Phase Change and Interaction Behaviour of Supercooled Water Droplets

V. Kunberger*[1], B. Weigand[1]
Institute of Aerospace Thermodynamics, University of Stuttgart, Germany
*Corresponding author: verena.kunberger@itlr.uni-stuttgart.de

Introduction

The phase change and interaction behaviour of supercooled water droplets is of great importance to understand the processes in clouds and precipitation. These micro-processes are relevant for the development and improvement of climate models and weather forecasts [1].
The ambient conditions in clouds are extreme with temperatures as low as -40° C and lead to the supercooling of water droplets. The following phase change and freezing of the droplets is one of main energy sources in clouds or more severe weather phenomena like cyclones [2] as latent heat is released during that process. This also influences the clouds on a macroscopic scale as droplets in the vicinity of the latent heat release evaporate and therefore the cloud size is limited [1].
Phase change is initiated by either homogeneous nucleation or by heterogeneous nucleation through particles like aerosols or ice crystals. Supercooled droplets are in a metastable state of the Gibbs Free Energy

$$G = U - TS + PV \qquad (1)$$

and for them to change into the stable solid phase, they have to overcome the energy barrier

$$\Delta G^* = \frac{16\,\pi\sigma^3}{3\,\Delta G_V^2} \qquad (2)$$

This is a statistical process and the probability overcoming the energy barrier is dependent on the degree of supercooling [3].

Due to convection, the supercooled water droplets are in constant motion in the clouds and interact and collide with each other. Thus, the question arises if the droplets themselves can trigger nucleation by colliding with each other. This is possible if the energy of the impact is large enough to overcome the energy barrier mentioned in equation (2) and therefore depends on the impact velocity and droplet size and the degree of supercooling. But also the ambient conditions need to be taken into account as the surrounding gas has to transport the latent heat released during the freezing process away which is dependent on the conduction and convection properties of the ambience. This means that the rate of freezing is dependent on the surrounding gas and its heat transfer ability.

Furthermore, the interactions of several droplets are of interest as this is the onset of precipitation. As the supercooled water droplets move through the clouds, they can collide with snowflakes and freeze onto them. This is called riming and snowflakes fully covered in rime are called graupel [4]. If the impact energy of the droplets is higher due to higher velocity in more severe weather conditions, clear ice and a denser structure is formed which can result in hail [1,5].

The experimental methods to research these micro-processes and the main influences of temperature and relative humidity on them will be explained briefly in the next section as well as some results will be shown.

Experimental methods

To research the binary collisions of supercooled water droplets with minimal disturbances, an optical levitation setup is used in which the laser holds the droplet in a stable position in a cooling chamber. Laser levitation is described in more detail by Ashkin [6]. The wavelength of the laser is chosen such that it is not absorbed by the water. The experimental setup is shown in Figure 1.

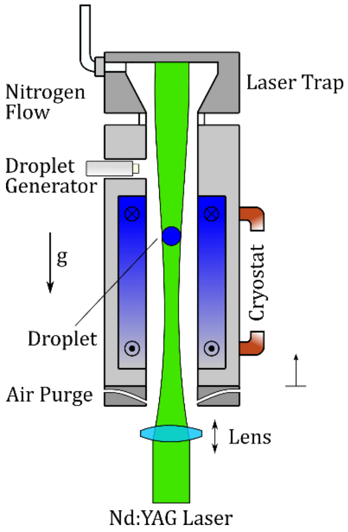

Figure 1. Experimental setup of the levitation of a supercooled water droplet

The droplets are produced by a droplet generator driven by a piezo-electric dispenser from microdrop Technologies and they have a size of 45 µm.

The chamber can be cooled down to -40°C and the relative humidity is controlled by a constant nitrogen flow within the chamber.

Through PMMA windows, optical access for backlight-imaging is given which is realised with a LED and a 12MP CCD camera. To detect the freezing point of the droplets, two PSD sensors record the scattered light of the laser and the change of polarisation which is caused by the freezing of the droplet.

As the laser beam is not strong enough to hold more than two small droplets, another setup is needed to look at the interactions of several supercooled water droplets. It looks similar to the levitation setup and is shown in Figure 2. This setup also consists of a cooling chamber, droplet generator and nitrogen supply, but a small aluminium surface is inserted on which the droplets impact. The surface is cooled down to ambient temperatures which is controlled by thermocouples in the surface. The droplet generator is moved randomly across the surface by two linear motors to simulate the random motions in clouds. The structures the form are observed with backlight imaging, a zoom-lens and a 12MP CCD camera. The droplet generator produces single droplets or groups of droplets on demand with a size of 75 µm.

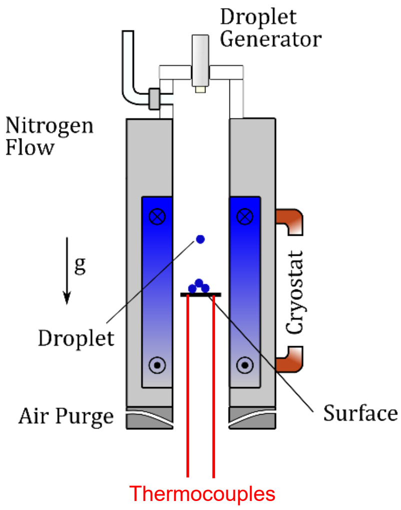

Figure 2. Experimental setup of interactions of several supercooled water droplets on a surface

Results and Discussion

Experiments were conducted at -37.5 °C and a relative humidity of 30% the first collisions observed are shown in Figure 3 below.

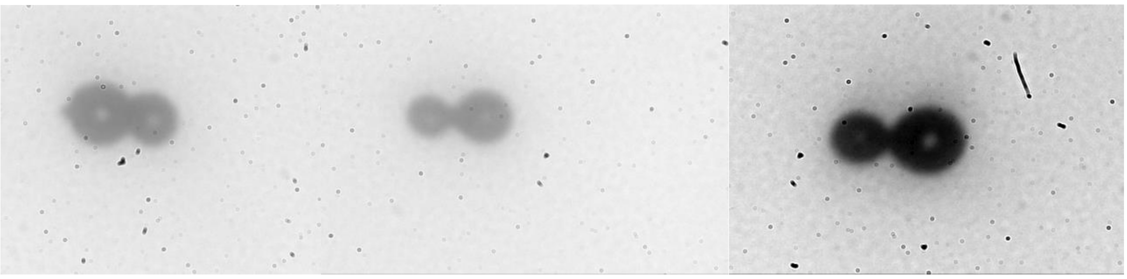

Figure 3. Binary droplet collisions of supercooled water droplets at 37.5°C

The smaller of the droplets is the one which is levitated first and subsequently evaporates and sublimates before the larger droplet collides.

It can be seen that the droplets freeze immediately on impact and remain mostly spherical. Therefore, the freezing process is very fast and the latent heat is transported away quickly. A spike in the larger droplet in the first image is also clearly visible which indicates a frozen droplet.

The sublimation process can also be observed in the collision product and the reduction of its size over time as shown in Figure 4.

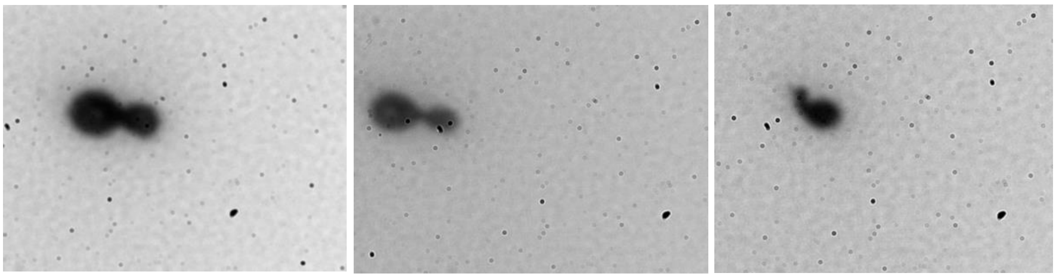

Figure 4. Sublimation of collided and frozen supercooled water droplets at 37.5°C

Future work will extend these experiments to different temperatures and also study the influence of relative humidity on the outcome of the collisions of supercooled water droplets.

In the setup to research interactions of several droplets on a surface, preliminary results were obtained. At different temperatures, structures of different densities were observed.

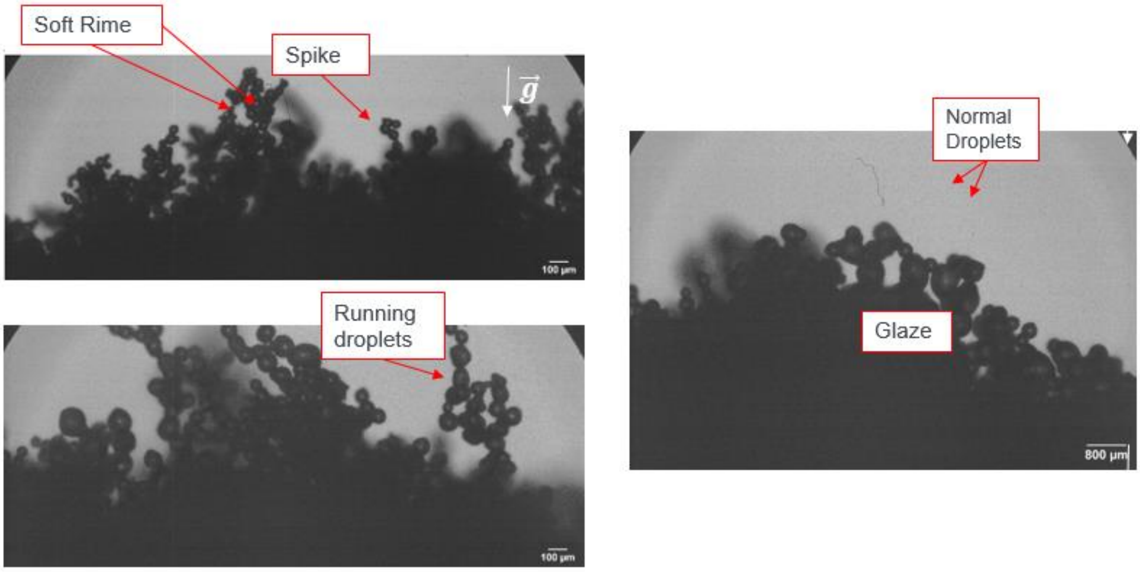

Figure 5. Freezing timescales of supercooled water droplets in different conditions

The images in Figure 5 show these structures that form if several droplets interact with each other. Depending on the conditions, they can form soft rime with larger gaps in between resulting in a loftier structure. But also denser structures and ‘running droplets’ that do not freeze instantly freeze could be observed for higher relative humidities. In very humid conditions, glaze ice that is very dense could be seen and larger deformed droplets.
But as these results are very preliminary, extended studies have to be performed in the future to relate the formation of different types of ice and rime to the conditions found in the chamber.

Nomenclature

G	Gibbs Free energy [J]
U	Internal Energy [J]
T	Temperature [K]
S	Entropy [J K^{-1}]
P	Pressure [Pa]
V	Volume [m^3]
σ	Surface Tension [N m^{-1}]
ΔG_V	Latent Heat [J]

References

[1] Pruppacher, H. R., Klett, J. D., 1997, ’Microphysics of clouds and precipitation’. Kluwer Academic Publishers.
[2] A. Khain, M. Ovtchinnikov, M. Pinsky, A. Pokrovsky, H. Krugliak, 2000, Atmospheric Research, 55(3), pp. 159-224.
[3] Kelton, K.F. and Greer, A.L., 2010, ‘Nucleation in condensed matter – Applications in Materials and Biology’, Elsevier
[4] A. Rango, J. Foster, E.G. Josberger, E. F. Erbe, C. Pooley, W. P. Wergin, 2003, SCANNING, 25, pp. 121–131.
[5] W.C. Macklin, 1962, Q.J.R. Meteorol. Soc., 88, pp. 30-50.
[6] A. Ashkin, 2000, IEEE Journal on Selected Topics in Quantum Electronics, 6 (6), pp. 841-856.