Experimental observations with ultra-speed imaging revealing new complex detail of drop separation processes from a large diameter drophead

D. Carbery¹, N.D. McMillan², M. O'Neill², S. Riedel², D.Kennedy³ and T.Nicholls⁴.

¹Institute of Technology Carlow, Kilkenny Road, Carlow, Ireland

² Drop Technology Ltd., Tallaght Business Park, Whitestown, Tallaght, Dublin 24

³School of Engineering, DIT, Bolton Street, Dublin 1

⁴Photron Ltd., Bottom Road, West Wycombe, HP14 4BS, Bucks, United Kingdom

Abstract. An ultraspeed (5000 frames a second) camera has been used to record the drop separating from the photometric instrument the tensiograph. New insights into drop separation processes which have been evaluated in terms of the correction factors used in tensiometric methods for measuring surface tension. An innovative approach to drop separation process drop satellite labelling is proposed and the paper ends with suggestions as to practical improvements to the photometric measurement of what is a very long-established method.

Keywords : drops; separation; Yildirim; Basaran; high-speed; imaging; water; methanol; satellite.

Contact details: des.carbery@itcarlow.ie

1. Drop separation

This study looked at the physical reality of drop separation with some of the highest performance camera system currently available with 5000 frames a second. Details and new analysis of the pendant drop separation process are given. These are the first observations of the drop separation process on such a large 8.5 mm drophead. The study is primarily quantitative but also highlights qualitative issues with regard to measurements related to pendant drops.

The details revealed in the study and the identification of the component drops are consistent with the theoretical work of Yildirim and Basaran et alⁱ. When a drop (hanging from a capillary or drophead) increases in size, due to a gradual liquid input, it reaches a stage when part of it separates (primary drop). Other separating parts thereafter are called satellite drops. What remains on the capillary or drophead is termed by these authors the pendant but by others more commonly referred to as the remnant drop. Figure 1 is a schematic of the pendant, satellite and primary drops.

The series of events leading to these separate drops can be termed a drop separation process. During such a process there are many reasons why a drop changes in shape, separates and continues to evolve. The physics of an equilibrium process would force the drop to form into a spherical shape, but is of course acted on by many forces including force of gravity, surface tension, surface waves, fields of flow within the drop and external friction. This study looks at this dynamic non-equilibrium process. Camera images of water and methanol drop separations are presented, commenting on significant stages.

2. Experimental arrangement

Figure 2 shows the experimental arrangement. A quartz drophead 8.5mm diameter was used. The drophead was held vertically and liquid continuously pumped to it through capillary tubing. The pump used was a Hamilton PSD/4 stepper motor driven syringe pump with a 250ul syringe. The pump speed was set at 200 which gave a flow rate of 1.25μ l/s. A drop was formed on the drophead and images of the drop were captured just before the time that the drop became unstable and continued to be taken through the drop separation process.



Figure 1 Pendant, satellite and primary drops

A Photron camera (Fastcam SA1.1 model) was used for capturing the images. The maximum resolution of the camera was 1000*1000 pixels and the images were taken at 5000 fps and 5400 fps (frames per second).

The drop liquids were at room temperature (20°C). Table 1 shows the liquid propertiesⁱⁱ of the Analar methanol and water drops.

Properties	Water	Methanol
Density (kgm ⁻³)	998.6	791.1
Surface tension (mNm ⁻¹)	73.05	24.05
Viscosity (cP)	1.002	0.6

Table 1 Liquid properties of the drops under test



Figure 2 Drophead with the capillary tubing connected at the top (on left). Drophead with the extended camera lens and a screen image(on the right)

The following camera images were taken:

- i. Individual camera images of separating water drops
- ii. Individual camera images of separating methanol drops
- iii. Camera images of the contact edge of the methanol drop at the drophead

The camera speed for the drop images was 5,000 fps.

3. Camera images

The general sequences of water and methanol drops Figure 3 shows images captured during the separating process of a water drop. The frame numbers are given so that the time between images can be calculated on the basis of the camera speed.

At first the full drop is shown, then the obvious necking showing positive and negative radii of curvature of the drop edge. Next, the almost cylindrical midsection reduces to a pinhead before the primary drop separates. At this stage the pendant drop is fully extended before pulling back upwards when the satellite drop separates. As the changing shape satellite falls, a daughter satellite separates upwards and continues until it contacts the pendant drop when it rebounds. A similar observation to the latter was made by Hauser et alⁱⁱⁱ who high-speed motion picture camera (1200 f.p.s) to observe the drop separations from a 4.58mm capillary.

Figure 4 shows images captured during the separating process of a methanol drop. The process is generally similar to the water set but the proportions of size are very different. A significant difference occurs however after the primary drop separates when there are greater number of satellite drops. The general asymmetrical shape of the methanol drop is noted but not explained. The drophead was levelled and rechecked but may not have been exact. (This issue has been addressed in the commercial tensiograph instrument^{iv} with a sensitive levelling system together with vibration control.).



Figure 3 Images captured during the separating process of water drop 2. Frames 0, 299, 399, 413, 414, 424, 434, 444, 471, 501, 541.



Figure 4 Images captured during the separating process of a methanol drop. Frames 0, 299, 399, 444, 445, 563, 564, 573, 594, 605, 636.

4. Images related to Basaran studies.

There is a direct correspondence between this study and the theoretical description of this author but the detail is too great to deal with here.

Convention to identify drops in a separating process

For clarity in the study of the separating drops, a convention is adopted to identify all drops in this process. The convention is similar to that used by Basaran but is extended further. Each drop is identified by an index d, the pendant drop is designated drop d 1. Figure 5 shows the convention used.



Figure 5 Water drops identified

The first frame in Figure 5 shows how the convention is used for the initial pendant, satellite and primary water drops. However, when a further drop separates from the satellite d 2, the convention is extended as shown in the second frame.

5. Satellite drop

The resulting complex effects on the satellite drop comes from the interplay of forces as the drop oscillates under their action. The lower part falls more slowly than the upper part. Generally, surface tension and gravity forces oppose each other on the lower part while they act together on the upper part. However this is not totally consistent as can be seen from the results that follow. Figure 6 shows the falling speeds of the top and bottom surfaces of the water satellite drop.



Figure 6 Speed of fall of top and bottom of water satellite drop.

6. Second satellite drop

Figure 7 shows the sequence of images leading to the separation of a second satellite water drop (d2-1) from the main satellite drop (d2), then re-designated (d2-2).



Figure 7 Images of the separation of the satellite drop d2 into d2-1 and d2-2. Frames 405, 406, 407, 410, 412, 413, 416, 417.

This second separated satellite makes two oscillation before successfully leaving the main satellite. The first attempt is shown in Frame 405. The first two images here show what we will term an attempt to separate, while the third and fourth two images show its return to the main satellite. There are possible explanations for this event; one is that the force of attraction increases because of the change of surface shape of the second satellite (becoming more pointed and therefore electrical charge increases). While another explanation is that the main satellite shape is vibrating and its top surface rises upward to contact the separating second satellite.

However, the second attempt at separation is successful and is shown in Frame 412 The second satellite drop separates and is projected vertically upwards. The size of this second satellite (less than 1% of the main satellite) is estimated from the camera image as a sphere of 0.35mm diameter (volume 0.022μ l). The gravitational force on it is approx 0.22μ N. The separating force needs to exceed this and also must overcome the force of attraction and then requires a further additional force to accelerate the satellite upwards. It can be seen that a third satellite almost separates but is attracted back. During all of this time the main satellite drop is vibrating and being pulled together into a more spherical shape as it continues its fall downwards.

7. Drop separation times

The times at which the drops separate in the process were recorded and one set of results are shown in Table 2.

Separation times of water drops					
	Water drop 1		Water drop 2		
	Frame	Time (ms)	Frame	Time (ms)	
Primary drop (d3) separates	350	0	413	0	
Satellite drop (d2) separates	373	4.3	435	4.4	
Satellite drop (d2-1) separates	412	11.5	471	11.6	

 Table 2 Separation times of water drops

8. Conclusion

This study set out to investigate the details of the process of separating drop with particular reference to measurements such as the diameter and height of the drop. Many observations were made and recorded, both experimental and theoretical. The full study is presented in Drop Metrology^v. Some are presented in this paper and others may be the subject of a further paper.

This study presents the ultra-detail of this separation process of large pendant drops.

It shows clearly:

- i. a pendant drop adapts to an increase in volume by changing shape, together with a change in contact angle at the drophead,
- ii. the drop reaches a maximum volume when it can no longer be supported on the drophead and thereby begins the process of drop separation (This point of instability or "scission" point has not been exactly identified in this particular study. It would be useful in a further study to identify this event.),
- iii. the separation of the primary drop,
- iv. the separation of the satellite drops,
- v. the change in shape of the separate drops when subjected to the various forces,
- vi. the similarities and differences between two liquids in the same process.

The measurements have delivered graphical analysis for the first time with the high speed resolution of the camera. Also, an identity system for the separating drops has been established.

The final point to make from this study is the camera images explain in a practical way why the correction factors used in tensiometry are not very precise, the small satellite drop that rebounds from the remnant drop can on other days fuse with the drop. The atmospheric electrical charge will vary daily and the smallest tilt of the apparatus will clearly make a difference. Whether a drop fuses or not will have implications for the volume of the remnant drop and as a consequence the 'correction factor'^{vi}.

^v Carbery, D. (2010), PhD Thesis IT Carlow

^{vi} Harkins, W. and F. Brown (1919). "The determination of surface tension (free surface energy), and the weight of falling drops.- The surface tension of water and benzene by the capillary height method." Journal of the American Chemical Society **41**(4): 499-524.

ⁱ Yildirim, O., Q. Xu, et al. (2005). "Analysis of the drop weight method." <u>Physics of Fluids</u> **17**: 062107

ⁱⁱ Lide, D., CRC handbook of chemistry and physics. 2004: CRC press.

ⁱⁱⁱ Hauser, E., H. Edgerton, et al. (1936). "The Application of the High-speed Motion Picture Camera to Research on the Surface Tension of Liquids." <u>The Journal of Physical Chemistry</u> **40**(8): 973-988

^{iv} McMillan, N., S. Smith, et al. (2008). "Quantitative drop spectroscopy using the drop analyser." <u>Measurement Science and Technology</u> **19**: 055601.