

An experimental study of droplet-particle collisions

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AN EXPERIMENTAL STUDY OF DROPLET-PARTICLE COLLISIONS

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ABSTRACT

When spray drying a liquid slurry such as milk, collisions between droplets, partially dried particles and completely dry particles are important because coalescence, agglomeration and breakup events influence the size and morphology of the produced powder. When modelling such a spray drying process, it is therefore important to be able to predict the outcomes of individual binary collisions. Both binary dry particle collisions and binary droplet collisions have individually been thoroughly researched over the years due to their widespread occurrence. The importance of understanding binary particle-droplet collisions has been emphasized more recently. However, the number of available studies is limited and simulation studies usually focus on relatively high capillary number. A theory explaining the transition between different regimes is still lacking. The goal of this study is to provide an experimental data set at low capillary number. These results can be used to validate future theories and simulations. To produce and record particle-droplet collisions, an experimental setup that enables synchronized release of both a particle and a droplet was used. One single hanging droplet was released from above onto a particle that initially was held in place by vacuum suction. A high speed camera was synchronized with the setup, and recorded the collisions. Image files were then analysed in Matlab to find velocities and sizes of the particle and droplet before and after impact. The contrast of particle and droplet against the illuminated background was a key factor in succeeding with this. Different collision outcomes were identified as either agglomeration (merging), where the whole droplet would stick to the surface of the particle, or a stretching separation (breaking), where the droplet collides with the particle in an oblique position and stretches out until a part of the droplet detaches from the liquid sticking to the particle. The formation of satellite droplets, i.e. droplets with a radius significantly smaller than the leaving droplet, was also detected. The relation of these collision outcomes to impact conditions such as Weber number and impact parameter was reviewed and put into regime maps.

KEYWORDS

Agglomeration, Break-up, Spray Drying, Particle Tracking Velocimetry.

1. INTRODUCTION

Spray drying is an essential unit operation for making powder from liquid slurry. It is widely used in different industries such as the chemical industry, the pharmaceuticals industry, the food industry etc. Generally, a spray dryer comes at the end of the processing line, as it is an important step to control the final product quality. It has some advantages, such as rapid drying rates, a wide range of operating temperatures and short residence times. The morphology of the powder can be

controlled to some extent, creating possibilities in several fields where powder production is central. Furthermore, composite particles with a microencapsulated core can be formed for controlled release of an active substance [1].

Spray drying is used frequently in the food industry for producing powders in the form of soup, instant coffee, and milk powder. The desired characteristics of these powders are different but controllable to a certain extent. The most important characteristics for milk powder are good flowability, water solubility, and a limited dustiness, i.e. a low amount of small particles in the final powder. What largely affects all of these characteristics is the degree of agglomeration in the final particles. This is the result of collisions between viscous droplets or primary particles formed from droplets, as well as collisions between viscous droplets and recycled fines, i.e. small dry particles [2].

Many investigations have been made of binary droplet-droplet interactions, see [2] and the references therein. The outcome of such collisions can conveniently be characterized using the Weber number We , the impact parameter b (Figure 1), and the size ratio Δ [2]. These parameters are calculated as:

$$We = \frac{\rho_a d v_{rel}^2}{\sigma} \quad Eq (1)$$

$$b = \frac{2B}{d_1 + d_2} \quad Eq (2)$$

where We is based on the smallest droplet diameter d . An example of how these two parameters determine the outcome of a droplet-droplet collision is shown in the regime map in Figure 2. The regime boundaries will however change for different small to large droplet size ratios Δ .

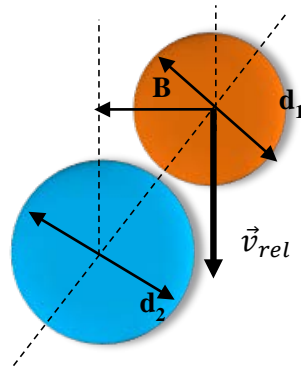


Figure 1: Geometric and kinetic parameters used to describe the impact parameter, b .

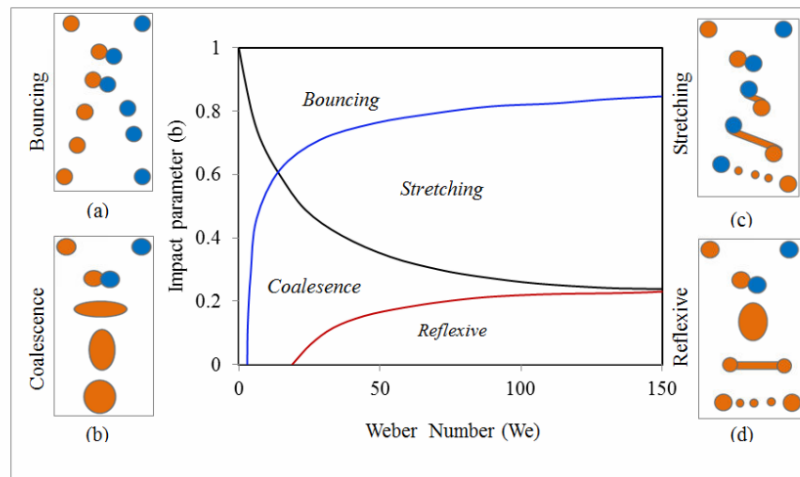


Figure 2: Schematic picture of the different outcomes for droplet-droplet collisions (here for equal droplet size) [3].

The amount of studies investigating binary particle-droplet collisions are very limited. Dubrovsky et al. [4] investigated droplet-particle collisions at relative velocities of 3.4-12.8 m/s, where the particle was smaller than the droplet [4]. A notable difference with droplet-droplet collisions is that no reflexive separation was observed at any velocity. Instead four different outcomes were observed for collisions with low impact parameter. These were particle capture, “shooting through” with satellite droplet formation, gas bubble formation after the particle shot through the droplet, and target destruction where the droplet is turned into fragments. No literature has been found on mid-air collisions where the particle is bigger than the droplet. However, some literature data exists for a fixed particle, where either agglomeration or droplet fragmentation against the surface of the particle is observed for head-on or near head-on collisions [4,5]. Dubrovsky et al. [4] did these experiments with a droplet Reynolds number ranging between 25 and 2500. It was found that coalescence increased with increased viscosity of the droplets and increased size ratio between the particle and the droplet. Shen [5] concluded that the amount of water attached to the particle decreased with increased velocity of the droplet. The experiment was made for two different sizes of droplets and was executed for a number of velocities. Furthermore, the impact parameter was found to have a bigger influence on the mass transfer, compared to the tested Weber numbers. Another difference from binary droplet collisions is that recoiling or bouncing is not as likely to occur. This requires certain conditions which are not met in this study. Specifically, it requires a high contact angle, for instance caused by a hydrophobic surface or a particle sufficiently heated, making the evaporation of the droplet take place in the Leidenfrost regime, causing a thin vapour film to prevent wetting of the particle surface [6,7].

The statistical distribution of water attachment and momentum transfer by particle-droplet collisions is examined in [8]. This work was not focusing on binary collision hydrodynamics, but more on the collision probability and mass transfer statistics. The study was executed using a set of free-falling particles, colliding with a horizontal spray of water and having several collecting bins in the direction of the falling droplets and hit particles. The model for liquid attachment does however neglect size and velocity distributions of the droplets and also the influence of the turbulence in the spray jet.

Mitra et al. [9] focused on the collision hydrodynamics of a small glass particle impacting into a larger stationary droplet. The experimental work was compared with a numerical investigation. The resulting particle sinking times, correlated to the transition from partial to complete penetration, were in good agreement between experiment and simulation. It was found that the effect of

capillary and pressure forces were dominant. The analysis was however limited to low We numbers in a range of 0.2-13.5.

The opposite case of small droplets impacting with large particles has also been investigated [10,11]. Hardalupas et al. [10] performed experimental work on liquid drops (160-230 μm diameter) colliding on the surface of a small solid sphere (0.8-1.3 mm diameter). They accurately analysed shape of the impacting droplet observing a retraction of the liquid crown at low droplet velocity and disintegration starting from the rim of the cups for high velocity. Bakshi et al. [11] performed an extensive experimental and theoretical investigation with particular attention to spatial and temporal evolution of film thickness on the target surface. Both these works had as main interest the understanding of the coating of particles. Moreover in both cases the spherical target particle was static, instead of freely moving.

A prime example of numerical investigation on droplet particle collision is given by Gac and Grado [12], who studied the impact of a droplet on differently shaped solid particles. Using the lattice-Boltzmann method (LBM) three collision regimes were identified: (1) coalescence, without droplet fragmentation, (2) ripping and coating, where one part of the droplet deposits and coats the particle and the other part detaches and continues the motion, and (3) skirt scattering, with the formation of a long conical surface "skirt" which breaks into small droplets. We note that this study was focusing on relatively large capillary numbers of order 1, whereas the capillary numbers studied in this work are about two orders of magnitude lower.

We conclude that studies that thoroughly evaluate how mid-air particle-droplet collision outcomes depend on the impact parameter, size ratio and the characteristic Weber number are limited. The main objective of this study is therefore to provide an experimental data set for the outcome of mid-air particle-droplet collisions at relatively low Capillary numbers, which can be used to validate future theoretical and simulation developments. The different collision outcomes include agglomeration and stretching separation, the latter both with and without the formation of satellite droplets. These outcomes are placed in regime maps based on parameters such as the impact parameter and the Weber number. The obtained experimental results will be based on the analysis of images from a high speed camera. We used water as the liquid phase and glass particles as the solid phase. Note that this means that we have limited ourselves to fully dry particles and low viscosity liquid droplets, leaving aside for the moment the complicating factors of partially wet and highly viscous particles which may agglomerate due to surface tack or stickiness [2,13].

2. EXPERIMENTAL SETUP AND PROCEDURE

Figure 3 shows a schematic picture of the setup. We used water as the liquid phase and glass particles as the solid phase, with an equilibrium contact angle between 70 and 80 degrees. The observation area is a piece of white paper, illuminated by the LED light. The main parts are a droplet syringe, a vacuum pump holding the particle and a high speed camera. The syringe connected to the pump can be moved horizontally and vertically to change the impact parameter and the velocity. The falling distance can be varied between 0.01 and 1 m. The size of the needle can be varied to create droplets of different size. We use a computer to synchronize the start of pump with the release of the particle, as well as the trigger for the camera to start recording. The accuracy for these signals is 1 millisecond. The temperature and humidity are also monitored. A HighSpeedStar camera (LaVision HS3G) is used to record the droplet-particle collision process with a 105 mm Sigma DG macro lens at f/2.8 aperture and an image frequency of 3200 - 4000 fps. Details of physical dimensions and properties are given in Table 1.

The contrast of the particle and the droplet is firstly increased by keeping them in the shadow and having an illuminated background. Secondly, further contrast is achieved by having black screens around and above the drop.

Once the needle was adjusted into position, the droplet's position in the depthwise direction (to and from the camera) seemed to be stable. There was no exact way of guaranteeing that the particle and droplet collided at exactly the same depthwise position. However, the very small depth of field of the fully open macro lens (0.8 millimeter) leads to a slightly blurred image of the droplet when it is slightly behind or in front of the plane of the (in-focus) particle. This allowed us to visually assess when the depthwise impact parameter was a noticeable fraction of the droplet diameter. If that was the case, the images of that experiment were discarded.

The size of the visual reference frame was sufficient to observe the complete collision outcome. In fact a first observation with a zoom out has been performed for every collision outcome in order to visualize the outcome and to determine the observation area size. The minimum detectable droplet size was about 1 pixel, corresponding to a real size of 53 micron at the used magnification of 0.38. The individuation of satellite droplets was done manually and their actual size was not calculated. The investigation was addressed to the distinction of collision outcomes and to provide a large number of results for different We numbers and impact parameters.

In order to get the desired information about the collisions, the recorded image files were analysed using Matlab R2013b. The parameters affecting the impact collision (i.e. Re , We , Δ and b) were identified, together with the velocity of the droplet and the particle the moment before and after collision. The fraction of the droplet that agglomerates to the particle is also calculated. With the purpose of getting this information, the analysis of the image files was divided into two parts, namely before and after collision.

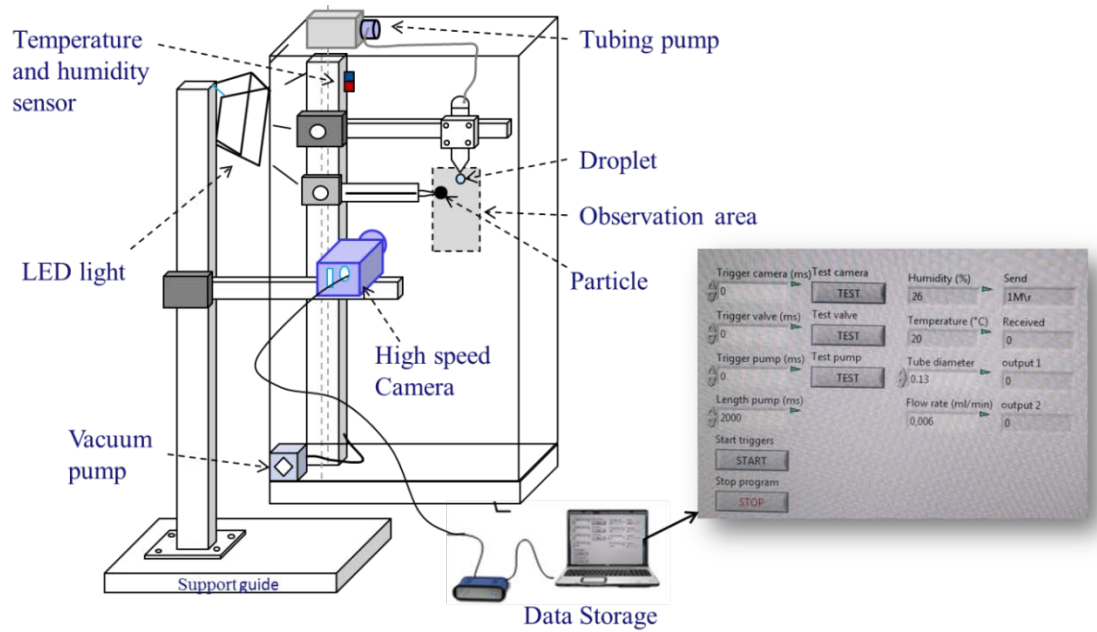


Figure 3: Schematic picture of the experimental setup. On the right hand side is a picture of the program used to synchronize the pump with the release of the particle and the camera.

Table 1: Values of parameters that are tested in this study. The column 'real' applies to typical conditions for freshly injected (undried) milk droplets in a spray dryer producing milk powder colliding with already dried milk particles. The column 'experiment' applies to our experiments using water for the liquid medium.

Parameter	Formula	Real (Milk)	Experiment (Water)	Unit
Relative velocity	v_{rel}	1 - 100	0.065-1.15	m/s
Density of droplet	ρ_d	1196	998	kg/m ³
Surface tension	σ	0.053	0.073	N/m
Diameter of droplet	d_d	40	2939 ± 125	μm
Diameter of glass particle	d_p	-	4000 or 2500	μm
Diameter of dried milk particle	d_p	20-100	-	μm
Viscosity of droplet	μ	0.002	0.001	Pa s
Density of glass particle	ρ_p	-	2526	kg/m ³
Density of dried milk particle	ρ_p	1100	-	kg/m ³

Weber number	$\frac{\rho_d d_d v_{rel}^2}{\sigma}$	0.55 - 5094	0.34 – 52	-
Reynolds number	$\frac{\rho_d d_d v_{rel}}{\mu}$	13 - 6750	258 – 3082	-
Capillary number	$\frac{\mu d_d v_{rel}}{\sigma d_p}$	$1.5 \times 10^{-2} - 7.5$	$6.7 \times 10^{-4} - 1.9 \times 10^{-2}$	-
Impact parameter	$b = \frac{2B}{d_d + d_p}$	0 - 1	0 - 1	-

Even though the maximum measured relative velocity was much lower than those in spray dryers, the bigger size of the droplets made the lower Weber number range similar. Obtaining Weber numbers in the higher part of the real range was not possible with this setup, but fortunately also not necessary because we observed that the interesting regime transitions took place within the investigated range, as we will show in the next section. Note that the relative velocities in our experiments may be comparable with conditions in a spray dryer at the edge of the spray. At the same time in a spray dryer the droplet viscosity varies from comparable to our experiments to much larger, depending on the moisture content of the milk droplet. As a consequence, our experiments are in the limit of low capillary number which corresponds to freshly injected (undried) milk droplets, where the viscosity is still close to that of water, colliding with solid particles at low relative collision velocities. This is representative of freshly injected milk droplets colliding with already dried milk particles near the edge of a spray jet, not too far from the droplet atomizer.

The number of images taken before collision normally ranges between 10 to 50 pictures depending on droplet and particle velocity. Finding the velocity and size of the particle and droplet is done by identifying them in the images, and by tracking their centre positions (particle tracking velocimetry). For the spherical glass particles, a script that identified circular shapes in each individual image was advantageously used for tracking both particle and droplet before impact, see Figure 4. This is done using `imfindcircles`, a function in Matlab's Image Processing Toolbox that returns circle centre positions and radii.

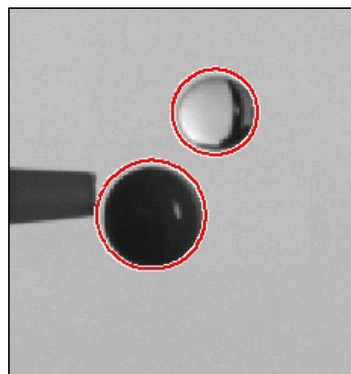


Figure 4: Detection of the glass particle and the water droplet using `imfindcircles` in Matlab before collision.

3. RESULTS & DISCUSSION

Regime map of single droplet-particle collision

The starting point for these experiments was collisions between droplets and 4 mm diameter glass particles, because collisions with a bigger target were more easily obtained. The average size ratio was $\Delta = 1.37$, where the ratio is defined as

$$\Delta = \frac{d_p}{d_d} \tag{Eq (3)}$$

A total of 214 collisions were recorded and analysed. The collision outcomes were identified as agglomeration (A) or stretching separation (SS). If satellite droplets were formed, the number of satellite droplets (SD) was counted. Examples of collision outcomes are shown in Fig. 5 (a), (b), (c).

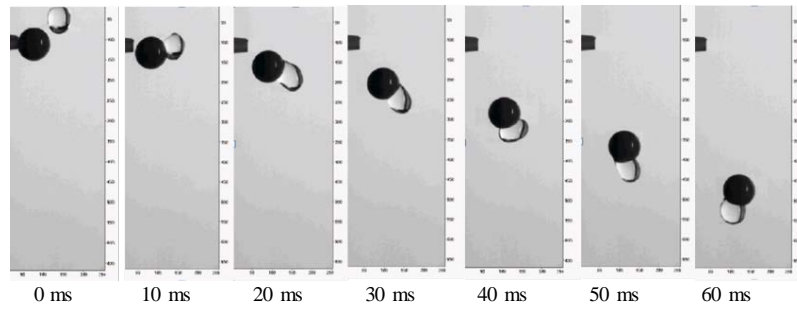


Figure 5(a): example of agglomeration (A)

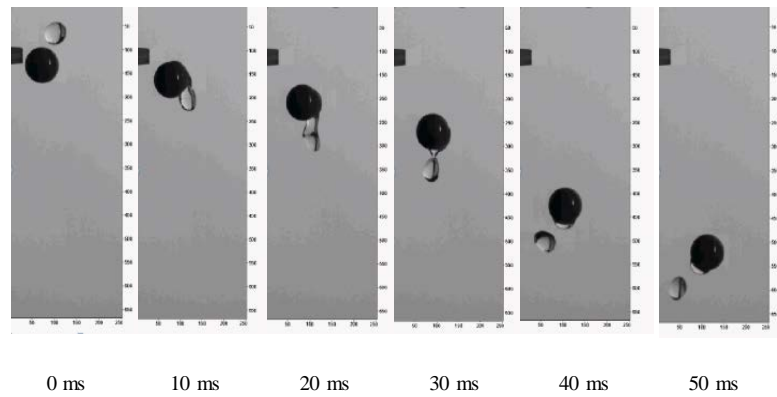


Figure 5(b): example of stretching separation (SS)

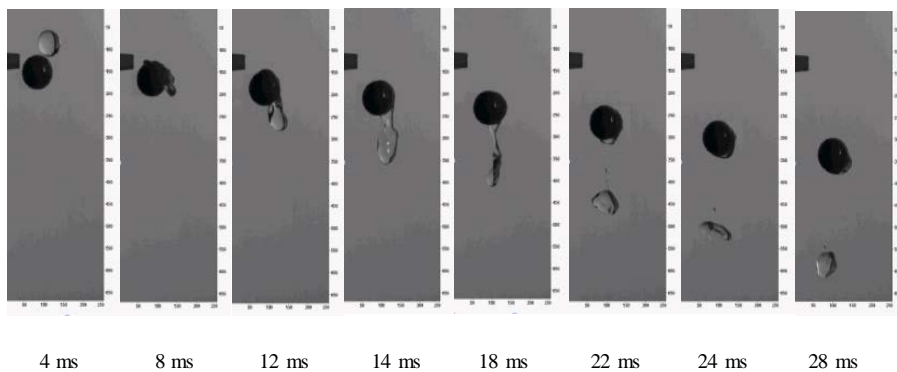


Figure 5(c): example of stretching separation with satellites droplets (SD)

A regime map based on the impact parameter and the Weber number is shown in Figure 6. A clear boundary between the agglomeration regime and the stretching separation regime can be seen.

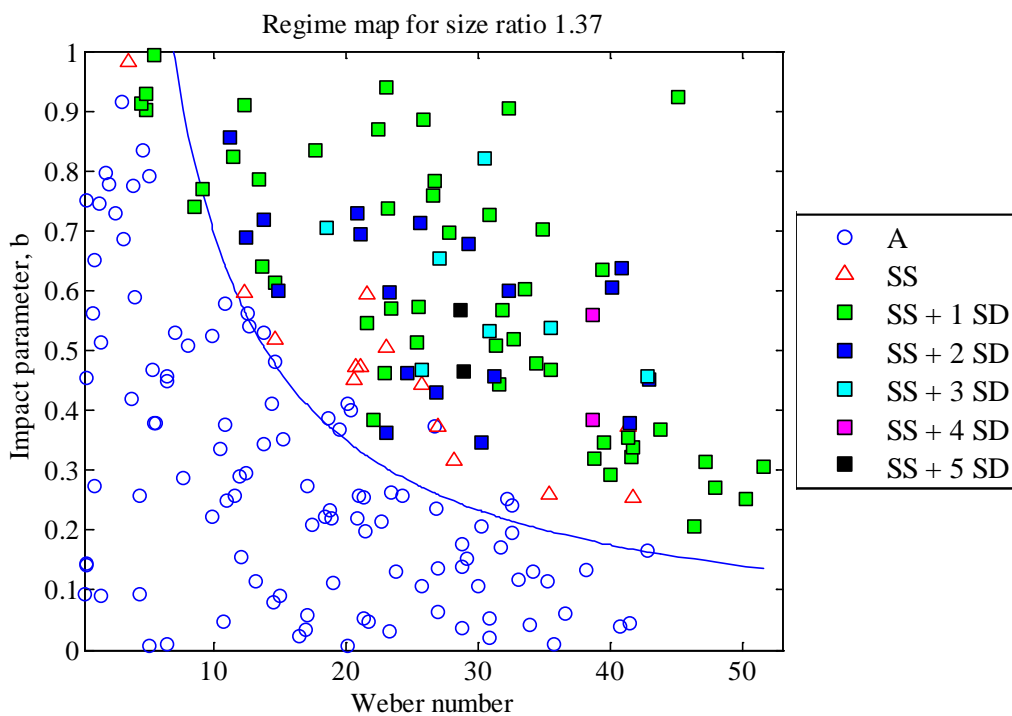


Figure 6: Regime plot of mid-air collisions for a glass particle diameter of 4 mm and an average droplet diameter of 2.9 mm. 214 collision outcomes are plotted. A = Agglomeration, SS = Stretching separation and SD = Satellite droplet. The blue line is E_{agg} given by Eq. (4).

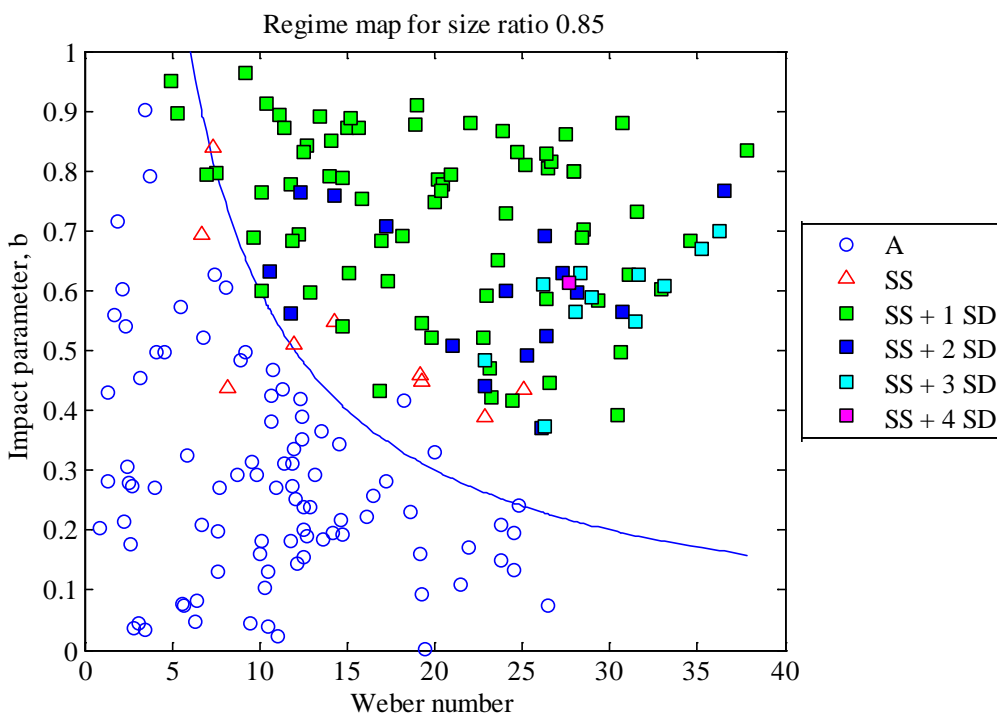


Figure 7: Regime plot of mid-air collisions for a glass particle diameter of 2.5 mm and an average droplet diameter of 2.9 mm. 201 collision outcomes are plotted. A = agglomeration, SS = stretching separation and SD = satellite droplet. The blue line is E_{agg} given by Eq. (4).

The number of satellite droplets are indicated with different colours. No obvious regimes are visible regarding the number of satellites but in general two or more satellite droplets are found for an impact parameter in the range 0.4 to 0.8, and the highest number of satellite droplets are found for an impact parameter of approximately 0.5. This is consistent with the droplet-droplet model predictions of [3] later used also by [4].

Next, experiments were conducted using smaller glass particles of 2.5 mm diameter, leading to a particle-to-droplet size ratio $\Delta = 0.85$. A total of 201 collisions were produced. For these experiments, the collisions were harder to achieve due to the smaller target size, as well as a faster initial horizontal velocity of the particle when the vacuum was released from the vacuum tip. This was improved by inserting wire sponge into the vacuum tip, stopping the reversed suction from blowing away the particle upon release. The regime map for these collisions is displayed in Figure 7.

The regime boundary between agglomeration and stretching separation is qualitatively similar to that found for the larger size ratio in the lower and higher range of Weber numbers and impact parameters. However, for Weber numbers between 8 – 20 (impact parameter values between 0.35 - 0.8) the boundary is shifted. For the size ratio 1.36, a Weber number of 15 requires a minimum impact parameter of approximately 0.55 for a stretching separation to occur (Figure 6), while for the smaller size ratio 0.85 the minimum impact parameter is 0.45 (Figure 7). During a collision, the surface contact area between the droplet and the particle is larger for a collision with a bigger particle for identical impact parameters. The droplet does not stretch out as easily due to surface tension forces, and therefore captures the particle without fragmenting.

When comparing Figures 6 and 7 with the droplet-droplet collision outcome regime map of Figure 2, we note that no bouncing between particles and droplets were detected in this study. This agrees with the numerical studies of Gac and Gradon [12, 14] who also made no direct observation of pure bouncing. We note that contrary to their work we also observed no reflexive separation. This may be due to the limited parameter range of our study, in particular with respect to the contact angle and capillary number. Gac and Gradon [12, 14] varied the contact angle from hydrophobic to extremely hydrophilic and studied impacts at relatively high capillary numbers of the order of 1 to 10. In that case, reflexive separation (called ripping and coating by Gac and Gradon) was found to occur when the liquid coats the particle surface, forms a ligament from the particle surface and then breaks up because of the high relative velocity. In our work the capillary number is at least 2 orders of magnitude lower, droplets are not sufficiently large and the particles not hydrophilic enough to allow the droplet to envelop the particle, and therefore it was not possible to observe this regime of coating and consequent formation of a liquid cone. For droplet-droplet collisions, the boundary between the coalescence regime and the stretching separation regime can be determined with the so-called coalescence collision efficiency, E_{coal} which is derived in [3]. A modified equation is shown below, which gives the boundary (in terms of impact parameter) between agglomeration and stretching separation:

$$E_{agg} = \min \left[1, \frac{2.4f(\gamma)}{We} \right], \quad Eq (4)$$

$$f(\gamma) = \gamma^3 - \gamma^{2.5} + \gamma + 1.2. \quad Eq. (5)$$

where $\gamma = \frac{d_l}{d_s}$ is the ratio of the larger sphere to the smaller one. Our modification is in Eq. (5) where we changed the coefficients to match our experimental results. The result is shown as a blue line in Figures 6 and 7. We find that Eqs. (5)-(6) give a good description of our observations.

Agglomerated fraction

In case of stretching separation, we calculate the liquid fraction agglomerated to the particle by measuring the size of the leaving droplet after it has relaxed back to a spherical shape. Loss of liquid to satellite droplets was neglected because the satellite droplets were generally much smaller than the main droplet, and also because tracking of the satellite droplets was not always possible due to low resolution of the image.

When the collision outcome is stretching separation, the velocity is measured from when the leaving droplet is completely detached from the particle or potential satellite droplets. With these velocities, the transferred mass fraction of the droplet is then calculated through a momentum balance. Knowing the mass and velocity of the particle, m_p and v_p , and the droplet, m_d and v_d , before collision, together with the velocity of the agglomerate, v_{agg} , and the leaving droplet, v_{ld} , it is possible to calculate how big is the fraction of the droplet that is transferred to the particle. The agglomerating fraction is then calculated as follows:

$$m_p * v_p + m_d * v_d = (m_p + m_{ad}) * v_{agg} + (m_d - m_{ad}) * v_{ld} \quad Eq (6)$$

$$\frac{m_{ad}}{m_d} = \frac{\frac{m_p}{m_d} * (v_p - v_{agg}) + (v_d - v_{ld})}{v_{agg} - v_{ld}} \quad Eq (7)$$

Note that the mass of the agglomerate or the leaving droplet are not needed in Eq. (7). Therefore, estimating the agglomerating fraction can be done even when the 3D distribution of droplet mass on the particle surface is not exactly visible or known.

Figures 8 and 9 show regime maps for the droplet-glass particle collisions, where the percentage of liquid that is agglomerated is indicated by color. Not surprisingly, we find that the lower the impact parameter, the more likely it is for more liquid to stick to the particle. This is due to the larger surface contact area that arises between the particle and droplet when the off-centre collision becomes more head-on. The Weber number is also a factor and for some collisions with a higher Weber number, a small agglomerated fraction was detected even if the impact parameter was relatively low. This is visible in Figure 8 for $35 < We < 50$. Comparing figures 8 and 9 we find that, at equal droplet size, a larger particle generally also leads to a larger agglomerated fraction of liquid. This can easily be understood to be a consequence of the larger available particle area for agglomeration.

These results should be quantitatively described and understood in terms of a theory involving kinetic and surface energy balances. This will be the topic of future work.

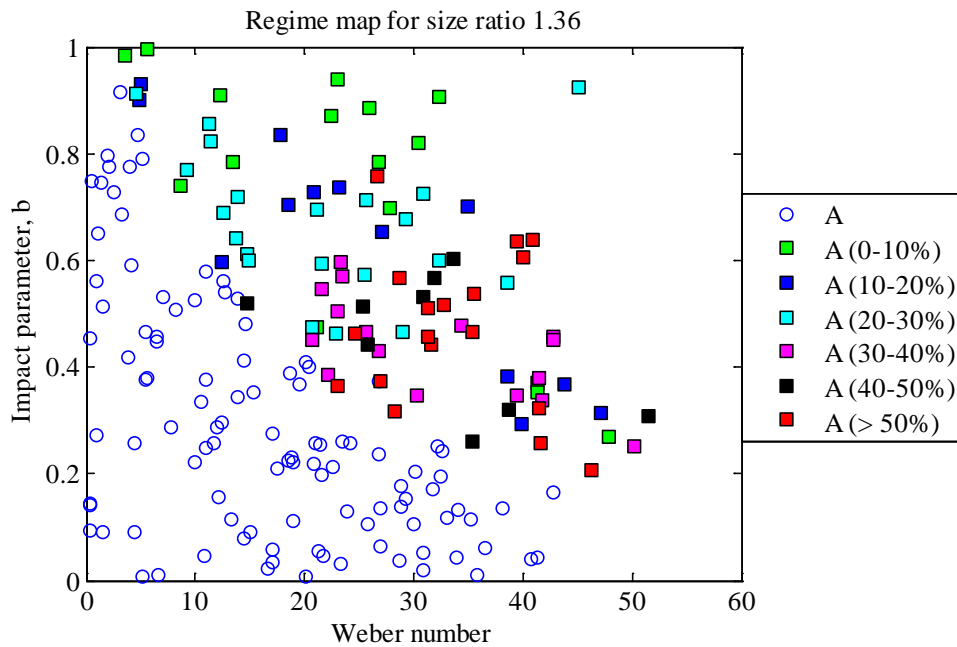


Figure 8: Regime plot of mid-air collisions for a glass particle diameter of 4 mm and a droplet diameter of 2.9 mm. Blue circles represents collisions with 100% agglomeration (A). The squares are coloured based on how much liquid sticks to the particle after collision.

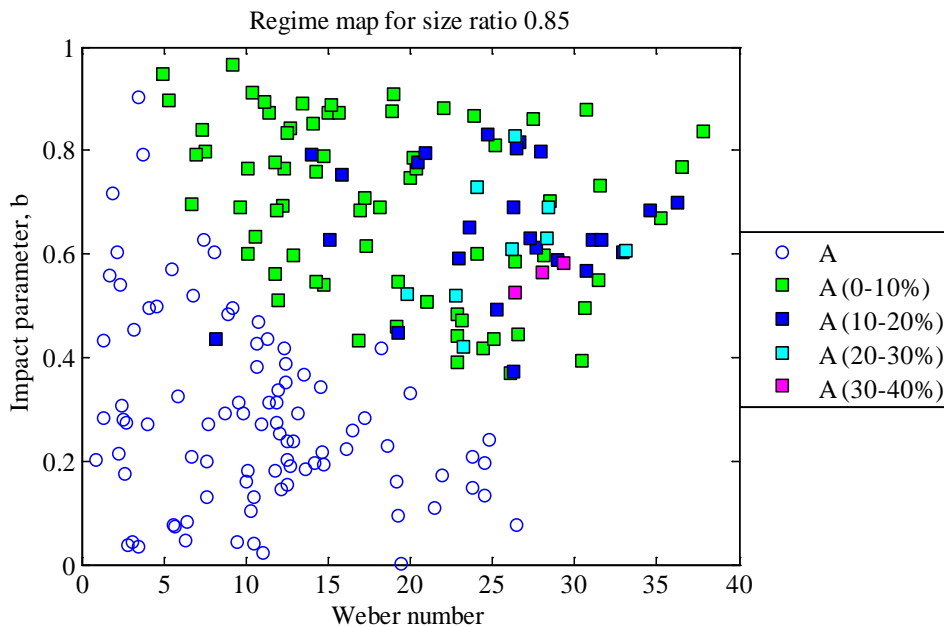


Figure 9: Regime plot of mid-air collisions for a glass particle diameter of 2.5 mm and an average droplet diameter of 2.9 mm. Blue circles represents collisions with 100% agglomeration (A). The squares are coloured based on how much liquid sticks to the particle after collision.

4. CONCLUSIONS

In this work we studied collisions between droplets and dry particles at relatively low capillary numbers. Different collision outcomes were identified as either agglomeration (merging), where the whole droplet would stick to the surface of the particle, or a stretching separation (breaking), where the droplet collides with the particle in an oblique position and stretches out until a part of the droplet detaches from the liquid sticking to the particle. The transition between the agglomeration

and stretching separation regime was determined (equation 4) and found to scale inversely with Weber number. The formation of multiple satellite droplets, i.e. droplets with a radius significantly smaller than the leaving droplet, was found to occur for impact parameters between 0.4 and 0.8, with the highest number of satellite droplets occurring for impact parameters near 0.5. When stretching separation occurred, the largest agglomerated fraction of liquid occurred for the largest particle. We hope that this set of experimental data and the obtained regime maps will be used to validate future theoretical and simulation studies of particle-droplet collisions.

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