A numerical study on the two-stage combustion behaviour of n-heptane droplet clouds

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Abstract

Two-stage combustion of one-dimensional spherical monodisperse n-heptane droplet clouds in hot ambient has been numerically investigated. The results reveal a complex autoignition and burning process with stages of: first-stage ignition, quasi-steady non-premix cool flame, second-stage ignition, premix hot flame. By varying the inner gas temperature T_i, ambient gas temperature T_a, droplet diameter d_0, droplet number density n, and cloud radius R_c, the resulted autoignition dynamics are studied. T_i has effects on flame structure. For T_i at 700 K, steady dual flame structure has been observed. It is also found that four distinct ignition regimes can be identified, i.e. from no ignition, single-stage ignition with temperature rise, single-stage ignition without temperature rise to two-stage ignition with increasing T_a. Besides, phase diagrams have been presented to show ignition modes under different ambient temperature and sub-parameters of droplet clouds. It is observed that ignition modes are sensitive to n and d_0, but remains the same when varying R_c. It has been found that classical group number $G = 2\pi n d_0 R_c^2$ cannot represent the transient ignition behaviour of droplet clouds. Instead, in this study, a new dimensionless number $G_{ig} = n^{2/3} d^2$ has been proposed, which has a much better ability to recognize different ignition modes.

Keywords

Droplet cloud; Two-stage ignition; n-Heptane; Group combustion

1. Introduction

Spray combustion is the common organization form in engines, like jet engines, gas turbines, and internal combustion engines [1]. Many fundamental problems (e.g. autoignition and flame propagation in spray) still call for better understandings due to the complicated multi-phase and multi-scale nature of spray combustion. For the microscopic view of spray, single droplets and droplet clouds are usually identified as basic components [2]. The latter considers wider effects of droplet group behaviour, which makes it different from single droplets, and hence has been paid attentions.

When droplets cluster as a cloud and burn, they will behave differently from single droplets. Pioneer work of such problem named group combustion was carried out by Chiu and co-workers [3, 4]. In their works, four possible combustion modes of droplet clouds are identified, i.e. single droplet combustion, internal group combustion, external group combustion, and external sheath combustion. A dimensionless group number G has been derived as criteria for modes transition. In convection-free ambient, $G = 2\pi n d R_c^2$, where n is the number density of droplet, d is the droplet diameter, and R_c is the cloud radius. However, some major assumptions in these works still need to be relaxed, including the quasi-steady process, one-step global mechanism in chemical reaction, and no convection etc., to better simulate the real case. It was found that quasi-steady assumption is only reasonable to the steadily combustion process, before which the flame has been well established, whereas the ignition of the droplet clouds is highly transient. After noticing this, transient ignition process was then investigated. Laster and Annamalai [5] studied the relationships between ignition delay of the droplet cloud and three cloud parameters, i.e. *n*, *d*, R_c mentioned above. Special structure was addressed as that ignition and vaporization happen at the edge of the cloud in this work for non-dilute droplet clouds, which conflicts with the traditional quasi-steady group combustion theory. Thibaut and Candel [6] investigated the ignition modes of dense droplet clouds in a homogeneously hot ambient, and found two ignition modes, i.e. internal homogeneous ignition and external diffusion ignition modes. Chiu and Lin [7] performed a numerical study on the transient ignition phenomena of an initially cold droplet clouds suddenly immersed into hot ambient. It was noted that anomalous combustion conflicting traditional group combustion theory happens. Internal ignition and combustion may happen even for a dense droplet cloud. These works all show the discrepancies between transient ignition phenomena and classical group combustion theory predictions, and help to gain more knowledge in such scenarios. However, in all their previous works, one-step global mechanism was still applied to deal with chemical reaction, which loses sense when considering the possible two-stage ignition in real fuel. In real engines, first-stage ignition happens before the final hot ignition, which is proved to be important in combustion control and is getting more and more attention nowadays [8, 9]. As a result, the competitions between vaporization and two-stage ignition require further study, which is the aim of this work.

2. Physical Model

One-dimensional spherical droplet cloud put into hot ambient has been simulated numerically. The governing equations here are similar to those in [7], and have been listed below.

$$\frac{\partial \rho_g}{\partial t}+\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\rho_g v_g\right)=S_\rho \tag{1}$$

$$\rho_g\left(\frac{\partial Y_{g,i}}{\partial t}+v_g\frac{\partial Y_{g,i}}{\partial r}\right)=-\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\rho_g Y_{g,i}V_{g,i}\right)+\omega_{r,i}+S_\rho\delta_{i,fuel}, \quad i=1\cdots N \tag{2}$$

$$\rho_g C_{p,g}\left(\frac{\partial T_g}{\partial t}+v_g\frac{\partial T_g}{\partial r}\right) =\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\left(k_g\frac{\partial T_g}{\partial r}\right)\right)-\rho_g\sum_{i=1}^{N}\left(Y_{g,i}V_{g,i}C_{p,g,i}\right)\frac{\partial T_g}{\partial r}-\sum_{i=1}^{N}\left(\omega_{g,i}H_{g,i}\right)+S_e \tag{3}$$

where *r*,*t* are the radial position and time. ρ, v, Y, T are respectively density, velocity, mass fraction and temperature. Subscript *g*,*i* represent gas phase and species index (total number *N*). $\delta_{i,fuel}$ is the Dirac delta for fuel species. C_p is the specific heat at constant pressure, and *k* is the thermal conductivity. ω means the sources from reaction, whereas *S* means source from vaporization. The former is calculated via Strang-Splitting method [10] using CHEMKIN II, with a skeletal mechanism of 88 species [11]. The latter is calculated using the models below.

$$S_\rho = n\dot{m}, \quad S_e=-H_v S_\rho \tag{4}$$

$$\dot{m}=4\pi k_g/C_{p,g}r_d\, ln(1+B) \tag{5}$$

$$B = C_{p,g}(T_g - T_b)/H_v \tag{6}$$

where H_v is the latent heat of liquid fuel. r_d is the droplet radius, B is the Spalding transfer number, and T_b is the boiling temperature of fuel liquid. It should be noted that in the quiescent environment, the Nusselt number equals to 2, which finally forms Eq. (5). This vaporization model is a simple Godsave-Spalding form as also used in many other researches [3, 4, 7].

3. Results and Discussion

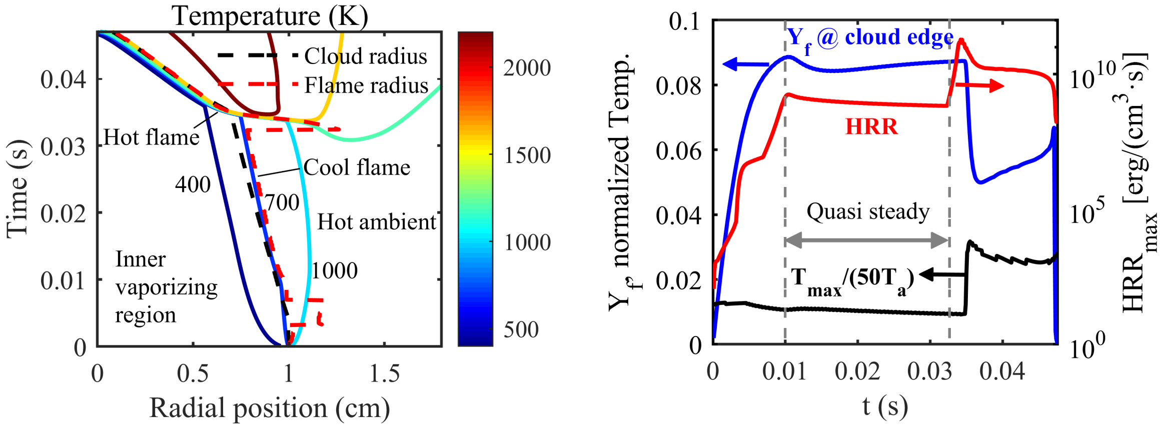
3.1 A typical droplet cloud two-stage ignition process in hot ambient

The simulative process is that a relatively cold droplet cloud has been suddenly put into an initially hot ambient. After that, due to the combining effects of vaporization and transport, there exists stratified temperature and species (fuel and oxidizer) profiles from cloud edge to undisturbed ambient. Spontaneous autoignition may occur in such condition. To illustrate the characters for the spontaneous autoignition behaviour, a representative case is detailly discussed in below. The conditions are listed in Table 1 for the convenience of reference.

Table 1 - Parameters and conditions for the representative case of two-stage ignition of a droplet cloud considered in this study

Name	Symbol	Unit	Value
Fuel	-	-	n-heptane
Initial diameter	d_0	μm	50
Number density	n	cm^{-3}	2000
Initial cloud radius	R_c	cm	1
Initial temperature inside the cloud	T_i	K	400
Initial ambient temperature	T_a	K	1200
Pressure	P	bar	1

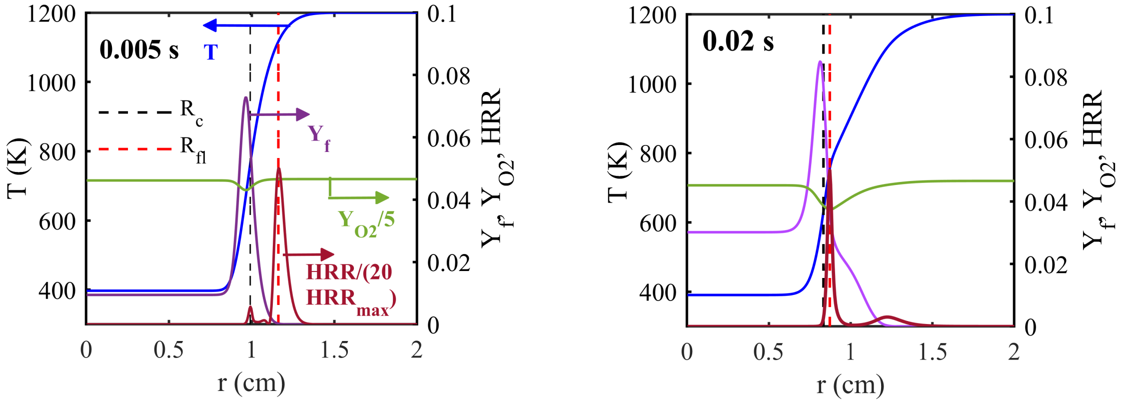
Figure 1 shows the computed field of temperature (1a) and some quantities' temporal variation (1b), where information of the ignition process at different radial position and time has been presented. In Fig. 1a, the ignition process can be divided into different regions in space-time axes: inner vaporizing region, hot ambient, cool flame front and hot flame front, as pointed out in the figure. The cloud radius (black dashed line) shrinks with time as vaporization goes, which is the boundary between inner vaporizing region and hot ambient. Inside the cloud, the inner region is slowly vaporizing due to the moderately high initial temperature, i.e. 400 K in this case. Because of the homogeneous nature for both liquid and gas phase inside the cloud, the vaporization behaviour of droplets should be the same, and hence there are no significant heat and mass transfers inside. In such case, the inner region should behave in zero-D manner. However, as a result of sharp temperature gradient from cold inner region to hot ambient, there must exist an intensely vaporizing boundary layer [12], which conflicts the zero-D feature. This thin layer structure can be observed qualitatively in Fig. 1a as between isotherm of 400 K and cloud radius. The entire processes can be described as follows by observing Fig. 1a and 1b. At the very beginning, there exists a short period of heating up and vaporization of the droplet cloud prior to first-stage ignition, during which the thin vaporizing layer is being established, which can be found by the isotherm 400 K and cloud radius. At approximate 0.01 s, first HRR peak appears and keeps almost stable (Fig. 1b), which indicates the existing of a stable cool flame front. At 0.03 s, the second HRR peak forms, where hot ignition happens. After that, hot flame burns until the cloud radius approaches zero, i.e. the burn-out of the droplet cloud. The flame radius (red dashed line) in Fig. 1a represents the maximum chemical heat release in gas phase. In the early stage transient process (before 0.01 s), the time scales for vaporization, diffusion and chemistry are close, thus strong competition among these processes exists. These competitions result in the stretching and shrinking behaviours of flame radius prior to the first stage ignition. After first stage ignition, the flame radius slightly exceeds the cloud radius, forming an external group combustion regime, which coincides the quasi-steady period in Fig. 1b. At the hot flame ignition instant, the flame radius stands out of the cloud, but soon shrinks back and coincides with cloud radius.



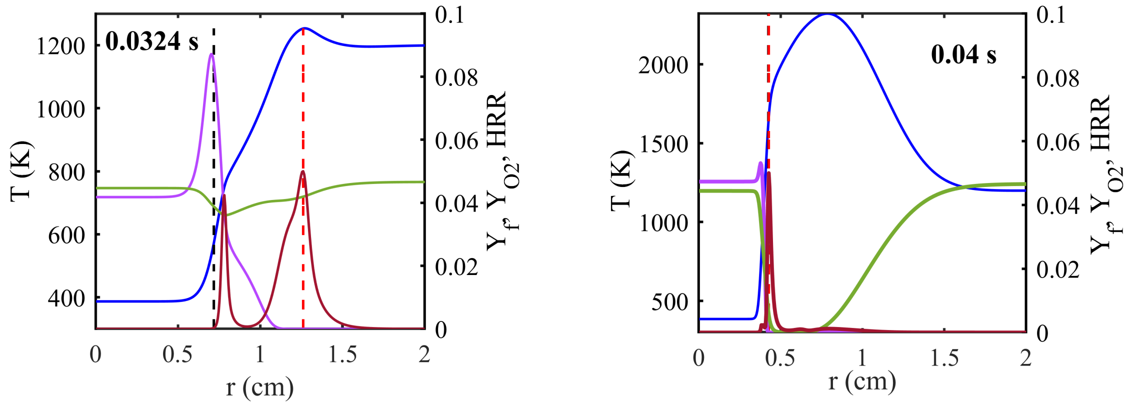
(a) (b)

Figure 1. (a) Temperature isotherms and different regions; (b) Fuel mass fraction at the droplet cloud edge, normalized temperature, and maximum heat release rate variation with time.

Figure 2 further selects representative instant to analyse the flame structure. At 0.005 s (Fig. 2a), after a short period of heating and vaporization, hot flame first gets auto-ignited by the hot ambient, as shown by the second peak of normalized HRR. However, it fails to burn steadily due to the lack of enough fuel vapor supply. It should be noted that the fuel concentration at the edge of the cloud is extremely low for droplet cloud, nearly 1/8 of the value of isolated droplet with liquid-gas interface. After this transient failure of hot flame, low temperature reaction adjacent to the edge of the cloud takes the lead, as shown by the first peak in Fig. 2b. At the same time, high temperature reaction still exists as observed by the second small peak of HRR curve. In this special flame structure, the products from low temperature reaction are transported outwards to a hot ambient, forming a high temperature reaction zone. Because of the low fuel consumption feature of cool flame, the overall performance of droplet cloud stays almost quasi-steady. After a short delay time, hot flame ignites again outside the cool flame front, which is evident in the HRR curve of Fig. 2c. Due to the limited fuel mass fraction (purple curve in the figure), the hot flame soon shrinks back and collapse to the cloud edge. Fig. 2d shows the profiles of the final stage. Such profiles of fuel and oxidizer species refers to a premix flame structure propagating inwards the droplet cloud.



(a) (b)



(c)

(d)

Figure 2. Radial profiles of temperature, species mass fraction (fuel and oxidizer), and heat release rate at different instant picked from Fig. 1: (a) 0.005 s; (b) 0.02 s; (c) 0.0324 s; (d) 0.04 s. The black dashed line points out the position of R_c, the cloud radius, while the red dashed line points out the position of R_{fl}, the flame radius.

Figure 3 provides proofs for cool flame and hot flame by showing the species distribution. Fig. 3a and 3b are respectively the contours of reactants and products, while Fig. 3c-Fig. 3f are the important intermediates for low or high temperature reactions. Fig. 3a shows a three-region vaporization structure. The inner region vaporizes homogeneously, and hence the mass fraction of fuel vapor rise gradually along time axis. A bright peak can be found near the edge of the cloud, which indicates a boundary layer like structure. Beyond this peak, the vapor stretches out, to meet with the hot ambient, where cool flame and hot flame occurs. As Fig. 3d-Fig. 3f tell, ket mainly stick with the cloud radius, showing an external cool flame burning mode. In the meanwhile, intermediate species like CH_2O and H_2O_2 will further stretch out together with fuel vapor. This feature indicates that beyond the cool flame front, there exist a transition zone from low temperature reaction back to high temperature reaction. After that, hot flame ignites suddenly beyond 0.03 s, where the fuel vapor (Fig. 3a) and intermediates (Fig. 3d-Fig. 3f) are depleted quickly. The burnt region still remains active elements (e.g. OH shown in Fig. 3c) for reaction due to the lack of fuel vapor supply.

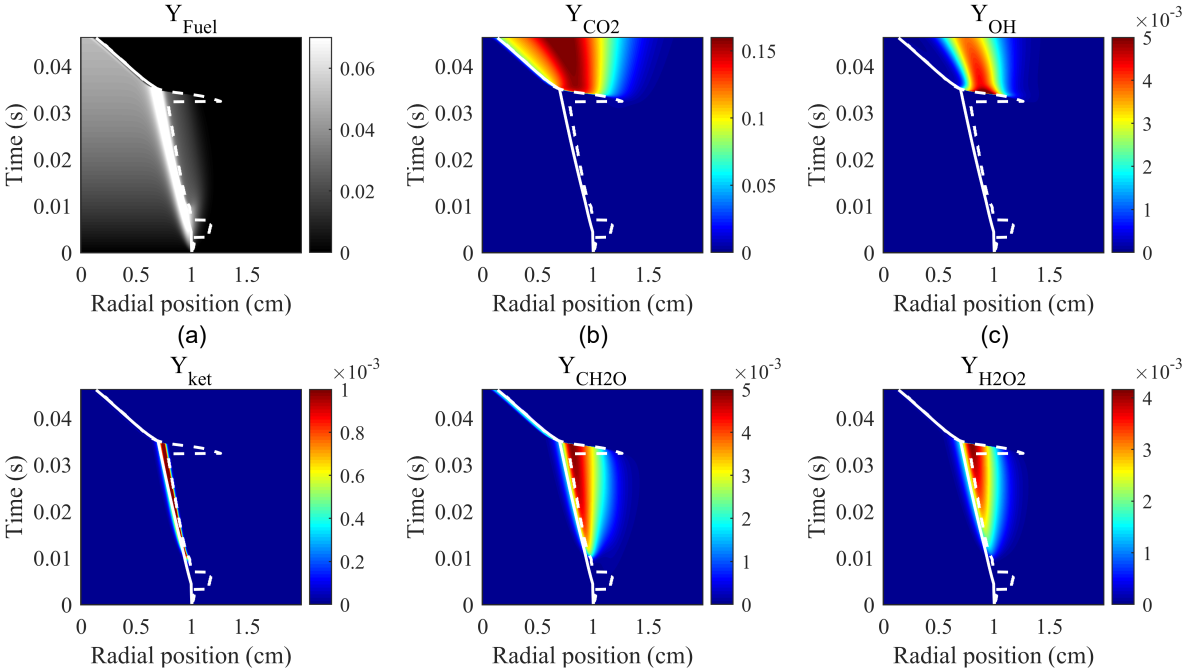
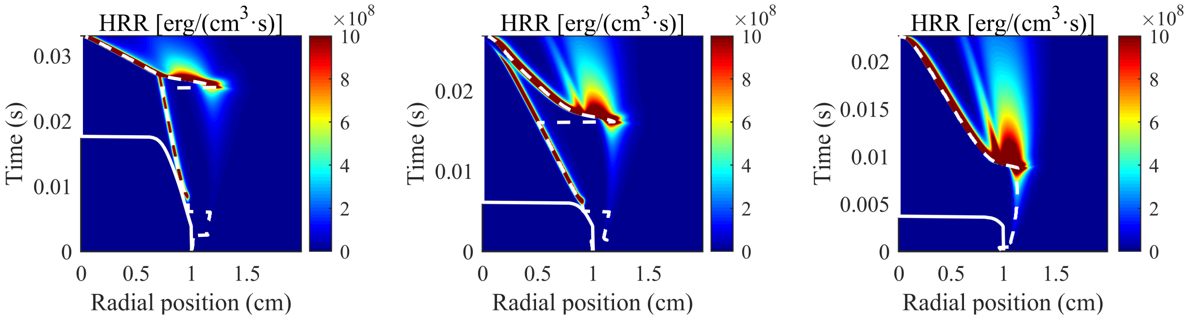


Figure 3. Species contours in radial position vs. time axes: (a) fuel vapour, (b) CO_2, (c) OH, (d) ketohydroperoxide, (e) CH_2O, (f) H_2O_2.

3.2 Influence of inner and ambient temperature

As usually the case in spray, the initial temperature inside the cloud may not be low, which is also the same with ambient temperature. Parametric studies have been done to check the influence of both inner temperature and ambient temperature with cloud parameters same with those in Table 1.

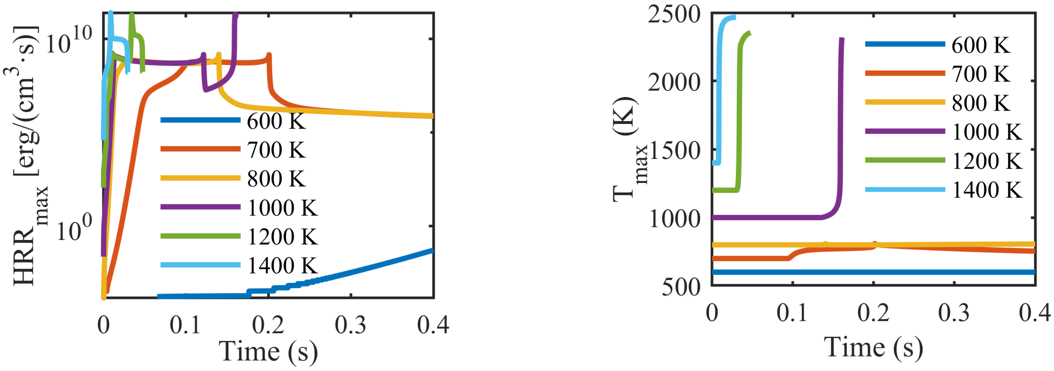
In Fig.4, heat release rate has been chosen to study the chemical reaction in both spatial and temporal axes. As observed in the figures, for the low inner temperature case (4a), the droplet cloud exhibits a typical two-stage ignition, which has been already described in detail above. However, in the moderate inner temperature case (4b), an independent inner cool flame front exists in the entire lifetime after first-stage ignition. In the meantime, high temperature reaction is developing and finally the flammable mixture gets ignited. After that, a steady dual flame structure is obvious. In the high inner temperature case (4c), no cool flame front exists, because the high inner temperature prohibits the establishment cool flame. As a result, only hot flame occurs, after the fast vaporization of the droplet cloud.



(a) (b) (c)

Figure 4. Heat release rate (HRR) contours of cases with different inner temperature: (a) T_i = 500 K, (b) T_i = 700 K, (c) T_i = 900 K. The ambient temperature of these three cases is 1200 K. The white solid lines in figures are cloud radius, while the white dashed lines in figures are flame radius.

To study the influence of ambient temperature, we fix the inner temperature to the low value, i.e. 400 K. Figure 5 shows the maximum heat release rate and temperature of the droplet cloud. From 600 K to 1500 K, one can find that droplet cloud experiences different ignition modes, i.e. no ignition (600 K), single-stage ignition with temperature rise (700 K), single-stage ignition without temperature rise (800 K), and two-stage ignition (>800 K). Apart from the typical two-stage ignition case, the second modes and transition point from single-stage ignition to two-stage ignition are of interest, and will be discussed in the following section.

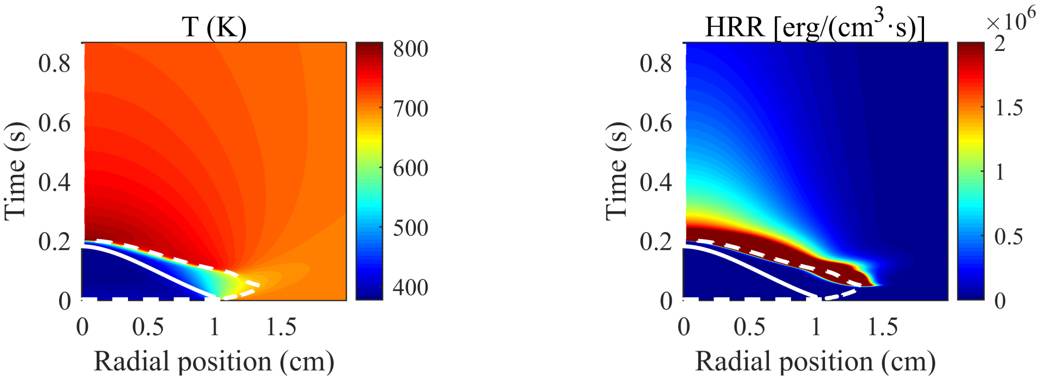


(a) (b)

Figure 5. Temporal variation of (a) maximum heat release rate and (b) maximum temperature at different ambient temperature. The inner temperature in these cases is 400 K.

3.3 Analysis of the near-limit cases

Figure 6a and 6b are the contours of temperature and heat release rate. From the temperature contour, we can observe that after the first-stage ignition at approximately 0.1 s, temperature rises abruptly. After that it goes up gradually to a peak value at nearly 0.2 s, which is because of the ongoing chemical reaction (Fig. 6b). The droplets have all been consumed after that, and the fuel vapor continues to react with oxidizer slowly, which can be tell from the lower value in heat release rate contour. However, hot ignition fails to occur due to the low ambient temperature, and the reaction slows down afterwards. As a consequence, the temperature falls back as shown in Fig. 6a.

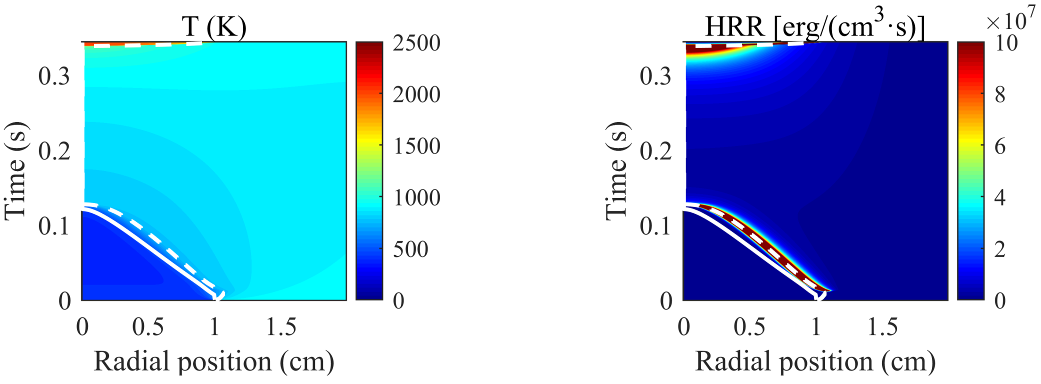


(a)

(b)

Figure 6. Contours of (a) temperature and (b) heat release rate for the single-stage ignition with temperature rise case, i.e. T_a = 700 K. The white solid lines in figures are cloud radius, while the white dashed lines in figures are flame radius.

If further increasing the ambient temperature, hot flame may occur due to the enhancement in high temperature reactivity. The transition point lies at 900 K in the present results, as shown in Fig. 7. Similar with the single-stage ignition case, cool flame has been ignited, and slow reaction holds after droplets are consumed. Difference lies in the successfully ignition at the end. Hot flame only exists for a very short period due to the finite amount of fuel vapor. After this critical temperature, hot flame ignition may happen earlier, even before droplet cloud has been depleted.



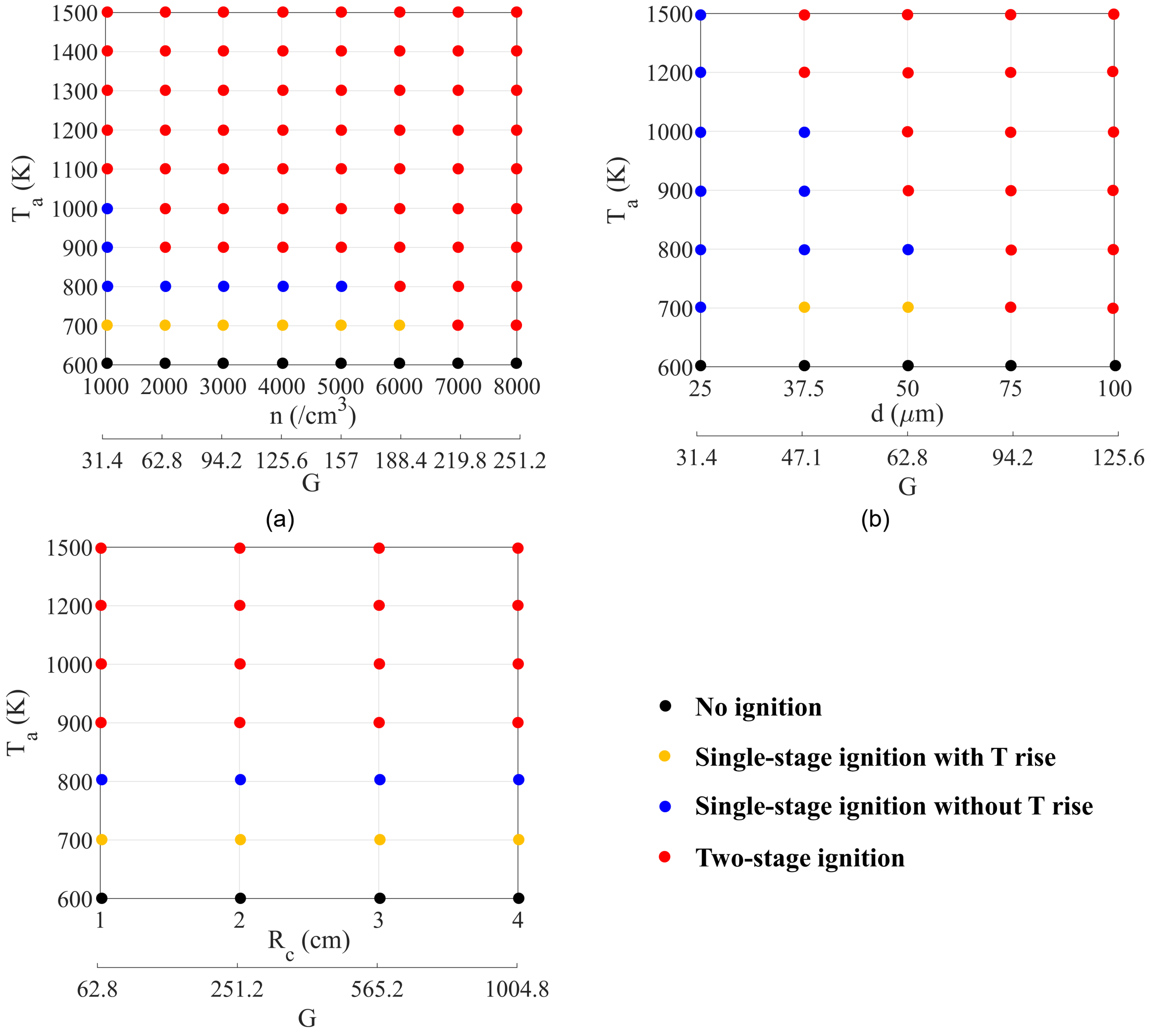
(a)

(b)

Figure 7. Contours of (a) temperature and (b) heat release rate for the transition point between single-stage ignition and two-stage ignition modes, i.e. T_a = 900 K. The white solid lines in figures are cloud radius, while the white dashed lines in figures are flame radius.

3.4 Influence of different cloud parameters on ignition modes

Phase diagrams have been presented in Fig. 8, where the respective group numbers *G* are also shown. From Fig. 8a, ‘single-stage ignition with temperature rise’ mode disappears when droplets number density is larger than 6000 cm^{-3}, while ‘single-stage ignition without temperature rise’ mode disappears when number density is larger than 5000 cm^{-3}. We can thus conclude that increasing number density favours two-stage ignition. This is because when the droplet cloud is denser, the cloud lifetime is longer, so more time is reserved for hot flame ignition. From Fig. 8b, no two-stage ignition mode occurs for 25 μm cases, but it finally transits to two-stage ignition with increasing initial diameter. It appears in Fig. 8c that increasing R_c has no direct effects on ignition modes, which is due to the self-similarity when we consider the heat and mass transfer in a spherical coordinate.



(a)

(b)

(c)

Figure 8. Phase diagrams for ignition modes under different ambient temperature and different cloud parameters: (a) number density, (b) droplet diameter, (c) cloud radius.

Cases with same group number but different sub-parameters can be picked out for comparison, e.g. G = 31.4, 94.2 and 125.6 in 8a and 8b, G = 251.2 in 8a and 8c. From these comparisons, it can be easily concluded that with the same group number, ignition modes are not exactly the same for different sub-parameters, which indicates that group number is not the controlling dimensionless number in ignition modes. As observed from these diagrams, modes depend more on number density and droplet diameter, and less on cloud radius. Both number density and droplet diameter determine the fuel concentration at the edge of the cloud, which shall influence the ignition modes. Besides, whether two-stage ignition occurs depends on the vaporization time of the droplet cloud prior to hot ignition. Number density has first order effect on the vaporization time through total droplets number, whereas droplet diameter has more severe effect on vaporization time, due to its d^3 dependence on droplets total volume. More specifically, the droplet cloud vaporizes in a manner of thin layer, where droplets' total surface area should be an important physical parameter instead of group number. As such, here we recommend a new dimensionless number $G_{ig} = n^{2/3}d^2$, which physically decouples the effects of cloud radius and pays attention to the total surface area in local vaporization layer. After converting the original group number in Fig. 8a and 8b to this new number G_{ig}, we can obtain new values as shown in Table 2. Close values of G_{ig} between two diagrams have been underlined in the table, and by comparing the ignition modes in Fig. 8a and Fig. 8b, we can conclude that this new dimensionless parameter has better predicting ability than classical group number.

Table 2 – Calculated G_{ig} to replace the *G* coordinate in Fig. 8

G_{ig} in 8a	G_{ig} in 8b
<u>0.0025</u>	0.0010
0.0040	<u>0.0022</u>
0.0052	0.0040
0.0063	<u>0.0089</u>
0.0073	0.0159
0.0083	
<u>0.0091</u>	
0.0100	

4. Conclusions

This work numerically studied the autoignition of droplet clouds put into hot ambient, covering wide conditions, i.e. the initial gas temperature inside T_i = 400-1200 K and outside T_a = 500-1500 K of the droplet clouds, droplet diameter d_0 = 25-100 μm, droplet number density n = 1000-8000 cm^{-3}, and cloud radius R_c = 1-4 cm. Major conclusions are listed below:

1) For different cloud parameters and ambient temperature, droplet clouds may exhibit four modes: no ignition, single-stage ignition with temperature rise, single-stage ignition without temperature rise, and two-stage ignition.
2) A typical two-stage ignition includes the following detailed processes: first stage ignition, quasi-steady non-premix cool flame, second stage ignition, premix hot flame.
3) Two near limit case has been analysed. These limit cases are mainly controlled by the competition between chemical reaction and vaporization.
4) Phase diagrams of ignition modes at different ambient temperature and cloud parameters have been presented. It has been found that classical group number cannot predict the ignition modes properly. As a result, a new dimensionless number $G_{ig} = n^{2/3}d^2$ has been raised in this study, which has a much better ability in recognizing different ignition modes.

In this work, the droplet cloud is assumed to be stationary without convection to do the decoupling research. However, droplet clouds may not be ideally spherical and homogeneous in real case due to convection. As a result, quantities such as number density and cloud radius of a real cloud in spray should be converted to those of an equal spherical and homogeneous droplet cloud in order to use these conclusions.

Acknowledgments

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