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The role of air entrainment on the outcome of drop impact on a solid surface

Dedicated to Professor Wilhelm Schneider on the occasion of his 70th birthday

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Abstract The characteristic conditions causing spreading or splashing after drop impact on solid surfaces are considered together with the underlying mechanisms. To this end, the results of the various studies published over the past few years that have addressed the issue of splashing after droplet impact, specifically in terms of the definition of a splashing threshold, are critically compared and synthesized. The discussion aims at clarifying some of the conflicting findings. Information drawn from these considerations is used to distinguish between various splashing thresholds and it is shown that there exists a distinct difference between splashing on smooth and on rough surfaces, both in terms of the splashing thresholds and in terms of the mechanisms. Finally, a physical mechanism akin to air entrainment in dynamic wetting is proposed that may be of primary importance for the inception of splashing as well as fingering on smooth surfaces.

1 Introduction

When a drop impinges on a dry solid surface it can stick to the surface, spread, splash, or bounce off the surface. The actual outcome depends on the particular impact conditions. Although these phenomena have received constant and significant attention since the seminal work of Worthington in 1876 [27,28], the mechanisms controlling the outcome of a drop impact are still only partially understood. This is particularly true if one considers critical conditions for the onset of splashing. Critical conditions of splashing inception, the so-called splashing thresholds, cited in the literature have often been obtained under slightly different experimental conditions and thus are partly contradictory. Furthermore, no consensus exists regarding the basic physical mechanisms that cause splashing. In the following we re-evaluate published splashing thresholds and propose a possible mechanism underlying the inception of splashing. We limit our considerations to drop impacts without phase change, i.e., the onset or enhancement of splashing resulting from evaporation or solidification processes is not considered.

A splash is defined as a drop impact that results in the disintegration of the drop, i.e., at least one secondary droplet is formed [14]. Different types of splashes, such as "corona" splash and "prompt" splash have been distinguished [15]. Right after impact a thin lamella begins spreading radially out on the solid surface. The advancing rim of the lamella may separate from the surface [11,15]. In the case of splashing, secondary droplets are normally first shed from the rim of the lamella. A corona splash is considered to occur if the rim of the lamella is lifted off the surface before secondary droplets are formed whereas in the case of a prompt splash

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droplets are ejected directly from the advancing contact line between the liquid and the solid. The shedding of droplets is usually preceded by the formation of disturbances on the rim of the lamella. This so-called fingering is likely connected with splashing [1]. The formation of "arms" and "fingers" was first reported by Worthington [27,28]. It is also possible for the expanding lamella to reach its maximum extension without disintegrating. During the subsequent contraction phase two other scenarios can lead to the formation of secondary breakup, receding break-up and partial rebound [15]. These scenarios are not considered splashing events hereinafter.

In the following, we first present a dimensional analysis of splashing based on the potential controlling parameters. A review and comparison of published experimental results pertaining to splashing thresholds is then conducted, including a discussion of the specific experimental setups used in those studies. Finally, information drawn from these considerations is used to distinguish between various splashing thresholds and a mechanism is proposed that may be of primary importance for the inception of splashing as well as fingering on smooth surfaces.

2 Critical conditions of splashing inception

2.1 Dimensional analysis

A very large number of factors potentially influences drop impact dynamics and thus splashing inception. In the basic configuration of interest however, normal impact of a single drop on a smooth dry substrate, the parameters typically considered are: the diameter *d* and the velocity *v* of the impinging drop as well as the density ρ , dynamic viscosity μ and surface tension σ of the liquid. As is well known, the Buckingham Π theorem allows these five parameters to be combined into two independent non-dimensional numbers. Typically, the Weber number (*We*) and the Ohnesorge number (*Oh*) are chosen [12,24]:

$$We = \frac{\rho v^2 d}{\sigma}, \quad Oh = \frac{\mu}{\sqrt{\rho d\sigma}}$$
 (1a)

but the Reynolds number (*Re*) and the capillary number (*Ca*):

$$Re = \frac{\rho v d}{\mu}, \quad Ca = \frac{v \mu}{\sigma}$$
 (1b)

have been used to interpret and analyze experimental data as well. These numbers are not independent; they are related to each other as follows:

$$Oh = \frac{We^{\frac{1}{2}}}{Re}, \quad Ca = \frac{We}{Re}, \quad Re = \frac{Ca}{Oh^2}.$$
 (2)

The splashing threshold is often expressed as a functional relation between two of these non-dimensional numbers. The relations can typically be rewritten so that only a single non-dimensional number describes the splashing limit. When this number exceeds or, respectively, falls below a critical value splashing occurs or (resp.) does not occur.

It is generally accepted [1,32] that in the case of oblique impacts the normal component of the drop incident velocity is most important and should therefore be used in forming the dimensionless numbers. This approach is also adapted in the following and will not be further discussed.

Additional parameters may have to be included in the analysis of droplet impacts as the configurations considered deviate from the ideal case described above. The roughness of the target surface, R_a , was varied in several studies. This requires the addition of another non-dimensional number, for example R_a/d , to the selected pair of independent non-dimensional numbers [12]. In some often cited cases a curved surface was used as a target [7,9]. Xu et al. [29] have shown recently in an important paper that the properties of the surrounding gas, especially its pressure and density, also play an major role in determining the impact outcome. Finally, the surface energy as well as the structure (heterogeneity) of the impacted plate was varied in a few cases possibly resulting in different dynamics of the contact angle between the solid and the liquid [16,30].

2.2 Review of drop splashing criteria after impact on a solid surface

Although drops splashing on dry solid surfaces are often addressed in the literature there are surprisingly few papers in which splashing conditions have been examined quantitatively. Perhaps the earliest correlations of splashing limits were given by Walzel [24] and Stow and Hadfield [20]. These correlations as well as more recent ones concerning the onset of splashing for drop impacts on dry solid surfaces are summarized in Table 1 (third column) together with the variables that were tested in each case and the ranges over which they were tested (second column). Splashing thresholds have often been expressed as a power law, typically the product of an exponentiated Weber number or Reynolds number, and the Ohnesorge number. Following Vander Wal et al. [22] in Table 1 all splashing thresholds have been rewritten in terms of a critical Ohnesorge number, Oh_{crit} , given as a function of the Reynolds number (fourth column). This function is always of the form

$$Oh_{\rm crit} = \alpha R e^{\beta},$$
 (3)

where α is a constant of proportionality and β an exponent. In this manner the various limits can be easily compared. Except for the splash threshold proposed by Walzel [24], splashing occurs for $Oh > Oh_{crit}$. However, as has already been noted by Rioboo et al. [15] a representation of splashing thresholds in this manner is oversimplified since it overlooks the influence of substrate roughness and material properties and fails to distinguish between the various splashing regimes. Furthermore, some of these correlations have been stated non-dimensionally although only few parameters occurring in the Ohnesorge and Reynolds numbers have actually been varied in the corresponding experiments. Consequently, we also provide the splash criteria in a dimensional form that is linear with respect to the impact velocity (cf. Table 1, fifth column). In these dimensional expressions of the splash criteria, the variables that were varied during the associated experiments are written on the left-hand side of the inequality while all others are included on the right-hand side.

As can be seen in Table 1, consensus prevails among studies on the splashing of drops in that a transition from spreading to splashing is hindered by an increase in the surface tension (only one reference conflicts with this consensus, it is discussed in more details in Sect. 2.3) and is promoted by an increase of either of the following parameters: impact velocity and diameter of the drop, or the roughness of the impacted surface. The latter is not included in the dimensional splash criteria of Table 1 because quantitative information is not always available. Note, however, that the effect of the viscosity of the liquid is not clear.

According to Walzel [24], Mundo et al. [12], Cossali et al. [6] and qualitatively also to Rioboo et al. [15], an increase in viscosity inhibits splashing. On the contrary, Range and Feuillebois [13] who used water–glycerol mixtures, found that a tenfold increase of the Ohnesorge number, realized by increasing the viscosity (all other parameters being kept constant) did not change the critical Weber number of splashing. It should be noted that both the range of Ohnesorge numbers and the water–glycerol mixtures used are similar to those in the experiments of Cossali et al. [6] who observed a hindering effect of the viscosity promotes splashing. Loehr [11] had reported the same trend earlier albeit qualitatively. This contradiction, which could be the result of slight differences in experimental conditions, appears not to have been previously discussed in the literature.

The various splash limits listed in Table 1 can be divided into two groups according to the effect of viscosity, i.e., to the value of the exponent β in Eq. (3). In the first group, $\beta \approx -5/4$ and Eq. (3) reduces to the often cited relationship of Mundo et al. [12], the coefficient α depending on the surface roughness. The second group contains relations with $\beta \ge -1$, i.e., cases in which the viscosity has no influence or its increase promotes splashing. Thresholds relating to these two groups are graphed in Fig. 1. As can be seen, a close agreement between different correlations is not obtained but the general trend is the same—regardless of $\beta < -1$ or $\beta \ge -1$, i.e., of the effect of the viscosity. Vander Wal et al. [22] proposed to approximate the exponent obtained from the fitting of their experimental splashing threshold, $\beta = -0.609$, by $\beta = -1/2$. In this way their splashing threshold could be simplified and expressed solely by a critical capillary number:

$$Ca_{\rm crit} = 0.35^2 = 0.1225,$$
 (4)

splashing being obtained for $Ca > Ca_{crit}$. The corresponding correlation in terms of the Reynolds and Ohnesorge numbers ($Oh_{crit} = 0.85Re^{-1/2}$) has also been included into Fig. 1. It approximates the experimental results of Vander Wal et al. [22] quite well. Although certain aspects such as the density and the size of the drop are no longer considered, the splash threshold expressed in terms of a critical capillary number is also quantitatively consistent with the other correlations shown in Fig. 1.

The primary importance of the capillary number is well-known for forced wetting as, for example, in coating flows [8]. There, air is entrained if the capillary number formed with the velocity of the moving

Source	Range of variables	Non-dimensional splash criterion	Splash threshold: $Oh_{crit} = f(Re)$	Dimensional splash criterion
Walzel [24] and Schmidt and Knauss [18]	ho: water, water/glycerol σ : 63 mN/m μ : 1.0–1.9 mPas R_a : smooth PVC plate	$We \times Oh^{-2.8} > 7.9 \times 10^{10}$	$Oh_{\text{crit}} = \alpha Re^{5/2}$ $\alpha = (7.9 \times 10^{10})^{-5/4}$ splash for $Oh < Oh_{\text{crit}}!$	$\rho^{6/5} \sigma^{1/5} \mu^{-7/5} d^{6/5} v > (7.9 \times 10^{10})^{1/2}$. It is not clear which parameters have been changed in the experiments
Stow and Hadfield [20]	ρ, $σ$, $μ$: water only d: 1.4–4.4 mm v: 2.1–4.2 m/s R_a : 0.05–12.0 μm	$Re^{0.31} We^{0.69} = \beta f(R_a)$ $\beta = (2\rho/\mu)^{0.31} (2\rho/\sigma)^{0.69}$	$\begin{array}{l} Oh_{\rm crit} = \alpha Re^{-1.225} \\ \alpha = (\beta f(R_a))^{0.725} \\ f(R_a) = 4.2 \\ -13.5 \times 10^{-3} {\rm m}^{2.69}/{\rm s}^{1.69} \end{array}$	$d^{0.59} v \approx d^{3/5} v > f(R_a)$
Mundo et al. [12]	$\rho: 789-998 \text{ kg/m}^3$ $\sigma: 22-72 \text{ mN/m}$ $\mu: 1.0-2.9 \text{ mPas}$ $d: 60-150 \times 10^{-3} \text{ mm}$ v: 12-18 m/s $R_a: 2.8, 78.0 \text{ µm}$	$Oh \times Re^{5/4} > K$ $K = f(R_a)$	$Oh_{\text{crit}} = K R e^{-5/4}$ $K = f(R_a) \approx 57.7$ for $R_a = 2.8$ and 78.0 µm	$\rho^{3/5} \sigma^{-2/5} \mu^{-1/5} d^{3/5} v > K^{4/5}$
Hardalupas et al. [7]	ρ : water/ethanol/ glycerol σ : 58–73 mN/m μ : 1.0–2.4 mPas μ : 1.60–230.10 ⁻³ mm ν : 6–13 m/s R_a : 35 mn, wetted surface	$Oh \times Re^{1.2} > K'$ $K' = f(d_{\text{sphere}})$ $K' \approx 90 \text{ for } d_{\text{sphere}} \to \infty$	$Oh_{\text{crit}} = K'Re^{-1.2}$ $K' = f(d_{\text{sphere}})$ $K' \approx 90 \text{ for } d_{\text{sphere}} \to \infty$	$ ho^{7/12} \sigma^{-5/12} \mu^{-1/6} d^{7/12} v > K^{'5/6}$
Cossali et al. [6]	Water and Water/glycerol $d :\sim 3.0-3.5 \text{ mm}$ $v :\sim 2.0-6.5 \text{ m/s}$ $R_a :\sim 0.2 \text{ µm}$	$Oh \times Re^{1.25} > K''$ K'' = 119.23	$Oh_{\rm crit} = K''Re^{-5/4}$ $K'' = 119.23$	$\mu^{-1/5} d^{3/5} v > K'^{4/5} \rho^{-3/5} \sigma^{2/5}$

Table 1 Selected published correlations of critical conditions for the inception of splashing for drops impinging on solid surfaces

Source	Range of variables	Non-dimensional splash criterion	Splash threshold: $Oh_{crit} = f(Re)$	Dimensional splash criterion
Range and Feuillebois [13]	ρ, $σ$: ethanol, water, water/glycerol μ: factor of 10 d:~3.9 mm v:~2–4 m/s R_a : 0.01544–2.765 μm (and higher)	$We > We_{\text{crit}} (\approx 310)$ (for $Oh \leq 0.02$)	$Oh_{\text{crit}} = We_1^{1/2}Re^{-1}$ for water/glycerol mixtures on a aluminum plate of roughness $R_a = 0.4365 \ \mu\text{m}$: $We_{\text{crit}} \approx 310$ for water at $R_a = 0.01544 \ \mu\text{m}$ $We_{\text{crit}} \approx 1000$	$ ho^{1/2} \sigma^{-1/2} \mu^0 v > W e_{ m cnit}^{1/2} d^{-1/2}$
Vander Wal et al. [22]	$\begin{array}{l} \rho: 684-1080 \mathrm{kg/m^3} \\ \sigma: 20-73 \mathrm{mN/m} \\ \mu: 0.5-3.4 \mathrm{mPas} \\ u: 2.17-4.22 \mathrm{m/s} \\ v: 2.17-4.22 \mathrm{m/s} \\ R_a: < 10 \mathrm{nm} \end{array}$	$Oh \times Re^{0.609} > 0.85$	$Oh_{\text{crit}} = 0.85Re^{-0.609}$ approximated by: $Oh_{\text{crit}} = 0.35Re^{-1/2}$ i.e., $Ca_{\text{crit}} = \text{const} = 0.1225$	$\rho^{0.179} \sigma^{-0.821} \mu^{0.642} v > 0.7658 d^{-0.179}$ approximated by: $\sigma^{-1} \mu v > 0.1225$
Xu et al. [29]	ρ , σ : methanol, ethanol, 2-propanol μ/ρ : 0.68–2.60 10 ⁻⁶ m ² /s d:~ 3.4 mm v: 2-7 m/s p_g : 1–100 kPa μ_g : 15.3–25.6 µPas M_g : 4–146 Dalton R_d : "very smooth" (glass microscope slide)	$\sum_{N=0.45} S_{C}/\Sigma_{L} > N = O(1)$	$Oh_{\text{cut}} = (2N\rho v/(\rho_g c_g))^{1/2} Re^{-3/4}$ here, $(2N\rho v/(\rho_g c_g))^{1/2} \approx 3.6$ is used (cf. the derivation in the appendix)	$\rho_g^2 c_g^2 \mu v > 0.81 \rho \sigma^2 d^{-1}$
Col. 1: reference, Col. 2: mai splashing threshold recast in during the associated experin	n parameters considered and their the form $Oh_{crit} = f(Re)$, Col. 5: c aents are written on the left-hand	values or ranges, Col. 3: non-dimen dimensional form of the splash crite side of the inequality while all other	sional splashing criteria as they appear in t ria expressed linearly with respect to the in rs are included on the right-hand side)	he referenced paper, Col. 4: non-dimensional npact velocity (the variables that were varied



Fig. 1 Comparison of experimentally determined splashing thresholds with the approximation $Ca_{crit} = 0.1225$ represented by the critical Ohnesorge number as a function of the Reynolds number

contact line exceeds a critical value. Expressing the splash boundary using only the capillary number is hence remarkable because it allows comparing the onset of splashing with the onset of air entrainment in coating flows, thus providing some insight regarding the nature of the mechanism potentially underlying the inception of splashing. Before this is discussed in more details (Sect. 3) the various experimental conditions under which splashing has been studied are considered.

2.3 Influence of the variability in experimental conditions on splashing criteria

The correlation of Walzel [24] fits into neither of the two groups introduced above and has therefore not been included in Fig. 1. It is based on only a few tests performed in the high Reynolds number range (15,000 < Re < 30,000, including also data of [18]) while all other thresholds listed in Table 1 relate to impacts at Reynolds numbers Re < 15,000. This may explain why Walzel observes splashing when the Ohnesorge number falls *below* a critical value, which contradicts all other splash limits of Table 1. What appears to be a slightly promoting influence on splashing of the surface tension may be an artefact of scaling since the surface tension was not really changed by Walzel. When Walzel's threshold is expressed in terms of a Weber number it results in a threshold that is one order of magnitude greater than thresholds determined at smaller Reynolds numbers (cf. [14]). It is not clear how Walzel determined the formation of secondary droplets and it may well be that the formation of the very first splash droplets escaped detection thus resulting in too high a threshold. Because of these uncertainties and the different Reynolds number range, we will not further consider Walzel's correlation.

The experiments of Stow and Hadfield [20] were performed with only one liquid (water) but for surfaces of different, well-defined roughness. The Reynolds number was varied in the range of $Re \approx 5,800-9,300$. Prior to each impact the surface was carefully cleaned and dried. The non-dimensional splash limit proposed by Stow and Hadfield [20] is in agreement with the form provided by Mundo et al. [12], with a threshold depending on the roughness. The lower threshold according to Stow and Hadfield [20] shown in Fig. 1 corresponds with a very rough surface ($R_a = 12 \,\mu\text{m}$) and prolongs the threshold of Cossali et al. [6] quite well to higher Reynolds numbers. For smooth surfaces the threshold is greater. The curve relating to a low surface roughness of $R_a = 0.05 \,\mu\text{m}$ that has also been included in Fig. 1 is much closer to the correlation of Vander Wal et al. [22] that was obtained with a smooth surface.

The importance of surface roughness on splashing was already observed qualitatively by Levin and Hobbs in 1971 [9]. They studied the impact of drops on dry rough surfaces (as well as on thin liquid layers). The Reynolds numbers considered ranged from $Re \approx 4,900-13,900$. They only varied the droplet impact velocity with all other parameters kept constant. Hence, the Ohnesorge number, Oh = 0.0022, stayed constant too. Impacts on rough surfaces always resulted in splashes while after impact on a "very smooth solid surface" drops "simply spread out radially". These findings thus agree well with the observations of Stow and Hadfield [20].

Mundo et al. [12] studied splashing for a wide variety of conditions. They used droplet streams formed by a forced disintegration of a liquid jet. The frequency f of the droplet stream was very high (f > 27 kHz). The droplets impinged onto a rotating cylinder the surface of which was wiped by a rubber lip to remove the liquid of previous drop impacts. Nevertheless, it is likely that some moisture remained at the impact site, which is known to influence wetting behavior (see Sect. 3). Similarly, Hardalupas et al. [7] let droplets impinge onto curved surfaces (spheres) that were wetted from preceding impacts. Here, the liquid simply flowed off the curved surface forming a film having a thickness of about 2.5% of the droplet diameter. Although this is much smaller than the typical film thickness seen in experimental investigations of droplet impacts on thin liquid films (for example, the film used in the experiments of Yarin and Weiss [33] had a thickness on a dry surface. The correlation of Hardalupas et al. [7] has nonetheless been included in the present considerations because in the limit of a target sphere of infinite diameter d_{sphere} , it agrees surprisingly well with another correlation that was obtained from experiments in which the state of the surface was not clearly stated thus providing some hint at the degree of wetness in the latter case (cf. last paragraph of Sect. 3.3).

Cossali et al. [6] were actually mainly concerned with splashing on thin liquid films. But they also considered the limit of vanishing film thickness. However they do not describe how the surface was prepared in this limiting case. In particular, it is not mentioned whether the surface was completely dry or still slightly wet. The surface itself was rather smooth (roughness $R_a \approx 0.2 \,\mu$ m).

Range and Feuillebois [13] let drops detach under the effect of gravity from the tip of a needle. Using water, they carefully checked a relation between the diameters of the drop and the needle, the surface tension and density of the liquid, and the acceleration of gravity. They then worked with drops of a constant size. However, it is not clear whether the drop diameter remained constant when the Ohnesorge number was changed by using water–glycerol mixtures of different viscosities since the size of drops detaching from the needle may depend on viscosity [26]. According to Range and Feuillebois [13] who changed the surface tension by using ethanol, the critical Weber number for splashing depends also on the particular combination of liquid and surface material. This is attributed to the variations in contact angles, but viscosity was also at least slightly changed in these experiments. For a particular combination of liquid and surface material the critical Weber number of splashing varies greatly with surface roughness. For example, for water drops impinging on glass $We_{crit} \approx 300$ at $R_a = 0.403 \,\mu\text{m}$ and $We_{crit} \approx 1,000$ at $R_a = 0.015 \,\mu\text{m}$, respectively.

A wide variety of different impact conditions was tested by Vander Wal et al. [22] with the intention of establishing an empirical splash limit for drops impinging on dry surfaces. The surface roughness was extremely small ($R_a < 10$ nm). Many parameters were changed appreciably but the drop size was constant for all liquids tested. An actual transition from spreading to splashing was observed for five liquids. In the case of the other liquids the spreading or splashing behavior agrees well with the correlations of Vander Wal et al. [22] both, using the Reynolds and Ohnesorge number ($\beta = -0.609$) and the capillary number alone ($\beta = -1/2$). Vander Wal et al. [23] also found that it is possible for the expanding rim of the lamella to separate from the surface without shedding secondary droplets during spreading on very smooth surfaces (roughness $R_a < 10$ nm).

Xu et al. [29] studied the inhibition of splashing by decreasing the pressure in the surrounding gas. They found that decreasing the ambient gas pressure can suppress splashing and that the gas pressure at which droplets cease to splash increases when the kinematic viscosity of the liquid is decreased. Very smooth glass surfaces (roughness not specified) were always cleaned and dried before impact. Methanol, ethanol and 2-propanol were used. Hence, the influence of changes in surface tension and density are negligible compared to that of the kinematic viscosity. Although this way of determining a splash threshold is not directly comparable to the other approaches in which the gas pressure is kept atmospheric and other variables were changed, the result that increasing the viscosity promotes splashing is remarkable. In subsequent tests Xu [30] showed that this behavior depends on the absolute value of the kinematic viscosity of the liquid. For more viscous liquids ($\mu/\rho \gtrsim 4 \times 10^{-6} \text{ m}^2/\text{s}$) the trend may become reversed and splashing can be suppressed by increasing the viscosity.

The preceding discussion has the following consequences. Existing correlations of splash thresholds can be divided into (at least) two groups. In addition to the roughness of the target surface, its degree of dryness may also be important. Finally, for very smooth surfaces there is evidence that the wetting behavior plays an important role in the inception of splashing. This is discussed next.

3 Air entrainment and the inception of splashing

In forced wetting experiments, for example, when a solid surface is plunged into a liquid pool, it is well-known that the liquid spreading on the solid surface cannot displace the receding fluid if the velocity of the contact line exceeds a certain critical value [8]. This phenomenon is of great importance in coating processes where it causes the entrainment of air into the coating layer as soon as the coating speed becomes too large. In non-dimensional form the critical velocity of the advancing contact line is primarily given by a capillary number, but other factors play a role as well. Phenomenologically, the spreading of the lamella during the initial phase of droplet impact is a forced wetting process. There may thus be a connection between the onset of splashing and air entrainment at the rim of the expanding lamella.

In the case of corona splashes the rim of the spreading lamella first separates from the solid surface and forms a free liquid sheet. Only then secondary droplets are shed from the rim [15]. In some respects this phenomenon, which was already reported to occur before corona splashes were even defined (cf. [14]), is similar to incipient air entrainment in coating processes. In the case of a spreading lamella that is formed after droplet impact, the film thickness and its radial extension are very small. Therefore air entrainment can easily lead to a detachment of the entire liquid film. In the following we will consider possible connections between air entrainment and corona splashes on smooth surfaces. Prompt splashes that are especially characteristic of drop impacts on rough surfaces will be addressed later.

3.1 Dynamic wetting

Air entrainment in coating flows is normally traced to the dynamics of the contact angle between the spreading liquid and the solid surface. Following Kistler [8] we will briefly review aspects of dynamic contact angles that may be of relevance to the inception of splashing. The dynamic contact angle Θ_d increases with the velocity of the advancing contact line. It is observed to be very close to $\Theta_d = 180^\circ$ when the critical velocity of air entrainment is reached. The velocity at which $\Theta_d = 180^\circ$ is reached has actually been conjectured to be the critical velocity of the onset of air entrainment. From experiments it is known that the dependence of the dynamic contact angle on the velocity of the contact line, u_{cl} , can well be described in a universal non-dimensional manner using a capillary number Ca_{cl} formed with u_{cl} and the viscosity and surface tension of the liquid, $Ca_{cl} = \mu u_{cl}/\sigma$, the specific form depending on whether the liquid wets the surface completely or partially. In the case of complete wetting the relation, usually connected with the name of Hoffman, is simply given as

$$\Theta_d = f_H(Ca_{cl}). \tag{5a}$$

In the case of partial wetting the static contact angle Θ_0 also enters the relation:

$$\Theta_d = f_H [Ca_{cl} + f_H^{-1}(\Theta_0)].$$
^(5b)

Many correlations between Θ_d and Ca_{cl} have been introduced to describe the relation between the dynamic contact angle and the velocity of the contact line. Another factor influencing this relation is $\lambda = \mu_g/\mu$, the ratio of the viscosity of the receding fluid, μ_g , (the receding fluid is a gas in the present consideration of splashing drops) to the viscosity of the advancing fluid. According to Kistler [8], the following correlations, which contain the dependence on the static contact angle in a normalized form only, work well for low and high viscosity liquids, respectively, when the capillary number is not too small ($Ca_{cl} > 0.01$):

$$\frac{\cos \theta_0 - \cos \theta_d}{\cos \theta_0 + 1} = \tanh(4.96 \, Ca_{cl}^{0.702}) \quad \text{(high-viscosity liquids)}, \tag{6a}$$

$$\frac{\cos \theta_0 - \cos \theta_d}{\cos \theta_0 + 1} = 2\sqrt{Ca_{cl}} \quad \text{(low-viscosity liquids)}.$$
(6b)



Fig. 2 Dependence of the dynamic contact angle (θ_d , θ_0 is the static contact angle) on the capillary number (Ca_{cl}) for low- and high-viscosity liquids wetting a solid surface (after [8])

The two relations are shown in Fig. 2. As can be seen, there exists a qualitative difference at greater values of Ca_{cl} . While the dynamic contact angle of low-viscosity liquids reaches the limiting value of $\Theta_d = 180^{\circ}$ at a well-defined value of $Ca_{cl}(=0.25)$, this limit is approached asymptotically for high-viscosity liquids. This qualitative difference in behavior corresponds with experimental observations. The ratio of the viscosities of the displaced and advancing fluids provides a criterion for distinguishing between high and low viscosity liquids, with low-viscosity behavior being observed for $\lambda = \mu_g/\mu > 10^{-5}$ and vice versa. According to Kistler [8] the "viscosity ratio is a key parameter in dynamic wetting, second in importance only to Ca_{cl} ". In all drop impact studies considered here $\lambda > 10^{-5}$, i.e., the behavior of the dynamic contact angle is described by Eq. (6b) for low-viscosity liquids.

Other aspects of dynamic wetting are potentially relevant to the outcome of drop impacts. If a surface has been previously wetted, dynamic contact angles on that surface are lower than on completely dry surfaces, "even when the excess liquid is removed with a scraper blade" [8]. In certain coating configurations a local decrease of the gas pressure behind coating beads (liquid bridges present in certain coating methods) has been observed to lower dynamic contact angles. Air entrainment has also been delayed by what has been termed hydrodynamic assist of coating [2]. For example, in the method of curtain coating a liquid sheet (the "curtain") impinges onto a moving substrate with a finite velocity. The critical speed of air entrainment, i.e., the highest substrate velocity without an onset of air entrainment, increases with increasing impact velocity (momentum) of the curtain. It has been conjectured that the increase in the stagnation pressure is responsible for the entrainment delay. Finally, experiments have shown that in coating flows air entrainment can also be delayed during spreading on rough surfaces. This has been tentatively traced back to the roughness of the surface that provides channels for the air to escape (cf. [8]) or to a sliding of the liquid over the crests of roughness elements [4].

3.2 Separation of the spreading lamella from the surface

In order to characterize under what conditions the air entrainment mechanism described above can induce lift off of the spreading lamella after droplet impact, we now focus on cases in which corona splashes have been observed. Xu [30] obtained nice corona splashes on smooth surfaces while prompt splashes occurred when drops impinged on rough surfaces. In Vander Wal et al. [22,23] the notion "prompt" splash is used to describe splash inception from the rim of the crown during the spreading phase as compared to a delayed splash where secondary droplets are shed from a receding crown [23]. Hence, here a "prompt" splash may actually be a "corona" splash according to the definition provided in the introduction. The approximation of the corresponding splashing limit by $Ca_{crit} = 0.1225$ [22] is now compared with a critical capillary number of air entrainment, $Ca_{cl,AE}$, at the moving contact line. In order to do so a relation between the impact velocity and the initial velocity of the spreading lamella is needed. Typical values reported for the latter velocity are between twice and ten times the impact velocity, the latter value being more realistic during the very initial phase. The ratio of the two capillary numbers is then $Ca_{cl}/Ca = O(10)$. Considering the splash threshold of $Ca_{crit} = 0.1225$ [22] yields a corresponding capillary number $Ca_{cl} = O(1)$ of the spreading lamella that is clearly greater than the critical capillary number of air entrainment, $Ca_{cl,AE} \approx O(0.1)$ of air–water systems with $\lambda = O(10^{-2})$ and still of the same order of magnitude as the critical capillary number $Ca_{cl,AE} \approx O(1)$ for higher viscosity liquids displacing air ($\lambda = O(10^{-5})$, cf. [8]). Hence, on smooth surfaces, the lift off of the lamella into a corona or crown-like form can well be caused by the same dynamics at the contact edge that leads to air entrainment in forced wetting. After the lamella has separated from the surface, a shedding of secondary droplets may occur in a way that is consistent with the mechanisms proposed in the literature [32]. For example, surface tension forces can lead to a thickening of the rim finally causing a break up by a mechanism analogous to the Rayleigh instability of a jet.

The order of magnitude analysis above shows that there is significant overlap between the criterion for air entrainment and that for splashing, thus supporting the hypothesis of air entrainment at the lamella's edge as a mechanism that induces splashing. However, the criteria were not found to match. For example, if $Ca_{cl} = 1.1Ca_{cl,AE}$ then $Ca = 0.1Ca_{cl} = O(0.01) < Ca_{crit}$ (with $Ca_{cl,AE} \approx O(0.1)$ as for $\lambda = O(10^{-2})$). This means that there is a range of impact conditions for which entrainment occurs but does not result in observable splashing. Several factors can explain this apparent contradiction. First, in contrast with forced wetting, the spreading lamellae in drop impact dynamics represent a highly unsteady flow—the rim of the lamella is quickly decelerating and its velocity quickly falls to values well below critical conditions of air entrainment. Second, while the lamella starts spreading out radially, the bulk of the drop is still approaching the surface. Initially, right after the first contact between the drop and the surface has been made, the gap between the approaching drop and the surface is very small. The thin, jetting and possibly uplifting lamella may therefore easily reconnect to the bulk liquid of the drop. This is similar to the reconnection of thin liquid sheets ejected after drop impact on liquid films [25]. Finally, high impact pressures initially prevailing in the impact region may also delay the onset of splashing by the effect of hydrodynamic assist.

3.3 Discussion

As has been shown in the last section, incipient air entrainment at the rim of the spreading lamella can well explain the onset of splashing in drop impacts. In dynamic wetting the critical velocity of air entrainment is known to be inversely proportional to the viscosity which is expressed in non-dimensional form by a critical capillary number of air entrainment, $Ca_{cl,AE}$. Although not all correlations of the splashing threshold listed in Table 1 display this dependency the general trend of all curves shown in Fig. 1 is the same. Deviations from this dependence on the viscosity may therefore be due to a certain insensitiveness of the curves to changes in the exponent β in Eq. (3).

A close inspection of Fig. 1 reveals that in the high Reynolds number range (for $Re \gtrsim 5,000$), all splash correlations obtained with impacts on smooth surfaces agree quite well with the concept of a critical capillary number for splashing, $Ca_{crit} = 0.1225$. However, thresholds determined from splashes on rough surfaces are characteristically lower. At smaller Reynolds numbers the only available experimental results for splashing on smooth surfaces are those published by Vander Wal et al. [22]. Again, the corresponding threshold for splashing is well described by a constant capillary number. In this case, however, curves relating to thresholds of splashing on rough surfaces (expressed in terms of a critical Ohnesorge number) lie above that corresponding to $Ca_{crit} = 0.1225$. This may be caused by greater (absolute) values of the slopes of the former curves, a point that will be considered next.

In Fig. 1 where the critical Ohnesorge number for splashing is plotted as a function of the Reynolds number, the splashing threshold corresponding to a constant critical capillary number ($Ca_{crit} = 0.1225$, i.e., $Oh_{crit} = 0.35Re^{-1/2}$) results in a curve whose slope differs slightly but consistently from the slopes of all other experimental curves. This may be due to the Reynolds number being formed with the impact velocity of the drop, v, but not with the initial spreading velocity of the lamella, u_{cl} , which would be more appropriate in the context of dynamic wetting. In the last section the latter velocity was approximated by $u_{cl} = 10v$, i.e., by a linear relation. However, spreading is driven by the stagnation pressure formed in the impact region after impact, which is proportional to v^2 . Accounting for this quadratic dependency would change the slope of the splashing threshold bringing it closer to the trend exhibited by the experimental curves.

Following the discussions above, it can be seen that splash thresholds in Fig. 1 can be grouped according to the finish of the surface on which the impact takes place: rough or smooth. This is particularly clear when comparing the thresholds of Stow and Hadfield [20] and Range and Feuillebois [13] obtained for rough and smooth surfaces, respectively. It is well-known that increasing the surface roughness enhances prompt splashing and decreases the likelihood of corona splashing (cf., for example, [15]). This no longer fits into the connection

between splash inception and air entrainment put forward above. In forced wetting roughness delays the onset of air entrainment. This effect has been connected to the channels present on the rough surface through which entrained air can escape or to a sliding of the liquid on top of the roughness elements. These mechanisms work only if the height of the coating layer is much greater than the roughness as it is usually the case in coating experiments. In the case of drop impacts, however, the thickness of the spreading lamella is initially very thin and comparable to the height of the elements of roughness. In the case of splashing on smooth surfaces the thickness h_c of the separating corona is $h_c/d = O(10^{-2})$ [31] and the height h_l of the spreading lamella is still only $h_l/d = O(0.1)$ at the late time tv/d = 1 [17]. Already Stow and Hadfield [20] had noted the connection between flow disturbances resulting from the interaction between the lamella and the elements of roughness and the inception and enhancement of splashing. This is corroborated by the fact that artificial geometrical structures on the surface have been found to have a profound influence on the outcome of splashing in different azimuthal directions [30].

According to the above discussion, it is likely that the thresholds shown in Fig. 1 that are associated with drop impact on a smooth surfaces correspond to corona splashes while those that are associated with drop impact on a rough surfaces correspond to prompt splashes. Information on the particular type of splashing event is not always reported however. A similar conjecture has also been put forward by Xu et al. [31] who decreased the ambient pressure to very low values. Under low pressures splashing was only observed on rough surfaces.

In plunging-tape experiments critical velocities of air entrainment are observed to depend only weakly on the wettability of the surface [8]. Rioboo et al. [16] studied the evolution in time of the lamella formed after drop impact for various combinations of liquids and surface materials (resulting in different static contact angles). They found that during the initial phase of spreading the dynamics of the lamella is also weakly influenced by the wettability of the liquid–solid system.

A correlation of the splashing threshold that has perhaps been more often cited than any other correlation is that proposed by Mundo et al. [12]. That expression clearly deviates from all other curves displayed in Fig. 1. One reason may be the relatively large values of roughness that were present in these experiments. According to the results of Stow and Hadfield [20], both values correspond to impacts on a very rough surface. The non-dimensional surface roughness (R_a/d) in the experiments of Mundo et al. [12] is larger than the largest non-dimensional surface roughness in any other experiment discussed in this paper. Another distinctive feature of the experimental procedure of Mundo et al. [12] is that drops impinged onto a rotating disk. This not only renders the impact effectively oblique—which should not make a difference as long as the normal velocity component is considered (cf. Sect. 2.1)—but also results in the formation of a boundary layer above the disk that needs to be traversed by impinging drops. This interaction causes a different impact behavior (cf. [14]) which may contribute to the comparatively low splashing threshold.

It is interesting to note that the well-known splashing threshold of Yarin and Weiss [33] obtained for droplet streams impinging on liquid layers can also be expressed in the form of Eq. (3) if the frequency f of the droplet stream is replaced by f = v/d. The exponent β is the same as in Mundo et al. [12], $\beta = -5/4$, while the coefficient α is about five times larger, $\alpha \approx 300$, i.e., splashing is delayed. Yarin and Weiss [33] supported their experimental findings by theoretical considerations that are based on the existence of a liquid layer prior to impact.

Finally, let us consider those cases where the degree of wetness of the surface is not clearly stated [6]. The good agreement between the threshold of Hardalupas et al. [7] (who used a prewetted surface) and Cossali et al. [6] suggests that the vanishing film thickness limit of Cossali et al. [6] was not achieved by a completely dry surface. It is interesting to note that these limits of splashing on wetted surfaces or thin films agree quite well with those of splash limits for rough surfaces.

4 Fingering at the rim of the spreading lamella

Finally let us address another point that has received much attention in drop impact studies, the so-called fingering at the advancing rim of the lamella. The "fingers" are not necessarily very pronounced and have therefore also been called lobes or undulations. They appear right after impact when the lamella is just starting to form [11,21]. A similar morphological phenomenon is found to occur in coating processes when the velocity of the contact line is close to or even slightly above its critical value of air entrainment; the originally straight contact line then exhibits a sawtooth like shape. This phenomenon has been studied in plunging-tape experiments. There, as shown in Fig. 3a, a tape is drawn with a velocity u_{tape} normally into a pool of liquid. In the



Fig. 3 a Formation of sawteeth at the wetting line in a plunging tape experiment when the velocity u_{tape} of the tape exceeds the critical velocity of air entrainment, $u_{cl,AE}$. b Undulations at the advancing contact line of a spreading lamella formed after drop impact ($u_{front} > u_{cl,AE}$)

reference frame of the tape the main front of the liquid wets the tape with a velocity $u_{\text{front}} = -u_{\text{tape}}$. Sawtooth patterns are formed when the front velocity exceeds the critical velocity of air entrainment. Blake and Ruschak [3] proposed that the angle Φ of the sawteeth are such that the velocity component normal to the contact line does not exceed the critical velocity of air entrainment, $u_{cl,AE}$. Then, $\cos \Phi = u_{cl}/u_{\text{front}} \le u_{cl,AE}/u_{\text{front}}$. This has more recently been corroborated experimentally [5].

In drop impacts, the velocity of the advancing contact line of the spreading lamella formed after impact can easily exceed the critical velocity of air entrainment. Then, as in the case of plunging tape experiments, undulations should form at the rim of the lamella before it lifts off the surface. Because the contact line quickly decelerates, deep sawteeth cannot develop and, therefore, in order to keep the normal velocity of the contact line below the critical velocity many small serrations need to be formed right away along the whole circumference. This is sketched in Fig. 3b where the radial velocity of the rim of the spreading lamella, $u_{\rm front}$, has been assumed to be greater than the critical velocity for the onset of air entrainment while the velocity normal to the contact line, u_{cl} , remains smaller than the critical value. For the normal velocity of the contact line not to exceed the critical velocity $u_{cl,AE}$, the sawteeth need to become more acute with increasing spreading velocity, i.e., with increasing impact velocity. Thus also the number of the undulations grows with the impact velocity. However, when the radial velocity of the spreading lamella remains below the critical velocity of air entrainment, i.e., when the impact velocity is small, the contact line should remain smooth forming no undulations. This behavior, the existence of a critical velocity for the onset of undulations and an increase of their number with the impact velocity, actually corresponds with experimental results obtained with smooth surfaces where mechanisms similar to those seen in forced wetting should occur [11,13]. Similarly, Xu [30] who suppressed splashing by decreasing the pressure in the surrounding gas observed that fingers are only formed if the gas pressure exceeds a critical value. Hence, the mechanism leading to air entrainment at advancing contact lines also provides a possible explanation for fingering after drop impact.

5 Conclusions

Although drops splashing after impact on solid surfaces have been studied for over a century, there is still no consensus on the mechanisms causing the well-known fingering at the rim of the spreading lamella and the onset of splashing. In order to shed some light on the conflicting findings of the various published studies that have addressed the issue of splashing after droplet impact, we have critically reviewed and synthesized the results of these studies. This was done in terms of the definition of a splashing threshold. It was found that there exists a distinct difference between splashing on smooth and on rough surfaces.

For impacts on smooth surfaces it has been shown that within an acceptable margin an approximation of the splashing threshold first introduced by Vander Wal et al. [22] also agrees with data of other authors. The approximation of Vander Wal et al. [22] predicts that splashing will occur if a capillary number formed with the impact velocity of the drop and its surface tension and viscosity exceeds a critical value. In this context it has also been shown that the splashing threshold proposed by Xu et al. [29] (who found splashing to depend on the pressure in the surrounding gas) can be expressed in terms of a critical capillary number. This critical capillary number is consistent with other experimental splashing thresholds for smooth surfaces. Based on this finding we have shown that splashing on smooth surfaces can be connected to the same mechanism that causes air entrainment at advancing contact lines in dynamic wetting.

For impacts on rough surfaces the splashing threshold is lower. In this case it is generally accepted that disturbances to the spreading lamella caused by roughness elements initiate splashing. Thus, there exist two different mechanisms of splashing for drops impinging on dry surfaces and, therefore, a distinction must be made between splashes on very smooth surfaces and those on rough surfaces. For millimeter-sized drops typically used in experiments, even a relatively small roughness of $R_a \approx 0.5 \mu m$, i.e., $R_a/d = O(10^{-4})$, is enough to cause transition from smooth to rough surface.

Finally, we argue that the fingering at the rim of spreading lamellae that has typically been observed during impacts on smooth surfaces, is akin to an effect characteristic of dynamic wetting, namely, the formation of serrations at contact lines moving at velocities above the critical velocity for air entrainment.

Many questions are still open in dynamic wetting. An interesting theoretical approach by Shikhmurzaev [19] for describing the physics at dynamic contact lines is controversial [10]. Therefore, it might be premature to try to describe quantitatively critical conditions for splashing on smooth surfaces on the basis of such a theory by comparing the inception of corona splashes to the onset of air entrainment in coating flows. However, the connection between these two phenomena can serve as a guide for designing novel experimental approaches such as that implemented by Xu et al. [29] that may also be useful for the investigation of dynamic wetting phenomena. In that endeavor, it is particularly important to consider the effect of the ambient gas.

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Appendix: Derivation of a splash threshold from the analysis of Xu et al. [29]

In their analysis Xu et al. [29] consider two stresses acting on the expanding lamella, one (Σ_G) being due to the restraining pressure of the gas, the other (Σ_L) to surface tension. These stresses are approximated by

$$\Sigma_G = \rho_g c_g (\mathrm{d}v/(4t))^{1/2} \sigma^{-1}, \quad \Sigma_L = \sigma (\mu/\rho t)^{-1/2}, \tag{7}$$

where t denotes the time after the instant of impact, and ρ_g and c_g are the density and sound speed of the surrounding gas. Splashing is expected to occur when these stresses are comparable. Experimentally, splashing is observed for

$$\Sigma_G / \Sigma_L = \rho_g c_g (\mathrm{d} v \mu / \rho)^{1/2} \sigma^{-1} / 2 > N \approx 0.45, \tag{8}$$

which can be rewritten as

$$Oh > (2Nv\rho/(\rho_g c_g))^{1/2} R e^{-3/4}.$$
(9)

We now provide an estimate of the factor $(2Nv\rho/(\rho_g c_g))^{1/2}$. The product of the density and sound speed of the gas is given by

$$\rho_g c_g = \left(pM_g/(k_B T)\right) \left(\gamma_g k_b T/M_g\right)^{1/2} = p\left(M_g/M_{\rm air}\right)^{1/2} \left(\gamma_g M_{\rm air}/(k_B T)\right)^{1/2},\tag{10}$$

where M_g and γ_g are the molecular weight and the ratio of specific heats of the gas, k_B is the Boltzmann constant, and p and T are the pressure and temperature in the gas. M_{air} is the molecular weight of air. Xu et al. [29] observed that with different gases, splashing always occurred at $p(M_g/M_{air})^{1/2} \approx 30 - 40$ kPa, the exact value depends slightly on the impact velocity. The ratio k_B/M_{air} is the specific gas constant R_{air} of air with $R_{air} \approx 287$ J/kgK for dry air. In providing an estimate for the factor $(\rho_g c_g/(v\rho))^{-1/2}$ we will assume that the experiments were performed at a temperature T = 292 K and use a value of $\gamma_g = 7/5$ for all gases. Because only the square root of γ_g is relevant this latter approximation results in an error of less than 5% if either a noble gas ($\gamma_g = 5/3$) or a gas with many degrees of freedom ($\gamma_g \approx 1.2$) is considered. The density of the liquids used by Xu et al. [29] is consistently about 800 kg/m³. For an impact velocity of $v \approx 3$ m/s Xu et al. [29] obtained p (M_g/M_{air})^{1/2} ≈ 40 kPa. Using these values the factor preceding the Reynolds number in the Ohnesorge versus Reynolds number relation of Xu et al. [29] is approximately

$$(2Nv\rho/(\rho_g c_g))^{1/2} \approx 3.6.$$
 (11)

This value was used in graphing the threshold of Xu et al. [29] in Fig. 1.

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