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Mass interminglement and hypergolic ignition of TMEDA and WFNA droplets by off-center collision

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ABSTRACT

Binary collision between a smaller N, N, N′, N′- tetramethylethylenediamine (TMEDA) droplet and a larger white fuming nitric acid (WFNA) droplet was investigated experimentally and computationally for understanding the influence of off-center collision on the hypergolic ignitability of the system, which is controlled by the mass interminglement and mixing subsequent to the droplet coalescence. The ignition delay time was experimentally found to non-monotonically vary with the impact parameter, which measures the degree of off-center collisions. This phenomenon was hypothetically attributed to the non-monotonicity of mass interminglement of colliding droplets with increasing the impact parameter—the increased droplet stretching by slightly off-center collision promotes mass interminglement, but the stretching separation by significantly off-center collision reduces mass interminglement. This hypothesis was computationally verified by a validated volume-of-fluid (VOF) simulation of a simplified problem, in which the transport phenomena and chemical reactions are neglected and the controlling physics of droplet mass interminglement is emphasized. Furthermore, a parametric study for wide ranges of controlling non-dimensional parameters, such as the collision Weber number of 20–140 and the droplet size ratio of 1.3–2.0, further confirms that the non-monotonicity of ignition delay time with the impact parameter is a general characteristic of the present hypergolic system.

1. Introduction

1.1. Hypergolic ignition by head-on collision of TMEDA and WFNA droplets

Spontaneous ignition of hypergolic liquid propellants occurs upon their contact without an external source of energy [1–4]. Hypergolic propellants and ignition have received lasting attentions in the past decades for their applications in rocket propulsion, particularly when multiple engine restarts are required [3]. Unlike the auto-ignition of a homogeneous gaseous mixture of non-hypergolic reactants, hypergolic ignition is intrinsically heterogeneous as it involves mixing and reaction of initially separated propellants in both liquid phase and gas phase [5–9].

Existing experimental methods for studying hypergolic ignition differ in the mode and extent of liquid-phase mixing [1,2]. Specifically, the piston-driven apparatus for rapid mixing of liquid reactants is designed to minimize the mixing effect in liquid phase [10–13]. Designed for variable testing conditions, the impinging jet apparatus [7,14–16] often employs partially mixing reactants. In recent years, the drop test [8,17] has been widely used as a fast tool to prescreen potential hypergolic propellants. In a typical drop test, a free or suspended propellant drop is made to impact the liquid pool of another propellant [18,19], which is either confined in a container or placed on a solid surface. Although these test systems help researchers obtain a lot of valuable understanding on hypergolic ignition [7,8,17,19], they have a common deficiency — it is generally difficult to quantify the system-dependent liquid-phase mixing owing to the unavoidable "wall effect" by the jet nozzle or the container or the surface.

Motivated by studying the hypergolic ignition initialized by the collision of freely moving propellant droplets and hence free from "wall effect", the authors recently established and conducted the experiment of binary droplet collision of N,N,N′,N′ – tetramethylethylenediamine (referred to as TMEDA hereinafter) and white fuming nitric acid (referred to as WFNA hereinafter) in atmospheric environment [20]. This preliminary experimental study was focused on verifying the applicability and reliability of the established experimental apparatus for studying hypergolic ignition triggered by binary droplet collision and on the special (also nontrivial) situation of head-on collision. The significance of the experiment can be appreciated by recognizing that binary droplet collision could be a frequent event in the combustion chamber of rocket engines, where liquid propellants

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Nomenclature

Physical quantities

D	droplet diameter
t	physical time
t_{ine}	characteristic collision time, $t_{ine} = D_L/U$
t_{osc}	characteristic oscillation time of the larger droplet, $t_{osc} = \sqrt{\rho_L D_L^3/\sigma_L}$
U	experimental relative velocity between the two colliding droplets in the x-direction
α	mass diffusivity
μ	dynamic viscosity
ρ	density
σ	surface tension coefficient
χ	projection of the distance between two droplet mass centers along the direction of U

Numerical parameters

C	time-dependent dye function concentration ϕ in the merged droplet, $C = \frac{\lvert\phi-0.5\rvert}{0.5}C_0$
C_0	dye function concentration ϕ in the initially unmixed droplets, $C_0 = \{\begin{matrix}0, & \phi = 0\\ 1, & \phi > 0\end{matrix}\}$
C_∞	dye function concentration ϕ in the fully mixed droplets, $C_\infty = 1/(1+\Delta^3)$
f	Volume-of-fluid (VOF) function
H	Heaviside function
R	local mesh refinement level
V	total volume of fluid droplets
ϕ	mass dye function with $\phi = 1$ in the smaller droplet otherwise $\phi = 0$

Experimental parameters

B	impact parameter, $B = 2\chi/(D_S + D_L)$
G	threshold value of the grayscale levels
N	pixel number of the entire image
N_d	total pixel number of the grayscale levels below G_{low}
N_b	total pixel number of the grayscale levels above G_{high}
r	ratio of the pixel number (N_b or N_d) to the total pixel number (N)

Non-dimensional and normalized variables

M	mixing index, $M = 1 - \frac{\int_V \lvert C-C_\infty\rvert H(f-1)\mathrm{d}V}{\int_V \lvert C_0-C_\infty\rvert H(f-1)\mathrm{d}V}$
Oh	Ohnesorge number, $Oh = \mu_L/\sqrt{\rho_L\sigma_L D_L}$
Pe	Peclet number, $Pe = UD_L/\alpha$
$\tilde{Q}$	dimensionless fluid-dynamical quantities of interest
T	non-dimensional time, $T = t/t_{osc}$
We	Weber number, $We = \rho_L D_L U^2/\sigma_L$
Δ	size ratio of the larger droplet to the smaller droplet, $\Delta = D_L/D_S$

Subscripts

b	species have the grayscale levels above G_{high} with $r_b = N_b(t\lvert G > G_{high})/N$
d	species have the grayscale levels below G_{low} with $r_d = N_d(t\lvert G < G_{low})/N$
g	fluid properties of the surrounding gas (air) environment
high	higher threshold value of the grayscale levels, $G_{high} = 250$
L	fluid properties of the larger (WFNA) droplet
low	lower threshold value of the grayscale levels, $G_{low} = 5$
S	fluid properties of the smaller (TMEDA) droplet

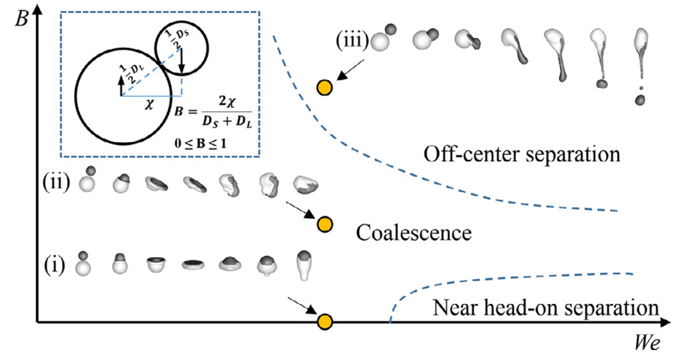

Fig. 1. Schematic of droplet collision regimes in the $We - B$ parameter space: (i) head-on coalescence, (ii) off-center coalescence, and (iii) off-center separation. Droplet bouncing occuring at smaller Wes and droplet splashing at higher Wes are not shown for clarity.

are injected into and quickly atomized into droplets. Furthermore, the recent interests in gelled hypergolic propellants (GHP) further stress the need for such an experiment because the substantially reduced volatility by gelling suppresses the droplet vaporization and therefore the ignition of GHPs heavily relies on the liquid-phase mixing [21,22]. It is known that the rapid mixing of reactants promotes the reaction rate of a non-premixed system. It is also true for the present system consisting of hypergolic fuel and oxidizer, where the enhancement of liquid-phase mass interminglement between droplets reduces the ignition delay time. The previous studies [1,7] have observed that the ignition delay time is closely related to the initial contact and subsequent mixing between hypergolic fuel and oxidizer droplets.

In order to focus on understanding the influence of droplet mixing on the hypergolic ignition and to avoid dealing with many factors at a time, the authors deliberately limited the scope of their previous experimental study [20] to the head-on collision at variable collision Weber number ($We = 20 - 220$) and droplet size ratio ($\Delta = 1.2 - 2.9$), but at a fixed Ohnesorge number ($Oh = 2.5 \times 10^{-3}$) by fixing the size of the WFNA droplet. The TMEDA/WFNA hypergolic system was chosen for the study because it has been recently proposed as a promising substitute for the highly toxic hydrazine propellants. The experimental method can be applied to study other hypergolic systems. The most notable experimental discovery in the study is the non-monotonic variation of hypergolic ignition delay time with increasing We. The underlying physics is the non-monotonic emergence of a "jet-like" internal mixing within the coalesced droplet with increasing We, a fluid dynamical phenomenon that was recently identified by Tang et al. [23] in unequal-size droplet collision. The "jet-like" internal mixing facilitates the exothermic liquid-phase reaction, $TMEDA + 2HNO_3 \rightarrow TMEDADN$, whose heat release is crucial to the subsequent droplet heating and vaporization, the decomposition of the propellants, and eventually the gas-phase ignition [7,8]. Another notable experimental discovery is that Δ significantly affects the hypergolic ignition mainly by changing the available mass of the propellants participating the chemical reaction, while it also affects the droplet mass interminglement and therefore liquid-phase mixing.

1.2. Off-center collision and mixing of binary droplets

In spite of the above findings about the hypergolic ignition by the head-on collision of TMEDA and WFNA droplets, it should be recognized that head-on collision is a rare event in reality and that off-center collisions are significantly more frequent. Fig. 1 illustrates the off-center collision of two droplets of diameters D_S and D_L (the subscript "S" denotes the smaller droplet and "L" the larger