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## Abstract

Directly using liquid ammonia in combustion devices can simplify the fuel supply system and reduce equipment costs. However, the detailed ignition and propagation mechanisms of liquid ammonia flames have not been fully understood. This study aims to investigate the ignition and propagation of liquid ammonia flames numerically under various conditions. Spherically expanding liquid ammonia flames are studied using detailed chemistry. The effects of initial temperatures, droplet diameters, equivalence ratios, and heat fraction are investigated. The results indicate that the rapid flash boiling of liquid ammonia droplets leads to a strong local heat loss, hindering the ignition and propagation of the flame. Preheating can significantly increase the reaction rate, which compensates for the heat loss caused by phase change, allowing the flame kernel to ignite and propagate outwards. While droplet-flame interaction is present but not significant due to the rapid evaporation of liquid ammonia. The effect of droplet on the flame surface curvature is only distinguishable at conditions of large diameter. Equivalence ratio and ammonia heat fraction ratio change the amount of liquid ammonia. The more liquid ammonia, the more heat is absorbed by the phase change process, making the flame more difficult to be ignited.

*Keywords:* Liquid ammonia combustion, Numerical simulation, Spherically expanding flames, Ignition and propagation

## 1 Introduction

In order to achieve the goal of reducing carbon emissions and sustainable energy use, ammonia, as one of the alternative and carbon-free fuels, has recently received more and more attention. Because of the well-established infrastructure for ammonia storage and transport [1,2], ammonia has broad prospects in the long-term goal of reducing carbon emissions. Remarkable progress has been made in recent decades. Much basic and applied research has been carried out on ammonia combustion. For example, the fundamental combustion characteristics of ammonia [3] and its co-firing with methane [4] and hydrogen [5] flames, such as flame speed and emission, have been advanced on various fronts. The investigation of using ammonia in many engineering devices, including internal combustion engines (ICEs), marine engines, and gas turbines, has also been explored in several experimental [6,7], analytical [8] and numerical studies [9,10] in terms of flame stability, emission control and flame characteristics. These studies have mainly focused on the combustion of gaseous ammonia. However, ammonia is usually stored in the liquid phase. Direct combustion of liquid ammonia can reduce the cost and complexity of fuel supply systems for engineering devices, which reduces about 7 - 9 % of the consumption of generated power for the vaporisation process and the start-up time in gas turbines (GTs) [11]. There is also no need to modify the fuel supply system of ICEs too much [12], and liquid ammonia is suitable for ICEs with high octane ratings [13]. However, relatively little effort has been devoted to the study of liquid ammonia combustion.

Several studies have investigated the liquid ammonia combustion in ICEs and GTs. Li et al. [12] investigated the dual-fuel modes of liquid ammonia combustion engines both experimentally and numerically, specifically the low- and high-pressure injection dual-fuel (LPDF and HPDF) modes. This study pointed out that the maximum ammonia fraction is about 80% and 97% in LPDF and HPDF modes, respectively. The thermal efficiency, exhaust loss, and emission characteristics were also compared with those of the pure diesel engines. The results shown that LPDF had the potential to achieve a higher efficiency and HPDF can reduce the emission. Zhou et al. [14] further studied the liquid ammonia/diesel dual fuel marine engines in HPDF mode experimentally and numerically. They indicated that the NO<sub>x</sub> emissions level can be reduced by about 47% in the optimized HPDF mode. Liu et al. [15] numerically investigated the combustion and emission characteristics of liquid ammonia preliminary based on a two-stroke marine engine. Zhang et al. [16] further studied the performance of a two-stroke liquid ammonia/diesel engine through experimental validation and evaluation under different conditions. They indicated that liquid ammonia flame can be enhanced by the ignition of diesel flame and the thermal efficiency was also increased. Meanwhile, several works about liquid ammonia combustion were conducted in the context of gas turbines. Zembi et al. [17] investigated the injection process of ammonia using Reynolds Averaged Navier Stokes (RANS) coupled with the Lagrangian particle method. The numerical results were validated by the experimental data. The flash boiling phenomenon was investigated in wide range conditions. The results indicated that there are challenges to capture the local details of liquid ammonia spray in all the regimes using a single model or setup. Okafor et al. [11,18] investigated the emissions and flame stability of liquid ammonia combustion in the single- and two-stage swirl combustors. Some techniques were employed to enhanced the combustion stability of liquid ammonia flame in terms of increasing the combustor inlet temperature (CIT), reducing the heat loss from wall, and blending methane. They pointed out that the pure liquid ammonia can not be ignited without the preheating because liquid ammonia has large latent heat. Liquid ammonia spray flame could stabilize after preheating, ever though the flame height was significant large. Blending methane can reduce the flame height, and the flammability limit was increased correspondingly.

In the past two years, several studies on liquid ammonia combustion have been conducted as discussed above [11–18]. However, most research analyzed the macroscopic characteristics, i.e. thermal efficiency, flame structure/stability, and emission phenomenon. More detail and fundamental works are still lacked. The mechanism of stability, ignition, and emission formation for liquid ammonia flames need to be investigated. The turbulent combustion dynamics of ammonia droplet-laden mixtures, which involve the interplay of flash boiling, heat loss, mass transfer, reaction thermal chemistry, and flame surfaces, require further exploration. Despite being of great interest, there have been limited studies on this subject. Hence, the objective of this work is to gain a deep understanding of the ignition and propagation mechanisms of liquid ammonia flames by investigating spherical flame propagation in droplet-laden mixture configurations, using numerical simulations with detailed chemistry. The main focuses of this study are as follows:

- 105 • To investigate the unique characteristics of liquid ammonia spray flames in  
106 comparison with those of gaseous conditions.
  - 107 • To clarify the mechanism of flame ignition and propagation caused by the high  
108 initial temperature, and explain experimental phenomena through the  
109 turbulent spherically expanding spray flames.
  - 110 • To study the effects of droplets diameter, equivalence ratio, and heat fraction  
111 on the ignition and propagation of liquid ammonia spray flames.
- 112 The rest of the paper is organized as follows. The mathematical method and  
113 computational details are introduced in Secs. 2 and 3, respectively. Based on these  
114 methods, the results are subsequently investigated and discussed in Sec. 4. The main  
115 findings are summarized in Sec. 5.

## 116 2 Mathematical framework

117 In the present study, simulations of the spherically expanding turbulent flames  
118 of liquid ammonia are performed using the Eulerian–Lagrangian framework [19–21].  
119 Conservation equations of the gas phase are directly solved without any filtering and  
120 averaging. For the dispersed phase, the Lagrangian method is used to capture the  
121 dynamics of droplets. The two-way coupling approach is used to consider the gas and  
122 particle interactions. This modeling stratagem is similar that in as our previous study  
123 of liquid ammonia spray simulation [22], and is extended to reacting flows in this  
124 study.

### 125 2.1 Governing equations of gas phase

126 The governing equations for mass, momentum, energy, and species mass  
127 fraction of the gas phase are solved as follows:

$$128 \quad \frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{u}) = \dot{S}_\rho, \quad (1)$$

$$129 \quad \frac{\partial(\rho \mathbf{u})}{\partial t} + \nabla \cdot (\rho \mathbf{u} \mathbf{u}) = -\nabla p + \nabla \cdot \boldsymbol{\tau} + \rho \mathbf{g} + \dot{\mathbf{S}}_{\mathbf{u}}, \quad (2)$$

$$130 \quad \begin{aligned} \frac{\partial(\rho h)}{\partial t} + \nabla \cdot (\rho \mathbf{u} h) &= \frac{\partial p}{\partial t} + \mathbf{u} \cdot \nabla p + \nabla \cdot (\rho D_h \nabla h) \\ &+ \boldsymbol{\tau} : \nabla \mathbf{u} + \dot{S}_{\text{reac}} + \dot{S}_h + \dot{S}_{\text{rad}}, \end{aligned} \quad (3)$$

$$131 \quad \frac{\partial(\rho Y_i)}{\partial t} + \nabla \cdot (\rho \mathbf{u} Y_i) = \nabla \cdot (\rho D_i \nabla Y_i) + \dot{S}_{\text{evap}, \text{NH}_3} + \dot{S}_{\text{comb}, i}, \quad (4)$$

132 where  $\rho$ ,  $\mathbf{u}$ , and  $p$  are the carrier phase density, velocity, and pressure, respectively.  $\boldsymbol{\tau}$   
133 is the viscous stress tensor.  $\mathbf{g}$  is the gravitational acceleration and  $h$  is the specific  
134 sensible enthalpy.  $\dot{S}_{\text{reac}}$ ,  $\dot{S}_h$ , and  $\dot{S}_{\text{rad}}$  are the source term for reaction, evaporation,  
135 and radiative heat, respectively.  $Y_i$  is the mass fraction of species  $i^{\text{th}}$ .  $D_h$  and  $D_i$  are

the thermal diffusivity and mass diffusivity, respectively.  $\dot{S}_{evap,NH_3}$  and  $\dot{S}_{comb,i}$  represent the source terms originated from evaporation and combustion, respectively. The reaction mechanism developed by Okafor et al. [4] is used to account for the gas phase chemistry (59 species and 356 reactions), which has been validated at various conditions [4,10,23]. The Particle-Source-In-Cell (PSIC) method [24] is employed to account for two-way coupling source terms, which are responsible for describing gas and particle interactions.

$$\dot{S}_\rho = -\frac{1}{\Delta V} \sum_i^N \frac{dm_d}{dt}, \quad (5)$$

$$\dot{\mathbf{S}}_{\mathbf{u}} = -\frac{1}{\Delta V} \sum_i^N \frac{dm_d \mathbf{u}_d}{dt}, \quad (6)$$

$$\dot{S}_h = -\frac{1}{\Delta V} \sum_i^N \frac{d(c_p m_d T_d)}{dt}, \quad (7)$$

$$\dot{S}_{evap,NH_3} = -\frac{1}{\Delta V} \sum_i^N \frac{dm_d}{dt}, \quad (8)$$

where, volume of the local computational cell is denoted by  $\Delta V$ , and  $N$  represents the number of droplets present in that cell.  $m_d$ ,  $\mathbf{u}_d$ ,  $c_p$ , and  $T_d$  are droplet mass, velocity, specific heat capacity, and temperature, respectively. In case of pure gaseous flames,  $\dot{S}_\rho = \dot{\mathbf{S}}_{\mathbf{u}} = \dot{S}_h = \dot{S}_{evap,NH_3} = 0$ .

## 2.2 Governing equations of dispersed phase

The equations of droplets position  $\mathbf{x}_{d,i}$ , velocity  $\mathbf{u}_{d,i}$ , and temperature  $T_d$  are solved as follows:

$$\frac{d\mathbf{x}_{d,i}}{dt} = \mathbf{u}_{d,i}, \quad (9)$$

$$\frac{dm_d \mathbf{u}_{d,i}}{dt} = m_d \frac{3\mu C_D Re}{4d^2 \rho_d} (\mathbf{u}_i - \mathbf{u}_{d,i}) + m_d \mathbf{g}_i, \quad (10)$$

$$\frac{dT_d}{dt} = f h_t \frac{A_d}{m c_p} (T_\infty - T_d) - \frac{dm}{dt} h_v + A_d \epsilon \left( \frac{1}{4} G_c - \sigma T^4 \right), \quad (11)$$

where  $m_d$  and  $\mathbf{u}_{d,i}$  are droplet mass and velocity component, respectively;  $C_D$  is the drag coefficient [25];  $d$  and  $\rho_d$  are the droplet diameter and density, respectively;  $\mathbf{u}_i$  is the gas phase velocity and  $\mathbf{g}_i$  represents the gravitational acceleration.  $f$  is the correction coefficient given in [26], calculated by  $f = \frac{-c_{p,v} \dot{m}}{\pi d k Nu} / \left( e^{\frac{-c_{p,v} \dot{m}}{\pi d k Nu}} - 1 \right)$ . Where  $A_d$  is each droplet's surface area;  $h_t$  is the heat transfer coefficient, where  $h_t = \frac{k Nu}{d}$ ; and

$k$  is the thermal conductivity. Nusselt number ( $Nu$ ) is calculated using the Ranz Marshall heat transfer model [27],  $Nu = 2 + 0.6Re^{1/2}Pr^{1/3}$ ;  $G_c$  is radiation coefficient,  $\epsilon$  is emissivity coefficient, and  $\sigma$  is scattering coefficient.  $dm_d/dt$  is calculated using the phase-change model.

The combined model [28,29] is used to describe the phase change process, as it has been validated under flash boiling conditions by comparing with the experimental data in our previous study [22]. It is briefly introduced in the following for completeness, and the interested readers can refer to Ref. [22] for the details. In case of evaporation conditions, the mass of phase change is calculated by

$$\frac{dm}{dt} = \frac{m_d}{\tau_d} \left( \frac{Sh}{3Sc} \right) \ln(1 + B_M), \quad (12)$$

where  $\tau_d$  is the droplet response time, calculated by  $\tau_d = \frac{\rho_d d^2}{18\mu_c}$ , where  $\rho_d$ ,  $d$ , and  $\mu_c$  are droplet density, diameter, and dynamic viscosity of the carrier gas, respectively. The Sherwood  $Sh$  and Schmidt  $Sc$  numbers are given by  $Sh = 2.0 + 0.552Re^{1/2}Sc^{1/3}$  and  $Sc = \mu/(\rho D_i)$ , respectively.  $B_M$  is the Spalding mass transfer number evaluated as  $B_M = \frac{Y_s - Y_\infty}{1 - Y_s}$ , where  $Y_s$  and  $Y_\infty$  represent the vapor mass fractions at the droplet surface and in the surrounding region, respectively. In case of boiling conditions, the mass of phase change is calculated by

$$\frac{dm}{dt} = \dot{m}_{flash} + \dot{m}_t. \quad (13)$$

The total phase change rate is contributed by superheated flash boiling rate  $\dot{m}_{flash}$  and the evaporation rate caused by outer heat transfer  $\dot{m}_t$ . The flash boiling part is given by

$$\dot{m}_{flash} = \pi d^2 \alpha_s \frac{(T_d - T_b)}{h_v}, \quad (14)$$

where  $T_d$  and  $T_b$  are the internal and boiling temperatures of droplets, respectively.  $\alpha_s$  and  $h_v$  are overall heat transfer coefficient and the latent heat of vaporization, respectively. The Adachi correlation is used to calculate the value of  $\alpha_s$ , which is then used to determine the overall heat transfer coefficient [30]. The other part of evaporation rate is evaluated by

$$\dot{m}_t = \pi d \frac{KSh}{c_p} \frac{1}{1 + \dot{m}_{flash}/\dot{m}_t} \times \ln \left( 1 + \frac{h_g - h_s}{h_v} \left( 1 + \frac{\dot{m}_{flash}}{\dot{m}_t} \right) \right), \quad (15)$$

where  $K$  represents the thermal conductivity.  $h_g$  and  $h_s$  represent the enthalpy of the carrier gas and vapor, and latent heat of liquid ammonia vaporization, respectively.

### 3 Computational setup and solution algorithm

Centrally-ignited spherically expanding liquid ammonia flames with different unburnt temperatures ( $T$ ), droplet diameters ( $d$ ), equivalence ratios ( $\phi$ ), and ammonia heat value fractions ( $\xi_{NH_3}$ ) are considered for the present investigation as listed in Table 1. The two-dimensional (2-D) computational domain has a size of  $L_x \times L_y = 20 \times 20 \text{ mm}^2$ , with a uniform mesh resolution of  $512 \times 512$ , as shown in Fig. 1(a). The mesh resolution is  $39 \mu\text{m}$ , which not only ensures more than 20 grid points within the flame thermal thickness but also ensures the minimum size of grid is larger 2.6 times than droplet diameter.

Table. 1: Summary of the conditions and parameters.

| I<br>D | $\phi$<br>[-] | $\xi_{NH_3}$<br>[-] | $T$<br>[K] | $d$<br>[ $\mu\text{m}$ ] | $n_d$<br>[-] | $S_l$<br>[cm/s] | $\delta_l$<br>[ $\mu\text{m}$ ] | $\eta_k$ [1]<br>[ $\mu\text{m}$ ] | $Re_t$<br>[2]<br>[-] | $Ka$<br>[3]<br>[-] | $Da$ [4]<br>[-] |
|--------|---------------|---------------------|------------|--------------------------|--------------|-----------------|---------------------------------|-----------------------------------|----------------------|--------------------|-----------------|
| 0      | 0.9           | 1.0                 | 500        | 10                       | 3720         | 16.83           | 1456.48                         | 118.47                            | 84.44                | 0.24               | 38.14           |
| 1      | 0.9           | 1.0                 | 500        | -                        | -            | 16.83           | 1456.48                         | 118.47                            | 84.44                | 0.24               | 38.14           |
| 2      | 0.9           | 1.0                 | 300        | 10                       | 6201         | 5.71            | 2295.17                         | 60.08                             | 208.81               | 1.76               | 8.21            |
| 3      | 1.1           | 0.7                 | 500        | 5                        | 25145        | 28.23           | 982.54                          | 118.53                            | 84.38                | 0.097              | 94.82           |
| 4      | 1.1           | 0.7                 | 500        | 10                       | 3143         | 28.23           | 982.54                          | 118.53                            | 84.38                | 0.097              | 94.82           |
| 5      | 1.1           | 0.7                 | 500        | 15                       | 931          | 28.23           | 982.54                          | 118.53                            | 84.38                | 0.097              | 94.82           |
| 6      | 0.9           | 0.7                 | 500        | 10                       | 2668         | 24.54           | 1039.89                         | 118.26                            | 84.63                | 0.12               | 77.88           |
| 7      | 1.0           | 0.7                 | 500        | 10                       | 2910         | 27.81           | 976.93                          | 118.40                            | 84.50                | 0.098              | 93.94           |
| 8      | 1.1           | 0.8                 | 500        | 10                       | 3555         | 25.58           | 1083.60                         | 118.61                            | 84.31                | 0.12               | 77.90           |
| 9      | 1.1           | 0.9                 | 500        | 10                       | 3958         | 23.36           | 1185.25                         | 118.69                            | 84.23                | 0.14               | 65.04           |

[1] Kolmogorov length scale,  $\eta_k = l_t Re_t^{-0.75}$ , where  $l_t$  is integral length scale measured directly from the initial turbulence field.

[2] Turbulent Reynolds number,  $Re_t = u' l_t / \nu$ , where  $u'$  is root-mean-square (RMS) turbulent fluctuation velocity.

[3] Karlovitz number,  $Ka = (\delta_l / \lambda)(u' / S_l)$ , where  $\delta_l$  is laminar flame thickness,  $\lambda$  is Taylor length scale,  $\lambda = l_t Re_t^{-0.5}$ , and  $S_l$  is laminar burning velocity.

[4] Damkohler number,  $Da = (l_t / \delta_l)(S_l / u')$ .

All boundaries have the Periodic boundary conditions. The left and right parts of Fig. 1(a) demonstrate the initial velocity and temperature fields with droplets, respectively. The initial flame kernel locates at the center with the radius of  $r_0 = 3.3$  mm, as seen in Fig. 1(a). The fresh mixture with ammonia droplets (randomly distributed) is filled the rest space. The ignition kernel is initialized by the burnt gas of fresh mixture in certain conditions. The initial values of temperature and species are transformed into smooth profiles according to  $\phi = \phi_0 + \frac{\Delta\phi}{2} \left( 1 - \tanh \left( s \cdot \frac{r-r_0}{r_0} \right) \right)$ , where  $\Delta\phi$  represents the difference between initial values of burnt and fresh mixture. The constant  $s$  measures the stiffness of interface between fresh and burnt, which is set to 10 following Refs. [31,32].

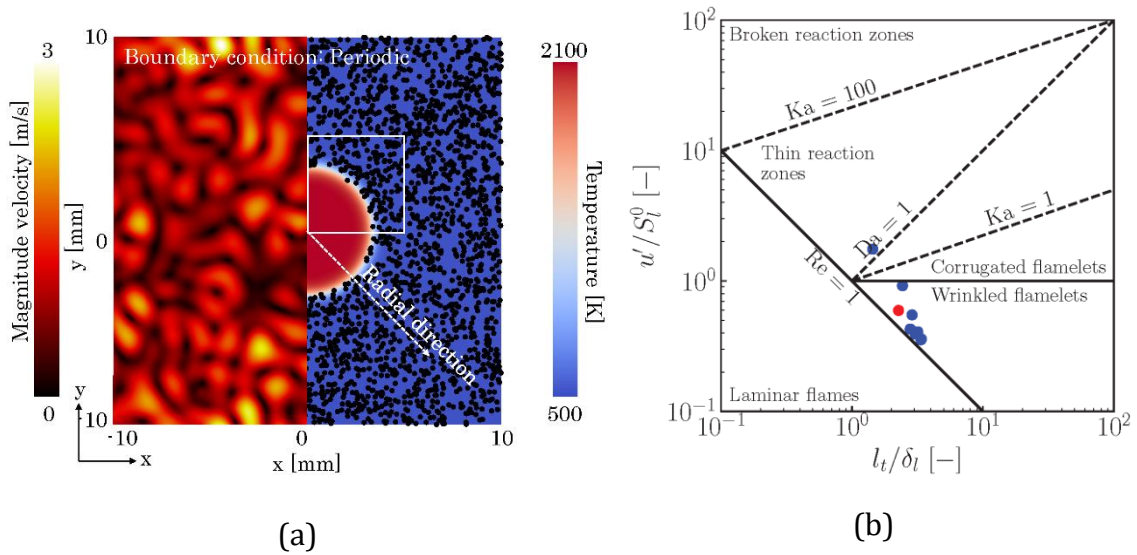


Figure1: (a) The computational domain and initial configuration of velocity (left) and temperature fields (right). The black dots and red field are liquid ammonia droplets and burnt gas, respectively. The burnt gas is used as ignition kernel. (b) Peters-Borghi's diagram contains the simulation conditions of liquid (blue) and gaseous (red) ammonia cases.

In total, ten cases are calculated as listed in Table 1, and the conditions are within the thin reaction and flamelets zones according to the Peters-Borghi's diagram as shown in Fig. 1(b). The initial turbulent flow is generated by using the Passot and Pouquet isotropic kinetic spectrum [33]. The turbulent kinetic energy is calculated by

$$E(k) = \frac{32}{3} \sqrt{\frac{2}{\pi}} \frac{u'^2}{k_e} \left( \frac{k}{k_e} \right)^4 \exp \left[ -2 \left( \frac{k}{k_e} \right)^2 \right], \quad (16)$$

where  $u'$  is the fluctuation velocity.  $k$  and  $k_e$  are the wave number and most energetic wave number, respectively. The turbulence integral length scale  $l_t$  and the initial turbulent fluctuation velocity  $u'_0$  are set 3.3 mm ( $L_i/6$ ) and 1 m/s, respectively. Therefore, the initial turbulent Reynolds number,  $Re_t = u' l_t / \nu$ , is from 84.38 to

208.81 for different conditions. The exact values are summarized in Table 1. It should be pointed that the turbulence in the present study is decaying, and the interactions of turbulence and combustion might be affected by this treatment. Specifically, decaying turbulence may decrease the mixing rate and thus slow down the combustion process, and cause the flame to transition from a turbulent flame to a laminar flame, and the flame propagation speed may decrease. The initial value of the minimum Kolmogorov length scale  $\eta_K$  is about  $60 \mu\text{m}$ . The maximum droplet diameter is set to  $15 \mu\text{m}$ , which is smaller than the Kolmogorov length scale for all cases and also the cell size, as suggested by Haruki et al. [34]. The mesh in all the calculations has a uniform size of  $39 \mu\text{m}$ , which could provide sufficient resolution to resolve the Kolmogorov and flame thickness. The Stokes number of droplet is calculated by  $St = \frac{\tau_p \sqrt{k}}{L}$ , where  $\tau_p$  is the particle time scale and  $L_i/\sqrt{k}$  is the turbulent time scale. The Stokes numbers are smaller than 1.0, indicating that droplets will follow the fluid motion well. Therefore, the initial droplet velocity is the same as that of carrier gas, with a temperature of  $T_d^0 = 273 \text{ K}$ . Ammonia flash boiling results in a strong local heat loss due to its large latent heat. Okafor et al. [11] reported that pure liquid ammonia flames could be achieved using an air inlet temperature of 500 K, and preheating the carrier gas is one of the efficient ways for flame stabilization. Therefore, the pre-heating temperature of carrier gas is set to 500 K for all conditions except Case 2, which is for comparison. The ammonia droplets will under the boiling condition in the initial conditions of all cases when the ambient pressure is 1 bar.

The blending ratio, ammonia fraction in the fuel stream is determined in a heat basis,  $\xi_{NH_3}$ , calculated by

$$\xi_{NH_3} = \frac{\chi_{NH_3} LHV_{NH_3}}{\chi_{NH_3} LHV_{NH_3} + \chi_{CH_4} LHV_{CH_4}}, \quad (17)$$

where  $\chi$  and LHV represent the mole fraction and lower heat value, respectively. Note that methane is premixed with the air and ammonia droplets are randomly distributed in the unburnt side. The overall equivalence ratio is calculated from

$$\phi_{overall} = \frac{\frac{m_{CH_4} + m_{NH_3}}{m_{O_2}}}{\left( \frac{m_{CH_4} + m_{NH_3}}{m_{O_2}} \right)_{st}}, \quad (18)$$

where,  $m_{NH_3} = n_d m_d$ .  $n_d$  and  $m_d$  are the droplet number and mass of single droplet, respectively.

Because of droplet evaporation, the mixture will be inhomogeneous. Bilger mixture fraction [35] is used here to identify the local fuel sources by

$$Z_{CH_4} = \frac{4}{3} Y_C, Z_{NH_3} = \frac{17}{3} \left( Y_H - \frac{1}{3} Y_C \right), \quad (19)$$

where  $Y_i$  is the atom mass fraction for element  $i$ . Therefore, the local mixture fraction  $Z$  can be defined by  $Z = Z_{CH_4} + Z_{NH_3}$ . Similarly, the progress variable cannot be defined through oxygen mass fraction and mixture fraction like Refs. [36,37]. Here, the progress variable  $c$  is defined based on the gas temperature to describe the flame evolution and the local reaction process from fresh mixture to burnt gas,

$$c = \frac{T - T_u}{T_b - T_u}, \quad (20)$$

where  $T_u$  and  $T_b$  are fresh and burned gas temperatures, respectively.  $T$  represents the local gas temperature. Flame index proposed by Takeno et al. [38] is used to access the local combustion mode,

$$FI = \frac{\nabla Y_F \cdot \nabla Y_{O_2}}{|\nabla Y_F \cdot \nabla Y_{O_2}| + \epsilon}, \quad (21)$$

where  $Y_F$  and  $Y_{O_2}$  represents mass fractions of fuel and oxidizer, respectively.  $\epsilon$  is an infinitesimal positive quantity to avoid dividing by zero.  $FI = -1$  and  $1$  represent the non-premixed and premixed combustion mode, respectively.

All the calculations were conducted using SprayDCFoam developed based on OpenFOAM-8 [39,40]. The second-order central differencing scheme is used to integrate spatial and transient terms, while the Euler-implicit scheme is utilized for integrating temporal terms. The Courant number is set to be less than 0.1 to ensure the numerical stability, resulting in a time step of about  $1.2 \mu s$ . All the simulations were performed on the supercomputer Fugaku, and each case costs about 14000 core hours by 960 cores parallel calculation.

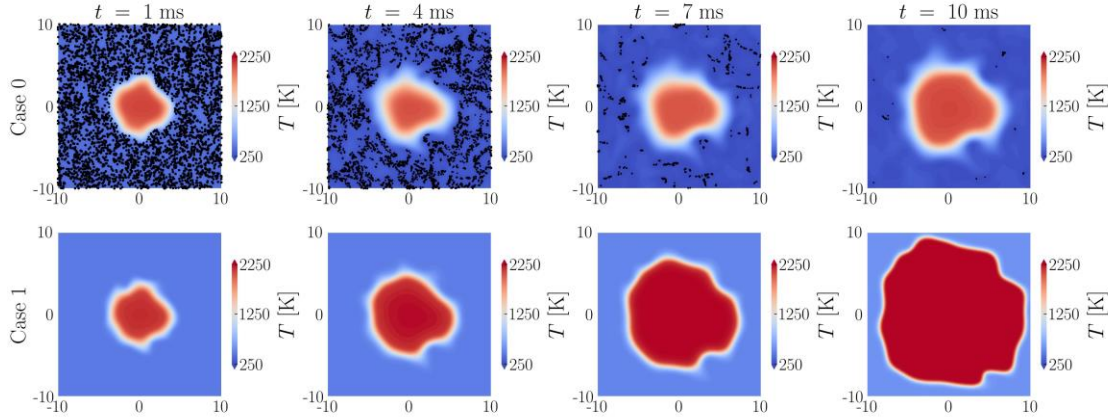
## 4 Results and discussion

In this section, a comparison between liquid and gaseous ammonia flames is performed first. Subsequently, the impact of various parameters such as initial temperature, droplet diameter, equivalence ratio, and heat fraction are also investigated.

### 4.1 Comparison of liquid and gaseous ammonia combustion

Figure 2 shows gas temperature and distribution of ammonia droplets for Cases 0 and 1 from  $t = 1-10$  ms. Cases 0 and 1 calculate the flames of liquid and gaseous ammonia, respectively, under the same initial conditions (see Table 1). The only difference is that the ammonia is present in liquid droplets in Case 0 but gaseous in Case 1. The propagation speed of the gaseous case is faster than that of the liquid case, and the flame kernel and its surrounding temperatures are also much higher in Case 1. This indicates that ammonia droplets significantly hinder the outward propagation of the flame kernel. Before the complete evaporation of liquid droplets in Case 0, the growth flame kernel is not obvious. The droplets nearly completely evaporate until

10 ms in Case 0. This flash boiling process leads to a strong heat loss, significantly reducing the surrounding gas temperature and further the flame kernel temperature.



*Figure 2: Temporal evolution of gas temperature for Cases 0 and 1. For Case 0, the droplets are shown as black dots, and their sizes are determined by their diameters (enlarged for around 40 times for a clearer observation).*

Figure 3 shows instantaneous gas temperature, ammonia mass fraction, heat release rate, and flame index for Cases 0 and 1 at  $t = 7$  ms. Because of the fast phase change process, temperature and composition stratification exist in liquid ammonia conditions. Furthermore, the surrounding temperature decreases from an initial value of 500 K to about 250 K. On the contrary, the surrounding gas temperature in the Case 1 increases to 530 K due to radiation, convection, and conduction heat transfer as shown in Fig. 3(a). In Case 0, the low temperature and high ammonia mass fraction regions are located in the droplet gathering places. In Fig. 3(b), the white isolines represent  $Y_{NH_3}/Y_{NH_3,st} = 0.913$ , corresponding to the mass fraction of ammonia when equivalence ratio is 0.9, which are isolated in the areas of droplets in Case 0 but continuous in Case 1. For the liquid flame, the local fuel/air mixture is under a relatively leaner condition compared with the gaseous flame due to the phase change delay, which also contributes to the lower burnt temperature in the liquid condition. The fields of HRR are shown in Fig. 3(c). The HRR in the liquid condition is lower than that of the gaseous condition owing to lower local temperature and equivalence ratio. The distribution of HRR shows the local low value which is related to the local extinction in the liquid flame, and high HRR zones are located near the regions where ammonia vapor accumulates (see Fig. 3(b)). However, the distribution of HRR is continuous and thin in the gaseous condition. As for the local combustion mode, only the premixed mode occurs in the gaseous condition as shown in Fig. 3(d). However, both premixed and non-premixed modes exist in the liquid ammonia flame, and the premixed mode is more dominant because of fast phase change process and well mixing of the ammonia vapor and oxidizer. This unique characteristic sets liquid ammonia apart from other conventional liquid fuels such as n-heptane and n-decane. It indicates that in addition to the flamelet/progress variable (FPV) model commonly used to deal with non-premixed flames, the flamelet-generated manifolds (FGM) model suitable for premixed and partially premixed flames may also be applicable for

simulating liquid ammonia spray combustion. This will serve as a research topic for the future work of model development.

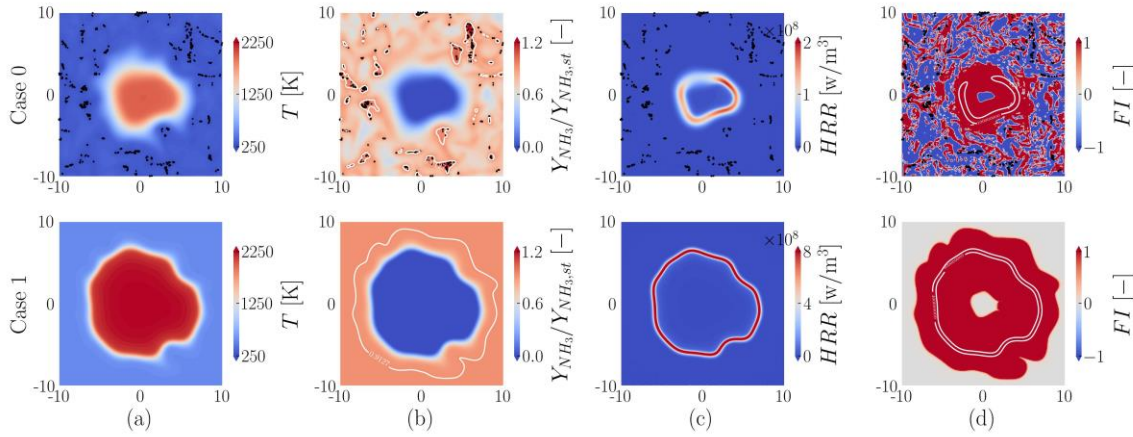


Figure 3: Distribution of (a) temperature, (b) normalized ammonia mass fraction, (c) heat release rate, and (d) flame index for Cases 0 and 1 at  $t = 7$  ms. The iso-lines in (d) represent regions of HRR values higher than 50% of the maximum HRR value.

## 4.2 Effect of initial temperature on ignition

To investigate the effect of preheating on ignition and propagation of liquid ammonia flames, another case with a low initial temperature condition (300K, Case 2) is also calculated. Figure 4 shows the temporal evolution of gas temperature. The temperature of flame kernel continues to decrease with time. The flame kernel is heavily wrinkled by the turbulence and is almost extinguished at  $t = 10$  ms. This indicates that the liquid ammonia cannot be ignited without preheating under conditions of normal temperature and pressure. Compared to Case 0 ( $\phi = 0.9$ ,  $d = 10$   $\mu\text{m}$ , and  $T_{ini} = 500$  K), as shown in the top row of Fig. 2, there are a greater number of droplets in Case 2 to maintain the equivalence ratio under a lower temperature (higher density). More droplets mean more latent heat is required and the fresh air/ammonia droplet mist is more difficult to be ignited. The reduction in the number of droplets is not significant before 7 ms in Case 2, indicating that the evaporation rate of ammonia droplet is slower due to the lower temperature.

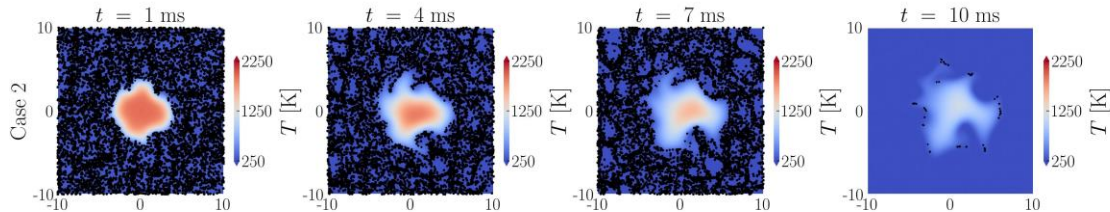


Figure 4: Temporal evolution of gas temperature for Case 2. The droplets are shown as black dots, and their sizes are determined by their diameters (enlarged for around 40 times for a clearer observation).

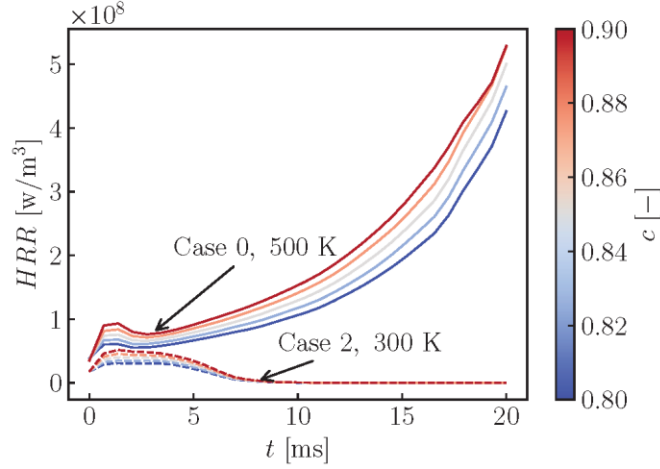


Figure 5: Temporal evolution of mean HRR calculated from the flame surface for Cases 0 and 2 (colored by progress variable).

Figure 5 visualizes the temporal evolution of HRR for Cases 0 (solid lines) and 2 (dash lines). The statistical values are extracted from the flame surface defined by a progress variable range of  $0.8 \leq c \leq 0.9$ . The HRR of Case 0 continues to increase after a slight drop, indicating that the flame has been ignited and is propagating outwards. This slight drop between  $t = 1$  ms - 2 ms is caused by the flash boiling of liquid ammonia. However, the HRR keeps decreasing when the initial temperature is 300 K (i.e., Case 2) and the flame is completely quenched after 8 ms. Temperature affects evaporation rate, reaction rate, and thermal properties of the species. High temperature promotes phase change, which enhances the heat loss. Note that when the progress variable is defined as the sum of mass fractions of the interested species, the overall evolution trends remains with a slight difference on the extracted HRR values.

To further understand why the liquid ammonia flames can be ignited with preheated carrier gas, the budget analysis of energy equation is conducted as shown in Fig. 6. The  $x$ - and  $y$ - axis denote the radial location and spatially integrated values along the radial direction, respectively. The radial direction is illustrated in Fig. 1(a). The energy involved in flame propagation phenomenon is mainly influenced by the heat release of reaction  $\dot{S}_{reac}$  (black), phase change  $\dot{S}_h$  (red), convection (blue), and radiation  $\dot{S}_{rad}$  (green), which are extracted from Eq. (3). The radiation source term of Case 0 is always larger than that of Case 2. However, its value is fairly small and negligible in the current conditions, being  $1/100^{th}$  of the convection heat transfer. The percentages of radiative heat flux may be significant when the wall surface is considered [20]. As for the reaction heat release  $\dot{S}_{reac}$ , in Case 0, with the preheated carrier gas, it is significantly enhanced compared with that in Case 2, and shows a trend where it decreases first and then increases with time, which is consistent with the trend of HRR along the flame surface as shown in Fig. 5. This indicates that the reaction heat release is dominant along the flame surface, and the droplets have a negligible effect. The heat absorbed from the phase change (i.e.,  $\dot{S}_h$ ) is also more significant in Case 0, indicating an enhanced phase change process. After the quick

evaporation process, the heat loss from phase change in Case 0 becomes close to 0 for  $t > 3$  ms. However, the heat loss of Case 2 continues to maintain a trim level over time. The value of convection heat transfer in Case 2 is similar to that of Case 0, but it is still commensurate with reaction heat release, which reduces the kernel temperature and makes the flame extinct.

The key point of the flame ignition and propagation is the competitive relationship between reaction heat production and phase change heat loss. Figure 7 demonstrates temporal evolution of the spatial-averaged heat source values. Radiation is omitted here. The convection shown by the blue color is zero in the global average. The reaction heat release of the preheating case is significantly larger than that of the non-preheating case. The value of the phase change sharply decreases with time. From the enlarged sub-figure of Fig. 7, the heat release and loss from reaction and phase change in the non-preheating case keep decreasing, and the heat loss from phase change is always larger than the heat release from the reaction. However, there are two stages under the preheating condition. In stage (i), the heat loss is larger than the reaction. In stage (ii), heat loss continues to decrease, but the heat release from the reaction increases and becomes larger than the heat loss from the phase change. After stage (ii), there is more net heat production, and the flame will be ignited later. Therefore, the most crucial reason the liquid ammonia flame can be ignited after preheating is that the reaction rate is significantly promoted. Although the heat loss from phase change is also accelerated at high temperatures, the heat loss from phase change can be compensated by the higher heat release of the reaction.

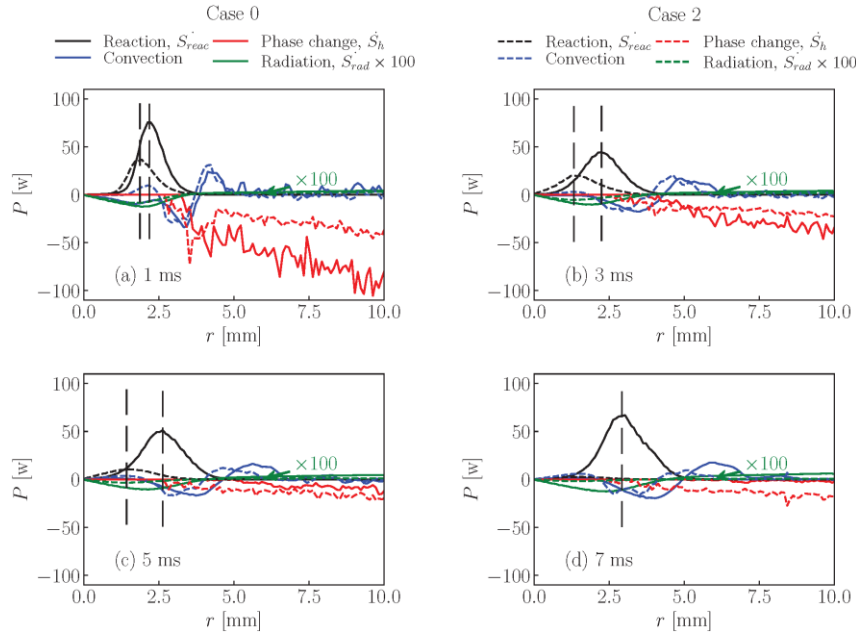


Figure 6: Budget analysis of the energy equation along the radial direction. The radiation term (green) is expanded 100 times for a clear visualization. (a), (b), (c), and (d) represent  $t = 1, 3, 5$ , and  $7$  ms. The long dash lines represent the peak locations.

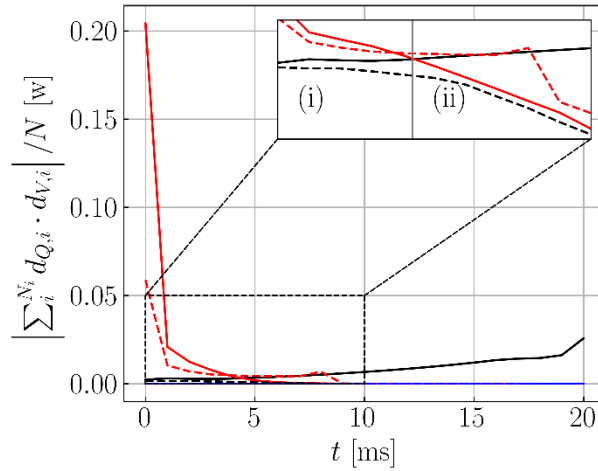


Figure 7: Temporal evolution of the spatial-averaged phase change heat loss and reaction heat release. The black, red, and blue lines represent the reaction term, the phase change term, and the convection heat transfer term in the energy equation, respectively. The logarithmic scale is used for y axis in the sub-figure.

### 4.3 Effect of droplet diameter on flame propagation and wrinkling

Figure 8 presents temperature and mass properties of ammonia droplets of different diameters (5  $\mu\text{m}$ , 10  $\mu\text{m}$ , and 15  $\mu\text{m}$  for Cases 3, 4, and 5, respectively) undergoing flash boiling at atmospheric conditions. As shown in Fig. 8(a), the temperature of the droplets rapidly decreases in the range of  $0 \text{ ms} \leq t \leq 2 \text{ ms}$ , with a more pronounced effect for smaller droplets. In this process, heat is transferred from the surrounding gas to the droplets, leading to an increase in droplet temperature once the heat loss from the phase change process is outweighed by the heat absorption from the outside. As a result, the mean temperature of droplets first decreases and then increases over time for larger droplets (i.e., Case 4 and Case 5), which is consistent with the findings of Gao et al. [41]. Figure 8(b) depicts the normalized mass of the ammonia droplets against with time, and the slope of the curves represents their evaporation rate. In the case of small droplets (Case 3), complete evaporation occurs rapidly, with a continuous and high evaporation rate. However, as the diameter increases (Cases 4 and 5), the evaporation rate is relatively slower than that of Case 3, and much slower with the time, resulting in a longer droplet lifetime. The gradual decrease in evaporation rate with time in these cases is a result of the reduction in the temperature difference between the droplets and their surroundings caused by the strong heat loss.

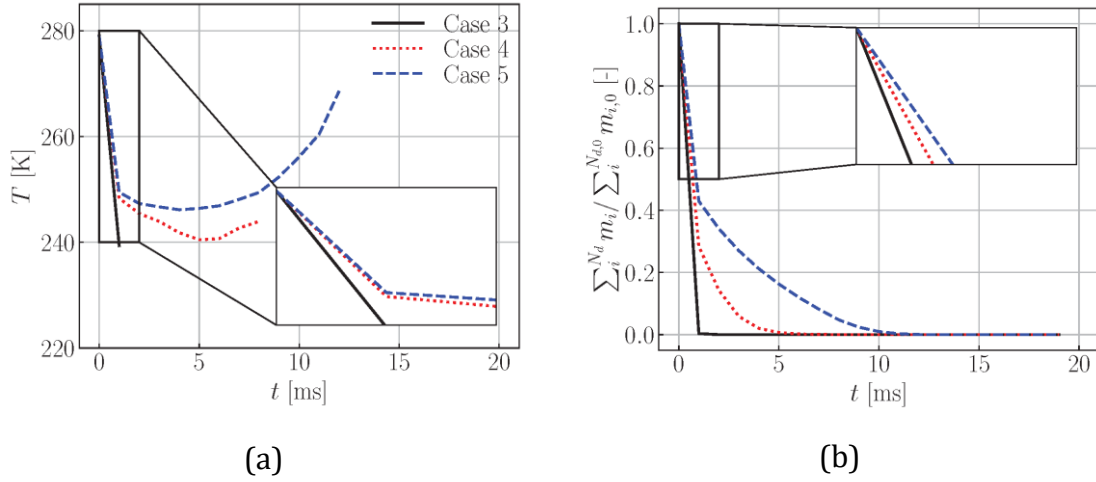


Figure 8: Temporal evolution of (a) mean temperature and (b) normalized mean mass of ammonia droplets for different diameter cases, with a close-up in the region  $0 \text{ ms} < t < 2 \text{ ms}$ .

Figure 9 compares the influence of different droplet diameter on flame propagation, with a specific focus on the liquid ammonia droplets with large latent heat. The equivalence ratio, initial temperature, and heat fraction of ammonia are set in accordance with the parameters specified by Okafor et al. [11]. When the droplet diameter is  $5 \mu\text{m}$  (Case 3), complete evaporation of the droplets is achieved and the released ammonia vapor is well mixed with air before arriving the flame surface. This results in combustion characteristics more similar to gaseous premixed mode for Case 3. On the other hand, for the cases with droplet diameters of  $10 \mu\text{m}$  (Case 4) and  $15 \mu\text{m}$  (Case 5), an interaction between the droplets and the flame surface is observed. In Case 5, droplets penetrate and evaporate near the flame kernel, causing unburned ammonia pockets near the  $c = 0.5$  isoline. The droplets also significantly wrinkle the flame surface. Due to flash boiling, there are no droplets present within the high temperature kernel ( $c > 0.8$ ), a phenomenon that is distinct from the behavior observed in n-heptane flames [42].

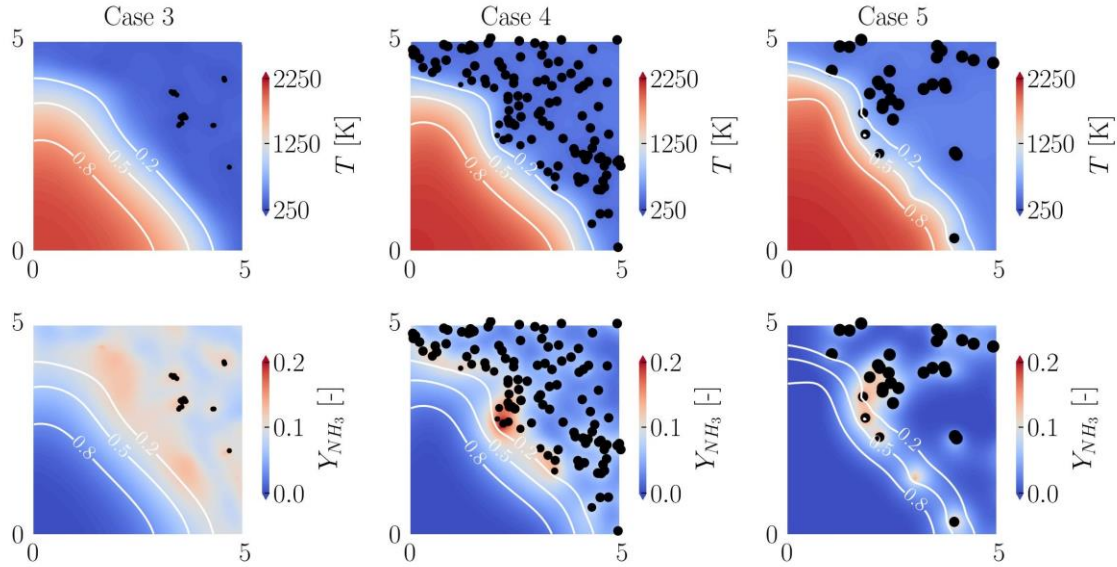


Figure 9: Local temperature (top) and  $\text{NH}_3$  mass fraction (bottom) fields at  $t = 1$  ms for different droplet diameter cases. White lines represent iso-value lines of progress

Figure 10 displays temporal evolution of the spatial-averaged values for different droplet diameter cases. As seen in Fig. 10(a), the rapid phase change rate of small-scale droplets results in a rapid decrease in the gas temperature (see the black line). Initially, the gas temperature slightly decreases before 5 ms due to strong phase change process, but gradually increases over time as heat release from the reaction becomes greater than the heat loss by evaporation. As for HRR shown in Fig. 10(b), in Case 5, it reaches its peak first due to a relatively shorter ignition delay time, which will be investigated later. The peak value for Case 4 is greater than that of the other two cases, suggesting that there is an optimal combination of mixture concentration and temperature to maximize the peak HRR. As the diameter of the droplets increases, the rate of evaporation decreases and the local equivalence ratio is reduced, resulting in a relatively higher local gas temperature. While a lower local equivalence ratio tends to result in a lower HRR, and the increase in surrounding gas temperature leads to a larger HRR. The interplay between mixture concentration and temperature creates a trade-off relationship, with the peak HRR observed for droplet diameters of approximately  $10 \mu\text{m}$ . The diameter has limited effect on the equilibrium product, primarily altering the evaporation rate of liquid ammonia. As a result, the sequence of spatial-averaged scalars for different species is also influenced by the droplet diameter as displayed in Fig. 10(c)-(i).

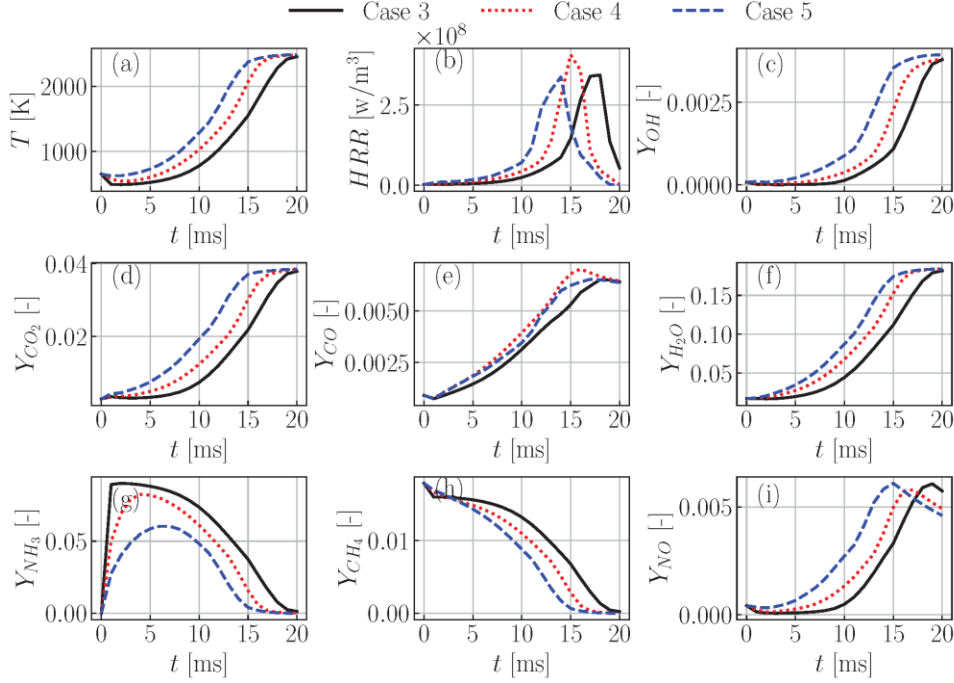


Figure 10: Temporal evolution of spatial-averaged (a) gas temperature, (b) heat release rate, (c) OH, (d) CO<sub>2</sub>, (e) CO, (f) H<sub>2</sub>O, (g) NH<sub>3</sub>, (h) NO, and (i) N<sub>2</sub>O mass fractions for different diameters of liquid ammonia droplets.

The analysis of the ammonia mass fraction along the flame surface in different cases has revealed distinct patterns in Fig. 9. In Case 3, the ammonia mass fraction is observed to be closer to the stoichiometric mixing fraction, which is a more favorable condition for ignition as found in previous study [43]. However, for Cases 4 and 5, the ammonia mass fractions along the flame surface exhibit a stratified distribution, leading to the reaction taking place at conditions farther away from the stoichiometric condition. Despite these regularities, it is discovered that the reaction rates for Cases 4 and 5 are higher than that of Case 3. This outcome is attributed to the significant impact that the temperature change associated with phase change has on the reaction rate, as well as the effect of local equivalence ratio. To better clarify their relative importance, ignition delay times (IDTs) under different temperatures and equivalence ratio conditions are calculated using the Cantera code package [44], as illustrated in Fig. 11. The configurations of initial pressure and heat fraction are held constant for the corresponding gaseous cases. It is found that temperature has a much more significant impact on the IDTs compared to that of the equivalence ratio. In the case of small diameter droplets (i.e. Case 3), the surrounding gas temperature is found to be much lower, leading to increased IDTs and makes it more difficult for the flame to propagate outwards.

In this study, the flame surface is defined as the locations where  $\psi(\mathbf{x}, t) = 0$ . The fresh mixture is represented by  $\psi < 0$  and the burnt gas by  $\psi > 0$ . The flame surface is described mathematically by the function  $\psi(\mathbf{x}, t) = c - c_{surface}$ , where  $c_{surface}$  is selected as 0.2, 0.5, and 0.8. The effect of droplets on the flame surface can

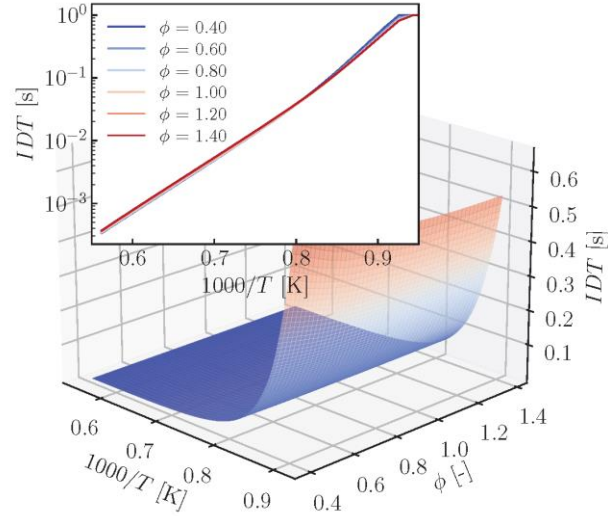


Figure 11: IDTs of  $\text{NH}_3/\text{CH}_4$  flames under the various temperatures and equivalence ratios conditions.

be analyzed by considering the flame curvature  $\kappa$ , which is defined as  $\kappa = -\nabla \cdot \mathbf{n}$ . Here,  $\mathbf{n} = \nabla\psi/|\nabla\psi|$  is the unit normal to the flame surface, pointing towards the burnt gas [45]. The probability density function (PDF) statistics of  $\kappa$  for Case 3, 4, and 5 are shown in Fig. 12. The values of  $\kappa$  are obtained from the iso-surfaces of  $c = 0.2$ , 0.5, and 0.8, respectively. The PDF of all cases indicates that the flame surface is primarily convex towards the burnt side, with a higher probability of positive curvatures than negative. This trend is more pronounced in the larger diameter droplets. The PDF of Case 5 ( $d = 15 \mu\text{m}$ ) shows a wider distribution of  $\kappa$  values than those of Cases 3 and 4 ( $d = 5$  and  $10 \mu\text{m}$ ) due to the greater wrinkling effect of the droplets on the flame surface. This phenomenon has been observed in previous experiments and numerical studies [42,46]. However, the flashing behavior of liquid ammonia results in differences in the wrinkling effect compared to studies of iso-octane and n-heptane flames, i.e., most of the droplets in the liquid ammonia flames quickly evaporate before reaching the burnt gas region, leading to a relatively insignificant influence of the droplets on the wrinkling of the flame surface on the burnt side ( $c = 0.8$ ).

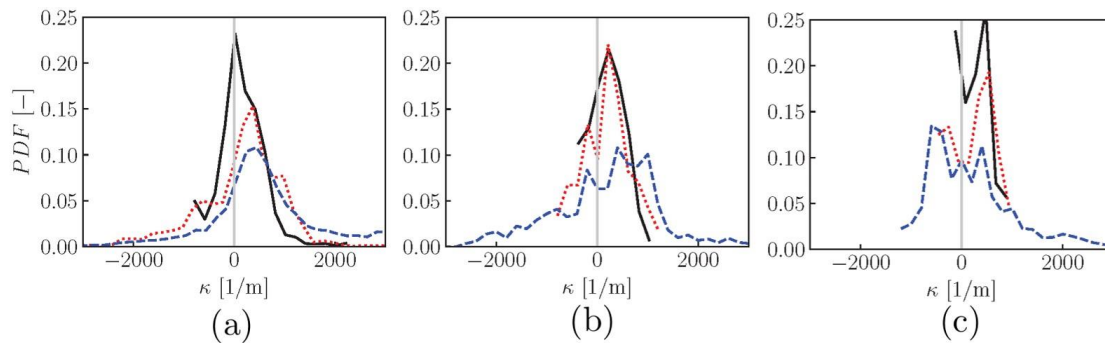


Figure 12: PDF distribution of  $\kappa$  along the flame surface which is defined by  $c =$  (a) 0.2, (b) 0.5, (c) 0.8 between three cases with different diameters. The color and styles of lines

are the same with Fig. 10. The black, red, and blue lines represent Cases 3 ( $d=5\ \mu\text{m}$ ), 4 ( $d=10\ \mu\text{m}$ ), and 5 ( $d=15\ \mu\text{m}$ ), respectively.

#### 4.4 Effect of equivalence ratios and heat fractions on flame propagation

The present study also examines the impact of equivalence ratios and heat fractions on the flame propagation. In Fig. 13(a), the propagation of flames is presented under different  $\phi$ . The initial temperature and heat fraction ratio are fixed at 500 K and 0.7, respectively, as the configuration of the experiment in Ref. [11]. It is observed that as  $\phi$  increases, the propagation ability, as indicated by the size and temperature of flame kernels, decreases. This trend is consistent with the findings in Fig. 9 of Okafor's work [11]. In gaseous conditions, the highest heat release rate, flame speed, and temperature are observed at stoichiometric conditions, which is in contrast to the liquid ammonia conditions. This difference is attributed to the dominant effect of heat loss due to phase change. As  $\phi$  increases, the total mass of ammonia increases, leading to a higher latent heat of vaporization and a lower ambient temperature, which slows down the rate of chemical reactions and reduces the flame propagation ability. A similar trend is evident in Fig. 13(b), which depicts the instantaneous temperature field of flame kernel evolution for different ammonia heat fraction ratios. The equivalence ratios and initial temperatures are fixed at 1.1 and 500 K, respectively, for all cases. It is observed that the size and temperature of flame kernels decrease as  $\xi_{\text{NH}_3}$  increases. Notably, the flame tends to be quenched at  $\xi_{\text{NH}_3} = 0.9$ . The reduction in flame propagation ability with increasing  $\xi_{\text{NH}_3}$  can be attributed to two main factors. Firstly, as observed in Fig. 13(a), a higher heat fraction ratio results in an increased amount of liquid ammonia, which leads to a higher latent heat and a lower temperature, thereby limiting flame expansion. Secondly, a decrease of methane content in the fuel blends leads to a reduction in the reaction intensity, as reported in Ref. [47]. These two factors work towards the same direction to reduce flame propagation ability. Therefore, under the same initial temperature conditions, flame propagation is limited and can even result in extinction with increasing  $\xi_{\text{NH}_3}$ .

A quantitative analysis of the difference between global heat release rate and phase change heat loss, as well as the mean temperature as shown in Fig. 14. As seen in Fig. 7, the heat transfer during the flame propagation process is primarily driven by chemical reactions and phase change. The process of flame propagation can be divided into two stages, as shown in Figure 14(a). The negative value in the difference represents stage (i), while the positive value represents stage (ii). The competition between the heat release from reactions, denoted by  $Q_r$ , and the heat loss due to phase change, denoted by  $Q_p$ , determines the ability of the flame kernel to expand. Hence, the difference between  $Q_r$  and  $Q_p$  can be used to quantitatively describe the flame propagation ability.

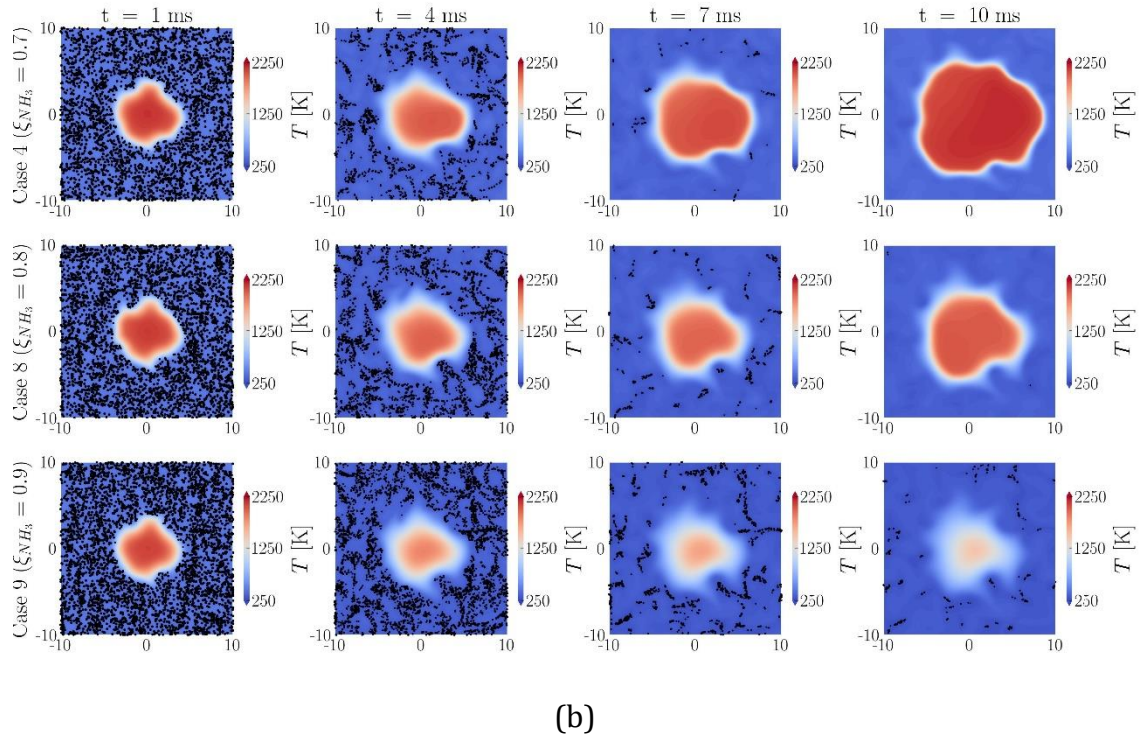
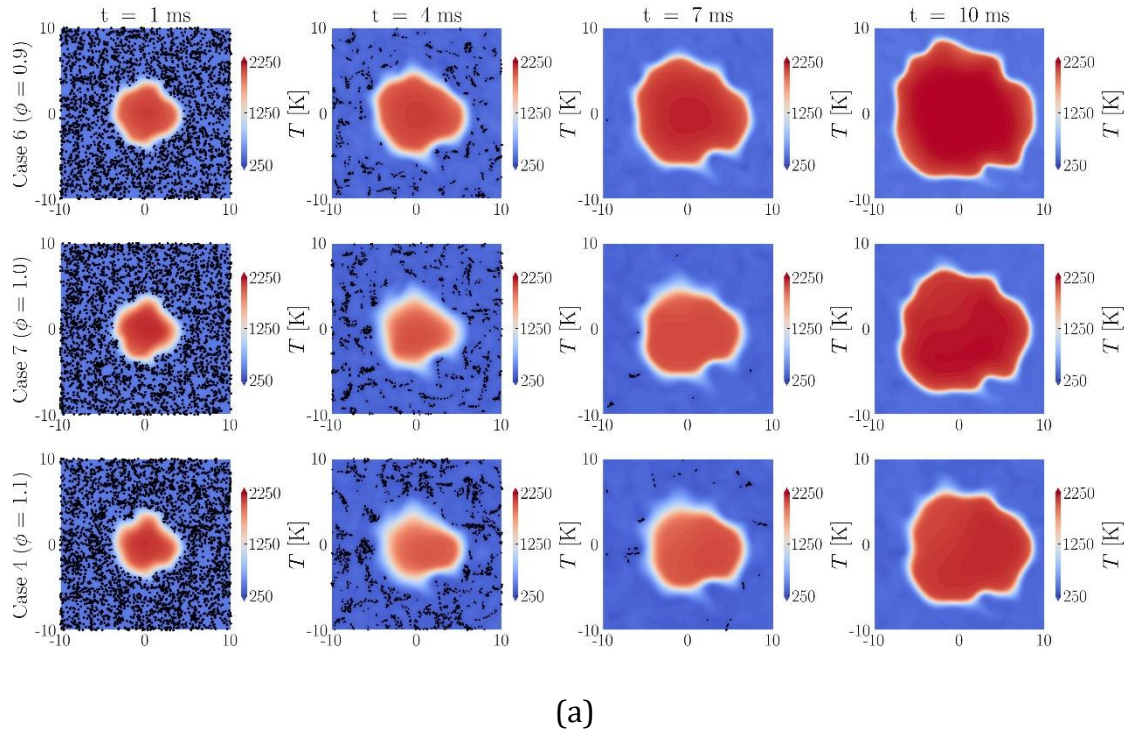


Figure 13: Instantaneous field of temperature: (a) The comparison between different equivalences and (b) ammonia heat fractions. The droplets are shown as black dots, and

their sizes are determined by their diameters (enlarged for around 40 times for a clearer observation).

A negative value of the difference indicates that the heat loss is dominant, leading to a decrease in the global temperature as shown in Fig. 7(b). As ignition progresses, the temperature difference decreases and reaches zero in sequence. The order is related to the total mass fraction of liquid ammonia. The higher the mass fraction, the longer it takes to reach equilibrium. After reaching equilibrium, the global temperature increases in the same order. However, due to the significant heat loss during stage (i), the temperature of Case 9 is too low to ignite, and the difference value and temperature remain at 0 and below 500 K, respectively. This result highlights the importance of ensuring that the temperature after the first stage of ignition is not too low, in order to allow the liquid ammonia flame to enter the second stage of ignition.

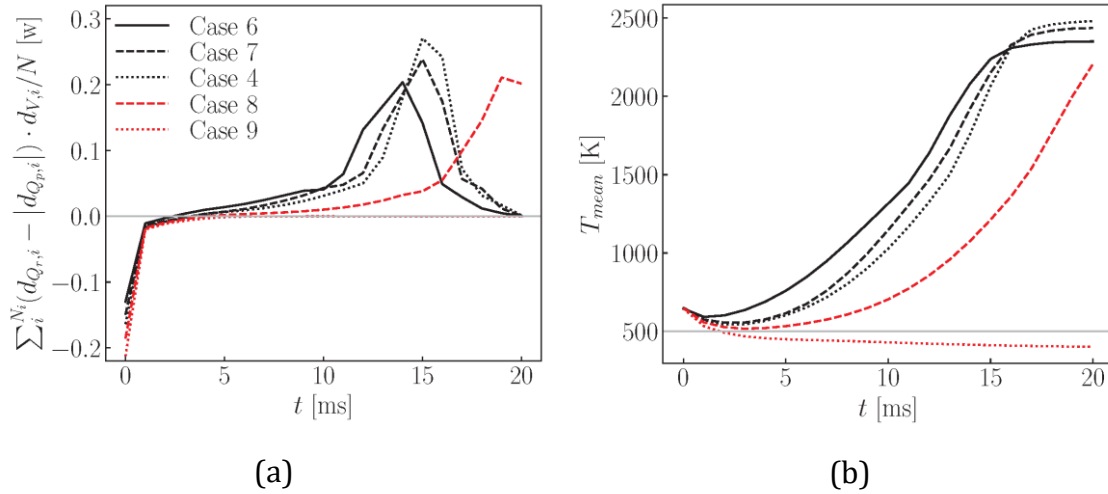


Figure 14: (a) Temporal evolution of the difference between global heat release rate and phase change heat loss. (b) Temporal evolution of the mean temperature of gaseous field.

The distribution of HRR in the mixture fraction space ( $Z$ , defined as  $Z = Z_{CH_4} + Z_{NH_3}$ ) is analyzed to investigate the contributions of methane and ammonia mass fractions to the overall HRR under various equivalence ratio conditions, as illustrated in Fig. 15. The scatter are colored with the ammonia fraction in the local stream, calculated by  $Z_{NH_3} / (Z_{NH_3} + Z_{CH_4})$ . The data is extracted at  $t = 7$  ms, which is a specific time instant during the process of ignition as shown in Fig. 13. At this time, the flame begins to spread outward significantly, and the temperature and heat release rate also increase significantly. With increasing  $\phi$  (from Fig. 15(a)-(c)), the maximum HRR decreases due to the significant heat loss from the phase change of flashing liquid ammonia. The percentage of  $Z_{CH_4}$  in the high HRR region becomes higher with increasing  $\phi$ , indicating that the contribution from methane to ignition becomes more pronounced. This is because the reactions related to methane are easier to activate, particularly in low-temperature conditions, and when  $\phi$  increases, the ambient

temperature decreases significantly due to the large latent heat from the phase change of liquid ammonia. The stoichiometric condition mixture fraction is 0.118, and most of the reaction heat is released on the lean side, which is consistent with previous studies [42,48]. This trend becomes more pronounced at higher  $\phi$  conditions. Overall, these results demonstrate that the optimal equivalence ratio is not a crucial factor for liquid ammonia flame propagation, the reaction primarily occurs on the lean side, and heat loss from the phase change has a significant impact on the propagation process.

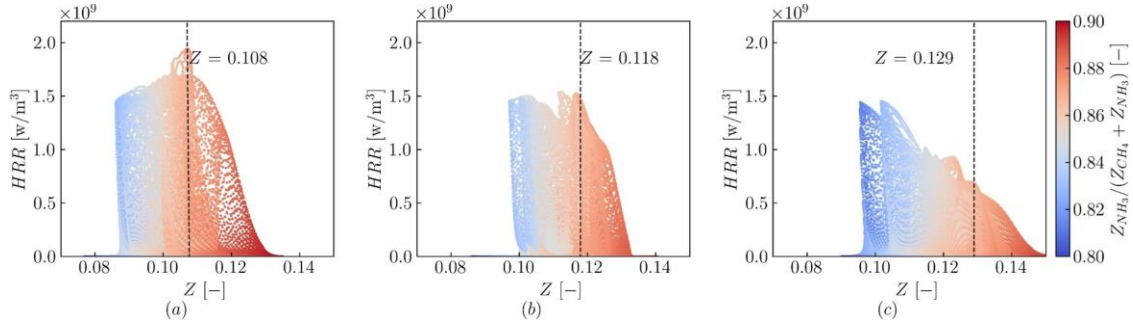


Figure 15: The HRR distribution in the space of mixture fraction  $Z$  for (a)  $\phi = 0.9$ , (b)  $\phi = 1.0$ , (c)  $\phi = 1.1$ . The scatters are colored by  $Z_{NH_3}/(Z_{NH_3} + Z_{CH_4})$ . The dash lines represent that  $Z$  for corresponding global equivalence ratio.

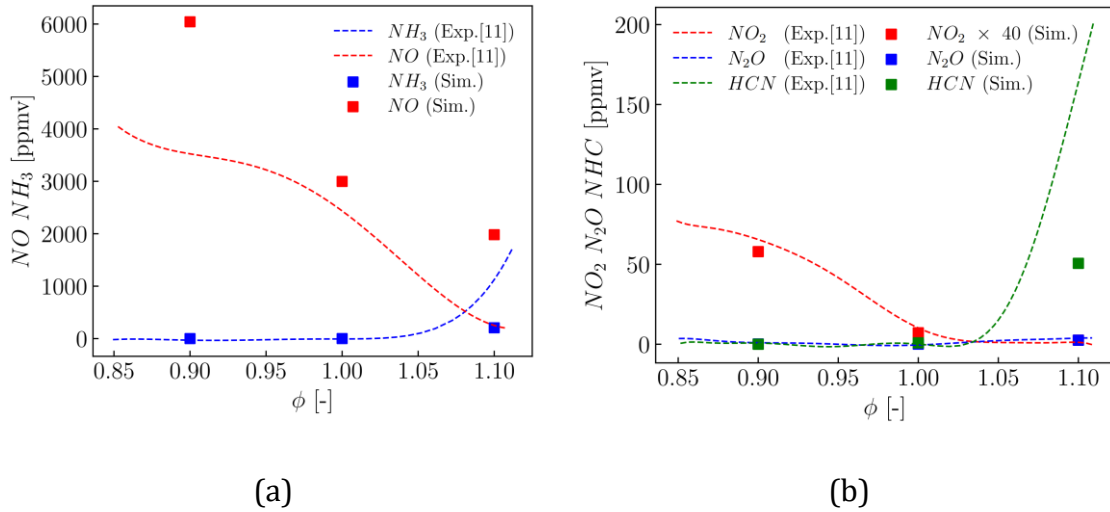


Figure 16: Variation of emission with the global equivalence ratio from experimental measurement (dash lines) and numerical simulation (square dots) in the same conditions, i.e.,  $\xi_{NH_3} = 0.7$ ,  $T_{initial} = 500$  K.

The emissions of  $NH_3$ ,  $NO$ ,  $NO_2$ ,  $N_2O$ , and  $HCN$  in the burnt region at  $t=10$  ms are depicted in Fig. 16. The experimental data under similar mixture conditions and initial temperature are included for comparison [11]. As seen in Fig. 16(a), the emission of  $NH_3$  is minimal due to complete combustion under fuel-lean conditions.

The emission of  $\text{NH}_3$  increases with increasing  $\phi$ . The emission of NO decreases with increasing  $\phi$  from a significantly high value to a low one. The trends of  $\text{NO}_2$ ,  $\text{N}_2\text{O}$ , and HCN emission with the global equivalence ratios are depicted in Fig. 16(b).  $\text{NO}_2$  shows the same trend as NO emission, while  $\text{N}_2\text{O}$  shows the opposite trend. The emission of HCN also shows a positive relationship with  $\phi$ . These results indicate that to control  $\text{NO}_x$  emissions, liquid ammonia should also be conducted in fuel-rich conditions.

## 5 Conclusions

In the present study, the spherical flame propagation in liquid ammonia-laden mixtures was numerically investigated with detailed chemistry. The verified combined model was employed to describe the flash boiling process of the liquid ammonia droplets. A comparison between liquid and gaseous ammonia flames was conducted. The effects of initial temperature, diameter, equivalence ratio, and ammonia heat fraction on the flame ignition, propagation, heat release rate, and structure were also investigated. The main results and findings are summarized as below:

1) Ammonia droplets significantly reduce the flame propagation speed and heat release due to the strong local heat loss caused by the flash boiling. As a result of the fast phase change, the premixed mode is more dominant than the non-premixed mode in ammonia spray flames.

2) Preheating plays a crucial role in ignition, and the liquid ammonia cannot be ignited at normal temperatures without preheating. The ignition process has two stages, and the flame kernel could only spread outward after entering the second stage. The competition between reaction heat production and phase change heat absorption is the dominant factor in the ignition of liquid ammonia, as identified from the energy equation budget analysis.

3) Smaller droplets quickly flash, significantly reducing the temperature but increasing the local mixture fraction. The flame-droplet interaction is more pronounced in large diameter cases, with local heat loss near the droplets causing local extinction, but without significant influence on the burnt side gas.

4) As for the effects of equivalence ratios and heat fractions, the main difference in flame propagation characteristics comes from the proportion of liquid ammonia, which brings different levels of heat loss. Stoichiometric conditions are no longer the most suitable condition for ignition. Under conditions of high equivalence ratios, the contribution of methane oxidization to the ignition process is significant.

## Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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