wavelength infrared thermometer. A circular acrylic platform, with a diameter of 6 mm, was installed centrally within the combustion chamber and was covered with a 1 mm thick layer of particles for the experiments. Between experiments, the heated particles on the platform were replaced by newly prepared particles and the container was resealed. The schematic diagram of the experimental apparatus is shown in Figure 1.

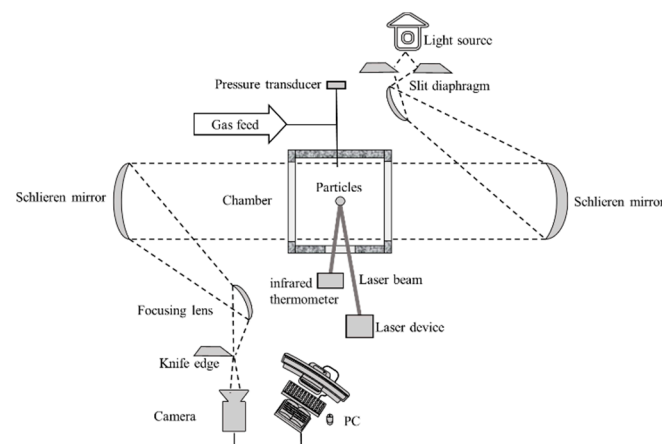


Figure 1. Diagram of the laser ignition experiment apparatus [21].

The chamber has several ports that are used as interfaces for precision pressure transducers (error $< \pm 0.5\%$) and fuel recirculation lines, which are used in combination with a vacuum meter to obtain the desired composition for the fuel mixture using the partial pressure method. After obtaining the desired equivalence ratio, ϕ , the fuel mixture is left to sit for 15 min to ensure its uniformity and to reduce the effect of turbulence. The ignition delay time was defined as the interval from particles illumination to ignition. The criteria for the start of ignition was marked by the appearance of a visible flame in the Schlieren images. To ensure the accurate measurement of particle temperatures, the semiconductor laser and two-wavelength infrared thermometers, both with response times of 2 ms, were synchronized using a combined switch. The thermal radiation source for the experimental apparatus is a 100 W semiconductor laser with wavelength and spot size of 915 nm and 5 mm, respectively. The laser is guided to the center of the heated particle zone by a combination of an aiming laser and CCD camera, while a laser power meter measures the power of the laser as it reaches the particle. Time-resolved temperature measurements were performed using a Sensor METIS M3 infrared thermometer, with operating wavebands of 1450–1650 nm and 1650–1800 nm. The emissivity of the particles is assumed to be constant; therefore, all temperatures are considered gray-body temperatures. According to [22], the emissivity value depends on the particle surface temperature, and the maximum ratio of emissivity of the two wavebands is 1.1. At a particle temperature of 2000 K, with a 10% emissivity change, the gray-body temperature error, which is calculated using a combination of instrumentation error and Planck's law, is ± 50 K. Due to the Gaussian distribution

of laser spot energy and non-uniform distribution of particle size, the recorded temperature values are the average measured temperatures of the particles in the heated particle zone.

Methane was selected as the fuel component of the fuel mixture because it is a common combustible gas, and this study elected to use silicon carbide as the targeted material because it is available in many different particle sizes. The particle preparation procedure is as follows: crushing of bulk specimens to create particles, sanding particles, and successive sieving of particles using meshes of varying grades. Though the batches of particles have undergone a series of sieves, the resultant particle diameters still follow a Gaussian distribution with a median particle diameter, which is experimentally determined using a laser particle size analyzer. Furthermore, silicon carbide has a 3000 K melting point and does not chemically react or change in size throughout the heating process. According to the empirical formula, as shown in Equation (1), given in [23], the trend of specific heat capacity change with temperature for silicon carbide is shown in Figure 2.

$$C_p = 925.65 + 0.377 \times T - 7.926 \times 10^{-5} \times T^3 - 3.195 \times 107 / T^2 \quad (200\text{K} < T < 2400\text{K}) \quad (1)$$

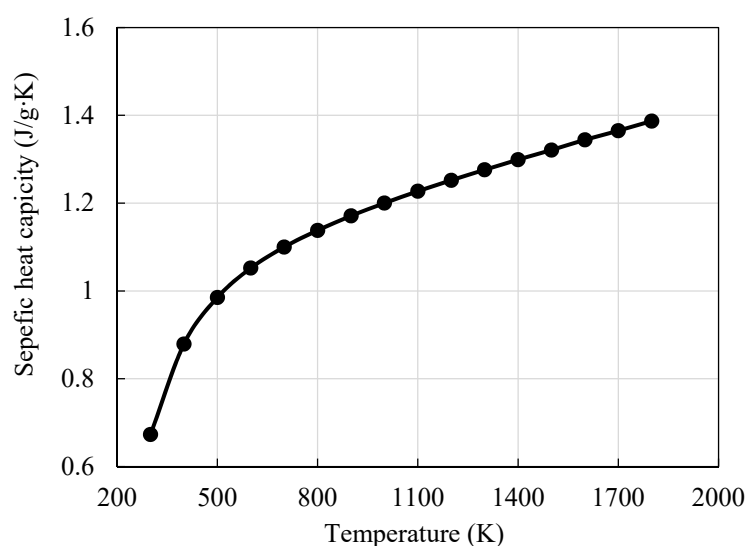


Figure 2. Specific heat capacity of silicon carbide as a function of temperature.

Ignition is considered successful when a self-sustained flame expands throughout the chamber. Ten repetitive tests under each experiment condition were conducted in order to achieve reliable results, and the experiment conditions were considered incapable of inducing ignition only after being consecutively and successfully repeated ten times.

3. Results and Discussion

3.1. Irradiance Threshold of Ignition

To ignite combustible fuel mixtures, enough energy needs to be provided in a very short period of time to the fuel mixture to ensure that it can be heated to the required ignition temperature [24]. During radiative ignition, the accumulation of energy in the fuel mixture surrounding the particle is a result of chemical reactions that take place within the fuel mixture and both thermal radiation and conduction from the particle. Whereas the dispersion of energy from the fuel mixture is a result of thermal convection and conduction. This study focuses on the influence of the fuel concentration and particle size on the irradiance threshold, wherein irradiance is obtained by dividing the power by spot area.

The irradiances of successful ignition and ignition failure experiments at varying equivalence ratios, with a sustained median particle size of $d_{50} = 145 \mu\text{m}$, are shown in Figure 3; wherein, the irradiance threshold is lowest around the stoichiometric ratio and increases as the equivalence ratio either increases or decreases. The difference in irradiance threshold is due to the difference in varied chemical reaction activity of fuel mixtures under varied equivalence ratios. Flame temperature decreases as the equivalence approaches the flammability limits, which causes a decrease in overall chemical reaction activity. Therefore, the particle has to provide additional energy to the fuel mixture to compensate for the reduced flame temperature and to induce and feed the self-sustained chemical reactions. The laser energy is the source of energy for the particle; hence the irradiation threshold increases as the fuel mixtures approach the flammability limits.

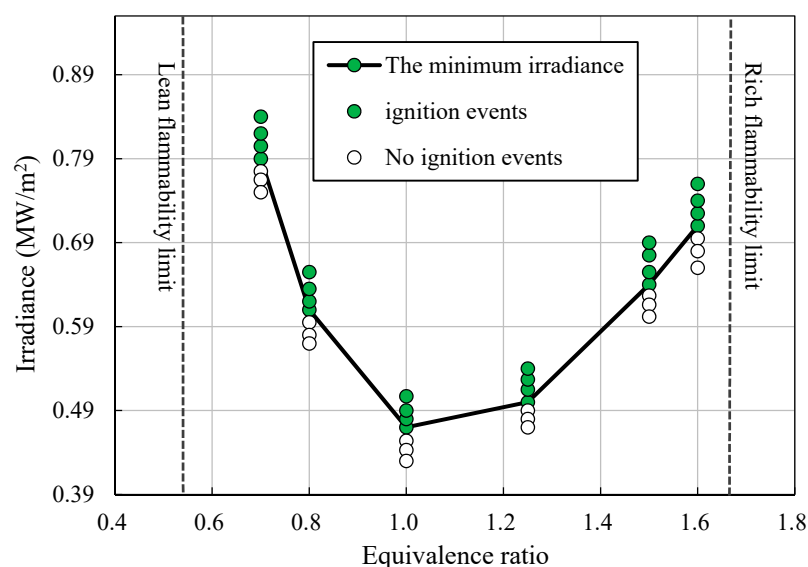


Figure 3. Ignition irradiance vs. equivalence ratio of methane/air mixtures with silicon carbide particles heated by thermal radiation, $d_{50} = 145 \mu\text{m}$.

Heated particles are the source of energy for the fuel mixture; therefore, the irradiance thresholds will be affected by changes in the properties of the particle. The experimental results indicated that the irradiance threshold increases as the median particle size increases when ϕ is kept constant, as shown in Figure 4. The direct proportionality between the irradiance and the median particle size is due to the differences in the physical properties, including heat capacity and surface area-volume ratio, of particles as the particle size is varied. Heat capacity increase as particle size increases, therefore higher irradiances are required to heat larger particles to the same temperature. Lower surface area-volume ratios will reduce the ability of the particle to transfer heat to the fuel mixture; therefore, larger particles have higher irradiance thresholds if the overall chemical reaction activity of the fuel mixture remains constant.

The possibility of successive ignition kernel formation is confirmed by the fact that the experimentally determined irradiance thresholds of ignition are lower than the observed thermal radiation output by dust explosions under similar conditions. Holbrow [25] measured the radiation produced by dust explosion fireballs that were caused by six types of dusts; wherein, the average emitted radiation ranged from 0.23 to 2.9 MW/m². Moore and Weinberg [6] calculated the heat flux to be approximately 1 MW/m² for flames that were the result of the ignition of stoichiometric fuel mixtures. Therefore, the majority of explosions in the field will produce enough thermal radiation to induce the ignition of fuel mixtures via particle heating because these thermal radiation values are higher than this study's irradiation ignition threshold values.

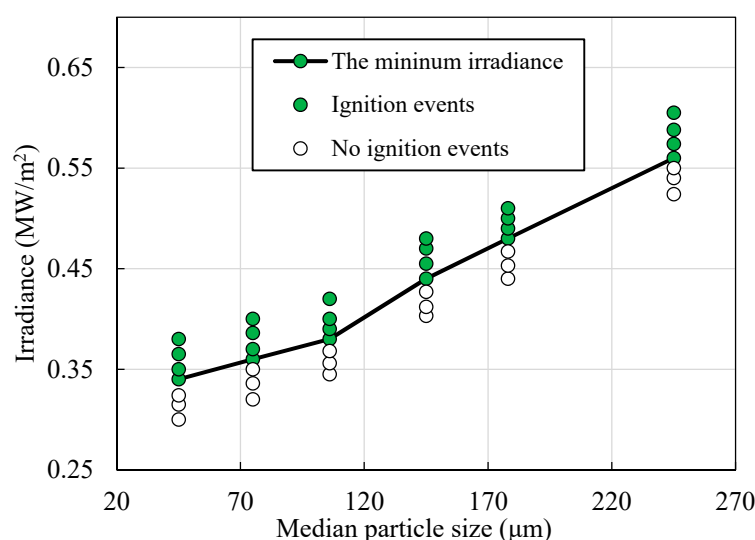


Figure 4. Ignition irradiance vs. median particle size of silicon carbide particles heated by thermal radiation, $\varphi = 1.0$.

3.2. Temperature Threshold of Ignition

The ignition threshold of temperature refers to the temperature of the particles at the instant of ignition. Figure 5 shows the particle temperature history for a successful ignition experiment with the fuel mixture at a stoichiometric ratio and a median particle size of 45 μm . When irradiance is constant, the heating rate of the particle decreases as the particle temperature increases due to the increase of the specific heat capacity combined with the particle heat loss. Ignition occurs when silicon carbide particles are heated to approximately 1700 K, and this particle temperature is significantly higher than the 900 K auto-ignition temperatures for the methane-air fuel mixture at the same equivalence ratios [26] because the large surface area-volume ratio causes the rapid dissipation of energy. When the irradiance is lower than its ignition threshold value, the particle temperature curve will gradually flatten and eventually fluctuate in a small range near a certain fixed temperature value and the heated particle-fuel mixture system will arrive at a thermal equilibrium rather than ignition due to the inability of the particle to reach the ignition temperature, resulting the inability of fuel mixture to accumulate enough energy.

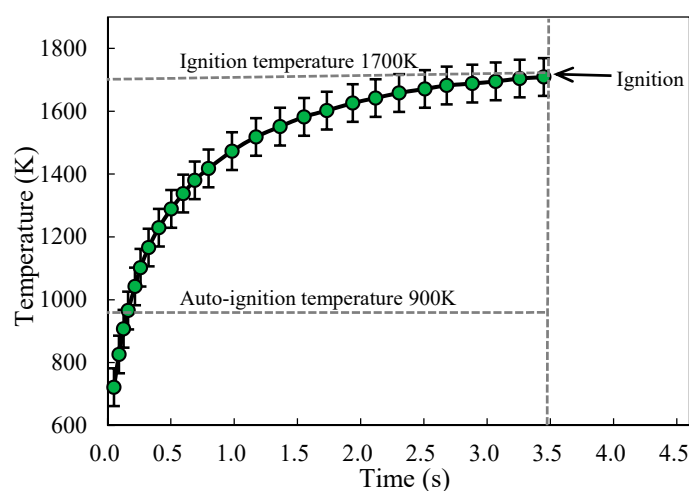


Figure 5. Temperature history of silicon carbide particles heated by thermal radiation in stoichiometric methane/air mixtures, ignition occurs at $t_{\text{ig}} = 3.45$ s, $d_{50} = 45$ μm , irradiance = 0.4 MW/m^2 .

The ignition threshold of temperature is significantly affected by the gas composition and particle size. As shown in Figure 6, when the median particle size remains unchanged at 145 μm , the distribution trend of the ignition temperature is the same as that of the ignition threshold of irradiance; that is, it is negatively correlated with the chemical reaction activity of the gas. As shown in Figure 7, when the fuel-air equivalence ratio remains unchanged at 1.0, the ignition temperature decreases with the increase of the particle size. The distribution of ignition temperature is opposite to that of the ignition threshold of irradiance due to the lower heat dissipation capacity of the larger particle size.

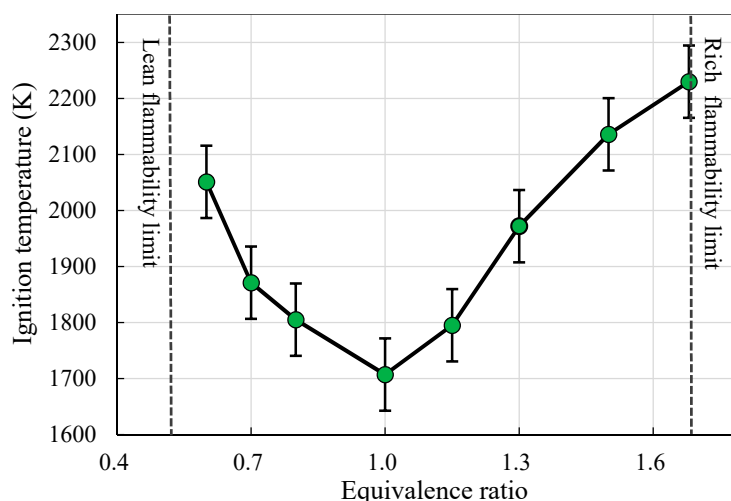


Figure 6. Temperature thresholds vs. equivalence ratio of methane/air mixtures ignited by heated silicon carbide particles, $d_{50} = 145 \mu\text{m}$, irradiance = 1.0 MW/m^2 .

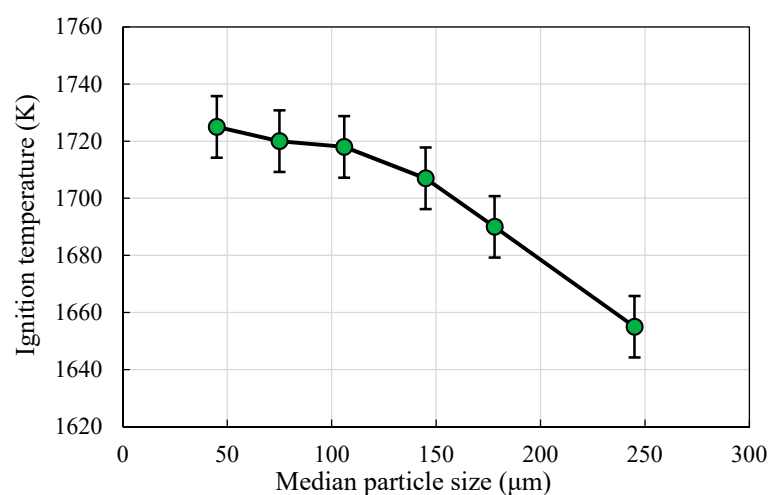


Figure 7. Temperature thresholds vs. median particles size of heated silicon carbide particles, $\varphi = 1.0$, irradiance = 1.0 MW/m^2 .

Irradiance and temperature thresholds are two of the deciding parameters for the achievability of ignition. These ignition thresholds are lowest when the fuel mixture is at the stoichiometric ratio and increases as the equivalence ratio approaches the flammability limits, as shown in Figures 3 and 6, respectively. Within the flammability limits, the increased sensitivity of the irradiance threshold compared to that of the temperature threshold can be seen in Figures 3 and 6, respectively; where the variabilities of the irradiance and temperature thresholds are 87% and 34%, respectively. This is because the ig-

nitron energy threshold is closely related to the chemical reaction activity of the fuel, however, the ignition temperature is affected by many factors such as the chemical reaction activity and the thermal conductivity of the fuel mixture [19]. Within the flammable limit, the narrow flammable limit range of methane-air mixtures makes the thermal conductivity of the fuel mixture less variable, causing the ignition irradiance thresholds to be more sensitive to changes in the fuel composition than the ignition temperature thresholds.

3.3. Formation and Propagation of the Flame

The Schlieren camera has the excellent ability to capture the formation and propagation of the flame, and ignition time is defined as the time interval from the heating of the particles to the appearance of the flame. In all experiment conditions presented in this research, the heat convection rate between the particles and the gas is much higher than the heat conduction rate inside the particles because the Biot number of SiC particles in this study is less than 0.1. As such, the characteristic time of heat conduction of the gas phase estimated by [27] is many magnitudes shorter than the ignition time of heated particle ignition in this research. Based on the above analysis, heat can be transferred repeatedly and continuously from the surface of the particle to the surrounding gas before the ignition of the fuel mixture. Subsequently, steadily growing heat convection currents will be formed because the rising heated low-density gases near the particle will push the unheated high-density gases downwards. Consequently, the initially entirely quiescent fuel mixture will be partially disturbed by a velocity field that affects an increasingly large area above the particle.

The growing heat convection current will likely affect the formation and propagation of the flame front; therefore, this study elects to utilize two experiments with significantly differing ignition times, 240 and 700 ms with corresponding equivalence ratios of 1 and 1.55, respectively, with constant irradiance of 1.0 MW/m^2 and $d_{50} = 45 \text{ }\mu\text{m}$. Henceforth, the ignition experiments with equivalence ratios of 1 and 1.55 shall be termed Case 1 and Case 2, respectively, to simplify their utilization. The movement of the gases prior to ignition and the propagation of the flame front for both experiments are shown in Figure 8.

There are two easily identifiable stages in the ignition experiment, the heating and ignition stages. The formation of heat convection currents and the consequent velocity fields occur during the heating stage, while the formation and subsequent propagation of the flame front occur during the ignition stage. The entire heating and ignition processes for Case 1 and Case 2 are shown in Figure 8, wherein the formation of the heat convection currents can be observed. In contrast to that of Case 2, the heat convection current of Case 1 is visibly smaller due to the interruption of the heat convection current formation process by its comparatively short ignition time of 240 ms. The additional heating time for Case 2 allows for the heat convection current to expand further upwards, as shown in the highlighted area in Figure 8. After ignition is induced, the initial flame front of Case 1 is both continuous and spherical because it is not significantly affected by the underdeveloped heat convection current. In contrast, the flame front of Case 2 initiates from the sides of the particle, becomes discontinuous, and deforms above the particle. The deformation of the flame front of Case 2 is due to the non-uniformity of the gas temperature and velocity surrounding the particle. The gases to the sides of the particle that are not affected by the heat convection current will have steadily increasing temperatures, whereas the heated low-density and non-heated high-density gases above the particle will rise and fall, respectively, under the effect of buoyancy. Therefore, the gases to the side of the particle will always have a higher temperature and lower location velocity than that of the gases above the particle. The above experimental phenomenon indicate that longer ignition times will result in a more drastic deformation of the flame fronts.

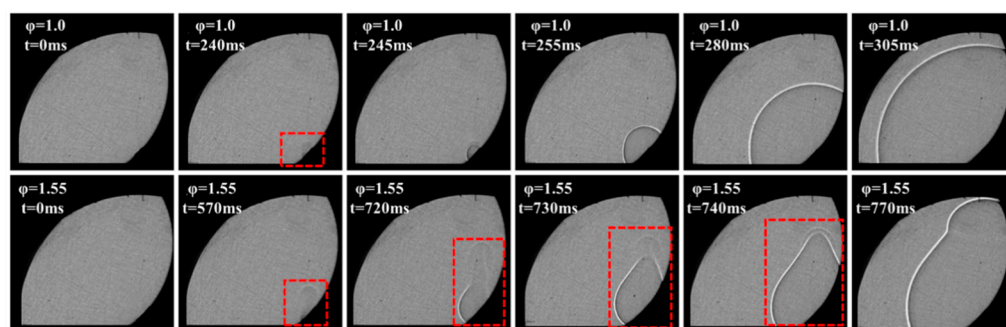


Figure 8. Flame formation and propagation of methane/air with an equivalence ratio of 1 and 1.55. The heat convection current is highlighted with a dotted line box.

4. Conclusions

The heating and ignition of methane–air fuel mixtures using stationary heated particles were studied in the following manner. The minimum irradiances and temperatures of ignition were measured and recorded for heated stationary particle ignition experiments to determine the variability of minimum irradiance and temperature to changes in the equivalence ratio and particle size. Furthermore, the locations of the ignition kernel formation and the propagation of the flame front were systematically observed in order to analyze the differences between the flame fronts of fuel mixtures with varying equivalence ratios. The following conclusions were drawn from the experimental results and comparative analyses of this study:

1. As the equivalence ratio approaches the flammability limits, the ignition threshold of irradiances increases, due to the decreases in the reactivity of the fuel mixture. Furthermore, the ignition threshold of irradiance increases as the particle size increases, due to increased heat capacities and decreased surface area-volume ratios at larger particle sizes.
2. The ignition threshold of temperature of the particles decreases with the increase of the particle size. Within the flammability limits, the trends of the temperature and irradiance ignition thresholds are similar; however, changes in the equivalence ratio affect irradiance more significantly than temperature. Due to the energy dissipation, the particle-ignition temperatures are significantly higher than the fuel mixture auto-ignition temperatures.
3. When irradiance is lower than the irradiance threshold, the inability to achieve ignition can be explained by the attainment of thermal equilibrium in the particle-gas system. At thermal equilibrium, the particles are unable to transfer additional heat to the surrounding fuel mixture, meaning that the temperature of the fuel mixture cannot reach the required ignition temperature.
4. Ignition always initiates from the surface of the particle because the temperature of the gases surrounding the particle is the highest. Increased ignition times allow for the formation of heat convection currents above the particle due to the effects of buoyancy, which will cause the non-uniformity of the gas temperatures surrounding the particle and a velocity field to form. The non-uniformity of gas temperature and the velocity field will cause the flame to be deformed.

Further research will focus on the utilization numerical simulation methods to quantitatively study the fuel mixture ignition via heated particles. Furthermore, additional experimentation should be conducted to obtain ignition parameters, including respective minimum ignition energy, ignition temperature, and the formation and propagation of the flames for corresponding numerical simulations.

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writing—review & editing, C.Z. Both authors have read and agreed to the published version of the manuscript.

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