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You-An Chen (陳宥安); Chih-Hsiang Chiang (蔣智翔); Chao-Yi Yang (楊詔亦); Shou-Yin Yang (楊授印) ⑩ ; Hsien-Hung Wei (魏憲鴻) ᢦ ₀

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You-An Chen (陳宥安),<sup>1</sup> Chih-Hsiang Chiang (蔣智翔),<sup>1</sup> Chao-Yi Yang (楊詔亦),<sup>1</sup> Shou-Yin Yang (楊授印),<sup>2</sup> (D) and Hsien-Hung Wei (魏憲鴻)<sup>1,a)</sup> (D)

### AFFILIATIONS

<sup>1</sup>Department of Chemical Engineering, National Cheng Kung University, Tainan 701, Taiwan <sup>2</sup>Department of Power Mechanical Engineering, National Formosa University, Yunlin 632, Taiwan

<sup>a)</sup>Author to whom correspondence should be addressed: hhwei@mail.ncku.edu.tw

### ABSTRACT

We show both experimentally and theoretically for the first time that the widely used  $D^2$ -law for describing the shrinking kinetics of a vaporizing droplet is not the true law for single burning droplets under the influence of gravity. Experimentally, the instantaneous diameter D for such a droplet is identified to obey a new kinetic law:  $D^n$  decreases linearly with time t, with the exponent  $n = 2.53 \pm 0.30 - 2.69 \pm 0.14$  for a variety of common liquid fuels such as alkanes and alcohols of low and high boiling hydrocarbons. Theoretically, we develop a phenomenological theory to show  $n = 8/3 \approx 2.67$  well capturing the experimental values. The burning rate constant in this new  $D^{8/3}$ -law no longer behaves like the thermal diffusivity  $\alpha$  of the gas phase but turns into  $\alpha^3 \ell^{-7/3}/g$  due to the additional inherent fluid-property-determined buoyancy length  $\ell$  under the influence of the gravitational acceleration g. This non-square power law is purely transport determined. It is a consequence of simultaneous momentum, heat, and mass transfer resulted from buoyant convection setup by the blazing flame around the droplet, insensitive to combustion chemistry and detailed reaction kinetics. This study provides not only renewed insights into the multifaceted droplet combustion phenomenon but also possibly new paradigms for a better understanding or characterization of actual fuel droplet combustion processes in normal gravity environments.

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### I. INTRODUCTION AND MOTIVATION

Combustion of liquid fuels is a widely used process to produce intense heat from rapid thermal oxidation, capable of generating enormous power to drive engines and mechanical devices. It often takes the form of spray combustion in which a liquid fuel is atomized into a cloud of fine droplets of size 100  $\mu$ m or smaller moving at speed 10 m/s or faster to render a massive heat release in a short period of time.<sup>1,2</sup> During burning, such droplets receive immense heat from the surrounding gas and then become quickly vaporized. For this reason, the burning potential of a liquid fuel may be evaluated by measuring how fast a single droplet reduces its size due to burning. The droplet in such test is typically mm in diameter, and its shrinking kinetics is customarily described by the  $D^2$ -law, stating that square of the instantaneous diameter D of the droplet (of the initial diameter  $D_0$ ) decreases linearly with time  $t_i^{3,4}$ 

$$D^2 = D_0^2 - Kt. (1)$$

Here, K is the burning rate constant often constituted by the fuel's properties and the ambient gas condition. So by simply plotting  $D^2$ 

against t using the monitored size history of a vaporizing fuel droplet, the slope will give the value of K to reflect the burning capability of the fuel.

While the  $D^2$ -law is known to be better suited for a vaporizing droplet in the absence of gravity without convection,<sup>3,4</sup> it is still routinely employed to characterize a burning droplet under the normal gravity condition because the corresponding  $D^2$ -t plot still displays a roughly linearly decreasing profile.<sup>5-9</sup> However, such a widely used law seems questionable in droplet combustion under gravity because of the following reasons. First of all, if the exponent in (1) is slightly deviated from (2), it still displays a roughly linear profile in the  $D^2$ -t plot. In other words, a seemingly linear looking curve in the  $D^2$ -t plot could be deceptive. Second, Eq. (1) is originally derived for a vaporizing droplet under the microgravity condition where conduction and diffusion are the sole transport mechanisms without convection. On the contrary, an actual droplet combustion is operated in a normal gravity environment. For such a droplet, a buoyant convection will inevitably be set off by the blazing flame, exhibiting a speed of a few tens of cm/s.<sup>10</sup> This can strongly affect the subsequent heat and mass transfer

processes around the droplet and, hence, alter the  $D^2$ -law. This is very likely because even for a purely evaporating droplet without combustion it no longer obeys the  $D^2$ -law under strong forced convection by an airflow at speed of a few m/s.<sup>11</sup> There also exists evidence that the shrinking kinetics of an evaporating droplet significantly departs from the  $D^2$ -law when natural convection is present.<sup>12</sup> Third, from a dimensional point of view, gravitational acceleration g must enter the description for such a strongly buoyancy intervened droplet, thereby unlikely to possess the same form as (1) for the case in the absence of gravity. The situation here is quite similar to a well-known example: motion at constant velocity V vs free fall under g. The travel distance of the former is Vt whereas the latter is  $gt^2/2$ . For the same reason, if one wishes to have consistent dimensions in a correct kinetic description by including g, the exponent in (1) cannot be (2) and has to be a different value.

Motivated by the above skepticisms on the applicability of the  $D^2$ -law, in this paper we aim not only to attest whether the  $D^2$ -law can faithfully describe the shrinking behavior of a burning droplet under gravity but also to look at how the droplet vaporizes under the influence of buoyant convection. As will be shown shortly by both experiment and theory, the classical  $D^2$ -law is indeed not the true law for such a droplet and a new law emerges to govern the shrinking kinetics of the droplet.

### II. METHODOLOGY AND EXPERIMENTAL PROCEDURES

Our approach begins by experimentally determining the exponent n in the assumed power-law form below for a vaporizing droplet,

$$D^n = D_0^n - Kt. (2)$$

Recognizing the droplet lifetime  $t_{life} = D_0^n/K$  which is measurable, Eq. (2) can be re-written as

$$(D/D_0)^n = 1 - t/t_{life}.$$
 (3)

So by monitoring how *D* reduces with *t* and then plotting  $(1 - t/t_{life})$  against  $D/D_0$  in log–log scale, the value of *n* can be readily obtained from the slope of the plot. To further reduce the fiber's heat transfer to the droplet, the fiber is a ceramic filament of low thermal conductivity 0.12 W/mK and has diameter of  $134 \pm 52.8 \,\mu\text{m}$  in average. To ensure a droplet to be held without sliding off, we tie a knot (of size 145–285  $\mu$ m) at one end of the fiber. The experiments are conducted within a chamber constructed by four vertical aluminum framing posts and surrounded with glass boards (of 25 cm × 15 cm in size) while the top and bottom sides are kept open.

We carry out droplet combustion experiments for a variety of pure liquid fuels such as alkanes and alcohols with different hydrocarbon contents. For each fuel, we repeat its droplet combustion experiment for 50 times. The starting droplet diameter for all the fuels is 1.34-2.11 mm. It should be noted that because there often exists a heat-up period at the early stage of burning, the initial droplet diameter  $D_0$  as well as the starting time t = 0 need to be re-defined at the moment when the droplet starts to decrease with time after that period. In each experiment, after a droplet is ignited with a lighter, we monitor and tape its burning and shrinkage using a high- speed CCD camera (Phantom v7.3) placed on the front side of the chamber. The video shooting is undertaken at 1000 frames/s and exposure time 1 ms. We further add an LED light source (LEDD1B, 14.4 W) behind the rear side of the chamber. This is to ensure the recording to be made in a backlighting manner. After recording, the video is stored and converted into a sequence of images of  $640 \times 480$  pixels in resolution. The length per pixel ranges from 0.03 to 0.05 mm/pixel, calibrated every time we use a new fiber or whenever we make adjustments in the experimental setup. The total number of the images is 500–3000, depending on the droplet lifetime. These images are then post processed using MATLAB to generate a series of snapshots for subsequent data analysis.

Because we use a fine hanging fiber to hold a droplet to keep it from falling due to gravity,<sup>13,14</sup> the droplet inevitably deforms away from spherical shape. To obtain a sensible value for *n*, it is necessary to take *D* to be the diameter of a volume-equivalent sphere.<sup>15,16</sup> This equivalent sphere approach is consistent with the notion that it is the droplet's volume (mass) responding to vaporization and the associated energy change. To obtain a correct volume of an equivalent sphere, it is necessary to subtract off the volume of the knot tied at the end of the suspended fiber, which guarantees *D* to vanish at  $t = t_{life}$ .

In order to more accurately determine the shrinkage exponent n from the log-log plot of  $(1 - t/t_{life})$  vs  $D/D_0$  using (3), the initial diameter  $D_0$  of a droplet is chosen at a re-defined starting moment t=0 after the early time heat-up period during which the droplet undergoes a simple thermal expansion prior to burning consumption. Furthermore, to avoid possible near-contact effects from the fiber, the data are taken only up to  $D/D_0 = 0.5$ .

### III. EXPERIMENTAL ATTESTATION OF VIOLATION OF THE $\mathsf{D}^2\text{-}\mathsf{LAW}$

Figure 1(a) displays sequential images for a typical droplet burning process using *n*-octane ( $C_8H_{18}$ ), revealing flame formation and diminishment due to the fuel droplet's supply and consumption. The droplet also responds to diminish its size, as shown in the corresponding zoom-in images.

Figure 2(a) displays log-log plots of  $(1 - t/t_{life})$  vs  $D/D_0$  based on (3) for a few representative alcohols and alkanes of low and high boiling hydrocarbons.<sup>17</sup> The measured average values of n for all the fuels are  $2.53 \pm 0.30 - 2.62 \pm 0.25$  (for the data up to  $D/D_0 = 0.5$ and 50 repeating runs), indicating a clear violation of the  $D^2$ -law. We also conduct the corresponding controlled droplet evaporation experiments (under the ambient temperature 70 °C) and found n = 1.96 $\pm 0.08 - 2.16 \pm 0.17$  (for the data up to  $D/D_0 = 0.5$  and 10 repeating runs), as illustrated in Fig. 2(b) for the representative fuels. This not only re-confirms the  $D^2$ -law, but also asserts that the nature of droplet evaporation is not altered by the fiber, and neither is that of droplet combustion. The latter is further supported by the measured droplet combustion values  $n = 2.57 \pm 0.19 - 2.69 \pm 0.14$  using the crossfiber technique,<sup>18,19</sup> which do not appear much difference compared to those using the hanging fiber technique above, as shown in Fig. 3. We have also repeated droplet combustion experiments with the use of a thinner fiber of  $\sim$ 35  $\mu$ m in diameter for hanging a droplet. It is found that the measured values of *n* also do not vary much compared to the values mentioned above using both the hanging fiber and the crossfiber techniques. All of the above evidence suggests that the shrinking kinetics of a burning droplet is not altered by the presence of fibers (nor by the knot). That is, although fibers can affect heat transfer to the droplet, the effects are not strong enough to alter the mechanisms governing the droplet vaporization process.<sup>21</sup>

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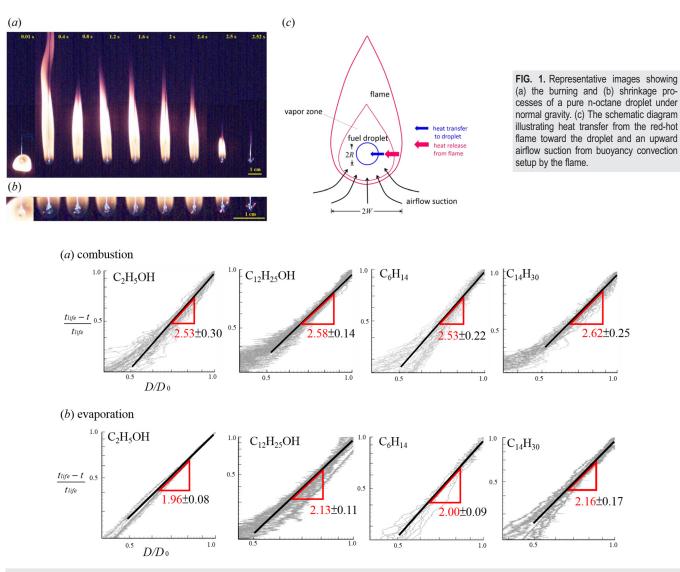


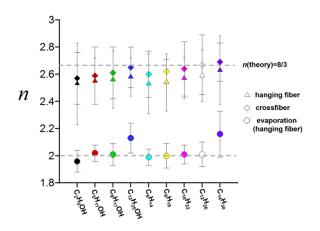
FIG. 2. Log-log plots of (1 - t/t<sub>life</sub>) against D/D<sub>0</sub> for determining the values of the shrinkage exponent n (indicated by the slopes) for representative alcohols and alkanes of low and high boiling hydrocarbons. n = 2.53 - 2.62 found for droplet combustion (a), showing a clear violation of the  $D^2$ -law with much higher values than n = 1.96 - 2.16for the corresponding evaporation results in (b).

### IV. THEORY FOR DROPLET COMBUSTION DUE TO FLAME-DRIVEN BUOYANT CONVECTION: NEW D<sup>8/3</sup>-LAW

We also develop a new theory to explain why the shrinking of a burning droplet under gravity displays an apparent departure from the  $D^2$ -law with n > 2. Here, we do not attempt a detailed modeling or rigorous mathematical formation to accurately describe every aspect of the droplet burning process. Rather, we aim to propose plausible mechanisms conceptually to capture key effects responsible for observed phenomena in scaling sense. We postulate that the process is mainly driven by buoyant convection set up by the flame around the droplet. As it involves simultaneous momentum, heat, and mass transfer, to better elucidate the effects at work we make the following assumptions. (i) transport processes are quasi-steady state,

(ii) non-ideal effects such as fiber effects and droplet asphericity only have minor impacts, (iii) heat transfer to the droplet is mainly through convective heat transfer from the heated air (plus vapor fuel) around the droplet, and (iv) while fluid properties often vary with temperature, the variations are not large enough to cause qualitative changes in the underlying physics. As will be shown shortly, with these assumptions, we are able to derive a new shrinking kinetic law with features successfully accounting for experimental observations. The success can then justify the reasonableness of these assumptions.

To begin with, the driving force comes from the immense heat released from the red-hot flame (of width 2W). As illustrated in Fig. 1(c), this heat sets up an upward natural convection due to buoyancy, sucking an airflow beneath the droplet at velocity<sup>21</sup>



**FIG. 3.** Measured values of the shrinkage exponent *n* for droplet combustion for a variety of pure liquid fuels, showing a close agreement with the theory prediction  $n = 8/3 \approx 2.67$ . They all significantly exceed those around n = 2 for the corresponding droplet evaporation experiments.

$$U \sim \left(g\beta\Delta TW\right)^{1/2}.\tag{4}$$

This airflow is inertially driven by buoyancy  $\rho\beta\Delta Tg$  due to the density reduction  $-\rho\beta\Delta T$  arising from the flame's temperature of  $\Delta T$  higher than the droplet's, where  $\beta(=1/T_{\infty})$  is the volume expansion coefficient of the air (at temperature  $T_{\infty} = 300$  K).

Such convection has profound impacts on both heat and mass transfers around the droplet. It begins by fuel burning within the flame (of volume  $V_f$ ), releasing an enormous heat at rate

$$(-r_{rxn}V_f)|\Delta H_{rxn}|,\tag{5}$$

due to a rapid fuel mass consumption at rate of  $r_{rxn}$  (<0 in mole/m<sup>3</sup>/s) with the heat of reaction  $\Delta H_{rxn}$  (in J/mole). The burning is mainly sustained by a continuous oxygen supply into the vapor zone (with a fraction of oxygen concentration decrease  $\Delta C_{O2}$ ) due to uptake by the upward airflow suction driven by the heating. Hence, the fuel mass consumption within the flame can be roughly represented by the oxygen consumption in the vapor zone

$$-r_{rxn}V_f \sim \Delta C_{\rm O2}UW^2. \tag{6}$$

Meanwhile, the fuel vapor (of diffusivity *D*) released from the droplet (with the concentration  $C_s$  at the surface) diffuses across the vapor zone, giving off a diffusive flux  $j_{fuel} \sim DC_s/W$  toward the flame. On the lower part of the flame where the burning is the most violent owing to the strong airflow suction, in particular, the burning happens so fast that  $j_{fuel}$  is immediately consumed by the supply oxygen convective flux  $j_{O2} \sim U\Delta C_{O2}$ . As a result of  $j_{fuel} \sim j_{O2}$  for such diffusion-convection-limited burning, the flame width is suppressed by the flow according to

$$W \sim (D/U)_{\gamma},\tag{7}$$

where  $\chi = C_s/\Delta C_{O2}$  is the concentration ratio reflecting the adverse impact by oxygen consumption.

The flow also promotes heat transfer from the flame to the droplet (of radius R), despite some heat loss out of the vapor zone because of advection by the upward flow. The net heat flow provides the heat transfer rate  $h\Delta TA$  over the droplet (with the heat transfer coefficient *h* and area  $A = 4\pi R^2$ ), sustained by the heat released from the flame through (5) and (6)

$$h\Delta TA \sim (-r_{rxn}V_f)|\Delta H_{rxn}| \sim \Delta C_{O2}UW^2|\Delta H_{rxn}|.$$
(8)

It is important to note that the droplet merely passively receives the above convective heat transfer from the upward vapor stream. That is, from the droplet's standpoint, since the natural convection here is not generated directly by the droplet but indirectly by the flame, the heat transfer to the droplet is of forced convection type. This is very distinct from a burning solid particle whose heat transfer is set up by the flame right next to it and hence is natural convection driven.<sup>22</sup> Therefore, the heat transfer coefficient *h* (on the air side of thermal conductivity *k*) in (8) can be evaluated from the Nusselt number Nu = hR/k according to<sup>21</sup>

$$Nu \sim Pe^{1/2},$$
 (9)

where the Péclet number  $Pe = UR/\alpha$  measures the strength of convection relative to conduction with  $\alpha (\approx 0.2 \text{ cm}^2/\text{s})$  being the thermal diffusivity of the air. Equation (9) is more applicable to the strong convection situation where *Pe* is large. In our case,  $Pe \sim 25$  for  $R \sim 1 \text{ mm}$  at  $U \sim 50 \text{ cm/s}$  under  $\Delta T \sim 10^3 \text{K}$  and  $W \sim 1 \text{ cm}$ . The exponent 1/2 in (9) comes from the slipping thermal boundary layer due to a possible fuel vapor film formed on the droplet surface.<sup>23</sup>

Such enhanced heat flow toward the droplet in turn raises the fuel mass vaporization rate  $\dot{m} = \rho_L \dot{V}/M$  (in mole/s through the fuel liquid density  $\rho_L$  divided by its molecular weight *M*) from the droplet, reflected by its volume reduction rate  $\dot{V} = 4\pi R^2 \dot{R} (< 0)$  due to a conversion of this heat into the latent energy to vaporize the fuel droplet

$$\dot{m}H_{vap} = -h\Delta TA,$$
 (10)

where  $\Delta H_{vap}$  (in J/mole) is the heat of vaporization. Note that if the heat transfer is purely by conduction, then h = k/R leads (10) to reduce to  $R\dot{R} = -k\Delta TM/\rho_L\Delta H_{vap}$ , recovering the  $D^2$ -law as (1). This also implies that a strong convection will render  $h \propto 1/R^m$  with  $m \neq 1$ , making n = m + 1 in (2) deviated from n = 2. For instance, a strong forced convection driven by a constant airflow gives m = 1/2 and n = 1.5.<sup>11</sup>

In droplet combustion under gravity, however, buoyant convection renders the flow speed *U* in *h* of (9) to no longer be constant but further to vary with both  $\Delta T$  and *W* that are functions of *R*, which can affect the value of *n* after an integration of (10). How  $\Delta T$  and *W* vary with *R* can be determined from (7), (8), and (9) under *U* given in (4). Specifically, we first re-write  $Pe = UR/\alpha$  in (9) as follows after substituting the velocity scale (4):

$$Pe \sim (\Delta T/T_{rxn})^{1/2} (W/\ell)^{1/2} (R/\ell).$$
 (11)

Here, we scale  $\Delta T$  with the temperature equivalent to  $\Delta H_{rxn}$ ,  $T_{rxn}$ =  $|\Delta H_{rxn}|/\tilde{C}_p$ . This temperature reflects the heat release capability of the fuel from the flame to the surrounding air (of molar heat capacity  $\tilde{C}_p$  and density  $\rho$ ). Note that it is not the adiabatic flame temperature  $T_{ad}$  since the latter measures the temperature rise when  $\Delta H_{rxn}$  is entirely released to air through complete conversion of the fuel into carbon dioxide and water.<sup>24</sup> The lengths W and R are scaled with the inherent buoyancy length determined solely by the fuel's and the gas's properties

$$\ell = (g\beta T_{rxn}/\alpha^2)^{-1/3}, \qquad (12)$$

which is about 100  $\mu$ m at  $T_{rxn}$  of typical value 10<sup>4</sup> K.<sup>24</sup> This inherent length is fixed for a given fuel-gas system, representing the ability to set off buoyant convection from heat release (via  $T_{rxn}$ ) from the flame with the necessity to work with gravity (via *g*). The shorter  $\ell$  the stronger the convective heat transfer from this flame-driven buoyant convection.

Next, we re-write (7) and (8) in terms of W and  $\Delta T$  by substituting (4) into (7) and replacing  $|\Delta H_{rxn}|$  by  $T_{rxn} = |\Delta H_{rxn}|/\tilde{C}_p$  in (8). This allows us to re-express (7) and (8) in the dimensionless form:

$$W/\ell \sim (\Delta T/T_{rxn})^{-1/3} (Le^{-1}\chi)^{2/3},$$
 (13a)

$$Nu(\Delta T/T_{rxn}) \sim f Pe(W/\ell)^2 (R/\ell)^{-2}, \qquad (13b)$$

where  $f = (M/\rho)C_s$  is roughly the mole fraction of the vaporizing fuel at the droplet surface and  $Le = \alpha/D(\approx 1)$  is the Lewis number. Substituting (9) and (11) into (13b), the resulting form of (13b) and (13a) provides coupled relationships for  $\Delta T$  and W. How  $\Delta T$  and Wvary with R can then be determined as

$$\Delta T \sim T_{rxn} f^{2/3} (R/\ell)^{-1} L e^{-1} \chi,$$
 (14a)

$$W \sim \ell (R/\ell)^{1/3} f^{-2/9} L e^{-1/3} \chi^{1/3}.$$
 (14b)

To derive the shrinking law, we re-write (10) as

$$\rho_L \Delta H_{vap} \dot{R}^2 = -2k N u \Delta T. \tag{15}$$

Substituting (9) along with (11) into (15), writing  $\Delta T$  and W in terms of R using (14a) and (14b), and integrating with respect to R = D/2, we arrive at a new law governing the true shrinking kinetics for a burning droplet under gravity

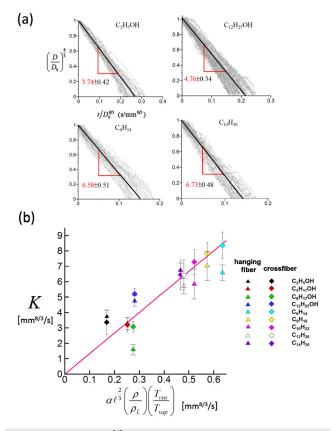
$$D^{8/3} = D_0^{8/3} - Kt, (16)$$

in which the exponent  $n = 8/3 \approx 2.67$  well captures the measured values for all the fuels, as shown in Fig. 3. More importantly, we find that the burning rate constant takes the form

$$K \sim \alpha \ell^{2/3} (\rho/\rho_L) \left( T_{rxn}/T_{vap} \right) f^{7/9} L e^{-4/3} \chi^{4/3}, \tag{17}$$

where  $T_{vap} = \Delta H_{vap}/\tilde{C}_p$  is the temperature equivalent to  $\Delta H_{vap}$ . Equation (17) clearly indicates that the units of *K*, rather than  $\alpha$ 's mm<sup>2</sup>/s in the  $D^2$ -law, now change to mm<sup>8/3</sup>/s due to the additional inherent buoyancy length  $\ell$  defined by (12). From this point of view, adding a buoyant convection correction term directly to *K* in the  $D^2$ -law (1)<sup>25,26</sup> may not be legitimate unless the effects are weak.

By plotting  $(D/D_0)^{8/3}$  against  $t/D_0^{8/3}$  according to (16), the value of *K* can be obtained from the slope of a virtually straight line displayed by the plot, as illustrated in Fig. 4(a). For a given fuel, the slope of each experimental run is obtained directly from the ratio between  $(D/D_0)^{8/3} = 1$  at the starting point t = 0 and  $t/D_0^{8/3}$  at the end point  $t = t_{life}$ , which gives  $K = D_0^{8/3}/t_{life}$  as defined in (16). The solid black line represents the result by averaging over those of 50 repeating runs (grey lines), giving the mean value of *K* plus standard deviation) indicated by the corresponding slope. Figure 4(b) shows that the measured mean values of *K* for all the fuels vary roughly linearly with  $\alpha \ell^{2/3} (\rho/\rho_L)(T_{rxn}/T_{vap})$  according to (17), which also supports our theory for deriving the 8/3 law (16).



**FIG. 4.** (a) Plots of  $(D/D_0)^{8/3}$  vs  $t/D_0^{8/3}$  for determining the average values of the burning rate constant *K* (indicated by the slopes) for representative fuels according to the new 8/3 law (13). (b) plots the measured values of *K* against  $\alpha \ell^{2/3}(\rho/\rho_L)$  ( $T_{rxn}/T_{vap}$ ) for all the fuels in Fig. 3, confirming (14). The line is the best fit of the data.

Equation (17) may give an expression that the burning rate constant under gravity is  $K(g \neq 0) \sim \ell^{2/3} K(g = 0)$  by having the burning rate constant K(g = 0) in the absence of gravity directly multiplied by the 2/3 power of  $\ell$ . However, this is not a correct interpretation because  $T_{rxn}$  in (17) is also contained in  $\ell$  given by (12). The more appropriate form of K in (17) should be written below by reexpressing  $T_{rxn}$  in terms of  $\ell$  using (12)

$$K \sim \alpha^{3} \ell^{-7/3} g^{-1} (\rho/\rho_{L}) \left(\beta T_{vap}\right)^{-1} f^{7/9} L e^{-4/3} \chi^{4/3}.$$
(18)

Because  $\ell \propto T_{rxn}^{-1/3}$  and  $T_{rxn} \propto |\Delta H_{rxn}|$  together with  $T_{vap} \propto \Delta H_{vap}$ , (18) indicates that impacts of  $\ell$  lead to

$$K \propto |\Delta H_{rxn}|^{7/9} / \Delta H_{vap},$$
 (19)

which provides an explicit dependence for how *K* increases with  $|\Delta H_{rxn}|$  due to heat release from flame combustion. It is clear that *K* is diminished by  $\Delta H_{vap}$  in an inversely proportional manner exactly like the way in droplet evaporation because the vaporization energy loss  $\dot{m}$   $\Delta H_{vap}$  determines the rate of the vaporizing fuel mass according to (10). Compared to droplet evaporation where  $K \propto (\Delta H_{vap})^{-1}$ , the droplet lifetime in droplet combustion will be much shorter because *K* 

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is significantly enlarged by a factor of  $|\Delta H_{rxn}|^{7/9}$  according to (19). This explains why the droplet lifetime in combustion is much shorter than that in evaporation. Although both  $|\Delta H_{rxn}|$  and  $\Delta H_{vap}$  grow with hydrocarbon content,  $\Delta H_{rxn}^{7/9}/\Delta H_{vap}$  may not, this may explain why the measured *K* values for alcohols roughly increase with carbon number whereas those for alkanes show the opposite trend, as shown in Fig. 4(b). At the same carbon content, the value of  $|\Delta H_{rxn}|$  for an alkane and that of an alcohol are about the same but  $\Delta H_{vap}$  for the latter is much higher, this explains why the value of *K* for the former is larger.

It should be noted that *K* may depend on  $D_0$  because the amount of the oxygen consumption  $\Delta C_{O2}$  in  $\chi$  should increase with the amount of the fuel supply through the droplet initial size  $D_0$ , depending on the detailed reaction kinetics whose rate constants are likely size dependent due to soot particle formation.<sup>27</sup> Nevertheless, given that the measured trend of *K* fairly agrees with (17) in Fig. 4(b), such a dependence may be weak. This also implies that the droplet's shrinking kinetics is mainly controlled by the bottom portion of the flame where the burning takes place most vigorously without serious sooting caused by incomplete burning.

Other fuel specific effects such as droplet heating<sup>28</sup> and water condensation effects<sup>29</sup> may also contribute, as occurring to high carbon fuels and low carbon alcohols respectively. In spite of this, we have shown in Fig. 4(b) that the measured values of K for the variety of fuels covering low and high carbons closely follow the predicted trend (17) according to the new  $D^{8/3}$ -law (16). From a physical point of view, these effects are likely unimportant when strong buoyant convection is present. This is because the droplet's lifetime would become much shorter due to enhanced heat transfer by buoyant convection, the droplet does not have time to build up energy inside,<sup>30</sup> making internal droplet heating less prominent. Also because the temperature around the droplet is high due to flame combustion, this might lower the degree of water condensation onto the droplet surface. Little influences caused by these fuel specific effects may also explain the slight variations of the measured K values among different fuels seen in Fig. 4(b). This also strongly implies that such fuel specific effects should not be so prominent to alter the dominant buoyancy-driven transport effects we propose here. This is also consistent with the result that the measured values of n for these fuels in Fig. 3 are roughly constant to display an invariance with respect to fuel types and hydrocarbon contents, in line with the notion that the droplet shrinking kinetics here is mainly transport determined and has little relevance to specific aspects of fuels.

### V. CONCLUSION AND PERSPECTIVES

In summary, we have demonstrated both experimentally and theoretically that the classical  $D^2$ -law is not the true law for a burning droplet under gravity. We also discover the new  $D^{8/3}$ -law, capable of capturing the shrinking kinetics of single burning droplets for a variety of pure liquid fuels such as alcohols and alkanes. This is also the first time that full impacts of flame-driven buoyant convection on heat and mass transfer around a droplet are taken into account.

The key aspect of our theory is that the droplet shrinking kinetics is mainly transport determined, insensitive to combustion chemistry and detailed reaction kinetics. We identify that the origin of the  $D^{8/3}$ -law is rooted in the inherent buoyancy length determined solely by fuel and gas properties according to (12). For this reason, the burning rate constant *K* no longer behaves like the thermal diffusive  $\alpha$  of the gas phase, but turns into  $\alpha^3 \ell^{-7/3}/g$  under the influence of the gravitational acceleration g. Therefore, to better characterize the behavior of a burning droplet in a normal gravity environment, this law should be more adequate than the  $D^2$ -law. More importantly, it also provides a self-contained recipe that is able to correlate the size reduction history of a fuel droplet with key factors such as the adiabatic flame temperature and the heat of vaporization to determine the features of shrinking kinetics.

We emphasize that the present theory is a merely scaling one that considers only key factors and dominant effects in a droplet combustion process under the influence of gravity. It does not mean that other subtle aspects (e.g., droplet heating or transient effects,<sup>28</sup> humidity,<sup>29,31</sup> radiation,<sup>32</sup> etc.) do not exist nor contribute. Since the theory's predictions agree with experimental observations shown in Figs. 3 and 4(b), it can be said that the theory successfully captures essential physics and that such subtleties may not have substantial impacts on the phenomenon. If they do appear to be as important as the effects we consider, it is not possible to attain an excellent agreement between experiment and theory shown here.

Another issue is that non-ideal effects such as droplet deformations,<sup>33</sup> fiber effects,<sup>34,35</sup> variable transport properties,<sup>6</sup> and fuel specific effects (e.g., soot formation<sup>27</sup> and water condensation in combustion of highly volatile alcohol droplets<sup>29</sup>) often exist and are inevitable. It is likely that variations caused by these effects should not span more than an order of magnitude changes; otherwise, we will observe completely different results or phenomena, making the success of the present theory impossible. Given the success of our theory on accounting for experimental observations, it can be said that the assumptions we made for the theory in the beginning of Sec. IV are mostly valid, or at least not seriously violated.

In any case, while variations or non-idealities caused by many other factors always exist, they can be tested by comparing experimental observations to theory's predictions to see if they can strongly intervene with or outweigh the proposed dominant effects governing the phenomenon. If not, they will not alter the predicted features that have been shown to agree with experimental observations. Quantitatively, such effects may only contribute to the dimensionless pre-factor of the scaling relationship (17) for the burning rate constant K without changing the scaling structure, which is responsible for slight variations in the measured values of K in Fig. 4(b).

We should also add that most of the subtleties and non-idealities are largely examined or discussed based on the notions of the  $D^2$ -law. This prohibits a simple extension to the present lack-of-study situation where buoyant convection prevails. Our theory at least provides a starting framework for future assessments on more realistic considerations for gaining a better understanding of actual droplet combustion processes under gravity.

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### AUTHOR DECLARATIONS Conflict of Interest

The authors have no conflicts to disclose.

### Author Contributions

You-An Chen: Data curation (equal); Investigation (equal). Chih-Hsiang Chiang: Data curation (equal); Investigation (equal). Chao-Yi Yang: Data curation (equal); Investigation (equal). Shou-Yin Yang: Investigation (equal); Resources (lead); Supervision (equal). Hsien-Hung Wei: Conceptualization (lead); Formal analysis (lead); Investigation (equal); Methodology (lead); Supervision (equal); Writing – original draft (lead); Writing – review & editing (lead).

### DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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